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Optimal lime materials for mitigating global warming potential with and without straw application in acidic upland soil

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Lime and crop straw are widely applied to mitigate soil acidification and improve soil fertility. However, how different lime materials interact with straw to influence greenhouse gas (GHG) emissions from acidic upland soils remains poorly understood. This study explored how different lime materials and their interaction with straw affect GHG emissions. Here, we conducted incubation experiments with acidic red soil to investigate the individual and combined effects of liming materials, including $Ca(OH)_2$, CaO, and $CaCO_3$, as well as rice straw addition on nitrous oxide (N_2O) and carbon dioxide (CO_2) emissions. Our findings demonstrated that in the absence of straw, liming increased N_2O emission by 20.3% (CaO) to 78.2% ($Ca(OH)_2$). $CaCO_3$ application raised CO_2 emissions by 182.7%, while CaO and $Ca(OH)_2$ decreased CO_2 emissions by 37.3% and 43.2%, respectively. Adding straw alone enhanced N_2O and CO_2 emissions by 80.69% and 302.7%, respectively. When combined with straw, liming further increased N_2O emissions by 85.0% to 140.1%, with $Ca(OH)_2$ causing the highest emissions. $CaCO_3$ increased CO_2 emissions by 37.3% when combined with straw, whereas CaO and $Ca(OH)_2$ reduced CO_2 emissions by 31.6% and 32.2%, respectively. Straw addition significantly increased global warming potential (GWP). Applying CaO and $Ca(OH)_2$ decreased GWP, whereas $CaCO_3$ increased it with straw application. Compared to $CaCO_3$, CaO and $Ca(OH)_2$ application resulted in a lower GWP, making them optimal lime materials for reducing acidification and mitigating GHG emissions. Linear regression and partial least squares path (PLS-PM) analyses indicated that soil carbon, nitrogen, and microbial biomass significantly influenced N_2O emissions under lime and straw application, while CO_2 emissions were unaffected by these soil properties. Both lime and straw addition increased microbial biomass carbon (MBC) and nitrogen (MBN), dissolved organic carbon (DOC), and NH_4^+–N contents, but decreased NO_3^-–N content, leading to higher N_2O emissions. CO_2 emissions were influenced by the chemical reactions of various lime materials in the soil. These findings suggest that selecting appropriate lime materials can significantly mitigate greenhouse gas emissions from acidic soils, contributing to more sustainable agricultural practices.

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Environmental significance

The application of lime and crop straw in acidic upland soils plays a critical role in mitigating soil acidification and improving fertility, and significantly influence greenhouse gas (GHG) emissions. This study highlights how different lime materials (CaO, $Ca(OH)_2$, $CaCO_3$), alone or combined with straw, distinctly influence GHG emissions. Increased N_2O emissions and decreased CO_2 emissions were observed under the combination of lime (CaO and $Ca(OH)_2$) and straw. Compared to $CaCO_3$, CaO and $Ca(OH)_2$ application resulted in a lower GWP, making them optimal lime materials for reducing acidification and mitigating GHG emissions. These findings support optimized lime and straw management to enhance soil health and reduce climate impacts, guiding sustainable agriculture in acidic soils.

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Introduction

Fertilizer application is one of the most efficient measures for improving agricultural productivity and ensuring food security.[1,2] However, recently, excessive use of chemical fertilizers, particularly nitrogen (N) fertilizers, has led to pronounced environmental consequences, such as decreased soil pH and increased greenhouse gas (GHG) emissions in arable land.[3–5] To overcome soil acidification and promote sustainable agriculture, management approaches like liming and straw return have been recommended, which have great potential to regulate GHG emissions.[6–8]

Nitrous oxide (N_2O) and carbon dioxide (CO_2) are the primary GHGs emitted from upland soils under aerobic conditions. These gases are produced during carbon (C) and N cycling and are regulated by soil pH, C, and N availability.[9–11] Lime application can increase soil pH and improve the availability of C and N for microbes, promoting microbial activities and release of N_2O and CO_2 in acidic soils.[12,13] The excessive emissions of N_2O and CO_2 into the atmosphere have escalated the global warming potential. In contrast, liming has also been found to mitigate N_2O emissions and reduce GWP by controlling the $N_2O/(N_2O + N_2)$ production ratio in acidic soils.[7,14,15] Several studies have demonstrated that liming reduced CO_2 emissions due to the physical protection of soil organic carbon (SOC) *via* improved soil structure,[16,17] and lowered the carbon requirements of soil microbes from increased soil pH.[18] These inconsistent findings obscure the overall understanding of GWP responses to liming, potentially due to variable soil properties, lime materials, and application rates. Prior research has mostly examined the impact of various soil conditions and lime application rates on GHG emissions,[19–21] often overlooking the importance of lime materials. Different lime materials cause distinct alterations in soil pH and nutrient availability,[22,23] producing different impacts on microbial activities and the associated emissions of N_2O and CO_2. Furthermore, the chemical reactions of various lime materials in acidic soils lead to distinct CO_2 fluxes. Quicklime (CaO) and hydrated lime ($Ca(OH)_2$) release OH^-, which directly neutralizes H^+ in soil without producing CO_2. As pH increases, OH^- can further react with soil CO_2 (derived from root and microbial respiration) to form CO_3^{2-}, which combines with Ca^{2+} to precipitate as $CaCO_3$, thus constituting a temporary carbon sink.[24] In contrast, the fate of carbonate materials (*e.g.*, $CaCO_3$) is more complex: under strongly acidic soil conditions, their dissolution directly releases CO_2; whereas under weakly acidic conditions (mainly caused by soil CO_2 dissolving into carbonic acid), the reaction $CaCO_3 + H_2CO_3 \rightarrow Ca^{2+} + 2HCO_3^-$ occurs, leading to CO_2 fixation and functioning as a carbon sink.[25,26] Therefore, the total greenhouse gas emissions following lime application to acidic soils are determined not only by microbial processes but also by these chemical reactions. However, the response and underlying mechanisms of greenhouse gas emissions to different liming materials remain unclear.

