

Environmental conditions rather than nitrogen availability limit nitrous oxide (N₂O) fluxes from a temperate birch forest

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Abstract. Forest ecosystems play an important role in the terrestrial nitrogen (N) cycle, accounting for over a quarter of the land area of the Earth. However, our understanding of nitrogen dynamics in forest systems is limited. The consequences of N
15 deposition to forest ecosystems are often overlooked. In this study, dry deposition of NH₃ was replicated over a two-year period in a temperate semi-natural birch forest via a unique custom-built automated NH₃ release system to investigate the impact on emissions of the greenhouse gas nitrous oxide (N₂O). This study provides evidence that in both natural forest soils (in-situ) and soils under controlled laboratory conditions (ex-situ), the substantial addition of reduced N compounds (NH₃/NH₄⁺) had no direct impact on N₂O emissions. Emissions of N₂O from these soils were dependant on the meeting of
20 several additional thresholds, below which N₂O producing activity was constrained. When environmental conditions in-situ were considered warm and wet (soil temperature >12 °C and volumetric water content >20%), emissions of N₂O were an order of magnitude higher than when either of these thresholds was not met, regardless of exposure to NH₃ deposition. Ex-situ experiments indicated that microbial activity in the soils was highly constrained by the availability of labile carbon. The addition of glucose to these soils resulted in a considerable increase in N₂O emissions after N application. While cumulative
25 NH₃ deposition to the in-situ soils was relatively large over the measurement period, there was no accumulation of mineral N observed in the soil, suggesting plant-uptake of N was able to mitigate N loading. The implication of these results is that forest ecosystems may be able to mitigate localised NH₃ pollution plumes, in the short-term at least, without incurring an N₂O penalty. However, the long-term impacts of N enhancement remain unclear and further long-term field experiments are required to examine the impact of prolonged exposure to high quantities of N deposition to forest soils.

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Short summary. The impacts of increasing nitrogen deposition on the fluxes of the greenhouse gas nitrous oxide from a temperate birch forest were investigated in-situ and ex-situ. Nitrogen levels only had a limited effect on emissions. Instead, emissions of nitrous oxide were modulated by soil carbon availability and meeting a dual temperature-moisture threshold. An implication of these findings is that forests could be used for mitigating nitrogen pollution without incurring a greenhouse gas penalty, at least in the short term.

1. Introduction

Reactive nitrogen (N_r) entering ecosystems due to anthropogenic activities has more than tripled at the global scale relative to 1961 (Galloway et al., 2021). Consequently, N_r pollution has contributed significantly to loss of biodiversity (Bobbink et al., 2010; Krupa, 2003), eutrophication (de Vries, 2021), soil acidification (Tian and Niu, 2015) and gaseous emissions in the form of nitrous oxide (N_2O) and nitric oxide (NO) (Butterbach-Bahl et al., 2013; Davidson et al., 2000; Pilegaard, 2013; Song et al., 2020). N_2O is a potent greenhouse gas (GHG) with a global warming potential approximately 273 times higher than carbon dioxide (CO_2) on a 100-year timescale (IPCC, 2023) and is also responsible for destruction of ozone in the stratosphere (Ravishankara et al., 2009). Natural soils are an important source of N_2O , representing approximately 35% (6.4 Tg N yr^{-1}) of the global N_2O budget (Tian et al., 2024). The production of both N_2O and NO gases in soils is generated as a by-product of microbial processes, predominantly via nitrification and denitrification (Baggs, 2011; Butterbach-Bahl et al., 2013; Pilegaard, 2013). These processes and the magnitude of resulting emissions from soils are affected by a wide range of environmental variables, such as temperature (Braker et al., 2010), pH (Weslien et al., 2009), soil moisture (Firestone and Davidson, 1989), and carbon (C) and nitrogen (N) availability (Butterbach-Bahl et al., 2013; Pilegaard, 2013; Skiba and Smith, 2000).

Nitrogen availability in soils is considered to be a major driver of N_2O fluxes. While generally considered a rate-limiting step in low-N natural environments, higher availability of N_r would be expected to result in increased microbial activity and higher gaseous losses from soils in the form of N_2O and NO fluxes (Firestone and Davidson, 1989). While this pattern is generally observed following the application of N-based fertiliser to agricultural systems (e.g. Cowan et al., 2020), this is not always the case in natural ecosystems, such as forests. The latter are typically exposed to chronic N inputs, primarily from atmospheric deposition (Sutton et al., 2004; Sutton et al., 2014), and often do not demonstrate a clear dose-response to elevated N levels (Du and de Vries, 2023). While some synthesis studies have attributed an increase in N_2O forest fluxes to elevated N levels (Aronson and Allison, 2012; Cen et al., 2024), others have reported a lack of response and high variability in the fluxes (Flechard et al., 2020; Liu and Greaver, 2009).

In addition to the amount of N_r , the form of reduced or oxidised N affects the rates of different microbial processes and their associated N_2O and NO emissions (Ding et al., 2023). In the UK, a shift in policies over the past several decades has significantly decreased emissions of oxidised N_r (e.g. NO_x) (Driscoll et al., 2024; Tomlinson et al., 2021) and shifted the ratio

of N deposition further towards reduced forms such as ammonia (NH_3) (Hicks et al., 2022; Tomlinson et al., 2021). NH_3 is the primary contributor to N_r pollution in the UK (Tang et al., 2018). An estimated 260 kt of NH_3 was emitted in the UK in 2022, with agricultural activities and fuel combustion (mainly from transportation) being the main sources (Mitchell et al., 2024). Volatilised NH_3 can deposit to natural ecosystems, such as forests, where it acts as a major source of N_r (Bobbink et al., 2010; Du and de Vries, 2023). It is estimated that between 7 to 50 kg N $\text{ha}^{-1} \text{yr}^{-1}$ are deposited to UK forests, with approximately 90% of forests being subjected to a critical load exceedance (the level of N deposition above which negative impacts occur) (Vanguelova et al., 2024).

Dry deposition of NH_3 is an often-overlooked source of N_r to forests (Du and de Vries, 2023; Flechard et al., 2011). It has been estimated that 63% of N_r deposition to European forests is due to dry deposition (Flechard et al., 2020). Dry deposition is likely to play a more important role in forests compared to other natural ecosystems (such as grasslands and moorlands) due to complex canopy structures and multiple surfaces for N_r deposition (Vanguelova et al., 2024). This has been demonstrated by the fact that the concentration of nitrate (NO_3^-) and ammonium (NH_4^+) in throughfall in the UK was higher in forested areas relative to moorlands and open grasslands (Sawicka et al., 2016).

While a large proportion of studies on N dynamics and the effects of elevated N levels in soils have focused on agricultural systems (Reay et al., 2012), forest ecosystems also play an important role in the terrestrial N cycle (Chapin et al., 2011). Forests cover approximately one third of global land area (Keenan et al., 2015; Ritchie, 2024). This proportion may increase as a result of global (DESA, 2018) and national (Westaway et al., 2023) efforts to increase tree cover as part of climate change mitigation measures. It is believed that increasing tree cover can contribute towards simultaneously mitigating climate change (by sequestering atmospheric CO_2) and reducing atmospheric NH_3 pollution (Kirschbaum et al., 2024; Tang et al., 2022; Verheyen et al., 2024). However, NH_3 deposition to forests can alter natural conditions, such as through changes in tree growth (fertilisation effect), soil C sequestration, understory vegetation diversity and changes in greenhouse gas (GHG) fluxes (such as CO_2 , methane (CH_4) and N_2O) (Tang et al., 2022; Vanguelova et al., 2024).

