Received: 12 January 2021 | Accepted: 17 March 2021
DOI: 10.1111/gcb.15607

PRIMARY RESEARCH ARTICLE

Global Change Biology WILEY

Potential benefits of liming to acid soils on climate change mitigation and food security

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Funding information
National Natural Science Foundation of China, Grant/Award Number: 41977282, 41675144 and 41807327; Chinese Academy of Sciences, Grant/Award Number: ZDBS-LY-DQCOO7

Abstract

Globally, about 50% of all arable soils are classified as acidic. As crop and plant growth are significantly hampered under acidic soil conditions, many farmers, but increasingly as well forest managers, apply lime to raise the soil pH. Besides its direct effect on soil pH, liming also affects soil C and nutrient cycles and associated greenhouse gas (GHG) fluxes. In this meta-analysis, we reviewed 1570 observations reported in 121 field-based studies worldwide, to assess liming effects on soil GHG fluxes and plant productivity. We found that liming significantly increases crop yield by 36.3%. Also, soil organic C (SOC) stocks were found to increase by 4.51% annually, though soil respiration is stimulated too (7.57%). Moreover, liming was found to reduce soil N_2O emission by 21.3%, yield-scaled N_2O emission by 21.5%, and CH_4 emission and yield-scaled CH_4 emission from rice paddies by 19.0% and 12.4%, respectively. Assuming that all acid agricultural soils are limed periodically, liming results in a total GHG balance benefit of 633–749 Tg CO_2-eq year^{-1} due to reductions in soil N_2O emissions (0.60–0.67 Tg N_2O-N year^{-1}) and paddy soil CH_4 emissions (1.75–2.21 Tg CH_4 year^{-1}) and increases in SOC stocks (65.7–110 Tg C year^{-1}). However, this comes at the cost of an additional CO_2 release (c. 624–656 Tg CO_2 year^{-1}) deriving from lime mining, transport and application, and lime dissolution, so that the overall GHG balance is likely neutral. Nevertheless, liming of acid agricultural soils will increase yields by at least 6.64×10^8 Mg year^{-1}, covering the food supply of 876 million people. Overall, our study shows for the first time that a general strategy of liming of acid agricultural soils is likely to result in an increasing sustainability of global agricultural production, indicating the potential benefit of liming acid soils for climate change mitigation and food security.

KEYWORDS
crop yield, greenhouse gas emissions, liming, meta-analysis, soil acidification, soil organic C, soil pH

1 | INTRODUCTION

Soil acidification, which promotes toxicities of aluminum (Al), manganese (Mn), and iron (Fe) and results in deficiencies of essential micronutrients for plant growth, such as calcium (Ca), magnesium (Mg), potassium (K), molybdenum (Mo), and phosphorous (P), is one of the most serious global issues of land degradation (Bolan et al., 2003; Goulding, 2016; Guo et al., 2010; Shi et al., 2019). Although soil acidification is a naturally occurring process, it has been accelerated remarkably over recent decades due to human activities. Increased deposition of acidifying compounds caused by emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) associated with fossil fuel combustion, usage of N fertilizers to support crop growth (Kunhikrishnan et al., 2016; Mahler et al., 2016; Meng et al., 2019; Qu et al., 2014), or deposition of reduced N compounds to natural land (Velthof et al., 2011) are the main drivers of soil acidification. Soil acidification due to fertilizer application or deposition of reduced N compounds to natural ecosystems is the result of the release of protons during nitrification and subsequent leaching of nitrates. For example, Guo et al. (2010) reported that overuse of synthetic N fertilizers, mainly in the form of urea, during the 1980s–2000s has resulted in a significant acidification of croplands in China, with soil pH decreases in the range of 0.13–0.80 pH units depending on the soil types and properties. Globally, acidic soils cover about 39.5 million km^2 or 30% of the global land surfaces (von Uexküll & Mutert, 1995). Dai et al. (2017) point out that approximately 50% of the world's arable soils are acidic, with areas further increasing in recent years. It is well known that acid soils can alter soil carbon (C) and nutrient cycling and adversely influence the growth of plant and soil biota, and threaten terrestrial ecosystem functions (e.g., net primary production and species richness; Meng et al., 2019; Stevens et al., 2010; Yang et al., 2012). Taking soil nitrous oxide (N_2O) fluxes as an example, soil acidification directly and/or indirectly aggravates emissions, particularly from N-fertilized agricultural soils (Kunhikrishnan et al., 2016; Qu et al., 2014; Raut et al., 2012).

Liming with Ca^{2+}- and Mg^{2+}-rich materials is a common soil amelioration practice worldwide. Liming promotes the immobilization of toxic heavy metals and alters the transformation and uptake of nutrients by plants, and consequently affecting the productivity of ecosystems (Fornara et al., 2011; Li et al., 2019). However, contradictory results of liming on plant growth and yields have been reported. While some studies found a significant enhancement of crop yields (Crusciol et al., 2019; Holland et al., 2019; Wang et al., 2015), other studies observed decreases in crop production, specifically if soils were limed or with excessive amounts (Hénault et al., 2019; Meng et al., 2004). Beyond affecting plant productivity, changes in soil pH caused by liming will also affect the soil microbial biomass as well as its composition and activity and soil C and N availability. Besides direct effects of pH on soil microbial processes such as denitrification (Bakken & Frostegård, 2020), changes in soil microbial parameters and substrate availability will also affect the production, consumption, and emission of GHGs, namely N_2O, methane (CH_4), and carbon dioxide (CO_2; Clough et al., 2004; Khaliq et al., 2019; Paradelo et al., 2015; Royer-Tardif et al., 2019; Shaaban et al., 2016).

The influence of liming on soil GHG fluxes has been controversial in previous studies (Kunhikrishnan et al., 2016; Page et al., 2009). For example, liming of soils may reduce N_2O emissions as the N_2O reductase (*nosZ*) activity of denitrifiers has been found to be increased at neutral pH values. This promotes the formation of N_2 as end product of denitrification and lowers the overall $N_2O/(N_2+N_2O)$ product ratio of denitrification (Brumme & Beese, 1992; Liu et al., 2010; Šimek & Cooper, 2002). In contrast, liming has also been reported to stimulate soil N_2O emissions, due to the stimulation of nitrification and nitrifier N_2O production (Baggs et al., 2010; Hink et al., 2017) as well as increased NO_3^- availability for N_2O production through denitrification (Clough et al., 2004). Liming effects on soil CH_4 fluxes are mainly linked to the pH effect on activities of two groups of microorganisms, known as methanotrophs and methanogens. Increasing soil pH through liming facilitates the growth and activity of methanotrophs and subsequently enhances CH_4 uptake by upland soils or CH_4 oxidation activity in predominating anaerobic paddy soils (Barton et al., 2013; Hütsch et al., 1994; Jiang et al., 2018). Conversely, liming has been observed to reduce CH_4 uptake or increase CH_4 emissions from arable and forest soils, due to complex effects of soil pH changes on mineral N and labile C availability as well as on plant growth (Butterbach-Bahl & Papen, 2002; Murakami et al., 2005; Page et al., 2009).