Crop straw can also exert a liming effect in acidic soils.[27,28] The acidified soil exhibits not only a low pH, but also poor soil structure and limited fertility. Although straw is less effective than lime in reducing soil acidification,[29] prior research has shown that straw return can enhance nutrient availability and improve soil structure, acting as a recommended strategy for improving acidic soils.[30] In highly acidic soils, combining straw with lime can elevate soil pH and enhance soil fertility.[31] As an organic amendment, crop straw can facilitate microbial mineralization, leading to the release of C and N and increased CO_2 and N_2O production.[32,33] Additionally, straw may also function as a potential energy source for denitrifiers, enhancing denitrification and N_2O emission.[34] The findings of a recent meta-analysis demonstrated that straw return generally results in higher GWP.[35] The impact of straw on soil GHG emissions can be impacted by soil properties, such as pH.[36] Therefore, straw incorporation and liming could potentially cooperate to influence GHG emissions. Liming increases soil pH and alters microbial communities, which influences the decay rate of crop straw and affects the availability of C and N for microbial mineralization.[37,38] The study conducted by Zhao *et al.* revealed that adding lime increased straw-derived C content and reduced straw-derived CO_2.[24] However, the combined effects of liming and straw incorporation on N_2O and CO_2 emissions remain unclear.

This study involved conducting incubation experiments using acidic red soil to evaluate the impact of different lime materials such as $Ca(OH)_2$, CaO, and $CaCO_3$ and straw incorporation on GHG emissions. The aim of this study was to (1) investigate the respective and combined effects of various liming materials and straw on N_2O and CO_2 emissions in acidic upland soil, (2) characterize the impact of soil pH, C, and N availability on N_2O and CO_2 emissions in the presence of liming and straw return, and (3) identify optimal lime materials with straw return for reducing GWP in upland soils. Results from this study may assist in developing appropriate management strategies for effectively mitigating GHG emissions and alleviating acidification.

Materials and methods

Site description and soil sampling

Acidified soil samples were obtained from a long-term field experiment in upland soil located at Zhanggong Town, Jinxian County, Jiangxi Province, China (28°21′6.08″ N, 116° 10′21.66″E). The research site experiences a subtropical climate, characterized by an average annual temperature of 17.2 °C and an annual precipitation of 1549 mm. The soil originates from quaternary red material and is classified as red soil according to the Chinese soil classification system, or as Ferralic Cambisol by FAO standards. This long-term experiment began in 1986, using a rotation of double maize. The field experiment employed a completely randomized block design, encompassing ten different fertilizer treatments and three replications. For this study, we chose NPK treatment, which involves applying chemical N, phosphorus (P), and potassium (K) fertilizers at rates of 60 kg N per ha, 30 kg P per ha, and 60 kg K per ha during each season. Prior to maize sowing, 40% of the chemical N and K fertilizer and 100% of the chemical P fertilizer were applied, with the 60% N and K fertilizer added during the vegetative stage (V12). As a result of the prolonged application of chemical N fertilizer, the soil experienced significant acidification,

with a pH decrease from 6.00 to 4.68.[39] Other fundamental soil properties prior to incubation were 7.86 g kg^{-1}, 86.63, 18.20, and 220.83 mg kg^{-1} for SOC, available N (AN), available P (AP), and available K (AK), respectively.

Following the harvest of the early maize in July 2018, samples were obtained from undisturbed surface soil (0–20 cm) using a soil auger. Five soil cores were combined into a single composite sample from each plot. The soil samples were air-dried in the laboratory, and visible roots, organic residues, and stone debris were manually removed. The dried soil samples were then sieved through a 2 mm mesh for physical and chemical analyses as well as for incubation experiments.

Liming materials and straw preparation

We used three different liming materials (CaO, $Ca(OH)_2$, and $CaCO_3$) obtained from Aladdin Chemicals Co. Ltd (Shanghai, China). Rice straw was harvested in November 2018 from a paddy field in Jinxian. The rice straw was dried at 60 °C to a constant weight, cut into 2 mm pieces, and then stored in a sealed jar. The total carbon and nitrogen contents of the straw were 381 and 8.83 g kg^{-1}, respectively.

Incubation experiment

The air-dried soils were adjusted to 45% of their water-holding capacity (WHC) before undergoing a 7-day pre-incubation period in darkness at 25 °C to stimulate and restore microbial activity. Subsequently, 150 g samples of dried soil were transferred to 500 mL glass bottles for further processing. Treatments were separated into two groups: those with straw addition and those without straw addition. Each group encompassed the following treatments: (i) CK (control, without lime); (ii) CaO; (iii) $Ca(OH)_2$; (iv) $CaCO_3$. This study utilized a completely randomized design with three replications for each treatment. The application rates for the liming materials were as follows: 3.00 g per kg dry soil for CaO, 3.81 g per kg dry soil for $Ca(OH)_2$, and 5.16 g per kg dry soil for $CaCO_3$, determined by a neutralization titration experiment targeting a pH of 7. The application rate of rice straw was 5 g per kg dry soil, equivalent to a field application rate of 12 t ha^{-1}. The amendments and soil were thoroughly combined, and each treatment was adjusted to 65% WHC. The mixtures were then placed in a dark environment at 25 °C for a period of 30 days. Soil moisture was regulated by the addition of deionized water every other day to maintain a consistent water content of 65% WHC during the entire incubation.

