Effects of liming on oxic and anoxic N2O and CO2 production in different horizons of boreal acid sulfate soil and non-acid soil under controlled conditions

Xu, Chang

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Effects of liming on oxic and anoxic N$_2$O and CO$_2$ production in different horizons of boreal acid sulfate soil and non-acid soil under controlled conditions

Chang Xu [a], Vanessa N.L. Wong [a], Anna Tuovinen [b], Asko Simojoki [b],*

[a] School of Earth, Atmosphere and Environment, Monash University, Wellington Road, Clayton, VIC 3800, Australia
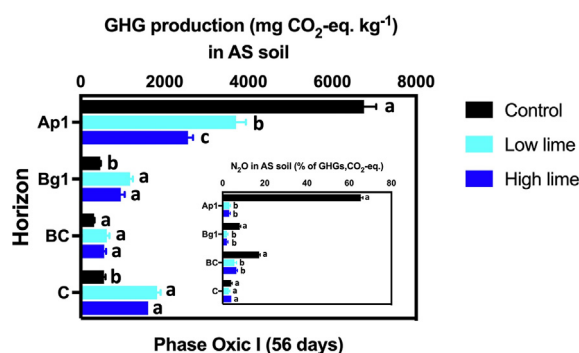[b] Department of Agricultural Sciences, University of Helsinki, P. O. Box 56 (Biocenter 1, Viikinkaari 9), FI-00014, Finland

HIGHLIGHTS

- Organic matter and nitrogen-rich AS soils are large sources of acidity and GHGs.
- Liming decreased oxic and anoxic N$_2$O production in most horizons in AS soil.
- Higher CO$_2$ and N$_2$O production were found in AS soil compared with non-AS soil.
- Liming reduced GHG production efficiently in topsoil but not in subsoil of AS soil.
- Liming often reduced N$_2$O/(N$_2$ + N$_2$O) but increased CO$_2$ production and denitrification.

GRAPHICAL ABSTRACT

ABSTRACT

In acid sulfate (AS) soils, organic rich topsoil and subsoil horizons with highly variable acidity and moisture conditions and interconnected reactions of sulfur and nitrogen make them potential sources of greenhouse gases (GHGs). Subsoil liming can reduce the acidification of sulfidic subsoils in the field. However, the mitigation of GHG production in AS subsoils by liming, and the mechanisms involved, are still poorly known. We limed samples from different horizons of AS and non-AS soils to study the effects of liming on the N$_2$O and CO$_2$ production during a 56-day oxic and subsequent 72-h anoxic incubation. Liming to pH $\geq$ 7 decreased oxic N$_2$O production by 97–98 % in the Ap1 horizon, 38–50 % in the Bg1 horizon, and 34–36 % in the BC horizon, but increased it by 136–208 % in the C horizon, respectively. Liming decreased anoxic N$_2$O production by 86–94 % and 78–91 % in Ap1 and Bg1 horizons, but increased it by 100–500 % and 50–162 % in BC and C horizons, respectively. Liming decreased N$_2$O/(N$_2$ + N$_2$) in anoxic denitrification in most horizons of both AS and non-AS soils. Liming significantly increased the cumulative oxic and anoxic CO$_2$ production in AS soil, but less so in non-AS soil due to the initial high soil pH. Higher carbon and nitrogen contents in AS soil compared to non-AS soil agreed with the respectively higher cumulative oxic N$_2$O production in all horizons, and the higher CO$_2$ production in the subsoil horizons of all lime treatments. Overall, liming reduced the proportion of N$_2$O in the GHGs produced in most soil horizons under oxic and anoxic conditions but reduced the total GHG production (as CO$_2$ equivalents) only in the Ap1 horizon of both soils. The results suggest that liming of subsoils may not always effectively mitigate GHG emissions due to concurrently increased CO$_2$ production and denitrification.

1. Introduction

Acid sulfate soils (AS soils) are soils or sediments that contain oxidizable or partly oxidized sulfide minerals (Pons, 1973). They have been estimated

to occupy an area of over 17 Mha worldwide, including in South and Southeast Asia, West and Southern Africa, Australia, Latin America, North America and boreal Europe, especially Finland (Andriesse and Mensvoort, 2006; Ljung et al., 2009). Sulfide-containing soil horizons that remain under anaerobic or waterlogged conditions usually have a near neutral pH and do not cause environmental problems. However, aerobic conditions promote the oxidation of sulfides and the production of sulfuric acid (Backlund et al., 2005; Boman et al., 2008). Aerobic conditions in AS soils can be caused by natural processes such as coastal regression, isostatic uplift, increased droughts caused by climate change (Boman et al., 2010; Mosley et al., 2017), or by anthropogenic interventions such as drainage for agriculture, construction or mining (Åström and Spiro, 2000). High soil acidity can lead to the mobilization of aluminium and heavy metals, low crop production and poor plant growth (Burton et al., 2008). Another potential problem caused by the drainage of AS soils is enhanced microbial decomposition of soil organic matter which can cause rapid production and high emissions of N_2O due to the inherently high organic matter content and large stocks of mineral N in hypoxic AS subsoils (Paasonen-Kivekäs and Yli-Halla, 2005) as well as large emissions of CO_2 (Gatland et al., 2014).

N_2O is produced in soils by two main biological processes: nitrification and denitrification (Davidson et al., 2000; Saggar et al., 2013). During nitrification, N_2O is produced as a by-product during the oxidation of NH_4^+-N to NO_3^--N by ammonia oxidizing bacteria under mostly aerobic but slightly O_2 deficient (hypoxic) conditions (Inubushi et al., 1996; Kowalchuk and Stephen, 2001). In denitrification, NO_3^--N and NO_2^--N are reduced to nitric oxide (NO), nitrous oxide (N_2O) and molecular dinitrogen (N_2) gases when O_2 levels are very low (Cavigelli and Robertson, 2001), *i.e.* under more severe hypoxia or anoxia.

Soil CO_2 production is a combination of microbial decomposition of soil organic matter (heterotrophic) and plant root (autotrophic) respiration (Amundson and Davidson, 1990). In uncropped soil, it is an indicator of microbial activity and decomposition (mineralization) of soil organic matter. Any input of decomposable organic matter into soils increases soil respiration to produce more CO_2 (Bruce et al., 1997). CO_2 emissions from oxic (aerobic) soils are much larger than those from anoxic (anaerobic) soil because oxic respiration by microbes and roots is more efficient compared with anoxic respiration (Poungparn et al., 2009; Xu et al., 2021). The maximum respiration rate occurs at intermediate moisture contents, often at around 40–60 % water filled pore space (WFPS) (Davidson et al., 1998; Schaufler et al., 2010; Scott-Denton et al., 2003).

Soil pH is a key factor that regulates microbiological processes and affects the end products of denitrification (Parkin et al., 1985; Samad et al., 2016). In a review including 50 years of research on soil pH influence on denitrification, Šimek and Cooper (2002) demonstrated a variable relationship between soil pH and denitrification but a consistent negative correlation between soil pH and N_2O/N_2 ratio. Low soil pH prevents the reduction of N_2O mainly by inhibiting the assembly of functional reductase (Liu et al., 2014). In acidic soils, N_2O dominates as a denitrification product over N_2 (Qu et al., 2014; Thomsen et al., 1994). Moreover, in AS soils, the interconnected reactions of nitrogen, iron and sulfur favoured by acidic conditions can contribute to denitrification and N_2O production (Macdonald et al., 2010).

Lime treatments have been widely applied to solve the acidity problems of AS soils, typically utilizing calcium carbonate ($CaCO_3$), calcium hydroxide ($Ca(OH)_2$) and dolomite ($CaMg(CO_3)_2$) to neutralise the acidity, including new methods for liming subsoil horizons (Dalhem et al., 2019). Liming concurrently promotes the reduction of N_2O to N_2 by enhancing the activity of N_2O reductase (Liu et al., 2010; Samad et al., 2016; Shaaban et al., 2018). Hénault et al. (2019) observed that N_2O reduction was inhibited below pH 6.8 but it was more efficient at pH above 6.8–7 after liming. Goulding (2016) reported that raising soil pH from 5.3 to 6.8 by liming can mitigate N_2O emissions in cropped land in the UK. Žurovec et al. (2021) found that liming of grassland soils resulted in a linear decrease in cumulative N_2O emissions in the pH range between 5.1 and 6.9.

Liming may also affect CO_2 emissions from soil by boosting microbial activity that is generally enhanced by increasing pH (Oertel et al., 2016).

In addition, the acid neutralizing reaction of carbonate in itself can also produce CO_2 (West and McBride, 2005).

Besides pH, many other soil and environmental factors also influence N_2O emissions and control the $N_2O/(N_2O + N_2)$ ratio, including soil type, organic carbon, soil NO_3^--N availability, gaseous diffusivity and soil water content. Soil NO_3^--N content is an important factor affecting N_2O emissions through denitrification. Soil water content is a key factor affecting soil aeration and N_2O emissions. Nitrification was usually the dominant source of NO and N_2O production under relatively well aerated conditions at WFPS below 50–60 % (Bateman and Baggs, 2005; Davidson et al., 2000). Conversely, the largest N_2O emissions were produced mainly by denitrification under more hypoxic conditions, with a maximum often occurring near 70 % WFPS (Denmead et al., 2011). The N_2O/N_2 ratio has often been found to decrease at moisture conditions close to saturation, especially above 80 % WFPS (Colbourn and Dowdell, 1984; Rudaz et al., 1999). For example, Guo et al. (2014) observed that the $N_2O/(N_2O + N_2)$ ratios were much higher in a clay loam soil rewetted to 75 % WFPS than to 90 % WFPS.

However, scientific knowledge on the magnitude and factors contributing to the production of N_2O and other GHGs in AS soils is currently still limited. In particular, studies on the effects of liming on N_2O production, GHG production (as CO_2 equivalents), total denitrification ($N_2O + N_2$) and the product ratio of denitrification $N_2O/(N_2O + N_2)$ in the different horizons of AS soils and non-AS soils are limited.

The aim of this study is to determine and compare (1) the effects of lime ($CaCO_3$) treatments on the N_2O and CO_2 production between different horizons of boreal AS and non-AS soils under oxic and anoxic conditions, and (2) the effects of lime on the total denitrification ($N_2O + N_2$) and the product ratio of denitrification $N_2O/(N_2O + N_2)$ between different horizons in AS and non-AS soils under anoxic conditions.

2. Materials and methods

2.1. Field site and soil characterization

The soil materials of AS and non-AS soils for the incubation experiment of this study were collected from two sites at the Viikki research farm of the University of Helsinki, located on the coast of the Gulf of Finland, which is part of the Baltic Sea. The sites have been previously studied by Mokma et al. (2000) and Šimek et al. (2011, 2014). The mean annual air temperature in this area is 5.9 °C and annual precipitation is 655 mm (a 30-year average in 1981–2010, Finnish Meteorological Institute). At 50 cm soil depth, the mean annual soil temperature is about 6 °C and the mean soil temperature in summer is about 14 °C (Mokma et al., 2000; Šimek et al., 2011). The AS soil (Patoniitty: 60°13′N, 25°0′E, elevation at sea level), formed on fine-textured (clayey) sediments, is representative of AS soils along the coast of Finland. The non-AS soil (Alaniitty: 60°13′N, 25°1′E with elevation about 1.5 m above average sea level), formed on fine-grained sediment, is poorly drained (Mokma et al., 2000). According to Soil Taxonomy (Soil Survey Staff, 2014), the AS soil of Patoniitty is a Sulfic Cryaquept and the non-AS soil of Alaniitty is an Aquic Haplocryoll.

Soils were sampled on 27th September 2019 from excavated large pits. After characterization of soil horizons at both sites, representative samples were taken from four selected horizons of the AS and non-AS soils. In the AS soil, the Ap1, Bg1, BC and C horizons were at depths of 0–22, 31–47, 66–115 and 115–135 cm, respectively. The groundwater table was at 1.1 m depth. In the non-AS soil, the Ap1, Bg1, BC and C horizons were at depths of 0–20, 31–68, 78–115 and 133–153 cm. The groundwater table was at 1.3 m depth. Soils were sampled with spades from the unsaturated soil horizons into 40-L plastic boxes. The samples from the saturated C horizons below the groundwater level were sampled similarly but stored submerged under water in plastic bags before further processing.