Moreover, liming might affect soil organic C (SOC) concentrations and stocks. Generally, liming is expected to increase soil microbial populations and their activities and hence to increase the mineralization of organic matter. As a result, increases in soil respiration (CO_2) as well as decreases in SOC stocks have been observed (Ahmad et al., 2013; Biasi et al., 2008). Conversely, the liming-induced promotion of plant growth increased C inputs via litter and root exudation, which might outweigh increased C losses due to the SOC mineralization and finally result in increases in SOC storage (Abalos et al., 2020; Liang et al., 2017). Besides, liming ameliorates soil structure and improves soil aggregate stability by strengthening the clay-organic matter bonds, thereby enhancing the efficiency of the physicochemical protection of SOC, while decreasing mineralization rates and microbial respiration (Fornara et al., 2011; Paradelo et al., 2015). Overall, the direction and strength of liming effects on crop productivity, soil GHG fluxes, and SOC stocks have been found to be highly variable. Obviously, other factors such as ecosystem type, management practice (e.g., liming parameters and fertilization conditions), climate conditions, and soil properties are affecting the ecosystem response to soil liming. In the last years, a few reviews assessed the effects of soil liming on crop yields (Li et al., 2019), soil GHG fluxes (Kunhikrishnan et al., 2016), and SOC stocks (Paradelo et al., 2015). However, these studies remained rather qualitatively and did not aim at a quantitative assessment of liming effects upon soil N_2O and CH_4 fluxes and changes in SOC stocks or an integration of findings on GHG fluxes and plant productivity.

In our review, we synthesize how liming affects crop yields and soil GHG fluxes as well as changes in SOC stocks. We also explore if and to what extent ecosystem types, management practices, or soil properties are modulating the response of soil GHG fluxes, changes in SOC stocks, and crop yields to liming. Finally, we test the implication with a simple theoretical experiment where we assume that all acid agricultural soils are limed in a proper way, thereby asking what this might mean for global crop yields, the soil GHG balance and associated food security and climate change.

To address our objectives, we adopt a meta-analysis approach based on 1570 paired individual experimental observations derived from 121 peer-reviewed publications on field experiments.

2 | MATERIALS AND METHODS

2.1 | Literature search and data extraction

We used several databases such as Web of Science, Google Scholar, and the China Knowledge Integrated Database (CNKI) to search peer-reviewed articles (before February 2020) reporting on the effects of liming on soil GHG (i.e., N_2O, CH_4, and CO_2) fluxes, crop productivity, and SOC stock changes. The search terms used for the initial literature screening included 'liming' OR 'soil amendment', AND 'soil', AND 'CO_2' OR 'soil respiration' OR 'N_2O' OR 'CH_4' OR 'soil carbon' OR 'yield' and combinations of those. The retrieved literature was assessed for links to further studies. The resulting database comprised studies which reported on liming effects upon various target variables including crop yields, N_2O emissions, CH_4 emissions, CH_4 uptake, SOC stock changes, soil respiration (CO_2), yield-scaled N_2O emissions, yield-scaled CH_4 emissions, and soil pH. Based on this overview, we applied additional criteria to include studies in our analysis (Dataset S1): (a) only field studies were included, while pot and laboratory experiments under controlled environmental conditions were excluded; (b) the control and liming treatments only differed with regard to the application of lime, but not with regard to other agronomic practices (e.g., cropping intensity, fertilizer management, and irrigation); (c) the studies investigated the effect of liming on at least one of the target variables outlined above; (d) the experimental duration was clearly specified and covered at least a full growing season for soil GHG flux measurements and at least 6 years for SOC determination in order to avoid the effect of short-term noise. If one paper reported multi-year data, we extracted the effect of liming on a parameter for each individual observational year, as the strength of soil liming effects is known to diminish over time. To allow for solid performance of meta-analysis procedures, studies that reported CH_4 emissions (but not CH_4 uptake; i.e., Maljanen et al., 2006; García-Marco et al., 2016) or N_2O uptake (but not N_2O emissions; i.e., Maljanen et al., 2006; Winsborough et al., 2017) from upland ecosystems under liming were excluded from this analysis. The raw data used in our meta-analysis were either obtained directly from tables and texts or extracted indirectly by digitizing graphs using GetData Graph Digitizer (http://www.getdata-graph-digitizer.com/).

Our database finally comprised a total of 121 research papers with 1570 paired observations reporting on liming effects on yields, soil GHG fluxes, or SOC stock changes for various terrestrial ecosystems, which we broadly characterized as upland arable land, grassland and forest, and rice paddy (Dataset S1). Most of the field studies included in our analysis were carried out in Europe, East Asia, and North America, while only a few studies reported on experiments in South America, Australia, or Africa (Figure S1). With regard to soil GHG fluxes, we extracted or calculated annual cumulative fluxes of N_2O (in kg N_2O-N ha^{-1} year^{-1}), CH_4 (in kg CH_4 ha^{-1} year^{-1}), and soil respiration (CO_2, in kg CO_2 ha^{-1} year^{-1}) on the basis of provided information, with unit conversions conducted where necessary. In addition, we also extracted information on crop yields (in Mg dry matter ha^{-1}). Studies that simultaneously reported N_2O and/or CH_4 flux and yield data were used to estimate the effects of liming on yield-scaled N_2O emissions (n = 36, in kg N_2O-N Mg^{-1}) as well as yield-scaled CH_4 emissions (n = 45, in kg CH_4 Mg^{-1}). Similarly, to compare SOC sequestration across studies, we converted reported changes in soil carbon concentrations (% or g C kg^{-1}) over a minimum of six treatment years to estimate annual SOC stock changes (Mg C ha^{-1} year^{-1}), thereby using reported values of soil bulk density (g cm^{-3}), sampling depth (cm), and the duration of the experiment, that is, based on the SOC stock calculation method suggested by Eze et al. (2018) and Liu et al. (2018). In addition to the target variables, other key characteristics (e.g., location, climate, soil properties, ecosystem type, liming parameters, experimental conditions, etc.) were also extracted as far as reported (Dataset S1).