There remains high uncertainty regarding the impacts of a potential increase in soil N availability on N_2O fluxes in forests, and experimental methodology is a major limiting factor when attempting to replicate the complexities of N deposition in forest ecosystems (Du and de Vries, 2023). For example, Jiang et al. (2023) reported that applying N directly to the soil rather than to the forest canopy can increase N_2O fluxes by 20 to 50%, thus highlighting the importance of tree canopies in modulating the impacts of N deposition on soil processes. The form of N (reduced versus oxidised) has also been reported to affect the diversity and abundance of microbial communities (Ding et al., 2023) and as such it might indirectly impact gaseous emissions from the soil. This indicates a research gap and highlights the importance of studying and understanding the impacts of reduced N_r dry deposition on N_2O fluxes from forest soils.

In this study, dry deposition of NH₃ was replicated in a temperate semi-natural birch forest via a unique custom-built automated NH₃ release system (Deshpande et al., 2024). Anhydrous NH₃ gas was released based on wind conditions at low concentrations over two years, thus mimicking a gradient of realistic levels of N deposition across forest soils from which N₂O emissions were measured. A complementary laboratory experiment further investigated the role of other environmental factors, such as N form and soil carbon availability, on N₂O fluxes. The primary aims of this work were to: (i) determine the impact of increasing NH₃ dry deposition on N₂O fluxes from forest soils, (ii) identify the environmental factors affecting N₂O fluxes in forest soils and (iii) explore the role of soil carbon availability in modulating N₂O flux in forest soils.

2. Methodology

2.1. Site description

This study was carried out at Glencorse forest, Midlothian, Scotland, United Kingdom (55°51'13" N, 3°12'56" W; 186 m above sea level) (Fig. S1). It represents a semi-natural temperate forest. The dominant tree species are silver birch (*Betula pendula*) and downy birch (*B. pubescens*) which were planted in 1984 (Billington and Pelham, 1991). Some natural regeneration has occurred, predominantly ash (*Fraxinus excelsior*) and rowan (*Sorbus aucuparia*). The ground vegetation is dominated by grasses (*Calamagrostis stricta*, *Festuca gigantea*, *Holcus lanatus*, *Dactylis glomerata* and *Poa nemoralis*). Previously, the site was used as an agricultural field, similar to the modern use of the fields that surround Glencorse. The site has not been managed and has not received additional nitrogen inputs, other than atmospheric deposition, since the 1990s. Background atmospheric deposition rates of NH₃ at the Glencorse field site before the start of the manipulation experiment were approximately 0.63 µg m⁻³ (Deshpande et al., 2024), which falls below the critical N level for the UK (Rowe et al., 2021). Soil type in Glencorse is classified as freely drained brown earth from the Darvel series derived from Carboniferous sediments (Levy and Clark, 2009). Soil physicochemical properties are summarised in Table 1. Mean annual temperature is approximately 9 °C and annual precipitation is approximately 993 mm for the period 1991 to 2020 (UK Met Office, 2023).

Table 1 Soil physicochemical characteristics at the Glencorse field site (n = 36).

	Mean	SD
Total carbon, %	3.40	0.79
Total nitrogen, %	0.26	0.05
C:N ratio	13.1	-
pH	5.32	0.31
Bulk density, g cm ⁻³	0.96	0.15

2.2. Automated NH₃ release system and atmospheric NH₃ concentrations

A unique custom-built automated NH₃ release system was used to increase the atmospheric concentrations of NH₃ in the Glencorse study site (Fig. S2) (Deshpande et al., 2024). Anhydrous NH₃ gas was mixed with air and blown down three 20-metre-long perforated uPVC pipes (d = 11 cm) at three heights above the ground (0.5, 1.35 and 2.2 m) to facilitate even N enhancement along the soil surface, underground vegetation and trees (Deshpande et al., 2024). Wind speed and wind direction were measured using a weather transmitter (WXT 536, Vaisala, Finland) located 2.3 m above the ground on a meteorological tower. Ammonia was continuously released when the wind was in the South-West sector (275° – 345°) and between 0.3 and 10 m s⁻¹. Concentrations of NH₃ were highest closest to the release line and decreased downwind with distance away from the source, as observed in the vicinity of chicken farms (Sommer et al., 2009; Pitcairn et.al, 2002), thus creating a gradient of realistic concentrations. The system was activated in September 2021 and has been active since then. The timings and amount of NH₃ release were recorded by a Micrologger (CR 3000, Campbell Scientific).

Adapted Low-cost Passive High Absorption ALPHA® samplers (Tang et al., 2001) were used to measure atmospheric NH₃ concentrations. These are passive diffusion samplers with a path length of 6 mm. A filter paper coated in 12% citric acid was enclosed by a polyethylene sampler body. A PTFE membrane was placed at the open end of the body, thus allowing for air to diffuse from the atmosphere towards the filter without interference from turbulence. The optimum range of NH₃ concentrations that these ALPHA® samplers could measure was between 0.03 and 100 µg m⁻³. Samplers were exposed for one month at a time, thus presenting a cumulative amount of atmospheric NH₃ for the specified period. Samplers were positioned within 0.5 m of each static flux chamber (see below) so that NH₃ atmospheric concentrations could be correlated to N₂O fluxes without the need for spatial interpolation. Alpha samplers were placed 0.5 m above the ground to capture NH₃ concentrations close to the level of the chambers, yet minimising interference from ground vegetation. Following exposure, filters were extracted in 3 ml of deionised water for one hour and analysed using a flow injection analyser based on the salicylate method (Seal AA3 HR AutoAnalyzer, Seal Analytical Ltd., Wrexham, UK).

2.3. Measurements of N₂O and CH₄

The in-situ study was designed as a “before-after-control-impact” (BACI) experiment (Christie et al., 2019; Smokorowski and Randall, 2017). A total of 36 static flux chambers were first deployed in March 2021 (six months prior to activation of the NH₃ release system) to capture the temporal and spatial variability in N₂O and CH₄ fluxes (Fig. S3). Pre-treatment gas fluxes from the soils were measured between March and September 2021. The rest of the flux measurements took place after the activation of the release system. Seven of the 36 chambers were positioned upwind from the NH₃ release line to act as a control, where NH₃ concentrations were close to background levels. The other 29 chambers were downwind of the release line and hence received elevated atmospheric NH₃ deposition to examine the effect of the additional N_r.

Gas samples for measuring N₂O concentrations were collected approximately once a month. Metal lids were placed on top of the polyvinyl chloride (PVC) static chambers (inner diameter = 0.38 m, h = 0.12 m on average) to create a headspace of approximately 0.013 m³ during sampling. Draught excluders and bulldog clips were used to ensure airtightness and thus minimise the chances for leakage. Gas samples were collected using a 100-ml syringe via a three-way tap and stored in 20 ml glass vials in line with the double-needle technique described in Drewer et al. (2021). Gas samples were collected every 20 min for a total enclosure time of 60 minutes, resulting in four samples per chamber per sampling event (t₀, t₂₀, t₄₀, t₆₀). This is longer compared to some previous studies, in order to ensure that detectable concentrations built up since N₂O fluxes in unfertilised forests are smaller than agricultural systems (Stehfest and Bouwman, 2006). There were no signs of saturation within the chambers.