A representative sample of the field moist soil taken from a given horizon was mixed and subsequently divided into several sets of subsamples. The subsamples for initial mineral nitrogen analysis were immediately frozen at −20 °C, whereas the subsamples for the chemical analyses were air-dried in a ventilated oven at 35 °C and sieved through 2-mm mesh,

and a third set of subsamples was used for the determination of initial soil water content.

The main properties of AS soil and the non-AS are shown in Table 1. In the AS soil, the depth profile of pH was typical of AS soils, with a slightly acidic pH in the Ap1 (topsoil) and close to neutral pH in the C (massive, reduced subsoil) horizons but with a pH below pH 4 in the BC horizon. In contrast, the pH of the non-AS soil was close to 6 in the topsoil and at, or above 7 in all subsoil horizons.

The AS soil was an organic rich soil, where the SOC content was high in the Ap1 (4.6 %) and C (2.7 %) horizons, and only slightly below 2 % in the organic poor Bg1 horizon. In the non-AS soil, the SOC content was high in the Ap1 horizon (2.7 %) but very low (0.23–0.52 %) in all subsoil horizons (Table 1). The distribution of total nitrogen content (TN) followed a similar pattern with soil depth as that of SOC. Both AS and non-AS soils showed broadly similar patterns of decreasing NO_3^--N and NH_4^+-N with depth, except for the large accumulation of NH_4^+-N in the BC and C horizons in the AS soil, and modest accumulation of NH_4^+-N in the BC horizon of the non-AS soil. The SOC, TN and mineral N (sum of NO_3^--N and NH_4^+-N) contents in all horizons of the AS soil were higher than those in the non-AS soil (Table 1).

2.2. Setup and design of the incubation experiment

Sequential oxic (day 0–56) and anoxic (day 56–59) incubations with soil samples taken from each horizon of AS and non-AS soils were carried out at three lime levels at room temperature (20 °C) and at the targeted 70 % WFPS with a time sequence as shown in Fig. 1. Soil samples were prepared as follows: 10 g of soil (dry mass basis) was packed into 120-mL glass bottles (diameter 3.5 cm, height 12.5 cm). In the incubation bottle, the average dry bulk densities were 0.82, 0.80, 0.57 and 0.53 g cm^{-3}, and 0.83, 0.80, 0.77 and 0.59 g cm^{-3}, in the soil taken from the Ap1, Bg1, BC and C horizons of the AS and non-AS soil, respectively.

The WFPS of the soil packed into the bottle was calculated according to Eqs. (1) and (2) (See Supplementary materials, Tables S3 and S4):

$$WFPS = (\theta/n) \tag{1}$$

where θ (m^3 m^{-3}) is the volumetric soil water content and n (m^3 m^{-3}) is the soil porosity.

$$n = 1 - (\rho_b/\rho_s) \tag{2}$$

The dry bulk density of soil, ρ_b, was calculated as the ratio of dry mass and volume of soil in each bottle. Total porosity was calculated by assuming an average soil particle density of $\rho_s = 2.65$ g cm^{-3}.

The samples were maintained a constant WFPS by weighing the incubation bottles twice per week and adding MQ water as needed during the oxic incubation (Phase Oxic I, Fig. 1).

The soil samples were subjected to three experimental treatments to adjust the pH. At the beginning of Phase Oxic I, pure calcite (CaCO$_3$, EMSURE® Reag. Ph Eur) was applied to the soil at the rates of 12.5 and 25 mg g^{-1} (dry mass basis) by pipetting either 10 mL of 0.125 M CaCO$_3$ or 10 mL of 0.25 M CaCO$_3$ as a solution into the soil in the low lime and high lime treatments, respectively. The control treatment was established with no added lime by pipetting a volume of 10 mL MQ water into the soil. The calcite additions were based on the pre-tests of lime requirement in all studied horizons of AS soil (see Supplementary materials). The target soil pH in the high lime treatment was pH 7.

A total of 96 soil samples (set 1) were used for the weekly measurements of oxic N$_2$O and CO$_2$ production during Phase Oxic I at days 0, 7, 14, 21, 28, 35, 42, 49 and 56 followed by the measurement of anoxic N$_2$O and CO$_2$ production during the 72-h Phase Anoxic II (Fig. 1). Another 96 soil samples (set 2) were similarly incubated but without gas production measurement during Phase Oxic I in order to be able to determine the total denitrification by the acetylene (C$_2$H$_2$) inhibition method during Phase Anoxic II (see section 2.3). In addition, during Phase Oxic I, a separate set of 96 soils samples (set 3) (Fig. 1) was incubated separately to monitor soil pH with time at 7-day intervals.
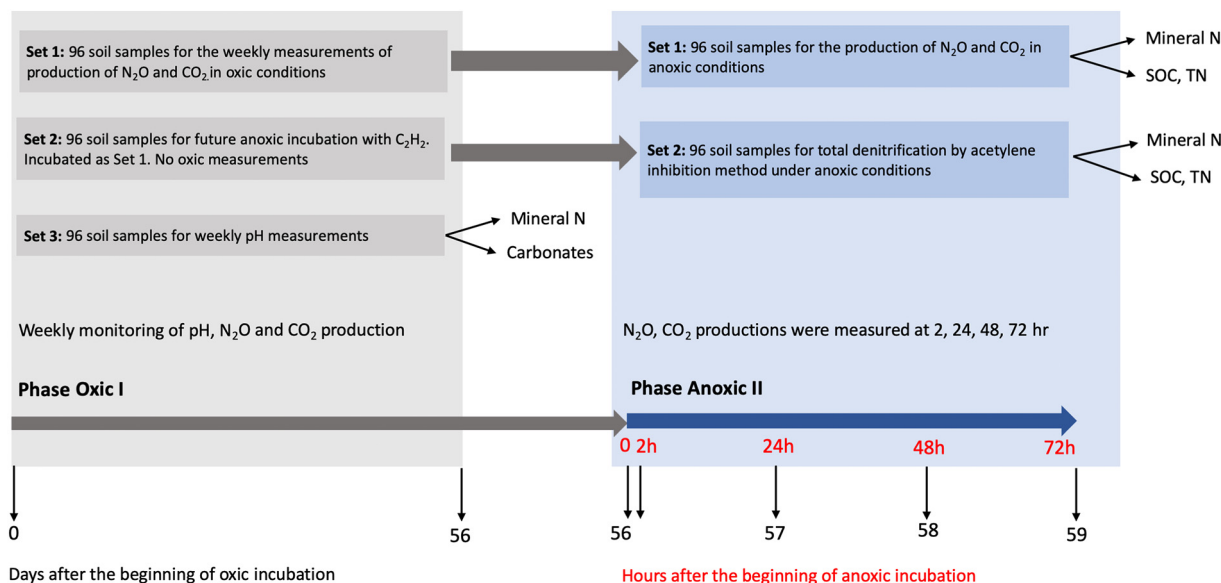
2.3. N$_2$O and CO$_2$ production determination

To determine the oxic gas production rates, the headspace of bottle was first flushed with compressed air, then the bottle was capped gastight with a butyl rubber septum until the gas samples were collected from the headspace after 24 h. A 9-mL gas sample was drawn within 10 s from the headspace of the incubation bottle using a gas-tight syringe and transferred into a He-flushed and pre-evacuated 3-mL Exetainer® vial capped with a double-wadded (PTFE/silicon-butyl) septum (VC329, Labco, UK). The gaseous composition of samples was determined by a gas chromatograph (Agilent Technologies 7890B GC custom, Santa Clara, CA, United States) connected to an autosampler and equipped with gas sampling valves and TCD, FID and ECD detectors. The air pressure in the headspace was monitored with a pressure meter (Tensimeter, Soil Measurement Systems, CA, United States) at each sampling to calculate the true gas concentration in the headspace, c = c$_s$ × p$_{hs}$/p$_0$, where p$_{hs}$ is the pressure in the headspace, p$_0$ is the normal atmospheric pressure, and c$_s$ is the gas concentration of sample at p$_0$.

The gas production rates F$_{N2O}$ and F$_{CO2}$ were calculated from the change of gas concentration in the headspace with time by using Eqs. (3) and (4). The N$_2$O and CO$_2$ production rates were corrected for air temperature during the measurement and presented as [μg kg^{-1} h^{-1}]

$$F_{N2O \text{ or } CO2} = \frac{\Delta c/\Delta t \times (V + V_w \alpha_B) \times M}{m_d \times MV_{corr}} \tag{3}$$

where $\Delta c/\Delta t$ is the temporal rate of concentration change in the headspace [$LL^{-1}h^{-1}$]; V is the volume of the headspace [L]; V$_w$ is the volume of water

Table 1
Selected physical and chemical properties[a] of AS soil at Patoniitty and the non-AS soil at Alaniitty in Helsinki.

Horizon	Depth (cm)	Bulk density (g cm^{-3})	pH (Fresh)[b]	pH (Incubated)[c]	SOC (Soil organic carbon) (%)	TN (Total nitrogen) (%)	C/N	NO_3^--N (N mg kg^{-1})	NH_4^+-N (N mg kg^{-1})
AS soil									
Ap1	0–22	0.85 ± 0.01	5.64 ± 0.005	5.57 ± 0.05	4.60 ± 0.01	0.38 ± 0.002	12.25 ± 0.11	27.76 ± 1.73	2.18 ± 0.57
Bg1	31–47	1.00 ± 0.01	4.15 ± 0.03	3.98 ± 0.005	1.89 ± 0.06	0.17 ± 0.007	10.87 ± 0.81	7.7 ± 0.21	3.82 ± 1.56
BC	66–115	0.64 ± 0.005	3.38 ± 0.04	3.08 ± 0.03	2.13 ± 0.01	0.21 ± 0.002	10.12 ± 0.03	0.11 ± 0.11	10.77 ± 2.08
C	115–135	0.54 ± 0.01	6.51 ± 0.10	4.28 ± 0.06	2.67 ± 0.01	0.29 ± 0.004	9.24 ± 0.09	0.005 ± 0.0001	13.28 ± 1.97
Non-AS soil									
Ap1	0–20	1.03 ± 0.02	6.30 ± 0.03	6.46 ± 0.05	2.69 ± 0.04	0.23 ± 0.003	11.83 ± 0.04	20.79 ± 1.39	1.79 ± 0.58
Bg1	31–68	0.97 ± 0.01	7.46 ± 0.02	7.45 ± 0.02	0.33 ± 0.001	0.02 ± 0.002	15.85 ± 1.26	5.67 ± 0.34	0.42 ± 0.20
BC	78–115	1.01 ± 0.02	7.68 ± 0.05	7.76 ± 0.03	0.23 ± 0.001	0.014 ± 0.0002	16.43 ± 0.28	2.08 ± 0.26	3.64 ± 1.72
C	133–153	0.81 ± 0.005	7.22 ± 0.02	7.30 ± 0.02	0.52 ± 0.004	0.03 ± 0.0001	16.61 ± 0.30	0.1 ± 0.07	0.22 ± 0.10

[a] Means ± standard errors (n = 4).
[b] Fresh pH refers to the pH of soils without aerobic incubation.
[c] Incubated pH refers to the pH of soils after Phase Oxic I (56 days).

Fig. 1. Time sequence of the oxic incubation (day 0–56) and anoxic incubation (day 56–59). A total of 288 soil samples were adjusted to 70 % WFPS at the beginning of incubation (2 sites × 4 horizons × 3 lime treatments x 4 replicates × 3 sets). The three lime treatments (CaCO₃) including unlimed control, low lime (12.5 mg g^{-1}) and high lime (25.0 mg g^{-1}) were applied at the beginning of Phase Oxic I.

in the soil sample [L]; α_B is the Bunsen's gas solubility coefficient ($\alpha_B = 0.629$ for N_2O and $\alpha_B = 0.942$ for CO_2 at 20 °C); m_d is the dry mass of soil (g); M is the molar mass of gas (12 g mol^{-1} for CO_2-C and 28 g mol^{-1} N_2O—N). MV_{corr} is the temperature-corrected molar volume of ideal gas [m^3 mol^{-1}]

$$MV_{corr} = 0.02241 \text{ m}^3 \text{ mol}^{-1} \times \left(\frac{273.15 + T}{273.15} \right) \tag{4}$$

where T is air temperature during the measurement [°C] and 0.02241 m^3 mol^{-1} is the molar volume of an ideal gas at the standard conditions of 1 atm (1013.25 hPa) and 273.15 K.