2.2 | Data and statistical analysis

For each side-by-side comparison, the response ratio (RR) of liming is used to calculate effect sizes (Hedges et al., 1999):

$$\mathrm{RR} = \ln\left(\frac{\overline{X_t}}{\overline{X_c}}\right) = \ln\left(\overline{X_t}\right) - \ln\left(\overline{X_c}\right),$$

where $\overline{X_t}$ and $\overline{X_c}$ are the mean values for the given variate in the liming treatment and control, respectively.

The pooled variance (v) of each individual RR is estimated by:

$$v = \frac{S_t^2}{N_t\overline{X_t}^2} + \frac{S_c^2}{N_c\overline{X_c}^2},$$

where N_t and N_c are the replication numbers for the liming treatment and control, respectively, and S_t and S_c are the standard deviation (SD) for the liming treatment and control, respectively. For literature sources where the standard error (SE) rather than SD was reported, we recalculated the SD by:

$$\mathrm{SD} = \mathrm{SE} \times \sqrt{N}$$

where N is the number of replications. When neither SD nor SE data were reported, we contacted the corresponding author and asked for information. Otherwise, SD values were estimated from the average coefficient of variation for the known data (de Stefano & Jacobson, 2018).

To derive the overall response effect of liming relative to the control, the weighted response ratio (RR_{++}) and 95% confidence intervals (CI) were calculated by bootstrapping (4999 iterations) using MetaWin 2.1 software (Rosenberg et al., 2000). Liming effects on a tested variable were regarded as significant at $p < 0.05$, if 95% CI values of the RR_{++} for a variable did not overlap with zero. For improved explanatory power, the mean effect size (i.e., RR_{++}) was transformed back to the percentage change for liming relative to the control, which is calculated by the formula: [exp (RR_{++}) − 1] × 100%.

To identify factors that regulate the direction and magnitude of liming effects on soil GHG fluxes, SOC stock changes, and crop yields, observations were further divided into subgroups according to ecosystem types, liming (e.g., liming rate, liming material, and time since liming) and fertilization (e.g., N fertilization and type) management, and soil properties (e.g., soil texture, soil pH, initial SOC, Total N (TN) and C/N ratio; Table S1; Dataset S1). Linear regression was used to explore if liming-induced changes in SOC stocks, GHG fluxes, and yields were correlated with these factors, thereby differentiating between different ecosystem types. Ecosystem types were categorized into upland (grassland, arable land) and lowland (rice paddy) agricultural ecosystems and upland forests. For each of these groups, between-group heterogeneity (Q_b) tests were used to examine whether different groups showed different responses under liming. Moreover, we tested if Q_b indicates significant differences among categorical groups.

Additionally, frequency distributions of RR were plotted to reflect the variability of liming effects among different studies by a Gaussian function (i.e., normal distribution; Table S2; Figure S2):

$$y = \mathcal{a} \times \exp \frac{(x-\mu)^2}{2\sigma^2}$$

where y is the frequency of RR values within an interval, x is the mean of RR for that interval, μ and σ^2 are the mean and variance of all RR values, respectively, and $\mathcal{a}$ is a coefficient indicating the expected number of RR at $x = \mu$.

Publication bias was tested by the Rosenthal's fail-safe numbers (N_{fs}; Rosenberg, 2005) to assess the robustness of the observed overall effects of liming (Table S3). If $N_{fs} > 5n + 10$ (where n is the number of studies), the result is considered robust despite the possibility for publication bias (Koricheva et al., 2013).

2.3 | Scaling-up estimation

Given the involved liming costs and that, for example, it is rather unlikely that lime will be applied at large scale to forests in the boreal or tropical regions, we limited our assumption to agricultural soils (upland grassland and arable soils as well as lowland rice paddies), and hypothesized that all agricultural soils will be limed in future to counteract acidification and to improve the crop and grassland growth. For estimating the global area with acidic soils currently used for agriculture, we used global land use information as provided by Ramankutty et al. (2008), which was supplemented with more specific information on the global distribution of rice paddies (IFPRI, 2019). This land use information was combined with information on soil properties from the WISE database in 30 by 30 arc-seconds resolution (Batjes, 2016). Based on this information, we estimated that the surface area of agricultural soils with a pH < 6.5 is 6.88 million km^2 (i.e., 0.23, 2.32, and 4.33 million km^2 for soil pH < 4.5, 4.5–5.5, and 5.5–6.5 level, respectively) for upland arable lands, 9.99 million km^2 (i.e., 0.34, 3.57, and 6.08 million km^2 for soil pH < 4.5, 4.5–5.5, and 5.5–6.5 level, respectively) for upland grasslands and 0.68 million km^2 (i.e., 0.009, 0.31, and 0.36 million km^2 for soil pH < 4.5, 4.5–5.5, and 5.5–6.5 level, respectively) for lowland rice paddies (see Figure 1 for the global distribution of land use and pH classes). Though we intensively searched the literature and also contacted