Gas samples were analysed within one week of collection using a gas chromatograph (GC) (7890B GC system, Agilent Technologies, California, USA). These were interspersed with sets of four standards of known concentrations for quality control, ranging from 208 ppb to 1040 ppb for N₂O and from 1.12 ppm to 98.2 ppm for CH₄. The GC system was fitted with a flame ionisation detector (FID) and a micro-electron capture detector (μECD) to measure CH₄ and N₂O, respectively. The analytical uncertainty in flux methodology was calculated to be ±0.05 nmol m⁻² s⁻¹ for N₂O fluxes and ±0.58 nmol m⁻² s⁻¹ for CH₄ fluxes (Cowan et al., 2025). Fluxes of N₂O and CH₄ were calculated from the change in concentrations during the enclosure period according to Eq. (1):

$$F = \frac{dC}{dt} \cdot \frac{\rho V}{A} \quad (1)$$

where F is gas flux from the soil (nmol m⁻² s⁻¹), dC/dt is the rate of change in concentration with time in nmol mol⁻¹ s⁻¹, ρ is the density of air in mol m⁻³, V is the volume of the chamber in m³ and A is the ground area enclosed by the chamber in m². Calculations were performed using the RCflux package in R (Levy et al., 2011). Linear regression was used to calculate dC/dt for all measurements as it presented the best fit model in the majority of cases, there were no signs of saturation within the chambers and is a commonly used flux calculation method (Levy et al., 2011).

2.4. Controlled laboratory experiments

A complementary laboratory experiment was performed in order to test for the effects of N form, N enhancement level, and C availability on N₂O and NO fluxes under controlled conditions. Thirteen soil samples from the top 0-10 cm were collected randomly from the Glencorse forest site in July 2022 from control areas which did not experience elevated NH₃ levels and combined to one bulk sample. Soil was dried at 25 °C and sieved through a 2 mm steel sieve. 800 g of dry homogenised soil were placed in Perspex chamber bottoms (d = 19 cm, h = 10 cm) and repacked to field bulk density (0.7 to 1.2 g cm⁻³).

Treatments simulated N deposition equivalent to 0, 40 and 100 kg N ha⁻¹ yr⁻¹ (hereafter labelled 0N, 40N and 100N, respectively). Target N deposition levels for the laboratory incubations were based on measured values from the in-situ field experiment to allow for continuity and comparability of the results. N was applied in the form of either NH₄⁺ in aqueous solution (Cowan et al., 2024) or NH₄NO₃ in order to study the effects of reduced and oxidised N forms. The application solutions were prepared by adding deionised water to a stock solution to reach a target concentration which simulates N deposition at the target level (0N, 40N or 100N). Consequently, 8.5 or 21.3 mg N were added either as NH₄⁺ or NH₄NO₃ to simulate the 40N and 100N treatments, respectively. Enhanced C availability was achieved through the addition of glucose. A 1% sugar solution was prepared by adding 20 g analytical grade glucose (Sigma Aldrich) to 1 L deionised water (Sanchez-Martín et al., 2008). This is the equivalent of 6.6 g of sugar per soil core (800 g dry soil on average). There were three replicate cores for each treatment (including control) (n = 15), from which N₂O and NO concentrations were measured.

All re-packed soil cores in the incubation chambers were initially saturated with deionised water. The cores were then allowed to gradually dry out to allow for any Birch effect and consequent artifact gaseous emissions to subside (Birch, 1964). Deposition of N was simulated following the method described in Song et al. (2020). First N dose (in the form of either NH₄⁺ or NH₄NO₃) was applied 7 days after the initial rewetting. The second dose (N plus glucose) was applied 34 days after the initial rewetting. N₂O and NO measurements were taken every day during the first week following the treatment application and every second day thereafter for a total of 42 days.

N₂O concentrations were measured using the dynamic chamber method described in Cowan et al. (2014). The experimental setup consisted of a quantum cascade laser (Tildas-FD, Aerodyne Research Inc., Billerica, MA, USA) and a pump used to circulate the air (SH-110, Dry Scroll Vacuum Pump, Agilent Technologies, Lexington, MA, USA). A Perspex chamber was attached via bulldog clips to each soil core for the duration of each measurement (on average 5 minutes). The flow rate of air through the system was around 3.5 L min⁻¹ on average. The total headspace of the soil cores plus the measuring chamber was 0.01 m³ on average. N₂O concentrations were recorded continuously with frequency of 1 Hz (once every second) for 5 min on average, which resulted in approximately 200 data points per measurement. Concentrations of N₂O were measured as dry mole fraction following an internal water correction within the Aerodyne software. Daily N₂O fluxes were calculated based on the change in concentrations, following the same principle as the in-situ experiment (Eq. (1)). Cumulative N₂O fluxes were calculated using linear interpolation (Cowan et al., 2019).

NO concentrations were measured using a NO-NO₂ ultrasensitive chemiluminescence analyser (Model T200UP, Enviro Technology Services plc) as part of a gas flow-through system (Drewer et al., 2015). A pump was used to circulate the air through the system at a flow rate of approximately 1 L min⁻¹. Filtered laboratory air was used to make up the volume needed for the analyser. This formed an open loop system, unlike the QCL setup which was a closed loop. The Perspex chambers used for measuring N₂O and NO fluxes were identical (d = 19 cm, h = 20 cm). NO concentrations were measured for 20 to 30

minutes, depending on how quickly the system reached equilibrium. Equilibrium concentrations were recorded for two to four minutes. Concentrations of O₃ were monitored (49C O₃ analyser, Thermo Environmental Instruments Inc, USA) to ensure that they were below 5 ppb and thus the probability of chemical reactions occurring in the system was low (Seinfeld and Pandis, 2016). Temperature and relative humidity inside the chamber were measured using an integrated transmitter (Humitter 50 YC Y 50 10002, Vaisala). These were recorded every 10 seconds using a data logger (CR1000, Campbell Scientific).

Fluxes of NO were calculated using equilibrium concentrations (as opposed to change in concentrations) following the formula outlined in Schindlbacher et al. (2004) (Eq. 2).

$$F = (C_{eq} - C_0) * \frac{M * Q * 10^6}{V_m * A * 10^9} * 60 \quad (2)$$

Where F is NO-N flux expressed in $\mu\text{g m}^{-2} \text{h}^{-1}$, M is atomic weight (N = 14.008 g mol⁻¹), Q is the mass flow rate of air through the chamber (1 L min⁻¹ on average), V_m is the standard gaseous molar volume (24.055 * 10⁻³ m³ mol⁻¹), C_{eq} is the mixing ratio of NO at equilibrium, C_0 is the mixing ratio of NO from an empty chamber (blank), and A is the soil surface area of the soil core (0.03 m²).

2.5. Soil properties and meteorological conditions

Inorganic N availability in soils was measured in the form of NH₄⁺ and NO₃⁻. During the in-situ experiment, soil samples were collected from the top 10 cm within 0.5 m of each static flux chamber once every season. All samples were frozen within two hours of collection and stored at -20 °C until analysis. 15 g of soil were extracted in 50 ml of 1M potassium chloride (KCl) solution to obtain mineral N. These were mixed at 100 rpm for 60 minutes on an orbital shaker (SSL1 orbital shaker, Stuart) and consequently filtered through Whatman No. 40 ashless filter paper (pore size = 8 μm). The concentrations of KCl-extractable NO₃⁻ and NH₄⁺, were measured using a discrete multi-chemistry analyser (Seal AQ 2, Seal Analytical Inc., Wisconsin, USA). Water content was corrected for by measuring and subtracting the gravimetric content of separate samples (dried at 105 °C for three days). A set of blanks (KCl solution only) were analysed alongside all samples to ensure there was no N contamination in the laboratory. Blank values were subtracted from sample values. When sample concentrations were low, this resulted in artefact negative concentrations.

The amount of N in the form of NH₄-N and NO₃-N was calculated per gram of dry soil according to Eq. (3).

$$N = \frac{c * v}{m} \quad (3)$$

Where N is the mass of N in the form of NH₄-N or NO₃-N expressed in mg N per g of dry soil; c is the concentration of NH₄-N or NO₃-N in mg L⁻¹; v is the volume of KCl solution used during soil extractions in L; m is the mass of dry soil in g.