Cumulative N_2O (µg N_2O-N kg^{-1}) and CO_2 (mg CO_2-C kg^{-1}) production were calculated with the trapezoidal rule for the production rates (µg kg^{-1} h^{-1}) of all individual sampling data during the incubation period (56 days). The cumulative GHG production rate as CO_2-equivalents (CO_2-eq) was calculated as the sum of CO_2 and N_2O with a global warming potential of 298 used for converting N_2O to CO_2-eq. CH_4 production was also measured but the results were always zero or very low (data not shown). For this reason, the sum of N_2O and CO_2 refers essentially to the sum of all GHGs in our study.

In Phase Anoxic II (after Phase Oxic I), the soil samples from the sets 1 and 2 were incubated for a further 72 h under anoxic conditions at 70 % WFPS to support denitrification (Fig. 1). The C_2H_2 inhibition method was used to block the reduction of N_2O to N_2 for the determination of the total denitrification ($N_2O + N_2$) (Nadeem et al., 2013; Yoshinari et al., 1977). The soil sample sets of 1 and 2 were capped with a butyl rubber septum and evacuated, and the air in the headspace was replaced with N_2 (set 1) or $N_2 + 10$ kPa C_2H_2 (set 2) to achieve strictly anoxic conditions. Soil sample sets of 1 and 2 were incubated simultaneously. Anoxic gas production was measured by sampling the headspace at 2, 24, 48 and 72 h after closing the bottle, applying the Eqs. (3) and (4) for each time interval (0–2, 2–24, 24–48 and 48–72 h) separately and summing up the contributions of time intervals.

2.4. Soil chemical and physical analyses

Soil analyses were performed with four replicates. The initial soil pH and electrical conductivity (EC) were measured in a 1:2 soil:water suspension (*w/v*) (Rayment and Lyons, 2011) using a combination electrode and

a pH meter (SCHOTT GLAS Mainz CG-843) and an EC meter (Radiometer Copenhagen, Meterlab CDM210, France).

Soil organic carbon (SOC) and total nitrogen (TN) contents were analysed using the initial soil samples, and soil sample sets 1 and 2 after Phase Anoxic II incubation. The analyses were done with oven-dried (105 °C) samples by Dumas combustion with a TCN-analyzer (Leco, CN828). Soil carbonate contents were determined by removing SOC by a muffle furnace and determining the remaining C by Dumas combustion to allow the calculation of the fractional conversion of carbonates (as detailed in Supplementary materials).

Mineral N (NO_3^--N and NH_4^+-N) contents were determined using the initial soil samples, Phase Oxic I soil samples (set 3) and Phase Anoxic II soils samples (set 1, 2 and 3) (Fig. 1). Samples used for mineral N determination were frozen immediately after sampling and stored at −20 °C before analysis. Mineral N extraction was carried out by shaking a soil sample (10 g dry mass of soil) and 40 mL of 2 M KCl solution in a 100-mL bottle with an orbital shaker (180 rpm) for 2 h (Esala, 1995). NO_3^--N and NH_4^+-N were analysed colorimetrically with a Gallery Plus discrete analyzer (Thermo Scientific).

2.5. Statistical analysis

The mean gas production rates in each horizon under different lime treatments in the AS and non-AS soils were based on four replicates. The cumulative N_2O production (µg N_2O-N kg^{-1}) and the cumulative CO_2 production (mg CO_2-C kg^{-1}) during Phase Oxic I were calculated by "area under the curve" analysis (trapezoidal method) with GraphPad Prism 8.0. One-way analysis of variance (ANOVA) and a post-hoc Tukey's test were conducted to detect significant differences ($p < 0.05$) in the fractional conversion of carbonates, the cumulative gas production rates (N_2O and CO_2) and the GHG production rates (as CO_2-eq). All the analyses were performed using GraphPad Prism 8.0.

3. Results

3.1. Soil pH, oxic N_2O production and mineral N contents during Phase Oxic I

In the AS soil, the target pH of 7 was achieved within 2–3 weeks by all low lime and high lime treatments, except for the low lime treatment in the BC horizon in which pH peaked at 6.5 after 1 week and steadily decreased

thereafter (Fig. 2a, c, e, g). In all horizons of the non-AS soil, the pH exceeded 7 immediately after application of the lime treatments (Fig. 2b, d, f, h).

The initial oxic N_2O production rates in the unlimed AS soil decreased with soil depth from 104 μg N_2O-N kg^{-1} h^{-1} in the Ap1 horizon to 0.13 μg N_2O-N kg^{-1} h^{-1} in the C horizon (Fig. 3 a, c, e, g). At the beginning of the incubation, the N_2O production rates in the low lime and high lime treatments of the Ap1, Bg1 and BC horizons were only 0.3 % and 0.15 % (Ap1, Fig. 3a), 77 % and 57 % (Bg1, Fig. 3c), and 19 % and 22 %

(BC, Fig. 3e), respectively, of those in the control treatment. In the C horizon, however, the oxic N_2O production rates in the low lime and high lime treatments were both about 90 % higher than those in the control (Fig. 3g). In the non-AS soil, the oxic N_2O production rates in the Ap1, Bg1 and BC horizons were much lower than those in the AS soil in all treatments (Fig. 3b, d, f). A decreasing temporal pattern of N_2O production rates and decreasing effects of lime additions were observed in Ap and C horizons (Fig. 3b, h), whereas the patterns and effects were variable in the Bg1 and BC horizons (Fig. 3d, f).
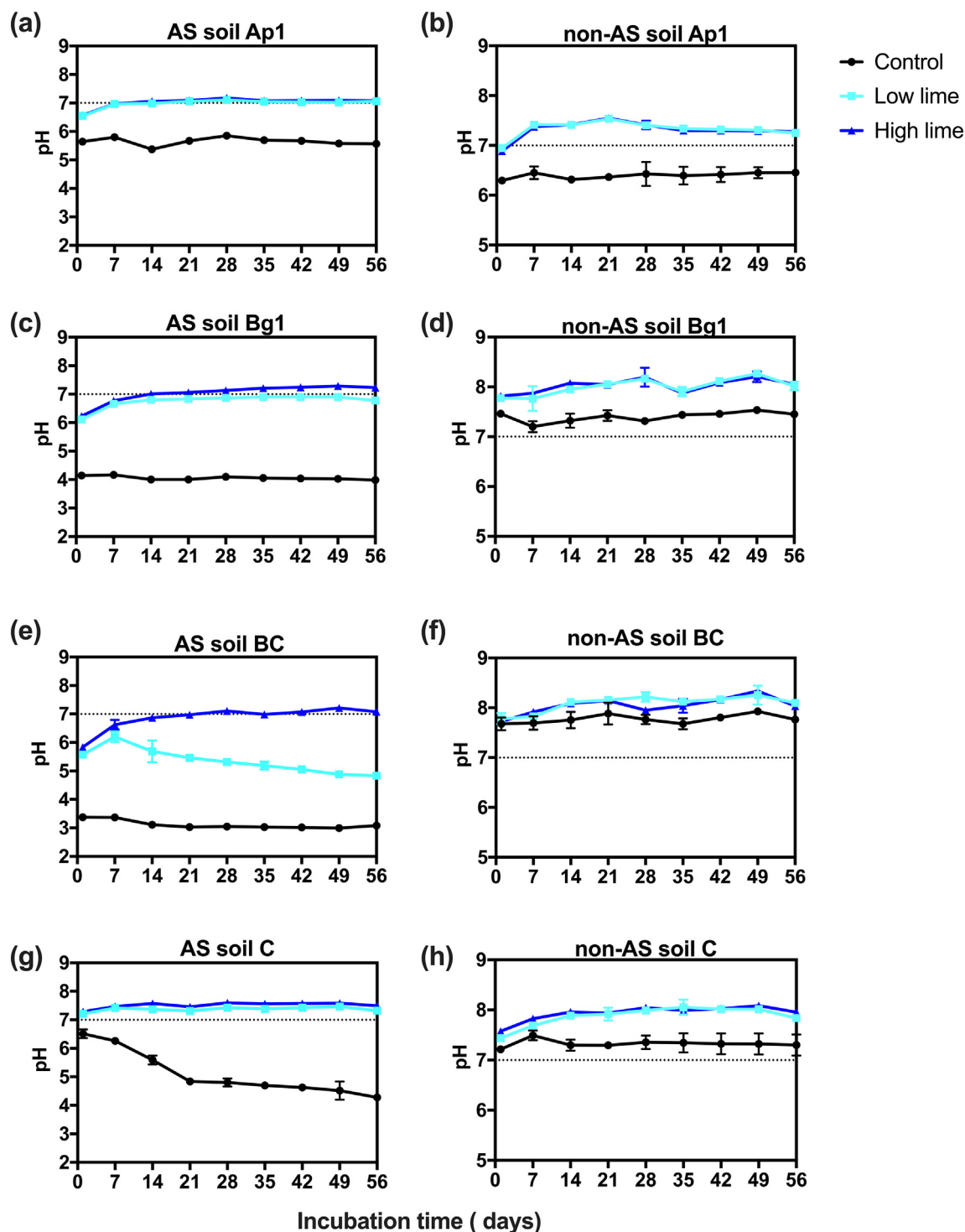

Fig. 2. Temporal variation of pH in the Ap1 (a, b), Bg1 (c, d), BC (e, f), and C (g, h) horizons and lime treatments of the AS and non-AS soils during Phase Oxic I (56 days). Values are the means and their standard errors ($n = 4$).
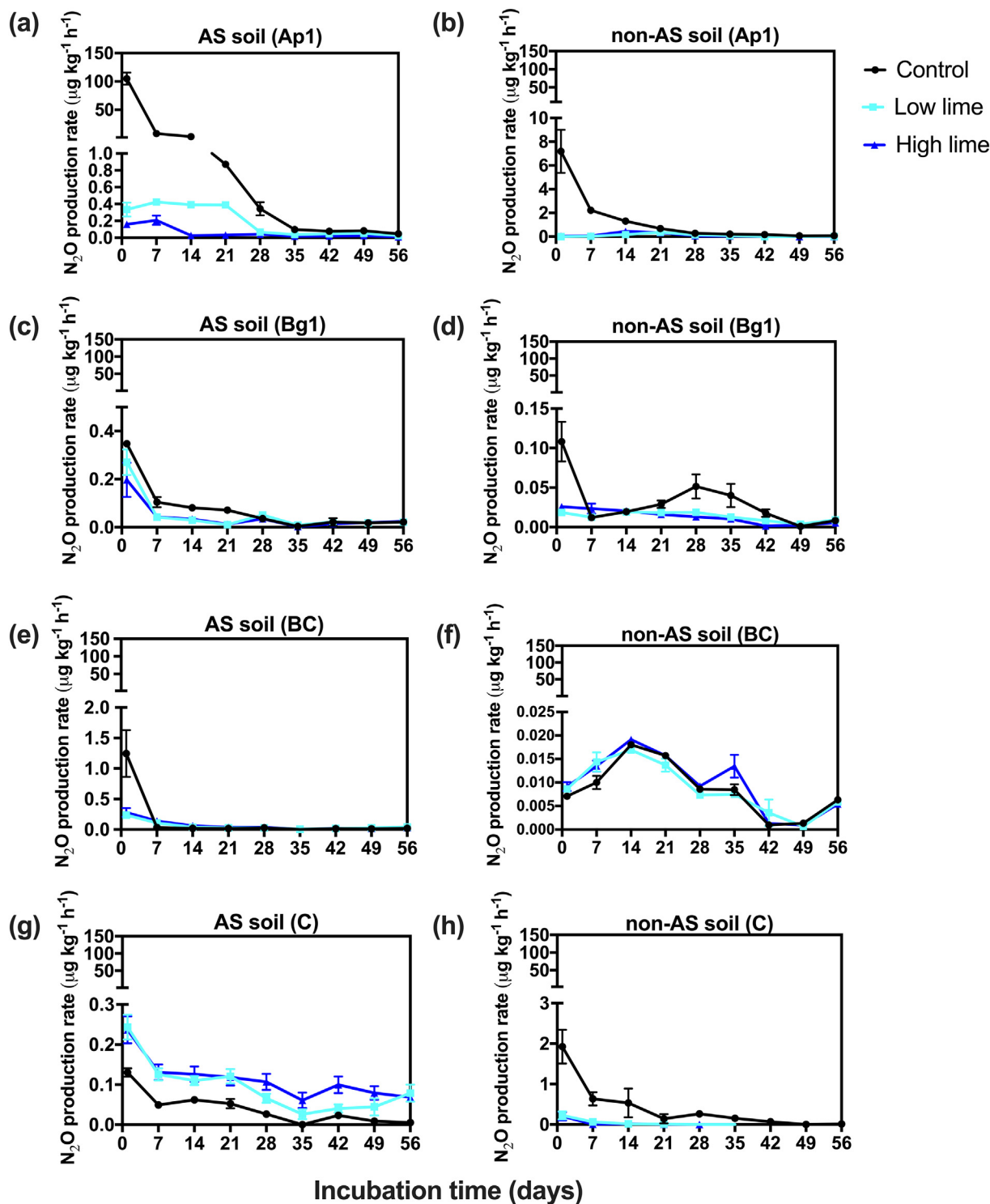
Fig. 3. Oxic N_2O production rate (μg N_2O-N kg^{-1} h^{-1}) in the Ap1 (a, b), Bg1 (c, d), BC (e, f) and C (g, h) horizons of the AS and non-AS soils during Phase Oxic I (56 days). Values are the means and their standard errors ($n = 4$).