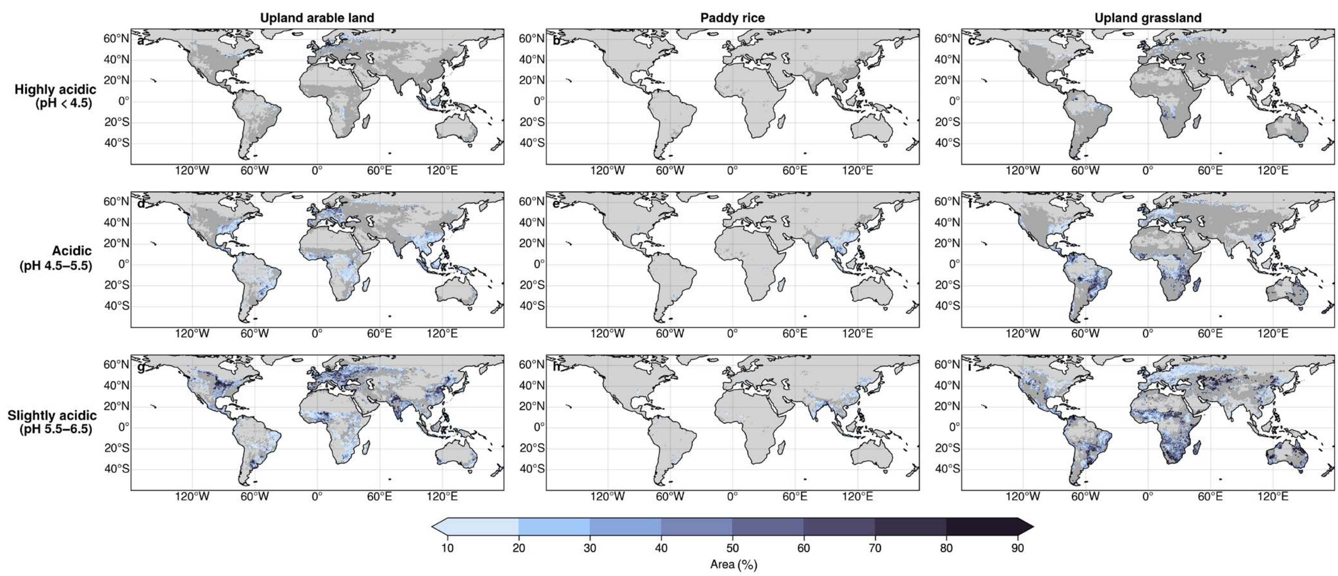

FIGURE 1 Global distribution of agricultural soils (i.e., upland arable land, paddy rice, and upland grassland) with highly acidic (pH < 4.5), acidic (pH 4.5–5.5), and slightly acidic (pH 5.5–6.5) conditions. Shaded areas indicate the spatial extent of the respective land use class for all pH conditions (data filtered to only include areas with >10% coverage for better readability). Information on soils was derived from Batjes (2016), while for land use the dataset of Ramankutty et al. (2008) and IFPRI (2019) for rice paddies were used

industry representatives in Europe and China, we were not able to obtain a robust estimate on agricultural soils currently already receiving liming. For example, the best estimate seems to be that about one third to 50% of all agricultural land in Germany is limed regularly. Given this uncertainty and for simplicity, we assumed in our calculation that acid agricultural soils are generally not limed. Based on the absolute mean positive or negative changes in soil GHG fluxes and the differences in crop yields and SOC stocks under liming, all expressed as area-scaled metrics (*U*-value), we scaled up results of this analysis by multiplying them for target variables with the corresponding liming-applied agricultural areas:

$$\Delta T = \overline{U} \times A$$

where ΔT is the liming-induced changes in target variables such as soil GHG (N_2O, in Tg N_2O-N year^{-1}; CH_4, in Tg CH_4 year^{-1}) fluxes, SOC stock (in Tg C year^{-1}), and crop yield (in Mg year^{-1}), and *A* is the assumed total area of agricultural land with acid soils, which may require liming. For calculating ΔN_2O, ΔCH_4, and ΔSOC as well as crop yield changes (ΔY), we used two methods: (a) Simply multiplying the effect size with the total area of agricultural soils with a pH <6.5, (b) Further discriminating by classes of soil pH using the following categories: highly acidic (pH < 4.5), acidic (4.5–5.5), and slightly acidic (5.5–6.5). To assess the overall importance of liming on the net GHG balance of acid agricultural soils (ΔNGHG, in Tg CO_2-eq year^{-1}), we summed up the liming-induced changes in soil N_2O and CH_4 fluxes as well as SOC stocks, that is, $\Delta NGHG = \Delta N_2O + \Delta CH_4 + \Delta SOC$. For this calculation, we referred to the IPCC global warming potential factors for CH_4 (34) and N_2O (298), respectively, over a 100-year time horizon (IPCC, 2013).

3 | RESULTS

3.1 | Yield response to liming

Overall, liming significantly increased yields by 36.3% (CI: 32.3%–40.5%), with an increase of 40.7% (CI: 34.9%–42.8%) for upland arable lands, 11.9% (CI: 9.26%–14.8%) for rice paddies, and 16.6% (CI: 9.43%–25.3%) for grasslands (Figures 2 and 3). Increases in biomass yields due to liming depended on ecosystem type, rate of lime application, type and number of years after lime application, and on soil properties and N fertilization practices (Table 1). The increase in crop yields was significantly and positively correlated with the RR of soil pH, liming rate, and years since liming application (Figure 3; Figure S3). However, the initial SOC, soil TN content, and C/N ratio showed significantly negative correlations with the response of yields to liming (Figure 3; Figure S3). When the initial SOC was >55 g C kg^{-1}, that is, for SOC-rich soils, liming even led to a reduction of crop yields by 8.88% (CI: −13.9% to −3.92%). All liming materials exhibited significantly positive effects on crop yields. Most positive effects of liming on crop yields were reported for $CaMg(CO_3)_2$ and $CaCO_3$, while application of other liming types such as CaO or $CaSiO_3$ was less effective with regard to increasing yields (Figure 3c). The positive effects of liming on crop and biomass yields of arable land and grassland were higher if fields received N fertilization, with combined liming and fertilization effects being most pronounced if synthetic N fertilizers were used (Figure 3e,f). Also, soil properties such as texture, soil pH, total soil N and C concentration, or soil C/N ratio affected the response of yields to liming (Figure 3g–k).

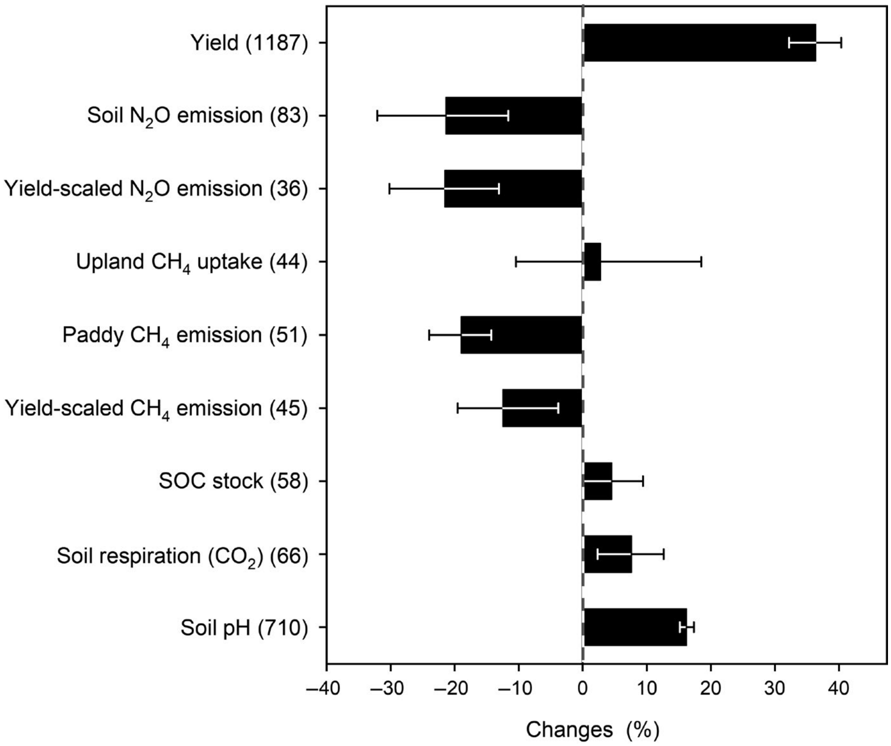

FIGURE 2 Effects of soil liming on yields, annual mean soil nitrous oxide (N_2O) and methane (CH_4) fluxes, yield-scaled N_2O and CH_4 emissions, soil organic C (SOC) stock changes, and soil respiration (CO_2). Number in brackets indicates the number of paired observations. Error bars indicate 95% confidence intervals. Variables were significant at $p < 0.05$, if error bars did not overlap with zero