Approximately 15 g of soil were collected from the top 10 cm within 0.5 m of each static flux chamber three times throughout the experiment (March 2021, November 2022 and March 2023) to measure total C and N content. Samples were dried at 105 °C for three days (until constant weight) and milled using a ball mill (MM200 ball mill, Retsch). Approximately 2 mg of soil sample were analysed using an elemental analyser (Flash SMART, Thermo Fisher Scientific).

Samples for measuring soil pH were collected and stored on the same dates and in the same way as the samples used for mineral N analysis. pH was measured using a pH meter (MP 200, Mettler Toledo GmbH, Schwerzenbach, Switzerland), where 20 ml of deionised water were added to 10 g of soil sample, shaken and left to rest for 60 minutes prior to measurement. A 2-point calibration using buffer solutions of pH 4 and 7 was performed at the beginning of each measurement day.

Bulk density was measured at the beginning of the experiment (March 2021). Soil was collected from the top 10 cm within 0.5 m of each static flux chamber using a metal ring of known volume ($d = 7.5$ cm, $h = 5$ cm). Care was taken to minimise compaction when sampling. Samples were oven-dried at 105 °C for three days or until constant weight was reached. Bulk density was calculated by dividing the dry weight of soil by the soil volume (Robertson et al., 1999).

Soil temperature and soil moisture were recorded in two distinct ways to capture both spatial and temporal variability. Soil temperature and soil moisture were recorded within 0.5 m of each static flux chamber every time gas samples were collected using a hand-held temperature probe and a Hydrosense II moisture probe (Campbell Scientific, Logan, Utah, USA), respectively. Measurements were also recorded continuously at different heights and depths at a meteorological tower (a single location) using a CS655 water content reflectometer (Campbell Scientific, Logan, Utah, USA) (Fig. S4).

2.6. Data analysis

All data were inspected, cleaned, transformed, statistically analysed and visualised using R software (R Core Team, 2022). Data from the in-situ field experiment were visualised following the BACI principle (before-after-control-impact). The aim was to more efficiently distinguish between environmental versus treatment effects (Christie et al., 2019; Underwood, 1992). A two-sample Welch t-test was used to statistically test for differences in N₂O fluxes before and after the start of the in-situ experiment. This was implemented via the `t.test()` function from the `stats` package in R. A Welch t-test was preferred over Student t-test as it provides more flexibility in case the variances of the different groups were not equal. Data on in-situ N₂O fluxes were split into subsets according to the observed meteorological conditions (air temperature and soil moisture). Conditions where air temperature was above or below 12 °C were labelled as “warm” and “cold”, respectively. Conditions where soil moisture was above or below 20% VWC were considered to be “wet” or “dry”, respectively.

280 The laboratory N₂O flux data were subset to include only the first seven days of measurements in order to balance the samples collected during the first and second application periods. Welch Two Sample t-tests were also performed upon the laboratory cumulative fluxes of N₂O and NO (n = 3 per treatment) to test for any significant effect of C availability. No outliers have been removed from any of the datasets. Upon further examination, these were considered to represent genuine variation in N₂O fluxes rather than measurement error.

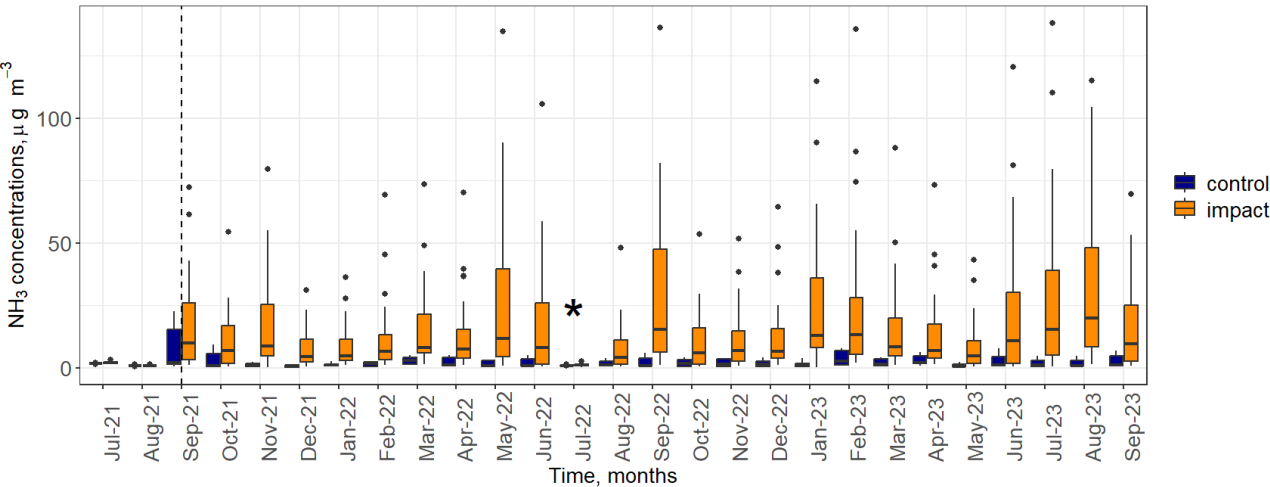
285 **3. Results**

3.1. Experimental NH₃ release

Between 1,000 and 10,000 standard litres of anhydrous NH₃ were released each month from September 2021 onwards, depending on wind conditions. Monthly NH₃ concentrations measured next to each static flux chamber at 0.5 m above the ground, increased from a maximum of 3.2 µg NH₃ m⁻³ before the start of the experiment (July and August 2021) to a maximum

290 of 146.7 µg NH₃ m⁻³ in response to NH₃ addition (Fig. 1). Experimentally increased concentrations of NH₃ were highest next to the chambers that were closest to the release line (median of 64.3 µg NH₃ m⁻³ at a distance of 0.9 m) and decreased to nearly background concentrations beyond 30 m away from the source (median concentrations <3 µg NH₃ m⁻³). Concentrations of NH₃ next to the control chambers were close to background levels (median range of 0.4 to 2.9 µg NH₃ m⁻³). During the experiment, the enhanced atmospheric NH₃ concentrations resulted in a deposition gradient from approximately 3.6 to 71 kg

295 N ha⁻¹ yr⁻¹ and 12 to 162 kg N ha⁻¹ yr⁻¹ for soil surface and total deposition to all canopy layers, respectively (Deshpande et al., 2024).



300 **Figure 1 Concentrations of NH₃ over the course of the experiment measured by ALPHA® samplers located at 0.5 m above the ground and next to each static flux chamber (n = 36). The vertical dashed line denotes the time the NH₃ release system became active.**

‘Control’ (blue) refers to chambers up wind of the NH₃ release system; ‘impact’ (orange) refers to chambers down wind of the system. An asterisk (*) in July 2022 indicates that the NH₃ release system was inactive for most of the month due to technical issues.

3.2 In-situ flux measurements

305 Soil fluxes of N₂O measured directly from the field site were generally low (maximum value of an individual flux was 1.2 nmol N₂O m⁻² s⁻¹) and 57% of flux measurements were lower than the minimum detectable flux (0.05 nmol N₂O m⁻² s⁻¹). Fluxes of N₂O were higher before the start of NH₃ release (March to August 2021) compared to after (Fig. 2). Individual chamber fluxes of N₂O after the start of NH₃ enhancement were below 0.6 nmol N₂O m⁻² s⁻¹ (except for a single data point at 1.2 nmol N₂O m⁻² s⁻¹), and there was little difference between chambers located in control or in impact areas. In contrast, individual fluxes of N₂O before the NH₃ enhancement ranged from 0.04 to 1.2 nmol N₂O m⁻² s⁻¹. The median N₂O flux values
310 for control and impact chambers were similar during the same month in most cases. Median N₂O “after release” fluxes were higher in control compared to impact chambers in February 2022. Median “after release” fluxes were approximately equal during October 2021, April 2022, July 2022 and September 2022. In all other cases, fluxes from impact chambers were slightly higher compared to the control chambers. Some of the highest fluxes of N₂O were observed in July and August 2021 (before the start of the NH₃ enhancement).