Similarly, the cumulative oxic N_2O production in the Ap1, Bg1 and BC horizons of the AS soil during Phase Oxic I was higher in the control than in the lime treatments ($p < 0.05$) (Table 2). The amounts of cumulative N_2O production in the low lime and high lime treatments of Ap1, Bg1 and BC horizons were only 2.7 % and 1.8 % (Ap1), 62 % and 50 % (Bg1), and 64 % and 66 % (BC), respectively, of those in the control treatment (Table 2). In the C horizon, however, the cumulative oxic N_2O production in the low lime and high lime treatments was 136 % and 208 % higher

compared with the control, respectively (Table 2). Significant differences in the cumulative N_2O production were observed between all horizons. The cumulative N_2O production in the Ap1 horizon was always higher compared to all other horizons ($p < 0.05$), followed by that in the C horizon in the limed treatments. The N_2O production in the BC horizon was generally higher than that in the Bg1 horizon (Table 2).

In the non-AS soil, the lime treatments decreased the cumulative oxic N_2O production in the Ap1, Bg1 and C horizons compared to the control

Table 2

Cumulative N_2O production and CO_2 production in different lime treatments and horizons of AS and non-AS soils during Phase Oxic I (56 days).

Soil	GHG	Horizon	Control	Low lime	High lime
AS soil		Ap1	9452.4[aA]	256.4[bA]	171.7[cA]
		Bg1	80.9[aC]	50.0[bD]	40.8[cD]
		BC	117.1[aB]	74.4[bC]	76.9[bC]
	N_2O production	C	46.7[cD]	111.8[bB]	142.2[aB]
	(μg N_2O-N kg^{-1})	Ap1	1320.6[aA]	140.6[cA]	167.2[bA]
		Bg1	35.6[aC]	17.5[bC]	16.2[bC]
Non-AS soil		BC	11.1[bD]	12.5[aD]	13.3[aD]
		C	431.9[aB]	35.4[bB]	26.7[cB]
AS soil		Ap1	636.0[bA]	977.6[aA]	675.8[bA]
		Bg1	116.6[cB]	314.3[aC]	255.1[bC]
		BC	71.7[bC]	160.0[aD]	142.4[aD]
	CO_2 Production	C	144.9[bB]	482.5[aB]	419.6[aB]
	(mg CO_2-C kg^{-1})	Ap1	693.1[aA]	639.4[bA]	678.4[aA]
		Bg1	21.8[cC]	41.3[aC]	30.6[bC]
Non-AS soil		BC	5.1[bD]	16.5[aD]	16.8[aD]
		C	57.7 5[cB]	83.3[aB]	74.4[bB]

Different small letters within each row indicate significant differences between the lime treatments. Different capital letters within each column of N_2O production and CO_2 production in the AS soil and non-AS soil, respectively, represent significant differences between different soil horizons (one-way ANOVA, Tukey test, $p < 0.05$; n = 4).

($p < 0.05$) (Table 2). The low and high lime treatments decreased the cumulative N_2O production by 89 % and 87 % in the Ap1 horizon, by 51 % and 55 % in the Bg1 horizon, and by 92 % and 94 % in the C horizon, respectively, while in the BC horizon, they increased it by 13 % and 20 % compared to the control (Table 2). Significant differences in the cumulative N_2O production were observed between all horizons, with the rates decreasing in the order of Ap1 > C > Bg1 > BC horizon in all treatments. Nevertheless, in a given horizon and treatment, the cumulative N_2O production in the AS soil was always larger, and in most cases even several times larger, than that in the non-AS soil (Table 2).

In both AS and non-AS soil, the NO_3^--N content decreased with time in the Ap1 and Bg1 horizons, and increased with time in the BC and C horizons, in all treatments during the oxic incubation (Table S5). In the AS soil, NH_4^+-N increased with time in all horizons and treatments, whereas in the non-AS soil, NH_4^+-N increased in the Ap1, Bg1 and C horizons but decreased in the BC horizon (Table S5).

3.2. Oxic CO_2 production during Phase Oxic I

In all horizons of the AS soil, the oxic CO_2 production rate during Phase Oxic I was much higher in limed soils than in unlimed soils (Fig. 4a, c, e, g). The CO_2 production rate in the lime treated soils decreased significantly during the first 14 days and then decreased more slowly or stabilised in the Ap1, Bg and BC horizons (Fig. 4a, c, e), whereas in the C horizon, following an initial decrease, the CO_2 production rate increased slightly after day 14 (Fig. 4g). In the Bg1, BC and C horizons of the non-AS soil, the CO_2 production rate in the low and high lime treatments were higher than that in the control treatment, especially during the first week of incubation (Fig. 4d, f, h), whereas the CO_2 production in the Ap1 horizon was similar in all treatments. The CO_2 production rate in the AS soil was much higher than that in the non-AS soil (Fig. 4).

In the AS soil, lime treatments in most cases increased cumulative oxic CO_2 production compared to the control ($p < 0.05$) (Table 2). As compared to the control treatment, the cumulative CO_2 production in the low and high lime treatments were 50 % and 6 % higher in the Ap1 horizon, and 170 % and 120 % higher in the Bg1 horizon, 120 % and 100 % higher in the BC horizon, and 230 % and 190 % higher in the C horizon, respectively (Table 2). Significant differences in the amount of cumulative oxic CO_2 production were observed between all horizons before and after lime treatment, with the highest cumulative oxic production in the Ap1 horizon, and the lowest in the BC horizon (Table 2).

In the Bg1, BC and C horizons of the non-AS soil, higher cumulative oxic CO_2 production was observed in the limed treatments compared with the control ($p < 0.05$), whereas liming did not increase CO_2 production in the Ap1 horizon (Table 2). Compared to the control treatment, the amounts of cumulative CO_2 production were 90 % and 40 % higher in the Bg1 horizon, 220 % and 230 % higher in the BC horizon, and 40 % and 30 % higher in the C horizon, respectively (Table 2). The cumulative CO_2 production was significantly different between all horizons in a given lime treatment, with the amounts decreasing in the order of Ap1 > C > Bg1 > BC (Table 2). There was much higher cumulative oxic CO_2 production in the Bg1, BC and C horizons of AS soil compared to the respective horizons of the non-AS soil in a given lime treatment (Table 2).

3.3. Anoxic N_2O production, total denitrification and mineral N contents during Phase Anoxic II

In the AS soil, significant differences in the cumulative anoxic N_2O production were observed between all horizons (Table 3). The low lime and high lime treatments decreased the cumulative anoxic N_2O production by 86 % and 94 % in the Ap1 horizon, by 91 % and 78 % in the Bg1 horizon, but increased it in the BC and C horizons by a factor of 3 and 2, respectively (Table 3). In the non-AS soil, the lime treatments significantly decreased the cumulative anoxic N_2O production in all horizons (Table 3). The cumulative anoxic N_2O production in the low and high lime treatments of Ap1, Bg1, BC and C horizons were only 34 % and 14 % (Ap1), 31 % and 33 % (Bg1), 37 % and 24 % (BC), and 43 % and 81 % (C), respectively, of those in the control treatment (Table 3).

The lime treatments significantly increased the total anoxic denitrification ($N_2O + N_2$ production) in all horizons of the AS soil (Table 3). The total denitrification ($N_2O + N_2$) in the low lime and high lime treatments of the Ap1, Bg1, BC and C horizons were 0.9 and 1.7 times higher (Ap1), 6 and 9 times higher (Bg1), 1.4 and 4.7 times higher (BC), and 28 and 64 times higher (C), respectively, than those in the control treatment (Table 3). Significant differences in the total denitrification ($N_2O + N_2$) were observed between the control and lime treatments of the non-AS soil as well (Table 3). In the non-AS soil, the total denitrification ($N_2O + N_2$) in the low and high lime treatments of Ap1 and C horizons were 140 % and 150 % higher (Ap1), and 30 % and 80 % higher (C), than those in the control. In the Bg1 and BC horizons, however, the lime treatments decreased total denitrification or had no effect (Table 3). The cumulative anoxic N_2O production and total denitrification ($N_2O + N_2$) were higher in the Ap1 horizon compared with all other horizons in all treatments ($p < 0.05$) (Table 3). The total denitrification ($N_2O + N_2$) in the Ap1 horizon was higher than that in all other horizons in all treatments of both AS and non-AS soils ($p < 0.05$) (Table 3). Lime treatments significantly decreased the product ratio of denitrification ($N_2O / N_2O + N_2$) in all horizons of both the AS soil and non-AS soil (Table 3).

During Phase Anoxic II, NO_3^--N content decreased in all treatments and horizons of AS and non-AS soils regardless of C_2H_2 addition (Table S6). In the AS soil, NH_4^+-N content mostly increased in all treatments and horizons both with and without C_2H_2 (Table S6). In the non-AS soil, NH_4^+-N content generally decreased in the Ap1 and Bg1 horizons, and increased in the BC and C horizons, in all treatments regardless of C_2H_2 addition (Table S6).

3.4. Anoxic CO_2 production during Phase Anoxic II

In the AS soil, the low and high lime treatments increased the cumulative anoxic CO_2 production in all horizons ($p < 0.05$) (Table S9). The cumulative CO_2 production in the low lime and high lime treatments of Ap1, Bg1, BC and C horizon were 9 % and 12 % higher (Ap1), 140 % and 160 % higher (Bg1), 28 % and 33 % higher (BC), and 90 % and 80 % higher (C), respectively, than that in the control treatment (Table S9).