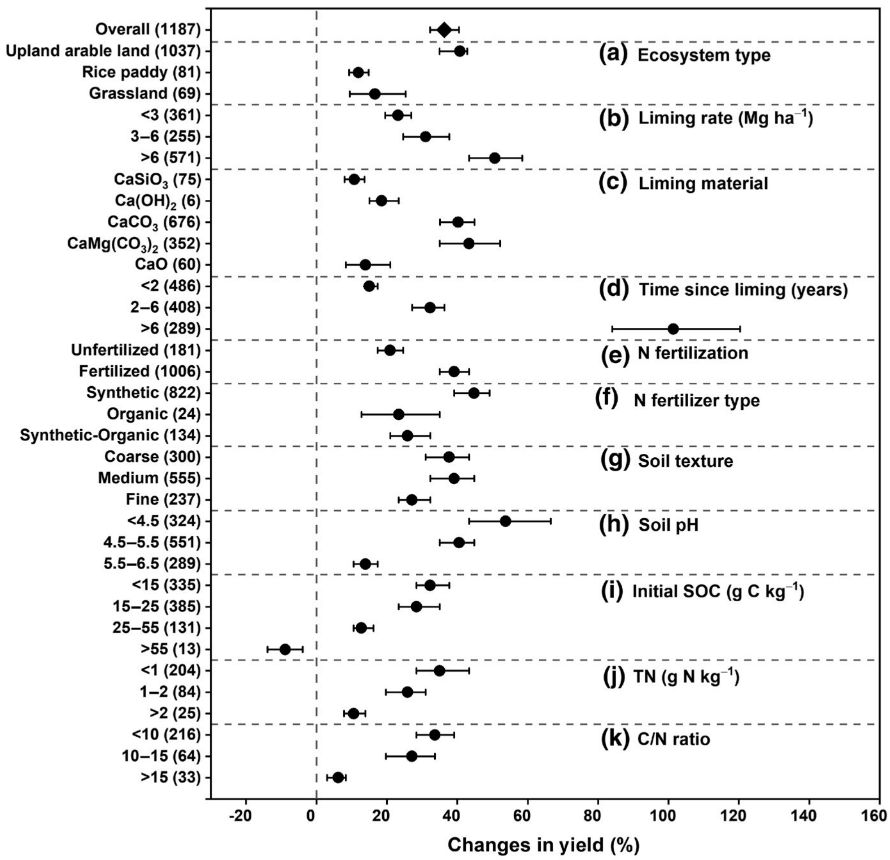

FIGURE 3 Response of crop (biomass for grasslands) yields to liming in dependence of ecosystem type (a), lime application rate (b), type (c) and number of years after lime application (d), as well as on N fertilization practices (e–f) and topsoil properties (g–k). Number in brackets indicates the number of paired observations. Error bars represent 95% confidence intervals. Variables were significant at $p < 0.05$, if error bars did not overlap with zero. SOC, soil organic C; TN, total N

TABLE 1 Effects of liming between group heterogeneity (Q_b) in relation to the response ratios of soil nitrous oxide (N_2O) emission, paddy soil methane (CH_4) emission, upland CH_4 uptake, soil organic C (SOC) stock, soil respiration (CO_2), and crop yield

	N_2O emission		CH_4 emission		CH_4 uptake		SOC stock		Soil respiration (CO_2)		Yield	
Categorical variable	Q_b	*p*-value	Q_b	*p*-value	Q_b	*p*-value	Q_b	*p*-value	Q_b	*p*-value	Q_b	*p*-value
Ecosystem type	2.75	0.25	—	—	0.02	0.89	**4.14**	**0.04**	2.75	0.25	**54.8**	**<0.001**
Liming rate (Mg ha^{-1})	**6.32**	**0.04**	**8.96**	**0.01**	3.33	0.19	1.38	0.50	1.30	0.52	**118**	**<0.001**
Liming material	5.78	0.22	1.88	0.39	1.56	0.46	1.72	0.19	2.69	0.44	**106**	**<0.001**
Time since liming (years)	4.15	0.13	1.07	0.30	2.70	0.26	—	—	**15.5**	**<0.001**	**743**	**<0.001**
N fertilization	2.54	0.11	—	—	0.070	0.79	1.56	0.21	0.45	0.50	**40.0**	**<0.001**
N fertilizer type	3.00	0.08	2.23	0.13	—	—	0.003	0.96	—	—	**32.4**	**<0.001**
Soil texture	0.48	0.78	0.33	0.56	2.20	0.33	1.56	0.21	4.66	0.097	**14.8**	**<0.001**
Soil pH	3.55	0.17	1.41	0.23	1.45	0.48	20.3	**<0.001**	4.45	0.11	**199**	**<0.001**
Initial SOC (g C kg^{-1})	**9.90**	**0.02**	**8.63**	**0.003**	3.77	0.29	**20.8**	**<0.001**	4.30	0.23	**58.5**	**<0.001**
TN (g N kg^{-1})	5.86	0.05	4.07	0.13	0.11	0.94	**22.5**	**<0.001**	5.10	0.078	**36.7**	**<0.001**
C/N ratio	4.26	0.12	**8.68**	**0.013**	1.30	0.52	**4.06**	**0.044**	0.60	0.74	**69.3**	**<0.001**

Note: p-values in bold indicate significance ($p < 0.05$).

Abbreviation: TN, total N.

3.2 | Liming effects on soil GHG fluxes

On average, liming significantly reduced soil N_2O emissions by 21.3% (CI: −31.0% to −10.6%); the negative effect was smaller for paddy soils (−11.6%; CI: −20.6% to 0.21%) than for upland agricultural soils (−20.6%; CI: −40.6% to 8.00%) and forest soils (−31.4%; CI: −47.8% to −9.52%; Figures 2 and 4). As yields generally increase for limed soils, while soil N_2O emissions decrease, a significant reduction was found for yield-scaled N_2O emissions (−21.5%; CI: −30.2% to −13.1%; Figure 2). The magnitude of reductions in soil N_2O emissions due to liming depended on various soil properties, N fertilization regimes, and liming management (Table 1; Figure S4). For example, most significant reductions in soil N_2O emissions were found if >6 Mg lime ha^{-1} was applied, or if soils with a pH < 4.5, initial SOC >55 g C kg^{-1}, TN > 2 g N kg^{-1}, or with a C/N ratio >15 were limed (Figure 4).