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The majority of CH₄ fluxes were negative, both before and after the start of the experiment (Fig. S5), thus indicating potential uptake of CH₄. Fluxes of CH₄ from control and impact chambers were typically within less than 0.2 nmol m⁻² s⁻¹ of each other. Apparent uptake of CH₄ was somewhat higher after the start of NH₃ enhancement, especially during July, August and September 2022. However, the majority of CH₄ fluxes (78%) fell below the analytical limit of detection (0.58 nmol m⁻² s⁻¹)
320 and hence no further statistical analysis was performed.

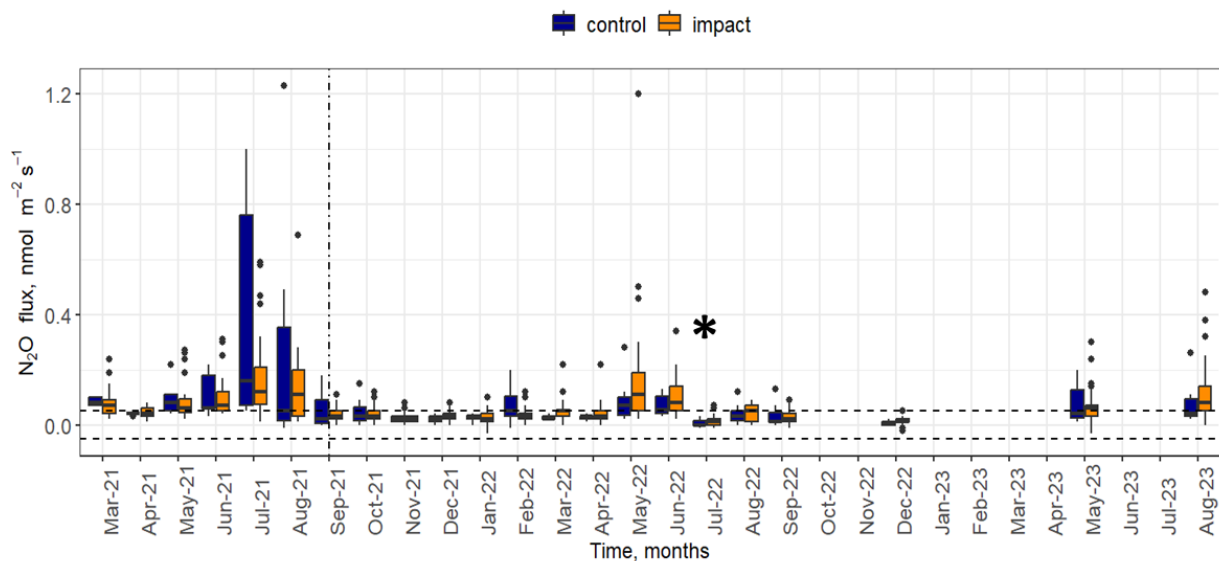


Figure 2 Fluxes of N₂O over the duration of the experiment. Fluxes measured from control area (blue) and area where the impact of NH₃ deposition was expected (orange) are shown. The horizontal dashed lines mark the minimum detectable flux. The vertical line denotes the start of the NH₃ release. An asterisk (*) in July 2022 indicates that the NH₃ release system was inactive for most of the month due to technical issues.

There was no clear relationship between NH₃ dry deposition and N₂O fluxes (linear regression, $R^2 = 0.1$, Fig. 3). Fluxes of N₂O at high levels of N deposition ($>100 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) were relatively low ($<0.4 \text{ nmol N}_2\text{O m}^{-2} \text{ s}^{-1}$), and similar in magnitude to fluxes measured from the control area. There was a single data point which represented a relatively high N₂O flux ($1.2 \text{ nmol N}_2\text{O m}^{-2} \text{ s}^{-1}$) at high N deposition (approximately $45 \text{ kg N ha}^{-1} \text{ yr}^{-1}$), however this was due to the commonly found lognormal distribution of natural N₂O fluxes rather than a treatment effect.

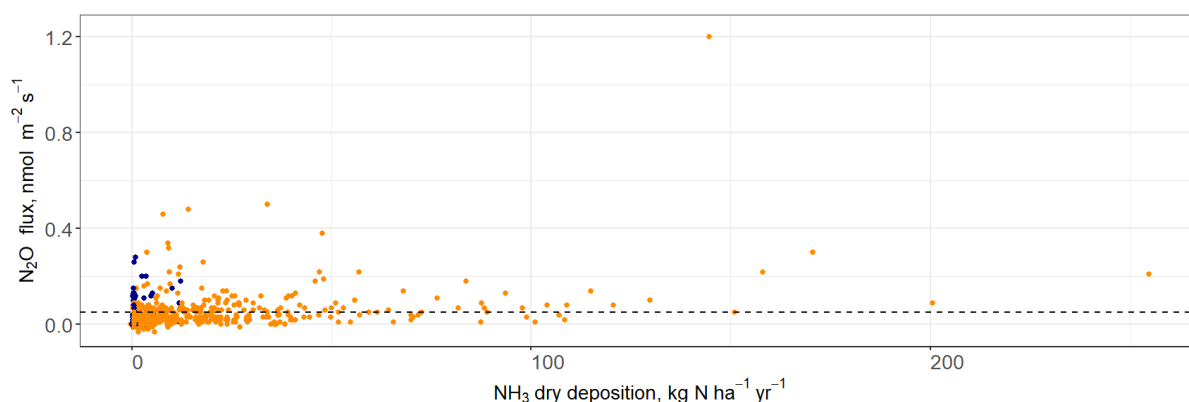


Figure 3 A scatter plot which represents the relationship between NH₃ dry deposition and N₂O fluxes. Horizontal dashed line marks the minimum detectable flux. Fluxes from control and impact areas are shown in blue and orange, respectively.

3.3. In-situ soil N availability

Soil inorganic N availability in the form of NO₃-N and NH₄-N changed little in response to the experimental NH₃ addition. The majority of soil NO₃-N concentrations (87%) were below 3 µg NO₃-N g⁻¹ (Fig. 4). There was little difference between inorganic N content in soils measured before and after the start of the experiment. The median values and the spread of the distribution of control and impact chambers were comparable during most months. One exception was March 2023 where median impact chamber NO₃-N concentrations were higher than the control chambers, though the spread of these values was large indicating high spatial variability.

Concentrations of NH₄-N were higher compared to NO₃-N (monthly medians ranged from 0.3 to 13.5 NH₄-N µg g⁻¹ and from 0.2 to 2.2 NO₃-N µg g⁻¹, for NH₄⁺ and NO₃⁻, respectively) and fluctuated more over the course of the experiment (Fig. 4). The highest median concentrations of NH₄-N were observed in April 2021, August 2022 and March 2023 (9.4, 10.0 and 13.5 µg g⁻¹, respectively). Median NH₄-N concentrations were higher for the control chambers compared to the impact chambers during all sampling months except for October 2021 and November 2022 (after the start of the experimental NH₃ addition).