Gas sampling and analysis and GWP calculation

Gas samples were collected on days 1, 2, 3, 5, 7, 9, 11, 13, 15, 18, 21, 24, 27, and 30 of the incubation period. For each sample, the glass bottle was left open for 30 minutes to allow fresh air to enter prior to sampling. Subsequently, it was sealed with a rubber plug for 60 minutes. Two gas samples were obtained at two different time points: immediately after closure and after 60 minutes. The CO_2 and N_2O concentrations in each gas sample were characterized using a Gas Chromatograph (Agilent Technology 7890 B, USA). The gas flux and cumulative emissions were calculated using eqn (1) and (2), respectively.

$$F = \rho \times \Delta C/\Delta T \times V/W \times \frac{273}{(273+T)} \times f \quad (1)$$

$$E = \sum_{i=1}^{n} \frac{F_i + F_{i+1}}{2} \times (t_{i+1} - t_i) \times 24 \quad (2)$$

where F denotes the emission rate of N_2O (μg N per kg soil per h) and CO_2 (mg C per kg soil per h), the symbol ρ represents the gas density under standard temperature conditions; ΔC indicates the variation in gas concentration between the initial and 60-minute time points during jar closure; ΔT refers to the duration of jar closure (in hours); V represents the volume of gas space within the bottle (m^3); W signifies the mass of soil (kg); T indicates the temperature of the experiment (25 °C); and f represents the conversion coefficient, which is 28/44 for N_2O and 12/44 for CO_2 when calculating emission fluxes. E is the cumulative N_2O (μg per kg soil) and CO_2 (mg per kg soil) emissions, and F_i and F_{i+1} are the emission rates of N_2O and CO_2 at time t_i and t_{i+1}, respectively.

GWP was expressed in terms of CO_2 equivalencies, and the GWP was 273 for N_2O (IPCC, 2021). The formula used was as follows:

$$\text{GWP (mg } CO_2\text{-eq per kg soil)} = 273 \times N_2O \text{ (mg } N_2O \text{ per kg soil)} + CO_2 \text{ (mg } CO_2 \text{ per kg soil)} \quad (3)$$

Soil analysis

Following incubation, soil samples were harvested to analyze various parameters, including soil pH, microbial biomass carbon (MBC) and nitrogen (MBN), dissolved organic carbon (DOC), soil organic carbon (SOC), and mineral nitrogen (NH_4^+–N and NO_3^-–N). Soil pH was determined through a pH meter (Mettler Toledo, Columbus, USA) from slurries of samples prepared with a soil to water ratio of 1 : 2.5. The concentrations of soil MBC and MBN were evaluated using the chloroform fumigation-extraction method.[40] Fresh soil was fumigated with chloroform, and both fumigated and unfumigated samples were incubated in darkness at 25 °C for 24 hours. Subsequently, the samples underwent extraction using a solution of 0.05 M K_2SO_4, and the extracts were analyzed using a TOC/TN analyzer (multi-C/N 3100, Germany). MBC and MBN were computed based on the difference in C and N concentrations between fumigated and unfumigated extracts. Soil DOC was the concentration of total organic C in the unfumigated sample. SOC was evaluated using the $K_2Cr_2O_7$ titration method.[41] Soil NH_4^+–N and NO_3^-–N were extracted using 2 M KCl and quantified using an automated flow injection analyzer (Tecator FIA Star 5000 analyzer, Foss Tecator, Sweden).[42]

Statistical analysis

Prior to analysis, the normality of all data was assessed using the Kolmogorov–Smirnov test. One-way analysis of variance (ANOVA) was conducted using SPSS software (SPSS, Inc., USA), with a significance level of $p \leq 0.05$ for the least significant difference (LSD). The relationship between gas emissions and

soil properties was examined through regression analysis. Additionally, partial least squares path modeling (PLS-PM) was performed using the "plspm" package in R 3.5.2 software (R Core Team, 2020), to identify pathways and key factors influencing gas emissions. Model quality was evaluated using the "goodness of fit" statistic.

Results

N_2O and CO_2 emissions under different lime materials and straw amendments over the 30-day incubation period

The N_2O fluxes increased following the addition of lime and straw compared to the non-amended soil, with peak emissions of N_2O occurring on the second day before declining exponentially thereafter (Fig. 1a and b). Liming enhanced cumulative N_2O emissions regardless of straw presence (Fig. 1c). Without straw, lime application led to a notable increase in N_2O emission by 20.3–78.2% ($p \leq 0.05$), with the $Ca(OH)_2$ treatment exhibiting the most pronounced effect. Incorporating straw alone increased cumulative N_2O emissions by 80.69% compared to non-amended soil. In the presence of straw, liming increased cumulative N_2O emissions by 85.0–140.1% compared to soil that received only straw, with the highest level in the $Ca(OH)_2$ and straw combination treatment. The cumulative N_2O emissions showed no significant difference between the CaO and $CaCO_3$ treatments, regardless of straw administration.