In the non-AS soil, the lime treatments increased the cumulative anoxic CO_2 production in the Bg1 and C horizons ($p < 0.05$), but no significant increases were observed in the Ap1 and BC horizons (Table S9). The cumulative anoxic CO_2 production in the low and high lime treatments in the Bg1
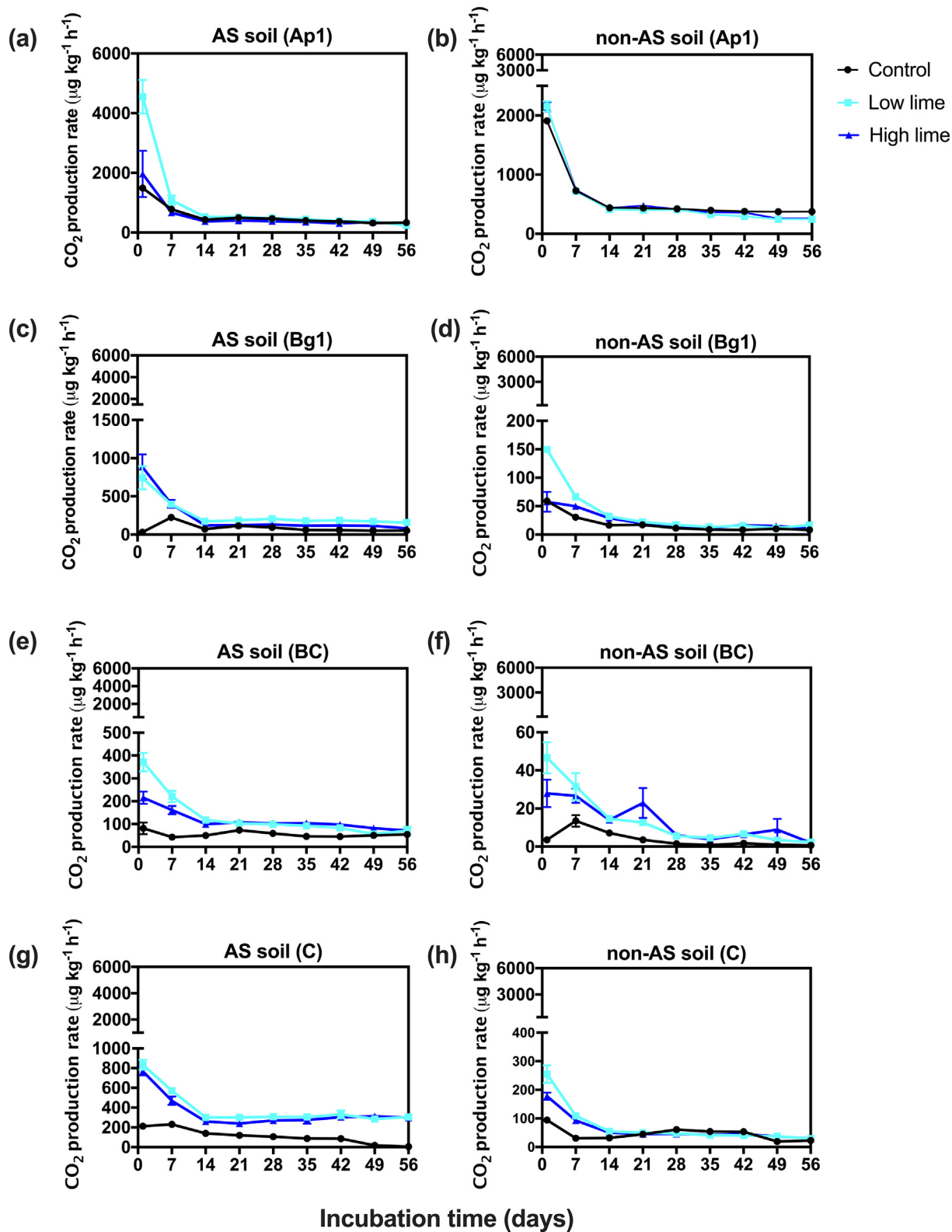
Fig. 4. Oxic CO_2 production rate (μg CO_2-C kg^{-1} h^{-1}) in Ap1 (a, b), Bg1 (c, d), BC (e, f) and C (g, h) horizons in the AS and non-AS soils during Phase Oxic I (56 days). Values are means and their standard errors (n = 4).

horizon were 26 % and 23 % higher, and those in the Bg1 horizon 70 % and 50 % higher, respectively, than those in the control treatment (Table S9). The cumulative CO_2 production in the Ap1 horizon was significantly higher than that in all other horizons in both the AS and non-AS soils (Table S9).

3.5. Oxic and anoxic GHG production based on CO_2-eq

During Phase Oxic I, the total oxic GHG production (N_2O + CO_2 as CO_2-eq) in the lime treatments was lower than that in the unlimed control

Table 3
Cumulative N_2O production, total denitrification and the product ratio of denitrification in the different lime treatments and horizons of AS and non-AS soils during Phase Anoxic II (72 h).

AS soil	Horizon	Control	Low lime	High lime
Cumulative N_2O production (μg N_2O-N kg^{-1})	Ap1	183.9[aA]	25.4[bA]	11.8[cB]
	Bg1	74.1[aB]	6.7[cB]	16.3[bA]
	BC	3.1[cC]	5.3[bB]	18.8[aA]
	C	0.8[bD]	1.2[cC]	2.1[aC]
Total denitrification ($N_2O + N_2$) (μg N_2O-N kg^{-1})	Ap1	2345[cA]	4343[bA]	6206[aA]
	Bg1	365[cB]	2544[bB]	3786[aB]
	BC	117[cC]	283[bD]	667[aD]
	C	15[cD]	435[bC]	975[aC]
Product Ratio of Denitrification ($N_2O/N_2O + N_2$)	Ap1	0.08[aB]	0.005[bB]	0.002[cC]
	Bg1	0.20[aB]	0.003[bC]	0.004[bB]
	BC	0.03[aC]	0.02[bA]	0.03[aA]
	C	0.05[aC]	0.002[bC]	0.002[bC]
Non-AS soil				
Cumulative N_2O production (μg N_2O-N kg^{-1})	Ap1	156.1[aA]	53.1[bA]	22.3[cA]
	Bg1	24.9[aC]	7.7[bC]	8.3[bB]
	BC	35.8[aB]	13.3[bB]	8.7[cB]
	C	5.6[aD]	2.4[bD]	4.5[aC]
Total denitrification ($N_2O + N_2$) (μg N_2O-N kg^{-1})	Ap1	1150[cA]	2714[bA]	2896[aA]
	Bg1	375[aB]	268[bB]	362[aB]
	BC	77[aC]	34[bC]	82[aC]
	C	20[cD]	27[bD]	36[aD]
Product Ratio of Denitrification ($N_2O/N_2O + N_2$)	Ap1	0.14[aC]	0.02[bC]	0.008[cC]
	Bg1	0.07[aD]	0.03[bC]	0.02[cC]
	BC	0.47[aA]	0.40[aA]	0.11[bA]
	C	0.28[aB]	0.09[bB]	0.13[bA]

Different small letters within each row represent significant differences between the lime treatments. Different capital letters within each column represent significant differences between different soil horizons (one-way ANOVA, Tukey test, $p < 0.05$; n = 4).

treatment, but only in the Ap1 horizon of AS soil ($p < 0.05$) (Fig. 5a). In the non-AS soil, the amount of total GHG production was not different between the lime treatments in most horizons ($p > 0.05$) (Fig. 5c), but liming decreased the proportion of N_2O in the GHG production in most horizons at both sites ($p < 0.05$) (Fig. 5e, g).

During Phase Anoxic II, the total anoxic GHG production (as CO_2-eq) in the lime treatments was significantly lower than that in the unlimed control in the Ap1 horizon of the AS soil (Fig. 5b), and in the Ap1 and BC horizons of the non-AS soil (Fig. 5d). Liming also significantly decreased the proportion of N_2O in the total GHG production in the Ap1, Bg1 horizons of AS soil (Fig. 5f), and in all horizons of the non-AS soil (Fig. 5h).

4. Discussion

4.1. Effects of liming on the oxic production of N_2O

Liming of soil to pH 7 or above clearly decreased the cumulative oxic production of N_2O in the Ap1, Bg1 and BC horizons in the AS soils. The acidic conditions represented a constraint on the reduction of N_2O to N_2 (Qu et al., 2014; Šimek et al., 2002; Thomsen et al., 1994) in the Ap1, Bg1 and BC horizons of the AS soil where the initial soil pH was in the range of 3.4 and 5.6. For this reason, the decrease of oxic N_2O production by liming can be mainly attributed to higher soil pH which increases the activity of N_2O reductase and thus causes a more rapid conversion of N_2O to N_2 (Guo et al., 2014). The results also agree with those by Shaaban et al. (2019) and Žurovec et al. (2021) who reported that liming can greatly decrease N_2O production and emissions through increased soil pH.

Contrary to the other AS soil horizons, liming was inefficient in decreasing the oxic N_2O production in the C horizon, where liming instead increased N_2O production. This can be explained by enhanced microbial activity, as indicated by the increased CO_2 production by liming, which likely also increased N_2O production by both nitrification and denitrification. In addition, soil structure and texture are also known to influence

N_2O emissions (Chapuis-Lardy et al., 2007). Due to the massive structure of C horizon, it is possible that the added lime suspension was distributed less evenly into contact with the soil in the C horizon than in the other horizons which had more developed, aggregated soil structure. Moreover, at the time of the highest N_2O emissions at the beginning of oxic incubation, the soil pH of 6.5 in the C horizon of the unlimed treatment was similar to the soil pH in the limed treatments of Ap1 horizon, and only decreased to low pH levels later during the oxic incubation. These factors may have decreased the efficiency of liming to mitigate the oxic production of N_2O in the C horizon of the AS soil.

In the Ap1 and Bg1 horizons of the AS soil, the initial large amounts of NO_3^--N (27.8 mg kg^{-1} in the Ap1 and 7.7 mg kg^{-1} in the Bg1) may have originated from the mineralization of soil organic matter and nitrification (Yli-Halla et al., 2020). The decreasing N_2O production rate in all horizons and treatments of the AS soil over time during Phase Oxic I was probably due to the gradual depletion of NO_3^--N. We attributed this decrease in NO_3^--N content in the Ap1 and Bg1 horizons to denitrification within anaerobic microsites of soil aggregates, which have been demonstrated to occur frequently, even in apparently oxic soils (Stevens et al., 1997), and to the immobilization of N (Case et al., 2015) into the microbial biomass, as there was no leaching from the incubation vessels. Nitrification and denitrification are known to occur concurrently in aerobic and anaerobic microsites in well structured soils (Ball, 2013). N_2O can be produced by nitrification, when the supply of O_2 is limited by diffusional constraints and the nitrifying bacteria reduce NO_2^--N to N_2O (Bollmann and Conrad, 1998), as well as by denitrification in anoxic microsites at high soil moisture contents (> 70 % WPFS) (Dobbie and Smith, 2001).

In addition, microbial immobilization of N can decrease the amount of NO_3^--N and NH_4^+-N in soil and convert them into organic forms of cellular components, such as proteins and amino sugars (Brady and Weil, 2016). As liming increased microbial activity (CO_2 production) in the limed AS soil horizons at the beginning of Phase Oxic I, the enhanced microbial growth could have caused some immobilization and contributed to the decrease of NO_3^--N as well. However, immobilization of N into microbial biomass did not cause net depletion of NH_4^+-N in soil as the net ammonification and net N mineralization during Phase Oxic I were positive, most likely due to high decomposition of organic matter in oxic conditions.

In the BC and C horizons of AS soil, the pool of NO_3^--N increased with time. The initial amounts of NH_4^+-N in the BC and C horizons were very high, and the large increases in NH_4^+-N in all horizons during Phase Oxic I indicated mineralization of organic N to NH_4^+. Oxic conditions promoted oxidation of NH_4^+-N to NO_3^--N by nitrification that likely also produced some N_2O as a by-product. Nevertheless, the fact that liming significantly decreased N_2O production suggested that the pH-sensitive N_2O reductase enzyme played an important role, and that N_2O was likely produced in anoxic microsites by coupled nitrification and denitrification (Ball, 2013; Šimek et al., 2011; Smith et al., 2003). As aerobic and anaerobic zones can develop even within the same aggregate in soil, coupled nitrification/denitrification can become the main contributor to N_2O production at high soil moisture contents (Kremen et al., 2005; Stevens et al., 1997).