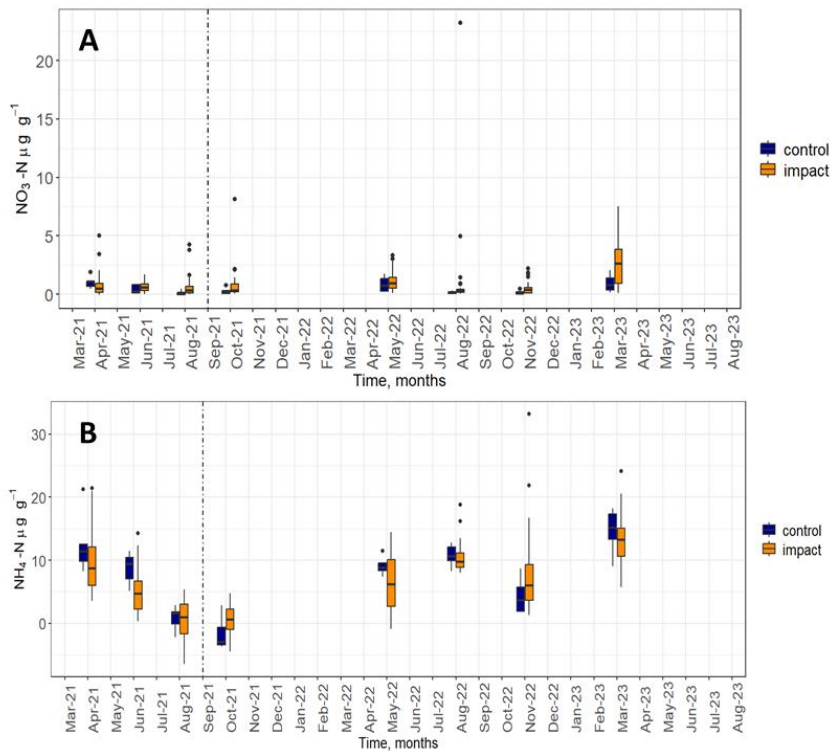


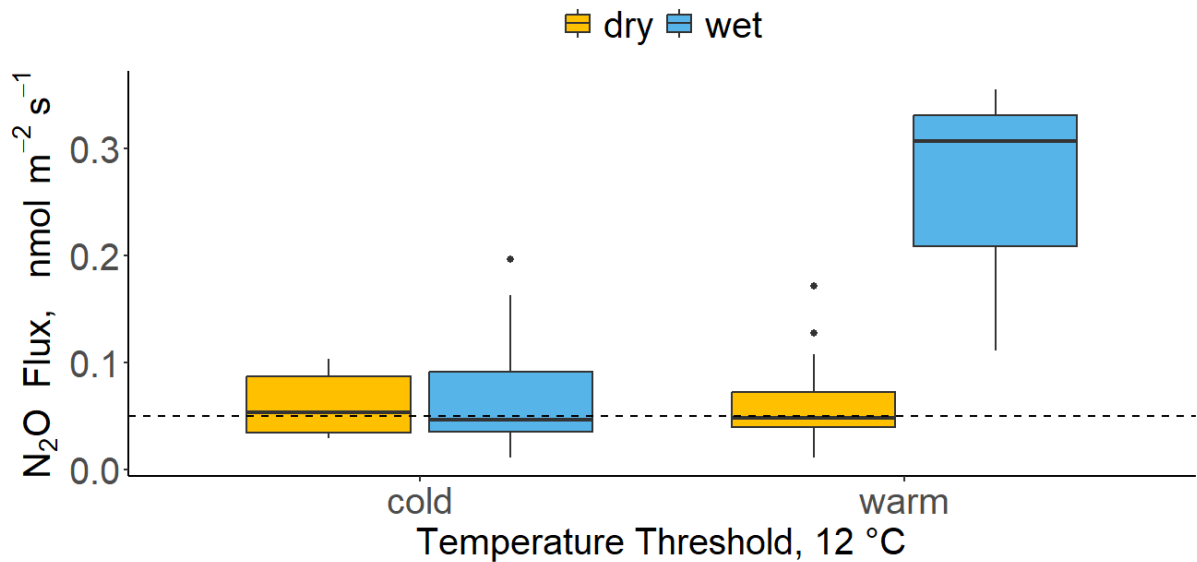
Figure 4 Soil NO₃-N (A) and NH₄-N (B) concentrations in control (blue) and impact (orange) chambers over the duration of the experiment.

3.4. In-situ soil temperature and soil moisture

355 Soil and air temperature as recorded by the meteorological station on site ranged between 2 and 16 °C and approximately -7 and 28 °C, respectively, and followed a seasonal pattern (Fig. S4). Values were consistent between the two years of the experiment.

360 Soil moisture also followed a seasonal pattern, although weekly variability was more pronounced compared to temperature (Fig. S4). There was a discernible temporal trend in soil moisture, whereby July and August 2022 were particularly dry (VWC <13%). Soil moisture was generally low throughout the entire study period (<45% VWC).

365 Soil temperature and soil moisture combined had a pronounced effect on N₂O fluxes (Fig. 5). Fluxes of N₂O were the highest when the environmental conditions were simultaneously “warm” and “wet” (median N₂O flux = 0.3 nmol m⁻² s⁻¹), which in this study corresponded to soil temperatures >12 °C and VWC >20%. There was no difference among N₂O fluxes when temperatures were <12 °C or when moisture was <20% VWC (median N₂O flux = 0.05 nmol m⁻² s⁻¹). This indicates a dual threshold which is likely determining N₂O fluxes.



370 **Figure 5** A boxplot presenting daily average N₂O fluxes at a dual threshold between soil temperature and soil moisture. Temperatures <12 °C were labelled as “cold” and >12 °C as “warm”. The moisture threshold was set to 20% VWC, whereby values below the threshold were labelled as “dry” (orange) and the rest were labelled as “wet” (blue). Horizontal dashed line represented the minimum detectable flux. Soil temperature and moisture were recorded next to each chamber.

3.5 Ex-situ impacts of soil carbon availability

375 The effects of C availability on N₂O fluxes were tested during a laboratory incubation experiment, where air temperature and soil moisture were kept constant at 19 °C and approximately 30% VWC. Similarly to the in-situ field experiment, N₂O fluxes did not increase in response to N addition under controlled laboratory conditions (N₂O fluxes <2 ng N₂O-N g⁻¹ d⁻¹). This was the case for both reduced (NH₄⁺) and oxidised (NH₄NO₃) forms of N at both medium (40 N) and high (100 N) levels of N addition and was consistent among the triplicate cores (Fig. 6). The highest N₂O flux across all cores was measured following
380 the application of a source of labile C (glucose) alongside N. It occurred on day 37 of the experiment and ranged between 1317 ng N₂O-N g⁻¹ d⁻¹ (NH₄ 40N treatment) and 1801 ng N₂O-N g⁻¹ d⁻¹ (AN 40N treatment) across all cores, including the 0N controls (Fig. 6). The magnitude (1531 ng N₂O-N g⁻¹ d⁻¹) and duration of the peak (3 days) of the control (0N) were not significantly different from the experimental ones (t-test, p value >0.5).

385 Cumulative N₂O fluxes were two orders of magnitude greater following the application of C and N together relative to the application of N only (t-test, p-value <0.01). These ranged from 13 (0N) to 71 ng g⁻¹ N₂O-N (AN 100N) and from 2416 (NH₄ 40N + C) to 3057 ng g⁻¹ N₂O-N (AN 40N + C) for the N and N+C applications, respectively. The mean cumulative emissions of application of 'N only' and 'N+C' were 2.5 ± 0.7 and 3091 ± 874 ng N₂O-N g⁻¹, respectively. Cores treated with AN exhibited higher cumulative N₂O flux relative to cores treated with NH₄⁺ during both application periods, although this
390 difference was not significant. In terms of N dose, 100N resulted in higher fluxes compared to 40N for all treatments except AN plus glucose.

The primary pathway for N gaseous losses following the application of N only was NO rather than N₂O fluxes, although their magnitude was relatively low (ranging from 1.2 to 13.8 ng NO-N g⁻¹ d⁻¹). Uncertainty around NO fluxes was higher compared
395 to N₂O fluxes, likely due to a higher variability among the triplicates (Fig. S6). Fluxes of NO were low (<1.2 ng NO-N g⁻¹ d⁻¹) following the application of glucose irrespective of the treatment. Overall, gaseous N losses occurred predominantly in the form of N₂O rather than NO during the laboratory experiment (two orders of magnitude higher cumulative fluxes) due to the effect of the glucose addition (Table 2).