The effect of liming on CO_2 emissions varied significantly contingent upon the lime materials used (Fig. 2a–c). The addition of $CaCO_3$ immediately increased CO_2 fluxes compared to non-limed soil, regardless of straw presence, with emission declining exponentially at later stages of incubation (Fig. 2a and b). In the absence of straw, the application of CaO and $Ca(OH)_2$ reduced CO_2 emissions to zero within the first five days of incubation, after which emissions slightly increased and stabilized. The incorporation of straw alone resulted in an increase in CO_2 fluxes when compared to non-amended soil, peaking on the second day. In the presence of straw, CO_2 emissions decreased to zero in the first four days following the application of CaO and $Ca(OH)_2$, then slightly increased, peaking on the seventh day. The application of $CaCO_3$ increased cumulative CO_2 emissions by 182.7% compared to the non-amended soil lacking straw, and by 37.3% compared to soil with only straw (Fig. 2c). Without straw supplementation, CO_2 emissions significantly decreased by 37.3% and 41.0% due to the application of CaO and $Ca(OH)_2$, respectively. Straw incorporation alone enhanced cumulative CO_2 emissions by 302.7% compared to non-amended soil. With straw addition, the application of CaO and $Ca(OH)_2$ decreased cumulative CO_2 emissions by 32.2% and 31.6%, respectively, relative to soil with only straw.

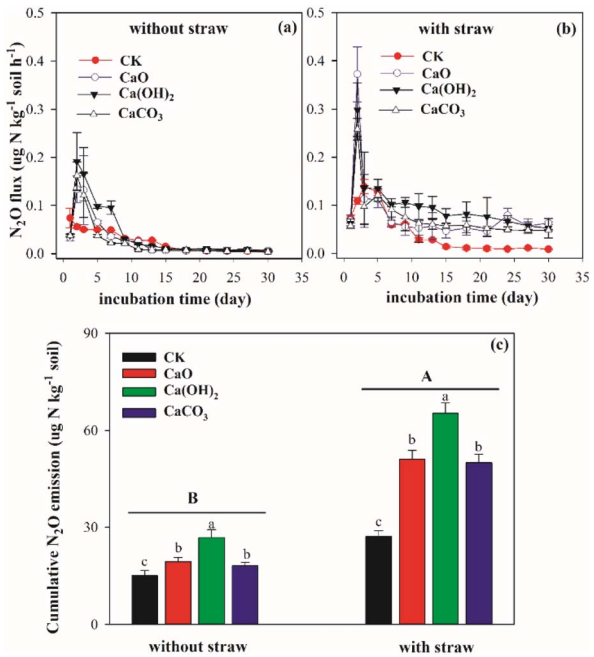

Fig. 1 N_2O fluxes and (a and b) and cumulative emissions (c) from different treatments over the 30-day incubation period. Error bars represent standard errors of the mean values ($n = 3$). Different uppercase letters and lowercase letters indicated the significant difference between straw and no straw treatments, and the difference among different lime materials treatments ($P < 0.05$), respectively.

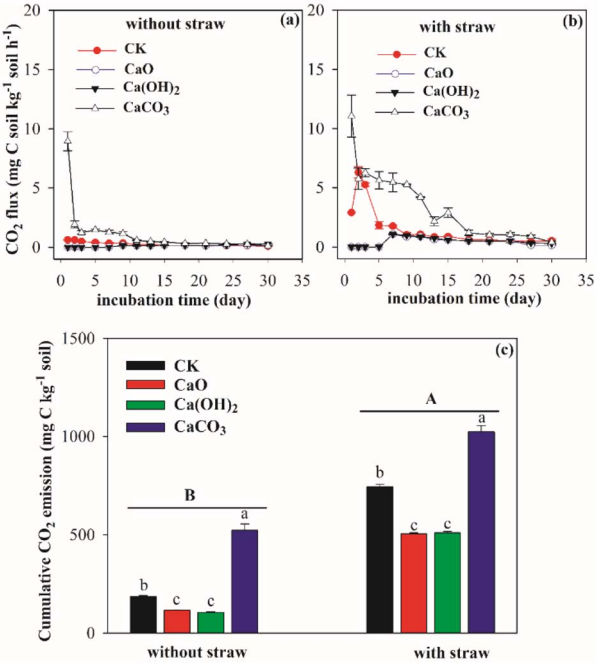

Fig. 2 CO_2 fluxes and (a and b) and cumulative emissions (c) from different treatments over the 30-day incubation period. Error bars represent standard errors of the mean values ($n = 3$). Different uppercase letters and lowercase letters indicated the significant difference between straw and no straw treatments, and the difference among different lime materials treatments ($P < 0.05$), respectively.

Global warming potential

Over the 30-day incubation period, in the absence of straw, the addition of CaO and $Ca(OH)_2$ did not significantly change GWP, whereas the addition of $CaCO_3$ increased GWP by 281.4% (Fig. 3). The addition of straw alone increased GWP by 301.3%.

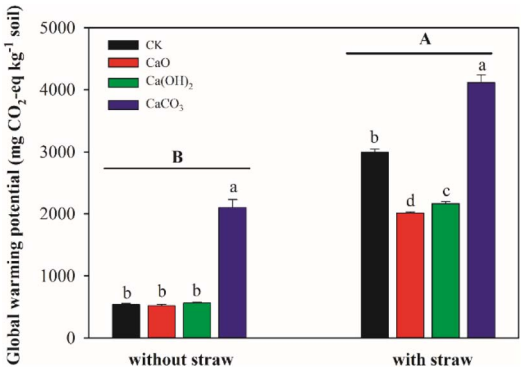

Fig. 3 Global warming potential in different treatments over the 30-day incubation period. Error bars represent standard errors of the mean values ($n = 3$). Different uppercase letters and lowercase letters indicated the significant difference between straw and no straw treatments, and the difference among different lime materials treatments ($P < 0.05$), respectively.

In the presence of straw, the application of CaO and $Ca(OH)_2$ reduced GWP by 32.8% and 27.7%, respectively, compared to soil amended only with straw. The GWP for the combination of $CaCO_3$ and straw reached the highest level at 4114.9 mg CO_2-eq per kg soil, representing a 37.4% increase compared to straw addition alone.