The AS soil had a much higher cumulative oxic N_2O production compared with the non-AS soil in nearly all soil horizons and lime treatments, which can be attributed to its higher contents of mineral N (NO_3^--N + NH_4^+-N). It was only in the unlimed C horizon of the non-AS soil where the cumulative oxic N_2O production was larger than that in the AS soil. Slightly higher bulk density, pH and NO_3^--N content in the non-AS soil compared with AS soil may have contributed to this exception, even if the underlying reasons remain uncertain and a possible topic of future research. Nevertheless, our results are generally in agreement with recent studies that have indicated larger N_2O emissions from AS soil than from non-AS mineral soils in Finland (Šimek et al., 2011; Šimek et al., 2014; Yli-Halla et al., 2020) and Australia (Galbally et al., 2010; Macdonald et al., 2008). Our results support the view that NO_3^--N and NH_4^+-N were important factors affecting oxic N_2O production and emissions from AS soils through nitrification and denitrification.
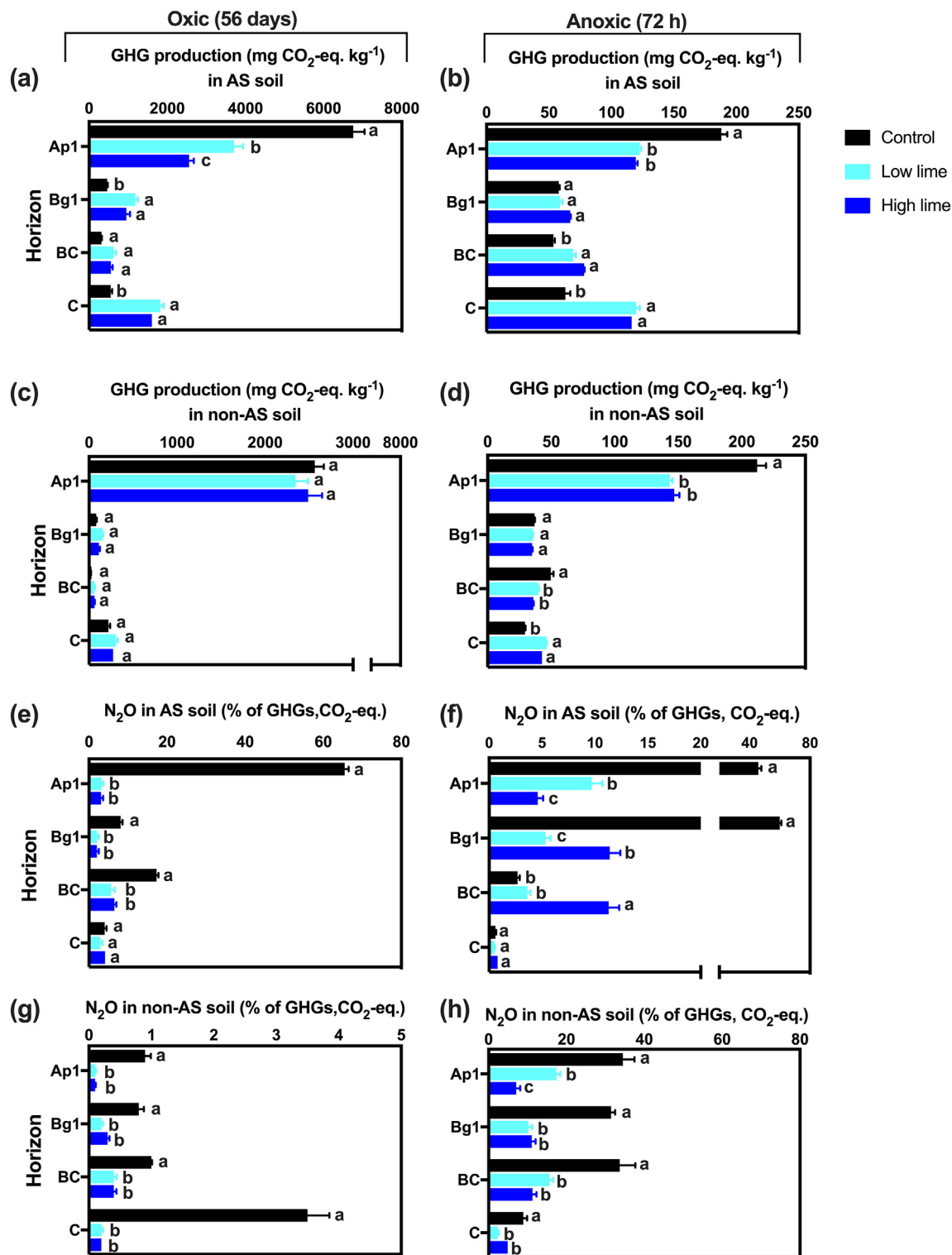
Fig. 5. GHG production (mg CO_2-eq. kg^{-1}) (a, c) and the proportion of N_2O-N in GHG production (% of GHGs, CO_2-eq.) in the different horizons and lime treatments of AS and non-AS soils (e, g) during Phase Oxic I (56 d). GHG production (mg CO_2-eq. kg^{-1}) (b, d) and the proportion of N_2O-N in GHG production (% of GHGs, CO_2-eq.) in the different horizons and lime treatments of AS and non-AS soils (f, h) during Phase Anoxic II (72 h). Different small letters within each horizon indicate significant differences between the lime treatments (one-way ANOVA, Tukey test, $p < 0.05$; n = 4). Values are the means and their standard errors (n = 4).

4.2. Effects of liming on the anoxic production of N_2O production and denitrification

Lime treatments decreased the anoxic production of N_2O in the Ap1 and Bg1 horizons of the AS soil, and in all horizons of the non-AS soil. However, exceptions were the BC and C horizons of AS soil. In these subsoil horizons,

liming proved inefficient in decreasing anoxic N_2O production but instead increased it. In the low lime treatment of the BC horizon, the results could be partly explained by the fact that the lime addition was too low to efficiently counteract the acidification of soil during Phase Oxic I. By the beginning of Phase Anoxic II, soil pH had decreased to 5. Low soil pH

may thus have limited the reduction from N_2O to N_2, which may partly explain why liming was not efficient to reduce anoxic N_2O production by liming in the low lime treatment in the BC horizon. Similarly, the massive structure of C horizon may have constrained the even contact of added lime with soil and thus reduced the lime efficiency. This was not the case for the high lime treatment of the BC horizon, where soil structure was not massive and the soil pH remained above 7 in Phase Anoxic II. However, this does not explain why liming increased the anoxic N_2O production in the BC and C horizons, which is discussed in more detail below.

The net N_2O production by denitrification is determined by the rate of total denitrification and the share of N_2O in the end products (the product ratio of denitrification). Even if total denitrification activity increases, the amount of N_2O produced relative to N_2 usually decreases with increasing soil pH (Šimek and Cooper, 2002). Thus, any liming-induced increase in the N_2O production, such as that in the BC and C horizons of the AS soil during Phase Anoxic II, indicates that liming induced a larger increase in the total denitrification than in the N_2O reduction. In contrast, any liming-induced decrease in the N_2O production indicates that the increase in the N_2O reduction was larger than that of total denitrification.

In the AS soil, both lime treatments decreased the anoxic product ratio of denitrification $N_2O/(N_2O + N_2)$ in all horizons of AS soils, except for the high lime treatment in the BC horizon. This supports the view that higher soil pH stimulated the activity of N_2O-reductase (Čuhel and Šimek, 2011) and agrees with the currently established view that the decrease of product ratio $N_2O/(N_2O + N_2)$ with increasing pH can be explained mainly by the pH-sensitivity of N_2O reductase enzyme. This enzyme is known to be inhibited by low pH (Liu et al., 2010; Qu et al., 2014; Šimek et al., 2002; Thomsen et al., 1994). Similarly, Liu et al. (2010) showed a strong negative correlation between soil pH and $N_2O/(N_2O + N_2)$ within a pH range of 4–7 in long-term lime experiments in peat and clay loam soils.

Soil pH not only affected the product ratio of denitrification but also the rate of total denitrifying activity by controlling the composition and activity of the denitrifying microbial community. Low soil pH limits the availability of organic C and mineral N to denitrifying bacteria (Bakken et al., 2012; McMillan et al., 2016), thus reducing their growth and activity.

In all horizons of the AS soil, liming increased total anoxic denitrification ($N_2O + N_2$) compared to the respective horizon of the unlimed control soil. In the non-AS soil, however, this only occurred in the Ap1 horizon with the lowest soil pH (6.3). The initially high soil pH (above 7.2) probably decreased the lime effects on $N_2O + N_2$ in the Bg1, BC and C horizons of the non-AS soil. Liming had raised the soil pH to the range of 7.0–8.0, the range at which total denitrification ($N_2O + N_2$) is generally considered to be the highest (Van Cleemput et al., 1975).

Total cumulative anoxic denitrification ($N_2O + N_2$) in the AS soil was always higher than that in the respective horizon of the non-AS soil irrespective of the lime treatments. This can be attributed to the higher total N and initial mineral N (NO_3^--N and NH_4^+-N) contents in the AS soil horizons compared with the non-AS soil. Large amounts of bioavailable N are known to favour denitrification and enhance the production of N_2O and N_2 under anaerobic conditions. Earlier studies on N_2O in AS soils have indicated that the presence of large amounts of NO_3^--N and NH_4^+-N in AS soils can cause large emissions of N_2O under anaerobic conditions (Čuhel and Šimek, 2011; Denmead et al., 2010; Denmead et al., 2011).

4.3. Effects of liming on the oxic and anoxic production of CO_2

Liming significantly increased the cumulative CO_2 production compared to the unlimed control in the AS soil in both oxic and anoxic conditions. Part of the CO_2 may have originated directly from the conversion of carbonates in lime ($CaCO_3$) to CO_2 with neutralization. Page et al. (2009) and Grover et al. (2017) also reported that raising the soil pH with the dissolution of lime can concurrently contribute to the release of CO_2 from soils. In our study, however, nearly all added lime had reacted in the soil by the end of the oxic and anoxic incubations, as the fractional conversion of carbonates always exceeded 90 % (Table S2), but even the largest differences in the total cumulative CO_2 production between the limed and

control treatment (about 340 mg CO_2-C kg^{-1}) were only a small fraction of the amount of added carbonate C (1500 or 3000 mg CO_2-C kg^{-1}). Thus, during the incubation, much of the dissolved carbonate must have remained in the soil. In the limed treatments, most of the added carbonates probably remained in porewater as dissolved bicarbonate, which is the dominant carbonate species in the circumneutral pH range.

Liming may have also increased the availability of organic carbon and nitrogen to the microbes which were initially limited by low soil pH in the AS soil (Zelles et al., 1990). Lime additions increased the biological activity in soil and therefore the CO_2 production by soil respiration in the same manner as in other studies (Badalucco et al., 1992; Biasi et al., 2008). In the non-AS soil, however, the effects of liming on the cumulative CO_2 production were less pronounced as the initial pH of non-AS soil was already above 6.3 in all horizons.

There was a significant difference in the cumulative oxic production of CO_2 between all horizons irrespective of lime treatment in the AS soil (Table 2). The oxic CO_2 production was highest in the Ap1 horizon, decreased rapidly with soil depth in the BC horizon, but then increased again in the C horizon. This confirmed the pattern observed in an earlier study with the AS soil from the same site (Šimek et al., 2011). Abundant carbon stocks in the top horizon (Ap1) and deep horizon (C) provided easily available substrates for enhanced microbial growth and respiration under oxic conditions. This agreed with many earlier studies where high amounts of organic carbon increased production of CO_2 in soil (Bruce et al., 1997; Gallardo and Schlesinger, 1994). The lowest oxic production of CO_2 occurred in the BC horizon due to the low soil pH (3.4) that limited microbial activity and CO_2 production. The cumulative oxic CO_2 production in the subsoil horizons was higher in the AS soil than in the non-AS soil, which can be attributed to the higher carbon stocks in the AS soil.

4.4. Effects of liming on the total GHG production based on CO_2-eq

Proper evaluation of the atmospheric impacts of pH mitigation on the total GHG production from soil in this study required taking into account the different global warming potentials of GHGs and summing up the production of all individual GHGs (as CO_2-eq). Increasing soil pH by liming caused opposite but varied effects on the oxic and anoxic GHG production in the different soils and horizons of our study. Liming enhanced reduction of N_2O to N_2, decreased the anoxic product ratio of denitrification $N_2O/(N_2O + N_2)$, which tended to decrease the net production of N_2O, but this was counteracted to varying extents by concurrently increased microbial activity and denitrification, which tended to increase the total CO_2 and N_2O production. Contrasting responses of GHG production to liming were found in different horizons of both AS and non-AS soils.