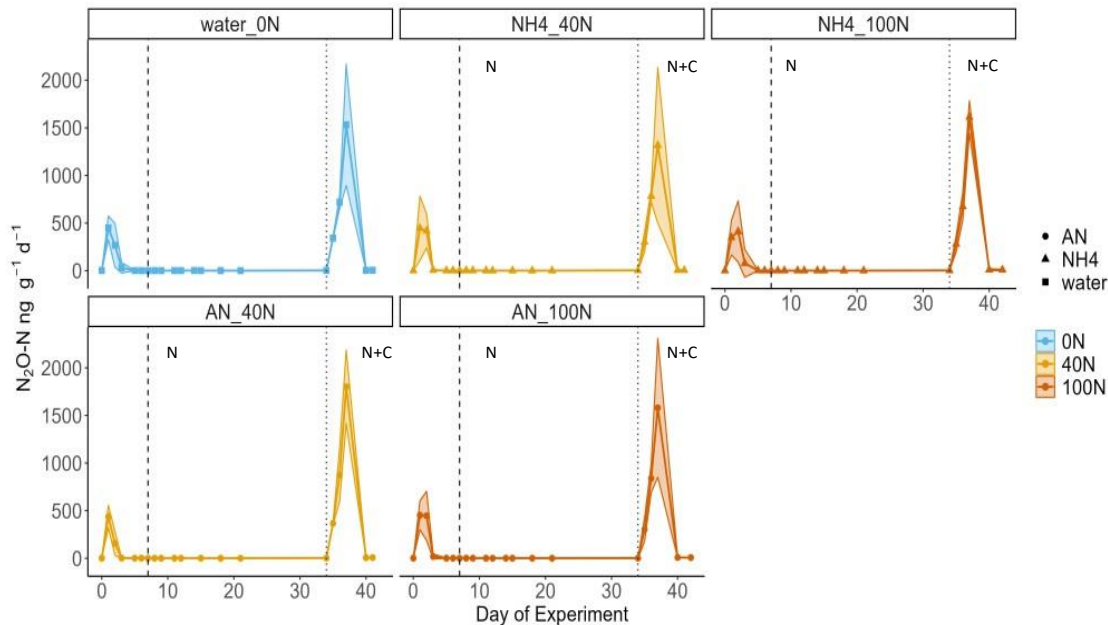


Figure 6 Fluxes of N_2O over the course of the laboratory experiment. Vertical dashed lines correspond to the first N application (in the form of NH_4^+ or AN, depending on the treatment); vertical dotted lines signify the second application of N and glucose (N+C). Blue, orange and red correspond to N addition of 0, 40 and 100 $\text{kg N ha}^{-1} \text{ yr}^{-1}$, respectively. Shaded areas correspond to the 95 confidence intervals (C.I.) as calculated based on triplicates. The initial increase in N_2O fluxes following rewetting on day 1 was attributed to the well-documented Birch effect (Birch, 1964) and is not discussed further. Fluxes of N_2O -N are presented per grams of dry soil rather than per area to better represent the laboratory nature of the experiment.

Table 2 Cumulative fluxes of N_2O and NO at three levels of N addition (the equivalent of 0, 40 and 100 $\text{kg N ha}^{-1} \text{ yr}^{-1}$). Nitrogen was applied either in the form of ammonium (NH_4^+) or ammonium nitrate (AN, NH_4NO_3). NDF stands for “non-detectable flux”.

N form	Treatment	Time (days)	Cumulative Flux, (N_2O -N, ng g^{-1})	Cumulative Flux, (NO-N, ng g^{-1})
Control (water)	0N	34	13	65
NH_4^+	40N	34	32	129
NH_4^+	100N	34	45	190
AN	40N	34	60	279
AN	100N	34	71	417
Control (water)	0N + C	42	2603	NDF
NH_4^+	40N + C	42	2416	NDF
NH_4^+	100N + C	42	2589	NDF
AN	40N + C	42	3057	NDF
AN	100N + C	42	2747	NDF

410 4. Discussion

4.1 Experimental NH₃ release

Experimentally increased atmospheric concentrations of NH₃ exhibited a comparable pattern to the ones observed in the Whim bog N addition experiment, where a similar NH₃ enhancement system was utilised (Leeson et al., 2017; Leith et al., 2005). Background monthly atmospheric NH₃ concentrations at the Glencorse field site ($<3.2 \mu\text{g NH}_3 \text{ m}^{-3}$) were of similar magnitude
415 as previously reported for non-agricultural areas across the UK and Europe (Sutton et al., 2011). Background dry deposition at the site was estimated to be approximately $0.7 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ before the start of the experiment (Deshpande et al., 2024), which fell below the critical loads for the UK which are currently considered to be around 10 and $12 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ (Vanguelova et al., 2024).

420 The experimental deposition values are high compared to naturally observed deposition in European forests (Flechard et al., 2020), however, they are representative of forests located close to point sources of NH₃, such as chicken farms (Pitcairn et al., 2002, 1998). Deposition values were also consistent with previous N manipulation studies in forests that often simulate N deposition levels of $100 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ or higher (Du et al., 2024; Liu and Greaver, 2009). The concentrations of total N in a commonly used bioindicator of N, moss tissue (Pitcairn et al., 2003; Salemaa et al., 2020), were also higher in the impact
425 relative to the control areas (data not presented), thus suggesting that the experimentally added N deposited within the field site. It can therefore be concluded that the NH₃ release system was working as expected resulting in elevated NH₃ concentrations at the Glencorse site.

4.2 Environmental factors affecting N₂O flux

Median N₂O fluxes were generally lower and with smaller variability after the start of the NH₃ enhancement experiment
430 compared to before, both for impact and for control chambers, regardless of the distance to the NH₃ source. This went against the expectations that N₂O fluxes would increase at higher levels of N deposition (Davidson et al., 2000; Deng et al., 2020). The classic Hole in the Pipe model proposed that N₂O fluxes would be higher at increased levels of N deposition as N acts as a substrate for the microbial processes of nitrification and denitrification (Firestone and Davidson, 1989). This has been supported by some experimental studies. For instance, in a meta-analysis of 33 studies from non-agricultural soils, Aronson
435 and Allison (2012) reported that N-amended plots released more N₂O relative to control plots. However, the measured N₂O response to N addition weakened over the 23 years covered by the meta-analysis. Bühlmann et al. (2015) and Horváth et al. (2006) also proposed that elevated atmospheric N deposition induced higher N₂O fluxes from Swiss and Hungarian forest soils, respectively.

440 The environmental factors that drive N₂O production and emission from soils are complex (Butterbach-Bahl et al., 2013) and increased N availability does not always enhance N₂O fluxes. Liu and Greaver (2009) reviewed global N₂O emissions from

agricultural and natural systems and how the magnitude and direction of fluxes are influenced by ecosystem type, N form and level of N addition. They reported a 215% increase in N₂O fluxes in response to N enrichment from agricultural systems. However, there was no clear dose-response in non-agricultural soils. Similarly, in a large study of 31 European forests, 445 Flechard et al. (2020) found that N₂O fluxes were not affected by levels of N deposition.