Soil properties under different lime treatments and straw amendments

The pH values of the limed soil (ranging from 6.65 to 7.15) were significantly higher than the unlimed soil (ranging from 4.55 to 4.65) regardless of the presence of straw, with the highest pH observed in the $Ca(OH)_2$ treatment (Table 1). Straw addition did not impact soil pH. In the absence of straw, the DOC content increased significantly by 14.5–43.1% due to lime application, with a more pronounced response in the $Ca(OH)_2$ treatment. Straw incorporation alone increased DOC by 15.9% compared to non-amended soil. In the presence of straw, liming elevated DOC content by 27.3–48.3% relative to the soil only subjected to straw, with the highest DOC found under liming with $Ca(OH)_2$ and straw combination. The trends for MBC and MBN mirrored those for DOC under lime and straw application. Liming increased MBC and MBN concentrations with or without straw addition, while the addition of straw alone led to a 38.2% increase in MBC and a 49.8% increase in MBN. However, the SOC content did not show significant differences across lime and straw treatments.

Liming significantly increased NH_4^+–N by 55.5% to 223.5% but decreased NO_3^-–N content by 62.5% to 80.2% across both straw and non-straw treatments, with the most pronounced effects in the $Ca(OH)_2$ treatment. The addition of straw alone did not affect NH_4^+–N but decreased NO_3^-–N content by 36.0% compared to non-amended soil.

Relationships between N_2O, CO_2 emissions, and soil properties

The results of the regression analysis indicated a significant positive correlation between N_2O emissions and soil pH, DOC, NH_4^+–N, MBC, and MBN, as well as a negative correlation with NO_3^-–N (Fig. 4a–f). Conversely, no significant correlation was identified between CO_2 emission and soil properties across treatments with lime and straw addition (Fig. S1). Soil N_2O emissions were significantly impacted by soil pH, soil C and N, and microbial biomass, collectively accounting for 87% of the variance in N_2O emission across treatments, as revealed by the PLS-PM (Fig. 5). Specifically, soil pH (standard coefficient = 0.553), soil C and N (standard coefficient = 0.649), and microbial biomass (standard coefficient = 0.553) exhibited direct positive effects on N_2O emission. Soil pH also exhibited an indirect effect on N_2O emission by positively modifying soil C and N (standard coefficient = 0.482) and microbial biomass (standard coefficient = 0.584). Furthermore, soil C and N indirectly regulated N_2O emission by influencing microbial biomass (standard coefficient = 0.816).

Discussion

Effects of different lime materials and straw application on N_2O emissions

In this study, the application of all three lime materials (CaO, $Ca(OH)_2$, and $CaCO_3$) resulted in an increase in N_2O emissions due to the higher DOC, NH_4^+–N concentration, and microbial biomass in the liming treatments (Fig. 1 and Table 1). The

Table 1 Soil properties in different treatments at the end of the incubation period[a]

	Treatments	pH	SOC (g kg^{-1})	DOC (mg kg^{-1})	NH_4^+–N (mg kg^{-1})	NO_3^-–N (mg kg^{-1})	MBC (mg kg^{-1})	MBN (mg kg^{-1})
Without straw	CK	4.65 ± 0.15^{c}	7.86 ± 0.15^{a}	29.85 ± 0.72^{d}	2.63 ± 1.38^{d}	19.81 ± 1.81^{a}	131.95 ± 3.20^{e}	13.07 ± 0.68^{d}
	CaO	6.75 ± 0.02^{b}	7.58 ± 0.11^{a}	36.53 ± 1.09^{c}	4.89 ± 0.19^{c}	$4.98 \pm 1.07^{c\ d}$	$164.70 \pm 9.62^{c\ d}$	$17.83 \pm 3.17^{b\ c}$
	$Ca(OH)_2$	7.15 ± 0.22^{a}	7.45 ± 0.32^{a}	42.72 ± 1.82^{b}	6.42 ± 0.74^{b}	3.92 ± 0.77^{d}	182.52 ± 8.33^{c}	20.37 ± 2.59^{b}
	$CaCO_3$	6.73 ± 0.04^{b}	7.56 ± 0.28^{a}	34.17 ± 1.61^{c}	4.69 ± 1.16^{c}	5.94 ± 1.46^{c}	154.37 ± 13.1^{d}	15.76 ± 0.74^{c}
With straw	CK	4.55 ± 0.12^{c}	8.18 ± 0.40^{a}	34.59 ± 2.45^{c}	2.72 ± 0.75^{d}	12.69 ± 0.83^{b}	181.88 ± 17.6^{c}	19.58 ± 0.75^{b}
	CaO	6.65 ± 0.16^{b}	8.26 ± 0.29^{a}	46.26 ± 3.24^{b}	$7.66 \pm 1.51^{a\ b}$	$4.23 \pm 0.42^{c\ d}$	243.17 ± 11.3^{b}	22.12 ± 0.77^{b}
	$Ca(OH)_2$	7.02 ± 0.08^{a}	7.73 ± 0.15^{a}	51.29 ± 2.08^{a}	8.80 ± 1.59^{a}	3.31 ± 0.46^{d}	282.02 ± 10.7^{a}	28.15 ± 2.67^{a}
	$CaCO_3$	6.68 ± 0.06^{b}	8.25 ± 0.17^{a}	44.05 ± 3.61^{b}	6.57 ± 0.30^{b}	$4.16 \pm 0.39^{c\ d}$	233.99 ± 12.1^{b}	23.50 ± 3.10^{b}

[a] SOC, soil organic carbon; DOC, dissolved organic carbon; MBC, microbial biomass carbon; MBN, microbial biomass nitrogen. The data presented in the table represent the mean and standard error values, with lowercase letters denoting a significant difference between treatments within each column ($P < 0.05$).