In terms of CO_2-eq, liming caused a net reduction in the oxic and anoxic production of GHGs only in the Ap1 horizons of both soils, and in the anoxic production of GHGs in the BC horizon of the non-AS soil, even if liming nearly always decreased the proportion of N_2O in GHG production, excluding the C horizon and anoxic BC horizon in AS soil.

Conversely, liming of acid subsoil horizons often increased the GHG production. In the BC horizon of the AS soil, liming concomitantly increased both anoxic GHG and N_2O production as well as the proportion of N_2O in the GHGs produced, whereas the product ratio of denitrification was almost unchanged. This result differed from all other horizons and may suggest a contribution of a different mechanisms of GHG production compared with the other horizons. It should be noted that in an earlier study, the highest anoxic N_2O production of all AS subsoil horizons was found in the BC horizon (Šimek et al., 2014). The mechanisms involved require further research.

The results suggest that liming can successfully reduce GHG production in AS soils by pH mitigation, provided that the topsoil dominates GHG production of the soil profile. Nevertheless, even if liming of acid AS subsoil horizons could reduce the proportion of N_2O in GHG production, it seems less efficient in reducing the total GHG production and involves the risk of increasing the GHG production by enhanced microbial respiration and

denitrification (Goulding, 2016; Wu et al., 2020). In the non-AS subsoils, high initial pH probably reduced the effects of liming.

In our incubations, we used mechanically disturbed soil samples at a constant soil moisture status (70 % WFPS) and at a higher temperature than typical in the field. The conditions were selected so as to have optimal conditions for oxic N_2O production and for its mitigation by liming. In the field, however, soil moisture content varies with time and soil depth. The degree of saturation may often exceed WFPS 70 % in the BC horizon and approach saturation in the C horizon of AS soil. This may increase the contribution of subsoil horizons to total GHG production in the soil profile.

On the other hand, the field temperatures in the subsoil horizons are much lower than the room temperature in this study, so that microbial activity and GHG production should be correspondingly slower in the field. Time-series of soil temperature data at our study sites were unavailable, but the subsoil temperatures were available in a nearby clay loam soil (Field Observatory, 2022), where the mean temperature at 100 cm was 6.7 °C (min −0.1 °C, max 14.9 °C; 1 Jun 2021–31 May 2022). According to Yli-Halla and Mokma (1998), Finnish soils mainly belong to the cryic temperature regime in Soil Taxonomy with the mean annual soil temperatures at the depth of 50 cm ranging from 6.4 °C at the warmest sites to 1.9 °C at the coldest sites. The mean annual soil temperatures are 2 °C to 5 °C higher than mean annual air temperature and related to snow cover (Yli-Halla and Mokma, 1998). Ground frost occurs typically from November/December to April/May, with the average maximum depth of ground frost of about 20 cm in southern Finland (Finnish Environment Institution, 2022).

Moreover, if lime suspension is added into a structured subsoil through subsurface drains in the field (Wu et al., 2015), such lime will only contact the surfaces of the largest macropores and cracks in the subsoil. The efficiency of such liming on CO_2 and N_2O production in different soil horizons and the contributions of different soil horizons to GHG emissions *in-situ* remain areas for future research.

5. Conclusions

In this study, liming nearly always decreased oxic and anoxic cumulative N_2O production in all horizons in the AS soil, except for the BC horizon in anoxic conditions and the C horizon in oxic and anoxic conditions. Liming decreased $N_2O/(N_2O + N_2)$ in all horizons of both AS and non-AS soils under anoxic conditions and supported the view that high soil pH stimulated the activity of N_2O-reductase. The higher cumulative oxic and anoxic production of N_2O and CO_2 in the horizons of AS soil as compared to those in the non-AS soil agreed with their higher total carbon and nitrogen contents. Significant lime-induced decreases in the total GHG production (as CO_2-eq) were observed only in the Ap1 horizon of AS (oxic and anoxic) and the Ap1 and BC horizons of non-AS (anoxic) soils. The results suggest that liming can reduce GHG production efficiently in topsoil by decreasing the product ratio of denitrification $N_2O/(N_2O + N_2)$ and, but not in subsoil. Liming of subsoil horizons would not effectively decrease GHG production as it may increase CO_2 production and total denitrification more than the reduction of N_2O, and can thus even lead to increases in the total GHG production. In the future, long-term field studies are needed to evaluate the potential of liming of different soil horizons as a pH-mitigating tool for N_2O production and facilitating the reduction of product ratio $N_2O/(N_2O + N_2)$ *in-situ*. At the same time, the viability of lime to mitigate GHG emissions should also account for the increased CO_2 emissions with lime application on agricultural land.

CRediT authorship contribution statement

Chang Xu: Conceptualization, Formal analysis, Funding acquisition, Methodology, Investigation, Visualization, Writing – original draft. **Vanessa N.L. Wong:** Conceptualization, Methodology, Writing – review & editing. **Anna Tuovinen:** Resources, Methodology, Writing – review & editing. **Asko Simojoki:** Conceptualization, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing – review & editing.

Data availability

Data will be made available on request.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.159505.

References

Amundson, R.G., Davidson, E.A., 1990. Carbon dioxide and nitrogenous gases in the soil atmosphere. J. Geochem. Explor. 38, 13–41.

Andriesse, W., Mensvoort, M.E.F.v., 2006. Acid sulfate soils: distribution and extent. In: Lal, R. (Ed.), Encyclopedia of Soil Science. CRC Press, Boca Raton, FL, pp. 14–19.

Åström, M., Spiro, B., 2000. Impact of isostatic uplift and ditching of sulfidic sediments on the hydrochemistry of major and trace elements and sulfur isotope ratios in streams, Western Finland. Environ. Sci. Technol. 34, 1182–1188.

Backlund, K., Boman, A., Fröjdö, S., Åström, M., 2005. An analytical procedure for determination of sulphur species and isotopes in boreal acid sulphate soils and sediments. Agric. Food Sci. 14, 70–82.

Badalucco, L., Grego, S., Dell'Orco, S., Nannipieri, P., 1992. Effect of liming on some chemical, biochemical, and microbiological properties of acid soils under spruce (Picea abies L.). Biol. Fertil. Soils 14, 76–83.

Bakken, L.R., Bergaust, L., Liu, B., Frostegård, A., 2012. Regulation of denitrification at the cellular level: a clue to the understanding of N2O emissions from soils. Philos. Trans. R Soc. Lond. Ser. B Biol. Sci. 367, 1226–1234.

Ball, B.C., 2013. Soil structure and greenhouse gas emissions: a synthesis of 20 years of experimentation. Eur. J. Soil Sci. 64, 357–373.

Bateman, E.J., Baggs, E.M., 2005. Contributions of nitrification and denitrification to N2O emissions from soils at different water-filled pore space. Biol. Fertil. Soils 41, 379–388.

Biasi, C., Lind, S.E., Pekkarinen, N.M., Huttunen, J.T., Shurpali, N.J., Hyvönen, N.P., Repo, M.E., Martikainen, P.J., 2008. Direct experimental evidence for the contribution of lime to CO2 release from managed peat soil. Soil Biol. Biochem. 40, 2660–2669.

Bollmann, A., Conrad, R., 1998. Influence of O2 availability on NO and N2O release by nitrification and denitrification in soils. Glob. Chang. Biol. 4, 387–396.

Boman, A., Åström, M., Fröjdö, S., 2008. Sulfur dynamics in boreal acid sulfate soils rich in metastable iron sulfide—the role of artificial drainage. Chem. Geol. 255, 68–77.

Boman, A., Fröjdö, S., Backlund, K., Åström, M.E., 2010. Impact of isostatic land uplift and artificial drainage on oxidation of brackish-water sediments rich in metastable iron sulfide. Geochim. Cosmochim. Acta 74, 1268–1281.

Brady, N.C., Weil, R.R., 2016. The Nature and Properties of Soils, Fifteenth edition Prentice Hall, Upper Saddle River, N.J, Upper Saddle River, N.J., pp. 1–1104.

Bruce, A.H., Elisabeth, A.H., Robert, B.J., Chapin, F.S., Harold, A.M., Christopher, B.F., 1997. The fate of carbon in grasslands under carbon dioxide enrichment. Nature 388, 576–579.

Burton, E.D., Bush, R.T., Sullivan, L.A., Johnston, S.G., Hocking, R.K., 2008. Mobility of arsenic and selected metals during re-flooding of iron- and organic-rich acid-sulfate soil. Chem. Geol. 253, 64–73.

Case, S.D.C., McNamara, N.P., Reay, D.S., Stott, A.W., Grant, H.K., Whitaker, J., 2015. Biochar suppresses N2O emissions while maintaining N availability in a sandy loam soil. Soil Biol. Biochem. 81, 178–185.

Cavigelli, M.A., Robertson, G.P., 2001. Role of denitrifier diversity in rates of nitrous oxide consumption in a terrestrial ecosystem. Soil Biol. Biochem. 33, 297–310.

Chapuis-Lardy, L., Wrage, N., Metay, A., Chotte, J.-L., Bernoux, M., 2007. Soils, a sink for N2O? A review. Glob. Chang. Biol. 13, 1–17.

Colbourn, P., Dowdell, R.J., 1984. Denitrification in field soils. Plant Soil 76, 213–226.

Čuhel, J., Šimek, M., 2011. Proximal and distal control by pH of denitrification rate in a pasture soil. Agric. Ecosyst. Environ. 141, 230–233.

Dalhem, K., Engblom, S., Stén, P., Österholm, P., 2019. Subsurface hydrochemical precision treatment of a coastal acid sulfate soil. Appl. Geochem. 100, 352–362.

Davidson, E.A., Belk, E., Boone, R.D., 1998. Soil water content and temperature as independent or confounded factors controlling soil respiration in a temperate mixed hardwood forest. Glob. Chang. Biol. 4, 217–227.

Davidson, E.A., Keller, M., Erickson, H.E., Verchot, L.V., Veldkamp, E., 2000. Testing a conceptual model of soil emissions of nitrous and nitric oxides. Bioscience 50, 667–680.

Denmead, O.T., Macdonald, B.C.T., Bryant, G., Naylor, T., Wilson, S., Griffith, D.W.T., Wang, W.J., Salter, B., White, I., Moody, P.W., 2010. Emissions of methane and nitrous oxide from Australian sugarcane soils. Agric. For. Meteorol. 150, 748–756.

Denmead, O.T., Macdonald, B.C.T., White, I., Byrant, G., 2011. Gaseous nitrogen losses from coastal acid sulfate soils: a short-term study. Pedosphere 21, 197–206.

Dobbie, K.E., Smith, K.A., 2001. The effects of temperature, water-filled pore space and land use on N2O emissions from an imperfectly drained gleysol. Eur. J. Soil Sci. 52, 667–673.

Esala, M.J., 1995. Changes in the extractable ammonium- and nitrate-nitrogen contents of soil samples during freezing and thawing. Commun. Soil Sci. Plant Anal. 26, 61–68.

Field Observatory, 2022. https://www.fieldobservatory.org/en/online-field-data/?site=haltiala. accessed 28 Aug 2022.

Finnish Environment Institution, 2022. http://wwwi3.ymparisto.fi/i3/paasivu/ENG/Routa/Routa.htm. accessed 28 Aug 2022.

Galbally, I.E., Meyer, M.C.P., Wang, Y.-P., Smith, C.J., Weeks, I.A., 2010. Nitrous oxide emissions from a legume pasture and the influences of liming and urine addition. Agric. Ecosyst. Environ. 136, 262–272.

Gallardo, A., Schlesinger, W.H., 1994. Factors limiting microbial biomass in the mineral soil and forest floor of a warm-temperate forest. Soil Biol. Biochem. 26, 1409–1415.

Gatland, J.R., Santos, I.R., Maher, D.T., Duncan, T.M., Erler, D.V., 2014. Carbon dioxide and methane emissions from an artificially drained coastal wetland during a flood: implications for wetland global warming potential. J. Geophys. Res. Biogeosci. 119, 1698–1716.

Goulding, K.W.T., 2016. Soil acidification and the importance of liming agricultural soils with particular reference to the United Kingdom. Soil Use Manag. 32, 390–399.