The small change observed in soil inorganic N availability in our experiments is a potential explanation for why N₂O fluxes did not change with increased NH₃ deposition in our study. While atmospheric N deposition is a commonly used proxy for predicting N₂O fluxes from soils (Hergoualc'h et al., 2019; IPCC, 2019, 2006), it does not always correlate with soil N 450 availability - which can be a major driver of N₂O production (Niu et al., 2016). This is consistent with the present findings, whereby inorganic N concentrations were not higher closer to the N source. Redding et al. (2016) suggested that it is soil N availability rather than atmospheric NH₃ concentrations that control N₂O fluxes from forest soils. They demonstrated this empirically by exposing clay and sandy soils to low levels of N deposition (the equivalent of 30 kg N ha⁻¹ yr⁻¹). Their findings proposed 70 mg N kg⁻¹ soil as a threshold below which N deposition did not induce N₂O flux. The concentrations of inorganic 455 N (NH₄-N and NO₃-N) that were measured in the current study were generally low (<35 µg N g⁻¹ or the equivalent of <35 mg N kg⁻¹) and so fell under this proposed threshold.

The fact that N₂O fluxes did not change in response to increased NH₃ concentrations (nor deposition) could be a result of N₂O production and emission being controlled by a wide range of environmental factors, other than N availability (Baggs, 2008; 460 Butterbach-Bahl et al., 2013). Notably, soil moisture, temperature, and C availability. This is consistent with the findings of this study which suggest that environmental factors had a more pronounced effect on N₂O fluxes relative to the experimental treatment (gaseous NH₃ addition). For instance, the highest in-situ N₂O fluxes were observed between June and August 2021 (Fig. 2) which corresponded to a relatively warm (soil temperature > 12 °C) and wet period (VWC > 20 %) (Fig. S4). In contrast, N₂O fluxes during June to August 2022 were relatively low (median flux < 0.05 nmol N₂O m⁻² s⁻¹), which could 465 potentially be explained by a period of drought that summer (VWC < 15%). These findings suggest a dual temperature-moisture threshold which could be controlling soil N₂O fluxes.

Soil temperature is generally considered to have an amplifying effect on N₂O fluxes (Braker et al., 2010). This is supported by the current observations where N₂O fluxes increased with temperature (especially above 12 °C) and were generally lower over 470 the winter (non-growing) season. In addition, soil moisture has also been documented to modulate N₂O, whereby fluxes typically occur between 30% and 90% WFPS, with a peak around 60% WFPS (Davidson et al., 2000). It is likely that the moisture levels at Glencorse during this experiment were generally too low for denitrification to occur as it is a well-drained sandy mineral soil. Even though N₂O can be produced through other microbial processes (such as nitrification), denitrification is considered to be a major pathway and it tends to occur in wetter soils, under anaerobic conditions (Butterbach-Bahl et al., 475 2013).

Carbon availability is another environmental factor which has been proposed to strongly influence N₂O production from forest soils (Chapin et al., 2011). Soil C acts as a major source of energy for microorganisms and as such plays an important role in modulating soil microbial processes (Fontaine et al., 2003). Glencorse is a young forest (<50 years old) which could explain the relatively low total C and C:N ratio that was observed (Luyssaert et al., 2008). For instance, the C:N ratio at Glencorse soils was 13.1 on average. Previously reported values for C:N ratio in the topsoil of European deciduous broadleaf forests ranged from approximately 10 to 20 (Flechard et al., 2020) and between 17.5 and 20.0 for European birch forest (Cools et al., 2014). In another study, Cleveland and Liptzin (2007) proposed that the stoichiometric C:N:P ratio is consistent in soils (similar to the “Redfield ratio” in oceans) and is approximately 186:13:1, which corresponds to a C:N ratio of 14.3. The C:N ratio in this study was lower than these stoichiometric values and hence it is possible that there was soil carbon limitation at the site. This hypothesis was further supported by the complementary laboratory experiment.

Under controlled laboratory conditions, a peak of N₂O appeared three days after applying N together with glucose to the soil but not when N was added alone. Furthermore, control cores, which did not receive any additional N, exhibited the same pattern after the application of glucose. Nitrogen form and dose did not have any significant effect on the peak, magnitude and duration of N₂O fluxes, whereby control cores were not significantly different from the experimental cores. This further highlighted that C limitation was more important than N availability in the study soils in terms of N₂O fluxes. This observation is consistent with previous research, where the role of soil carbon availability has been highlighted. For example, Weier et al. (1993) demonstrated that denitrification rates were low at high concentrations of N (100 kg N ha⁻¹) but in the absence of a labile source of C (denitrification rate <5 g N ha⁻¹ d⁻¹ at 60% WFPS) in a laboratory incubation experiment. In contrast, denitrification rates increased with increasing levels of C in the form of glucose (the equivalent of either 180 or 360 kg ha⁻¹) (maximum denitrification rate of 1309 and 2606 g N ha⁻¹ d⁻¹ for low and high levels of C, respectively). In another laboratory incubation study using temperate forest soils, Haohao et al. (2017) reported higher cumulative fluxes of N₂O when N was added (either as NH₄Cl or KNO₃) together with a source of C (glucose). This was further supported by a study of Scottish grassland soils where N₂O fluxes were higher after the application of glucose, especially at lower soil moisture levels (Sanchez-Martín et al., 2008). These observations could be due to the fact that organic C acts as an electron donor during the anaerobic process of denitrification (Morley et al., 2014; Zumft, 1997). This is consistent with the present findings from the laboratory experiment whereby N₂O fluxes were low (<2 ng N₂O-N g⁻¹ d⁻¹) before the application of C.

5. Conclusions

This study highlights carbon limitation as a major factor which modulates the impacts of elevated N levels on gaseous N fluxes in forest soils. Fluxes of N₂O increased after applying a source of C but not in response to N addition alone. Another implication of this study is the potential of forests or tree belts to mitigate NH₃ pollution without incurring an N₂O penalty, given the very

limited response of N₂O fluxes to elevated levels of N during both the field and laboratory experiments. However, impacts on sensitive forest species and ecosystems must be considered when designing NH₃ mitigation strategies, to ensure they remain below NH₃ critical levels and N critical loads. In this study, it has been demonstrated that chronically adding small amounts of N, which is consistent with a nearby source of NH₃, did not increase N₂O from forest soils. However, the role of environmental factors, such as soil temperature, moisture and carbon availability had a more pronounced effect on N₂O fluxes in comparison to the experimental treatment (NH₃ deposition). This might suggest that N₂O emissions at the study site, and in similar temperate forests, are unresponsive to increased N_r, at least in the short term. However, the long-term impacts of N enhancement on N₂O fluxes remain unclear. The current observations are not inconsistent with the IPCC emission factor (EF) for non-agricultural systems of 1% (IPCC 2023) and a longer-term dataset from this experiment would be beneficial to better constrain an emission factor. The response of forest ecosystems to increased N levels might change with more prolonged exposure (Skiba et al., 1999). Overall, there is a need for more long-term field experiments on the effects of increased N availability on N₂O fluxes from natural and semi-natural ecosystems.

520 **Acknowledgements**

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Data availability

Data are currently undergoing preparation for submission to the Environmental Information Data Centre (EIDC; <https://eidc.ac.uk/>, EIDC, 2025).

Competing interests

The contact author has declared that none of the authors has any competing interests.

Author contributions

GYT – conceptualisation, data curation, formal analysis, investigation, methodology, visualisation, writing (original draft)

535 DR – conceptualisation, funding acquisition, methodology, resources, supervision, writing (review and editing)
 MJ - conceptualisation, funding acquisition, methodology, resources, supervision, writing (review and editing)
 AD – methodology, investigation, resources, visualisation, writing (review and editing)
 NC – formal analysis, methodology, investigation, resources, visualisation, writing (review and editing)
 PL – formal analysis, methodology, resources, software, writing (review and editing)

540 DH – methodology, investigation, resources, writing (review and editing)
 AI - investigation, resources, writing (review and editing)
 JD - conceptualisation, funding acquisition, methodology, resources, supervision, writing (review and editing)

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