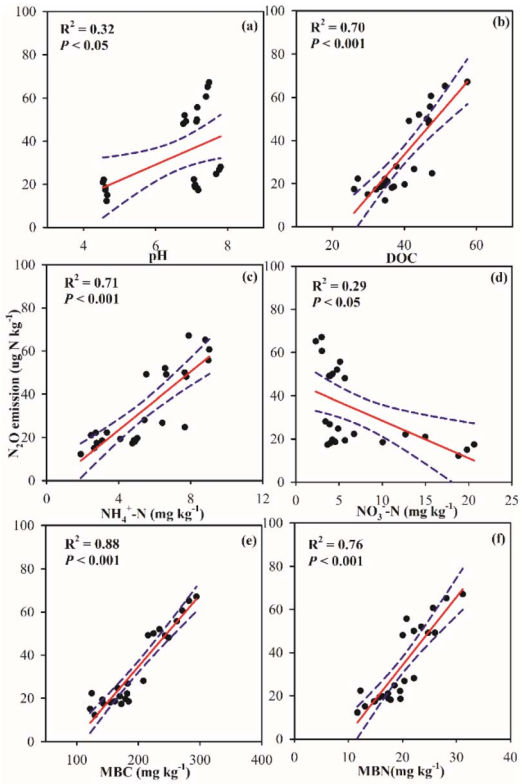

Fig. 4 Relationship between N_2O emission and soil pH (a), dissolved organic carbon (DOC) content (b), NH_4^+–N (c), NO_3^-–N (d), microbial biomass carbon (MBC) (e) and microbial biomass nitrogen (MBN) (f). The blue dashed lines represent the regression lines along with a 95% confidence interval ($P < 0.05$).

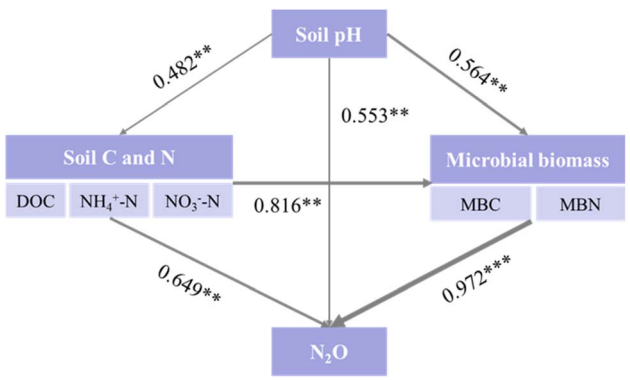

Fig. 5 Directed graph of the partial least squares path model (PLS-PM) of the effects of soil properties on N_2O. The width of arrows is proportional to the strength of path coefficients. Numbers adjacent to arrows denote standardized path coefficient, with ** and *** denoting $P < 0.01$ and $P < 0.001$, respectively. DOC, dissolved organic carbon; MBC, microbial biomass carbon; MBN, microbial biomass nitrogen. The Goodness of Fit (GoF) of model was 0.87.

positive impact of lime application on soil DOC mirrored previous studies,[12,43] attributed to the pH-induced changes in soil organic matter solubilization.[44] Enhanced carbon availability and favorable pH conditions led to higher microbial biomass with lime addition,[45] thus resulting in enhanced organic N mineralization and increased NH_4^+–N content. The increase of substrate and enhanced microbial activity promoted nitrification, causing higher N_2O emissions.[46] The decrease in NO_3^-–N may be due to its consumption during denitrification.[7] However, previous studies have reported that the application of lime decreased the release of N_2O in acidic soils by enhancing N_2O reductase activity at higher pH during denitrification,[47,48] which contrasts with our findings. This discrepancy can be explained by: (i) the soil used in the incubation experiment was seriously acidified (pH < 5), where lower pH could strongly inhibit the metabolic process of nitrifying and denitrifying microorganisms, decreasing N_2O emissions.[49,50] (ii) The responses of N_2O emissions to liming can differ significantly over short-term incubation experiments and long-term field experiments.[13,51] The immediate response to liming is a notable increase in N_2O emissions, likely attributed to a temporary enhancement of net N mineralization and nitrification resulting from increasing nutrient availability.[51] In the short term, pH changes have limited influence on the microbial communities and N_2O reduction genes. Over longer periods, liming can significantly impact the ratio of N_2O to N_2, influenced by changes in the microbial community and increased activity of N_2O reductase in response to higher soil pH, causing a decrease in N_2O emissions.[7,52] The results of linear regression and PLS-PM also suggested that soil C and N, along with associated microbial biomass, had more significant direct effects on N_2O than pH in the short term (Fig. 4 and 5). The magnitude of the liming effect on N_2O emissions varied with liming material. The greatest rise in N_2O emissions was observed in the $Ca(OH)_2$ treatments in this study (Fig. 1). This can be attributed to the highest values of soil pH, DOC, NH_4^+–N, and MBC observed in the $Ca(OH)_2$ treatments compared to CaO and $CaCO_3$ (Table 1), as documented in previous studies.[23] $Ca(OH)_2$ is highly effective in rapidly improving the pH and enhancing the availability of C and N in acidic soil, leading to higher substrates, microbial activities, and thus elevated N_2O emissions.[53,54]