Overall, liming of rice paddies also reduced soil CH_4 emissions by 19.0% (CI: −23.7% to −13.9%) and yield-scaled CH_4 emissions by 12.4% (CI: −18.9% to −3.82%; Figures 2 and 5a). Liming management as well as soil properties also affected the magnitude of the response of paddy CH_4 emissions to liming (Table 1; Figure 5a; Figure S5). In contrast, the effects of liming on CH_4 uptake by upland soils were insignificant (2.74%; CI: −10.4% to 18.5%), even if data were further analyzed to possibly identify interactions with ecosystem type, soil properties or liming, and fertilization practices (Figures 2 and 5b). However, linear regression analysis showed that the response of soil CH_4 uptake to liming was significantly and positively correlated with the rate of lime application (Figure S5a), that is, that uptake of atmospheric CH_4 by soils increased following soil liming.

Contrary to reducing rates of upland soil N_2O and lowland soil CH_4 emissions, liming increased soil respiration by on average 13.9% (CI: 6.62–21.5%) and 6.82% (CI: −0.02% to 13.9%) for

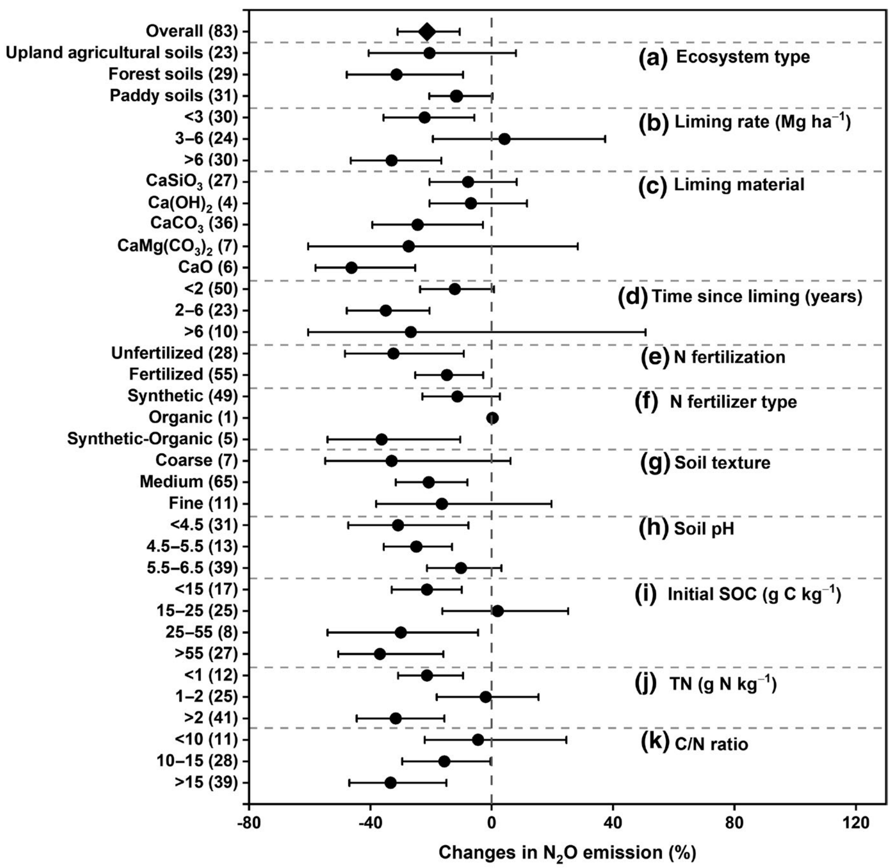

FIGURE 4 Response of annual mean soil nitrous oxide (N_2O) emissions to liming in dependence of ecosystem type (a), lime application rate (b), type (c) and number of years after lime application (d), as well as on N fertilization practices (e–f) and topsoil properties (g–k). Number in brackets indicates the number of paired observations. Error bars represent 95% confidence intervals. Variables were significant at $p < 0.05$, if error bars did not overlap with zero. The datasets from upland arable land and grassland were integrated into the upland agricultural soils grouping category. SOC, soil organic C; TN, total N

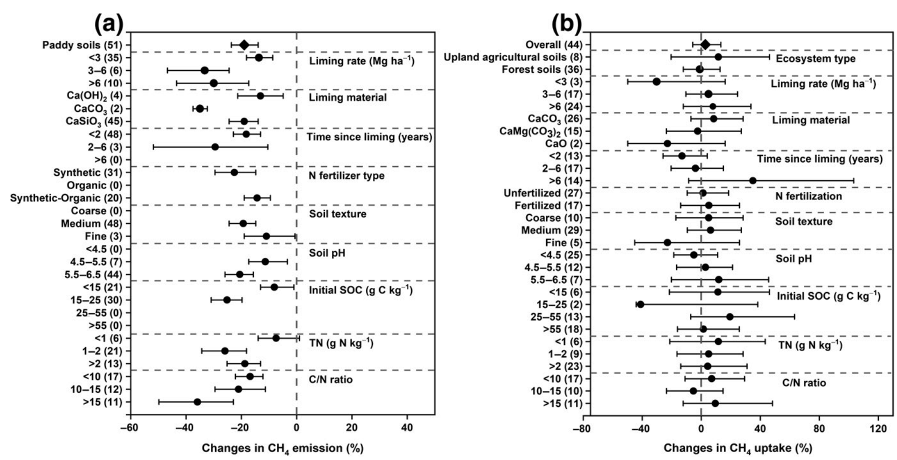

FIGURE 5 Response of annual mean soil methane (CH_4) emissions from paddy soils (a), and CH_4 uptake by upland soils (b) following liming. Number in brackets indicates the number of paired observations. Error bars represent 95% confidence intervals. Variables were significant at $p < 0.05$, if error bars did not overlap with zero. The datasets from upland arable land and grassland were integrated into the upland agricultural soils grouping category. SOC, soil organic C; TN, total N