Grover, S.P., Butterly, C.R., Wang, X., Tang, C., 2017. The short-term effects of liming on organic carbon mineralisation in two acidic soils as affected by different rates and application depths of lime. Biol. Fertil. Soils 53, 431–443.

Guo, X., Xueming, Y., Drury, C.F., Reynolds, W.D., Fan, R., 2014. The extent of soil drying and rewetting affects nitrous oxide emissions, denitrification, and nitrogen mineralization. (Report). Soil Sci. Soc. Am. J. 78, 194–204.

Hénault, C., Bourennane, H., Ayzac, A., Ratié, C., Saby, N.P.A., Cohan, J.-P., Eglin, T., Gall, C.L., 2019. Management of soil pH promotes nitrous oxide reduction and thus mitigates soil emissions of this greenhouse gas. Sci. Rep. 9, 20182.

Inubushi, K., Naganuma, H., Kitahara, S., 1996. Contribution of denitrification and autotrophic and heterotrophic nitrification to nitrous oxide production in andosols. Biol. Fertil. Soils 23, 292–298.

Kowalchuk, G., Stephen, J., 2001. Ammonia-oxidizing bacteria: a model for molecular microbial ecology. Annu. Rev. Microbiol. 55, 485–529.

Kremen, A., Bear, J., Shavit, U., Shaviv, A., 2005. Model demonstrating the potential for coupled nitrification denitrification in soil aggregates. Environ. Sci Technol. 39, 4180–4188.

Liu, B., Mørkved, P.T., Frostegård, Å., Bakken, L.R., 2010. Denitrification gene pools, transcription and kinetics of NO, N2O and N2 production as affected by soil pH. FEMS Microbiol. Ecol. 72, 407–417.

Liu, B., Frostegård, Å., Bakken, L.R., 2014. Impaired reduction of N2O to N2 in acid soils is due to a posttranscriptional interference with the expression of nosZ. MBio 5, e01314–e01383.

Ljung, K., Maley, F., Cook, A., Weinstein, P., 2009. Acid sulfate soils and human health—a millennium ecosystem assessment. Environ. Int. 35, 1234–1242.

Macdonald, B., Denmead, O., White, I., Wilson, S., Griffith, D.W.T., Bryant, G., Naylor, T., Wang, W., Moody, P., 2008. Greenhouse gas emissions from acid sulphate and non-acid sulphate canelands. Proceedings of the Joint Conference of the 6th International Acid Sulphate Soil Conference and the Acid Rock Drainage Symposium Guangzhou edition. Guangdong Press Group, Guangzhou, China, pp. 126–131.

Macdonald, B.C.T., White, I., Denmead, O.T., 2010. Gas emissions from the interaction of iron, sulfur and nitrogen cycles in acid sulfate soils. In: Gilkes, R.J., Prakongkep, N. (Eds.), 19th World Congress of Soil Science, Soil solutions for a changing world. 1 - 6 August 2010, Brisbane, Australia. International Union of Soil Science, Wien, Austria, pp. 80–83. https://www.iuss.org/19th%20WCSS/Symposium/pdf/0975.pdf.

McMillan, A.M.S., Pal, P., Phillips, R.L., Palmada, T., Berben, P.H., Jha, N., Saggar, S., Luo, J., 2016. Can pH amendments in grazed pastures help reduce N2O emissions from denitrification? – the effects of liming and urine addition on the completion of denitrification in fluvial and volcanic soils. Soil Biol. Biochem. 93, 90–104.

Mokma, D., Yli-Halla, M., Hartikainen, H., 2000. Soils in a young landscape on the coast of southern Finland. Agric. Food Sci. Finland 9, 291–302.

Mosley, L.M., Biswas, T.K., Cook, F.J., Marschner, P., Palmer, D., Shand, P., Yuan, C., Fitzpatrick, R.W., 2017. Prolonged recovery of acid sulfate soils with sulfuric materials following severe drought: causes and implications. Geoderma 308, 312–320.

Nadeem, S., Dörsch, P., Bakken, L.R., 2013. Autoxidation and acetylene-accelerated oxidation of NO in a 2-phase system: implications for the expression of denitrification in ex situ experiments. Soil Biol. Biochem. 57, 606–614.

Oertel, C., Matschullat, J., Zurba, K., Zimmermann, F., Erasmi, S., 2016. Greenhouse gas emissions from soils—a review. Chem. Erde-Geochem. 76, 327–352.

Paasonen-Kivekäs, M., Yli-Halla, M., 2005. A comparison of nitrogen and carbon reserves in acid sulphate and non acid sulphate soils in western Finland. Agric. Food Sci. 14, 57–69.

Page, K.L., Allen, D.E., Dalal, R.C., Slattery, W., 2009. Processes and magnitude of CO2, CH4, and N2O fluxes from liming of Australian acidic soils: a review. Aust. J. Soil Res. 47, 747–762.

Parkin, T.B., Sexstone, A.J., Tiedje, J.M., 1985. Adaptation of denitrifying populations to low soil pH. Appl. Environ. Microbiol. 49, 1053–1056.

Pons, L.J., 1973. Outline of the genesis, characteristics, classification and improvement of acid sulphate soils. In: Dost, H. (Ed.), Acid Sulphate Soils. Proceedings of the International Symposium on Acid Sulphate Soils. International Institute for Land Reclamation and Improvement, Wageningen, The Netherlands, pp. 13–20.

Poungparn, S., Komiyama, A., Tanaka, A., Sangtiean, T., Maknual, C., Kato, S., Tanapermpool, P., Patanaponpaiboon, P., 2009. Carbon dioxide emission through soil respiration in a secondary mangrove forest of eastern Thailand. J. Trop. Ecol. 25, 393–400.

Qu, Z., Wang, J., Almøy, T., Bakken, L.R., 2014. Excessive use of nitrogen in chinese agriculture results in high N2O/(N2O + N2) product ratio of denitrification, primarily due to acidification of the soils. Glob. Chang. Biol. 20, 1685–1698.

Rayment, G.E., Lyons, D.J., 2011. Soil chemical methods. CSIRO, Melbourne, Australia, pp. 1–482.

Rudaz, A.O., Wälti, E., Kyburz, G., Lehmann, P., Fuhrer, J., 1999. Temporal variation in N2O and N2 fluxes from a permanent pasture in Switzerland in relation to management, soil water content and soil temperature. Agric. Ecosyst. Environ. 73, 83–91.

Saggar, S., Jha, N., Deslippe, J., Bolan, N.S., Luo, J., Giltrap, D.L., Kim, D.G., Zaman, M., Tillman, R.W., 2013. Denitrification and N2O:N2 production in temperate grasslands: processes, measurements, modelling and mitigating negative impacts. Sci. Total Environ. 465, 173–195.

Samad, M.S., Biswas, A., Bakken, L.R., Clough, T.J., de Klein, C.A.M., Richards, K.G., Lanigan, G.J., Morales, S.E., 2016. Phylogenetic and functional potential links pH and N2O emissions in pasture soils. Sci. Rep. 6, 35990.

Schaufler, G., Kitzler, B., Schindlbacher, A., Skiba, U., Sutton, M.A., Zechmeister-Boltenstern, S., 2010. Greenhouse gas emissions from European soils under different land use: effects of soil moisture and temperature. Eur. J. Soil Sci. 61, 683–696.

Scott-Denton, L.E., Sparks, K.L., Monson, R.K., 2003. Spatial and temporal controls of soil respiration rate in a high-elevation, subalpine forest. Soil Biol. Biochem. 35, 525–534.

Shaaban, M., Wu, Y., Khalid, M.S., Peng, Q.-A., Xu, X., Wu, L., Younas, A., Bashir, S., Mo, Y., Lin, S., Zafar-ul-Hye, M., Abid, M., Hu, R., 2018. Reduction in soil N2O emissions by pH manipulation and enhanced nosZ gene transcription under different water regimes. Environ. Pollut. 1987 (235), 625–631.

Shaaban, M., Hu, R., Wu, Y., Younas, A., Xu, X., Sun, Z., Jiang, Y., Lin, S., 2019. Mitigation of N2O emissions from urine treated acidic soils by liming. Environ. Pollut. 1987 (255), 113237.

Šimek, M., Cooper, J.E., 2002. The influence of soil pH on denitrification: progress towards the understanding of this interaction over the last 50 years. Eur. J. Soil Sci. 53, 345–354.

Šimek, M., Jíšová, L., Hopkins, D.W., 2002. What is the so-called optimum pH for denitrification in soil? Soil Biol. Biochem. 34, 1227–1234.

Šimek, M., Virtanen, S., Krištůfek, V., Simojoki, A., Yli-Halla, M., 2011. Evidence of rich microbial communities in the subsoil of a boreal acid sulphate soil conducive to greenhouse gas emissions. Agric. Ecosyst. Environ. 140, 113–122.

Šimek, M., Virtanen, S., Simojoki, A., Chroňáková, A., Elhottová, D., Krištůfek, V., Yli-Halla, M., 2014. The microbial communities and potential greenhouse gas production in boreal acid sulphate, non-acid sulphate, and reedy sulphidic soils. Sci. Total Environ. 466–467, 663–672.

Smith, K.A., Ball, T., Conen, F., Dobbie, K.E., Rey, A., 2003. Exchange of greenhouse gases between soil and atmosphere: interactions of soil physical factors and biological processes. Eur. J. Soil Sci. 54, 779–791.

Soil Survey Staff, 2014. Keys to Soil Taxonomy. 12th ed. USDA-Natural Resources Conservation Service, Washington, DC.

Stevens, R.J., Laughlin, R.J., Burns, L.C., Arah, J.R.M., Hood, R.C., 1997. Measuring the contributions of nitrification and denitrification to the flux of nitrous oxide from soil. Soil Biol. Biochem. 29, 139–151.

Thomsen, J.K., Torben, G., Raymond, P.C., 1994. Mass spectrometric studies of the effect of pH on the accumulation of intermediates in denitrification by Paracoccus denitrificans. Appl. Environ. Microbiol. 60, 536–541.

Van Cleemput, O., Patrick, W.H., McIlhenny, R.C., 1975. Formation of chemical and biological denitrification products in flooded soil at controlled pH and redox potential. Soil Biol. Biochem. 7, 329–332.

West, T.O., McBride, A.C., 2005. The contribution of agricultural lime to carbon dioxide emissions in the United States: dissolution, transport, and net emissions. Agric. Ecosyst. Environ. 108, 145–154.

Wu, X., Sten, P., Engblom, S., Nowak, P., Österholm, P., Dopson, M., 2015. Impact of mitigation strategies on acid sulfate soil chemistry and microbial community. Sci. Total Environ. 526, 215.

Wu, H., Hao, X., Xu, P., Hu, J., Jiang, M., Shaaban, M., Zhao, J., Wu, Y., Hu, R., 2020. CO2 and N2O emissions in response to dolomite application are moisture dependent in an acidic paddy soil. J. Soils Sediments 20, 3136–3147.

Xu, C., Wong, V.N.L., Reef, R.E., 2021. Effect of inundation on greenhouse gas emissions from temperate coastal wetland soils with different vegetation types in southern Australia. Sci. Total Environ. 763, 142949.

Yli-Halla, M., Mokma, D., 1998. Soil temperature regimes in Finland. Agric. Food Sci. 7 (4), 507–512.

Yli-Halla, M., Virtanen, S., Regina, K., Österholm, P., Ehnvall, B., Uusi-Kämppä, J., 2020. Nitrogen stocks and flows in an acid sulfate soil. Environ. Monit. Assess. 192, 751.

Yoshinari, T., Hynes, R., Knowles, R., 1977. Acetylene inhibition of nitrous oxide reduction and measurement of denitrification and nitrogen fixation in soil. Soil Biol. Biochem. 9, 177–183.

Zelles, L., Stepper, K., Zsolnay, A., 1990. The effect of lime on microbial activity in spruce (Picea abies L.) forests. Biol. Fertil. Soils 9, 78–82.

Žurovec, O., Wall, D.P., Brennan, F.P., Krol, D.J., Forrestal, P.J., Richards, K.G., 2021. Increasing soil pH reduces fertiliser derived N2O emissions in intensively managed temperate grassland. Agric. Ecosyst. Environ. 311, 107319.