Straw application also increased N_2O emission (Fig. 1). The decomposition of straw provided essential substrates for microorganisms. The net N mineralization or immobilization following straw addition is largely dependent on the C : N ratios of the straw.[55,56] Recent research has indicated that when the C : N ratio of straw is below 45, it can supply sufficient N to support soil microbial community development following straw amendment, leading to a net increase in N mineralization.[36] In this study, the C : N ratio of rice straw was 43, resulting in net N mineralization and increased NH_4^+–N content in the soil, promoting nitrification and N_2O emissions. Simultaneously, straw decomposition consumes substantial oxygen, triggering denitrification in anoxic microsites and increasing N_2O emissions.[57] Although liming can reduce N_2O release during denitrification by enhancing the reduction of N_2O to N_2, it simultaneously stimulates microbial activity and straw decomposition,[58,59] which in turn promoted N mineralization and nitrification. Consequently, the simultaneous application of lime and straw led to greater N_2O emissions compared to using lime or straw separately due to the higher substrate availability and increased microbial activity.

It is important to note that our experiment was conducted without fertilizer application. Under such conditions, the low nutrient availability (especially N) in the soil was the main reason for the strong N mineralization and nitrification induced by lime and straw. However, fertilizer application is a common practice in agricultural systems. The addition of fertilizer (especially N fertilizer) may amplify the effects observed in this study. Research has shown that in strongly acidic soils (pH < 5), the combined application of lime and N fertilizer triggered a sharp increase in N_2O emissions compared to lime application alone, which was associated with a rise in AOB-amoA gene abundance and enhanced ammonia oxidation.[60] Under fertilized conditions, straw incorporation generally increases N_2O emissions,[61] with the magnitude of increase being positively correlated with N application rate,[62] due to elevated N substrate (especially NO_3^-) availability and enhanced microbial activity.[61] These findings indicate that the combined use of lime, straw, and N fertilizers in cropland should be carefully managed to avoid stimulating excessive N_2O emissions.

Effects of different lime materials and straw application on CO_2 emissions

The liming effect on CO_2 emissions varied significantly with the lime materials used. $CaCO_3$ application increased CO_2 emissions, while CaO and $Ca(OH)_2$ application reduced CO_2 emissions, regardless of the presence of straw (Fig. 2). Our findings indicated that soil properties did not significantly influence CO_2 emissions across treatments with different lime materials and straw additions (Fig. S1). This suggests that the mechanisms by which lime affects CO_2 emissions differ from those affecting N_2O emissions. The increased CO_2 emissions in $CaCO_3$ treatments may be linked to the dissolution of carbonates in limed acidic soils.[10,63] Research has suggested that CO_2 emissions from carbonates may account for up to 77% of the total CO_2 emissions.[25] Although Rousset *et al.* reported the potential for carbon sequestration reaction ($CaCO_3 + H_2CO_3 \rightarrow Ca^{2+} + 2HCO_3^-$) may occur during $CaCO_3$ amendment, this requires sufficient H_2CO_3 (dissolved CO_2) in the soil solution, which usually happens under relatively high pH conditions where the system approaches equilibrium.[26] In the present experiment, although the rise in soil pH due to neutralization in the later stage may have created conditions favorable for CO_2 fixation, the cumulative CO_2 flux indicate that the carbon emissions generated in the early stage by acid neutralization reactions were significantly greater than the minor CO_2 sequestration that may have occurred later. Therefore, in strongly acidic soils (pH < 5), $CaCO_3$ application primarily exhibits a net carbon emission effect in the short term. On the contrary, the application of CaO and $Ca(OH)_2$ led to lower CO_2 emissions, attributed to the chemical reaction of CaO with CO_2 to form $CaCO_3$.[64] This reaction is more likely in neutral or alkaline soils where $CaCO_3$ is more stable at higher pH levels. However, during short-term incubation experiments, CaO may also decrease CO_2 emissions in acidic soils by reacting with CO_2,[64] attributed to the incomplete release of potential acidity in the soil during the short-term. Overall, our results indicate that the type of lime material significantly influences CO_2 emissions. $CaCO_3$ increases CO_2 emissions, while CaO and $Ca(OH)_2$ decrease them, driven by specific chemical reactions and soil conditions during the incubation period.

Straw addition significantly increased CO_2 emissions (Fig. 2), consistent with prior studies.[65,66] The decomposition of straw released substantial CO_2.[67] Additionally, incorporating crop straw into nutrient-deficient acidic soil provided available C and N, stimulating microbial activity and leading to a positive priming effect of SOC and, consequently, greater CO_2 emission.[65] With straw addition, the application of CaO and $Ca(OH)_2$ decreased CO_2 emissions relative to soil with only straw, which attributed to the chemical reaction of CaO with CO_2 to form $CaCO_3$.

Field fertilization can also exacerbate the short-term CO_2 emissions induced by lime and straw incorporation. Chemical N fertilizers promote the dissolution of carbonate-based lime and the release of CO_2 by acidifying the soil.[25] Fertilizers and lime may also interact to indirectly enhance CO_2 emissions by stimulating the microbial decomposition and mineralization of SOC.[68] The mineralization of straw, as well as the associated priming of SOC mineralization, mainly depends on soil stoichiometry. Most studies have shown that the application of N and P fertilizers alleviates microbial nutrient limitations, thereby increasing straw mineralization and CO_2 production.[36,69] Some studies also suggest that this alleviation of nutrient limitations similarly enhances the mineralization of SOC.[70,71] Nevertheless, in the long term, optimizing fertilizer application can improve crop growth, enhance carbon inputs and straw-derived carbon sequestration, and ultimately contribute to SOC accumulation.[72]

The optimal lime material with straw return for ameliorating acidification and mitigating global warming potential in upland soil