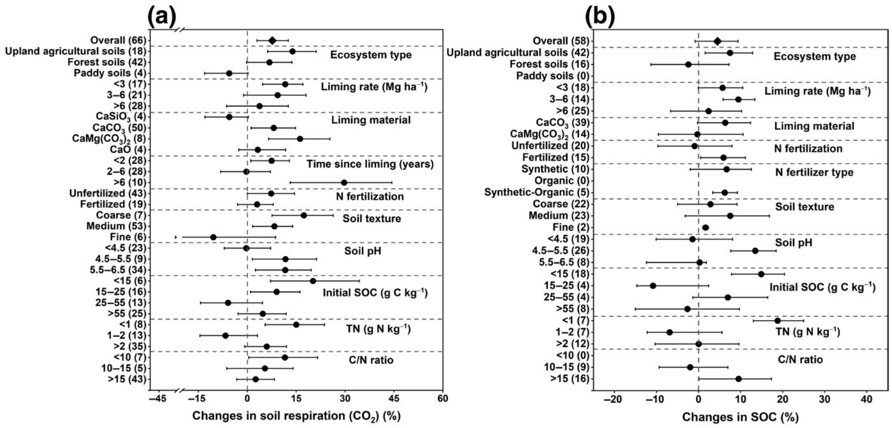

FIGURE 6 Response of annual mean soil respiration (CO_2) (a), and soil organic C (SOC) stocks (b) following liming. For the latter, only reports that effects of liming on SOC stocks were analyzed >5 years following application were used. Number in brackets indicates the number of paired observations. Error bars represent 95% confidence intervals. Variables were significant at $p < 0.05$, if error bars did not overlap with zero. The datasets from upland arable land and grassland were integrated into the upland agricultural soils grouping category. SOC, soil organic C; TN, total N

upland agricultural and forest soils, respectively, while decreasing soil respiration by 5.54% (CI: −11.3% to 2.01%) for rice paddy soils (Figure 6a). The response of soil respiration to liming significantly changed with the number of years since lime application (Table 1). Liming significantly stimulated soil respiration in the year following lime application or in studies where liming was applied >6 years earlier. However, in studies reporting on soil respiration where liming was applied 2–6 years earlier, no significant effect was found. For other management variables such as N fertilization, liming rate, and material, soil respiration was generally stimulated by liming, except that the application of $CaSiO_3$ decreased soil CO_2 emissions. With regard to soil properties, liming of soils with coarse or medium texture significantly increased soil respiration, while liming of fine-textured soils resulted in a decrease in soil respiration (Figure 6a). Furthermore, significant increases in soil respiration were observed when liming was applied to soils with a pH in the range of 4.5–6.5, initial SOC < 25 g C kg^{-1}, TN < 1 g N kg^{-1}, and a C/N ratio <10.

3.3 | Liming effects on SOC stock

To assess whether liming affects SOC stocks of terrestrial ecosystems, we only included long-term field studies, which compared soil C stocks of paired sites earliest 6 years after liming. Across all studies, liming resulted in a small increase in SOC stocks (4.51%; CI: −0.83% to 9.38%), with an increase of 7.48% (CI: 1.56%–12.9%) for upland agricultural soils and a decrease of 2.43% (CI: −11.3% to 7.25%) for forest soils (Figures 2 and 6b). Statistical analysis of between-group heterogeneity showed that the response of SOC stocks to liming was significantly affected by the ecosystem type, soil pH, initial SOC, TN, and C/N ratio (Table 1). As shown in Figure 6b, liming of soils with pH in the range of 4.5–5.5, initial SOC <15 g C kg^{-1}, TN < 1 g N kg^{-1}, or with a C/N ratio >15 resulted in significant increases in SOC stocks. However, for soils with highly acidic (pH < 4.5) or slightly acidic (5.5–6.5) conditions, larger initial SOC (≥15 g C kg^{-1}), TN (≥1 g N kg^{-1}), or with a C/N ratio in the range of 10–15, no significant positive effect of liming on SOC stocks was observed, or liming even resulted in a decline in SOC stocks. Moreover, the response of SOC stocks to liming also varied with various liming and fertilization practices (Figure 6b).

3.4 | Liming-induced net changes in GHG fluxes and crop yields on a global scale

Assuming that all acid agricultural soils including upland arable and grassland ecosystems and lowland rice paddies would receive liming amendment, global soil N_2O emissions may decrease by 0.60–0.67 Tg N_2O-N $year^{-1}$ for upland agricultural soils and by 0.0015–0.002 Tg N_2O-N $year^{-1}$ for rice paddies (Table S4; Figure S6). For the same scenario, liming of all acid paddy soils would reduce soil CH_4 emissions by 1.75–2.21 Tg CH_4 $year^{-1}$, while liming of acid upland agricultural soils would increase soil CH_4 uptake by 0.06–0.13 Tg CH_4 $year^{-1}$. Moreover, liming of acid upland agricultural soils would increase SOC stocks by 65.7–110 Tg C $year^{-1}$. Overall, our estimates reveal that liming of all acid agricultural soils at global scale would improve the net GHG balance by up to 633–749 Tg CO_2-eq $year^{-1}$ due to (a) decreasing soil N_2O emissions, (b) decreasing CH_4 emissions from rice paddies, and (c) increasing SOC stocks (Table S4; Figures S6). Moreover, we estimated that liming of all acid agricultural soils would increase yields by 6.64×10^8–14.1×10^8 Mg $year^{-1}$, consisting of 5.21×10^8–7.70×10^8 Mg $year^{-1}$ for upland arable lands, 0.48×10^8–0.56×10^8 Mg $year^{-1}$ for rice paddies and of 0.87×10^8–5.90×10^8 Mg $year^{-1}$ for grasslands (Tables S5 and S7; Figure 7; Figure S7).