The lime materials used in this study (CaO, $Ca(OH)_2$, and $CaCO_3$) significantly elevated the pH of acidic soils (Table 1). However, the straw application alone did not significantly increased soil pH (Table 1). This mirrored previous reports showing that lime is more effective than crop straw in the soil acidification amelioration.[28,29] Liming quickly raises soil pH through chemical pathways, producing rapid and impactful results.[28] While straw can also ameliorate soil acidity by releasing alkalinity during mineralization, this process can be time-consuming. This suggests that straw return should be integrated with lime for optimal acidic soil improvement. Combining lime and straw increases soil pH and enhances soil carbon sequestration to improve soil fertility.[31] However, we found that SOC did not significantly change with the addition of lime and straw (Table 1). This could be attributed to the relatively short duration of the incubation period, during which complete decomposition and transformation of straw C might not have occurred. Long-term incorporation of straw into fields not only enriches nutrients but also promotes improved soil structure.[73] Therefore, it is recommended that lime be applied together with straw to ameliorate acidic soils.

Straw application increased the GWP due to increased CO_2 and N_2O emissions. Although all three lime materials increased N_2O emissions, both CaO and $Ca(OH)_2$ treatments reduced CO_2 emissions, whereas the $CaCO_3$ treatment further increased CO_2 emissions. Regardless of straw addition, CaO and $Ca(OH)_2$ treatments exhibited lower GWP compared to $CaCO_3$. In the presence of straw, CaO exhibited a lower GWP than $Ca(OH)_2$, though the difference was minor. Therefore, compared with $CaCO_3$, CaO and $Ca(OH)_2$ represent more optimal materials for ameliorating soil acidification while mitigating greenhouse gas emissions.

Study limitations

While this study provides valuable insights into the effects of different lime materials and straw incorporation on GHG emissions, the reliance on short-term (30-day) incubation experiments may limit the generalizability of the findings to field conditions. The absence of fertilizer input in the incubation experiment, along with the controlled laboratory environment that simplifies field factors such as seasonal temperature fluctuations and rainfall variability, may affect the extrapolation of the results, as fertilization and environmental factors play critical roles in GHG emissions in agricultural ecosystems. Furthermore, transient responses observed over the 30-day period may not fully represent long-term trends. Studies have indicated that CO_2 emissions from lime dissolution were much lower under field conditions, and after four months of lime application, emissions were nearly negligible.[74,75] Additionally, the effects of prolonged pH alterations and immediate pH adjustments due to liming on N_2O emissions also differ.[52] The lack of field validation further emphasizes the need for caution when extrapolating these results to practical agricultural management. Future research incorporating long-term field trials and integrated assessments of crop productivity and environmental effects would significantly enhance the applicability of the conclusions. Moreover, long-term lime application may increase the risks of soil compaction, nutrient imbalance, and soil re-acidification.[76] Therefore, attention should be paid to the frequency and dosage of lime application, or co-application with organic fertilizers should be considered in agricultural practice.

Conclusions

The application of lime and straw, individually or in combination, increased N_2O emissions. Notably, the combined application of lime and straw resulted in higher emissions compared to individual applications. Among the various lime materials tested, $Ca(OH)_2$ exhibited the most pronounced effects on N_2O emissions, while CaO and $CaCO_3$ showed comparable impacts. The positive correlation between N_2O emissions and soil pH, DOC, NH_4^+–N, MBC, and MBN, coupled with the negative correlation with NO_3^-–N across different lime materials and straw application, underscores the complex interplay between soil properties and greenhouse gas dynamics. These findings highlight the immediate response to liming and straw amendments, characterized by a significant increase in N_2O emissions, attributed to the enhancement of net N mineralization and nitrification processes. Conversely, straw application significantly increased CO_2 emissions, while the liming effect on CO_2 emissions strongly relied on the lime material used. $CaCO_3$ contributed to elevated CO_2 emissions through its solution in acidic soil, whereas CaO and $Ca(OH)_2$ reduced CO_2 emissions through their interactions with CO_2. Our investigation identifies CaO and $Ca(OH)_2$ as the optimal choice of lime materials for addressing acidification and mitigating N_2O and CO_2 emissions in acidic upland soils, leading to a lower global warming potential compared with $CaCO_3$. Nonetheless, given the potential variability between the impacts of long-term lime-induced pH changes and short-term pH adjustments on N_2O and CO_2 emissions, and the inherent complexity of field conditions relative to incubation experiments, further research is required under realistic field conditions, to gain a comprehensive understanding of the effects of liming and straw return on greenhouse gas emissions in agricultural ecosystems, thereby informing sustainable management practices and environmental stewardship. Additionally, excessive and long-term application of lime may induce adverse effects such as soil compaction, nutrient imbalance, and a tendency toward re-acidification. Therefore, in agricultural practice, careful consideration should be given to the frequency of lime application, and integrating lime with organic amendments may represent a more sustainable strategy.

Author contributions

Zhe Shen: investigation, methodology, formal analysis, writing original – draft, writing – review & editing. Kailou Liu: investigation, data curation, funding acquisition. Jiwen Li and Nano Alemu Daba: investigation, writing – review & editing. Md Ashraful Alam and Kiya Adare Tadesse: methodology, writing – review & editing. Tianfu Han: writing – review & editing, funding acquisition. Huimin Zhang: methodology, writing – review & editing, funding acquisition.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data used in this study were obtained from a soil incubation experiment in the laboratory, and all data supporting the findings are presented in figures (Fig. 1–5, S1) and table (Table 1) in this paper. Supplementary information: the correlation between CO_2 emissions and soil properties. See DOI: **https://doi.org/10.1039/d5em00494b**.

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