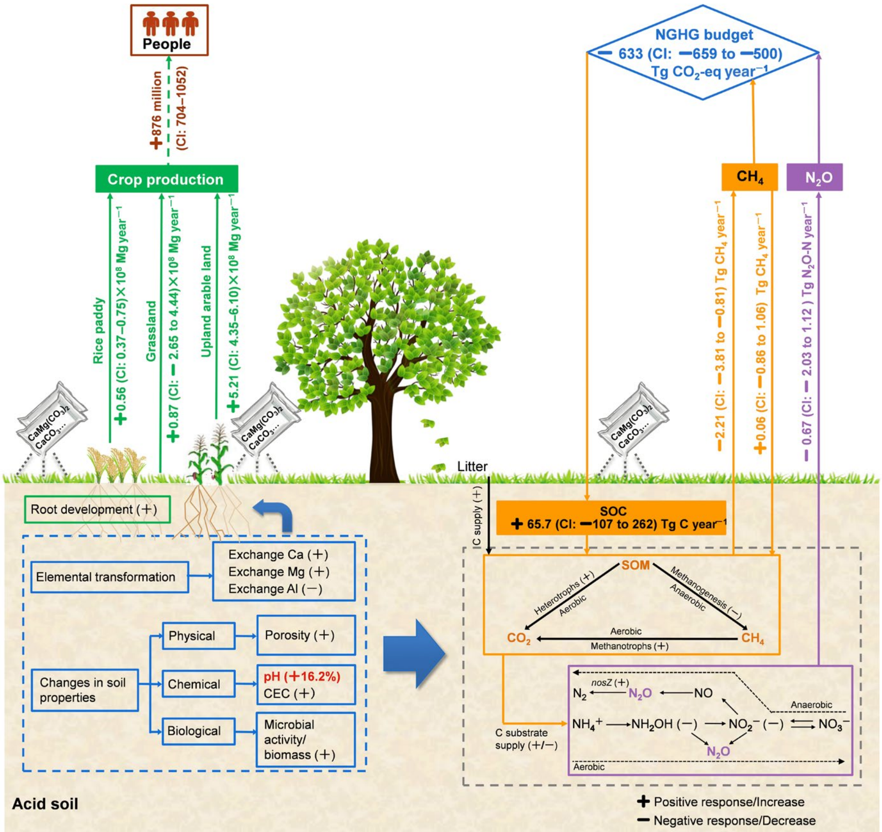

FIGURE 7 Conceptual diagram illustrating liming-induced changes in soil processes and associated changes in soil organic C (SOC) stocks, soil nitrous oxide (N_2O) emissions, soil methane (CH_4) fluxes, and crop production. Here liming-induced changes in SOC stocks, soil N_2O and CH_4 fluxes, and crop yields were calculated based on the areal extent of agricultural soils with highly acidic (pH < 4.5), acidic (pH 4.5–5.5), and slightly acidic (pH 5.5–6.5), that is, the area-weighted estimates. Net greenhouse gas (NGHG) fluxes were estimated by summing up the effects of liming on soil N_2O and CH_4 fluxes and SOC stock changes based on the 100-year horizon global warming potential factor of 298 for N_2O and 34 for CH_4 (IPCC, 2013). Numbers in brackets represent 95% confidence intervals (CI). CEC, cation exchange capacity; SOM, soil organic matter

4 | DISCUSSION

4.1 | Lime application increases plant productivity

Overall, our results show that liming of acid soils and associated increases in soil pH (on average 0.9 pH units in our study; Figure 2; Dataset S1) stimulates plant growth and yields (Figure 3). The increase in plant growth has been linked to liming-induced increases in the availability and mobility of plant nutrients (e.g., Ca^{2+}, Mg^{2+}, and K^+) and reductions or removal of Al^{3+} toxicity (Goulding, 2016; Holland et al., 2019; Li et al., 2019). Our study also shows that the magnitude of yield increases was strongly modulated by the quality and quantity of liming materials used as well as number of years following liming application. Several studies show that after several years following high rates of lime application, the soil physical structure is improved, which not only facilitates root growth, but also promotes soil nutrient retention (Basu et al., 2008; Fageria & Baligar, 2008). This also explains why in our meta-analysis maximum yield responses were observed at lime application rates of >6 Mg ha^{-1} and >6 years after lime application (Figure 3b,d). Our meta-analysis also showed that there was a significant negative relationship between the response of yields to liming and the initial concentrations of SOC and TN or the C/N ratio (Figure S3), that is, that the magnitude of yield increases in response to liming decreased with increasing concentrations of SOC and TN. We assume that soils with higher concentrations of SOC and TN already do have a high soil cation exchange capacity as well as a high pH buffering capacity (Briedis et al., 2012; Higgins et al., 2012; Li et al., 2019), so that additional lime application only results in minor improvements in soil physicochemical properties with regard to plant growth. Accordingly, liming is also more effective with regard to yield improvements in coarse- to medium-textured soils (Figure 3g). For light-textured soils, liming increases subsoil pH due to the transport of carbonates with the soil water percolation stream, which as well contributes to improved nutrient supply to plants and yield increases (Meng et al., 2004).

4.2 | Liming decreases soil N_2O and CH_4 emissions and increases SOC stocks

In our meta-analysis, we found that on average, liming significantly reduced soil N_2O emissions by 21.3% (Figure 4). Liming-induced increases in soil pH are known to significantly affect key soil N transformation processes involved in N_2O production, namely nitrification and/or denitrification (Barton et al., 2013; Holland et al., 2018; Stevens et al., 1998; Wang et al., 2018). For example, at increased soil pH, the conversion of NO_2^- to NO_3^- is facilitated, which reduces the concentration of NH_2OH and NO_2^- in soils, both well-known precursors of N_2O formation (Barton, Gleeson, et al., 2013; Vázquez et al., 2020). More generally, liming has been found to stimulate nitrification activity in soils and, thus, NO_3^- production. The increased availability of soil NO_3^- might finally lead to increased N_2O production by denitrification (Clough et al., 2004). At increasing soil pH the product stoichiometry of denitrification has been found to favor N_2 production. The reason for this is that at higher pH values the assembly of the final enzyme in the denitrification chain, that is, the

N_2O reductase (*nosZ*), which catalyzes the reduction of N_2O to N_2, is facilitated (Liu et al., 2010; McMillan et al., 2016). Consequently, even at stimulated denitrification rates, N_2O production by denitrification might decrease on the costs of increased N_2 production in limed soils (Abalos et al., 2020; Qu et al., 2014; Raut et al., 2012; Šimek & Cooper, 2002). Our observation that the response of soil N_2O emissions to liming negatively correlates with the rate of lime application (Figure S4) indirectly supports the abovementioned soil pH-regulating mechanisms on N_2O production in soils, as high rates of lime application usually results in significant increases in soil pH. Also, higher plant productivity under liming and hence increased plant N uptake and reduced N availability in soils might contribute to the observation of reduced N_2O emissions from limed soils (Abalos et al., 2020).

It is well accepted that CH_4 fluxes from soils are a product of the balance between CH_4 production and oxidation, that is, the balance between soil methanogenic and methanotrophic activities (Le Mer & Roger, 2001; Nazaries et al., 2013). Nevertheless, some studies reported that different types of CH_4-oxidizing microorganisms, that operate in different soils, displayed varying sensitivity to changes in soil pH (e.g., Page et al., 2009; Sitaula et al., 1995). Several studies found that the growth and activity of methanotrophs in soils are highly sensitive to changes in pH, with the soil CH_4 oxidation capacity being highest in pH neutral soils (Brumme & Borken, 1999; Knief et al., 2003; Zheng et al., 2012). However, other studies found that the soil methanotrophic population is relatively insensitive to low pH values or shows high resilience to acidic conditions, so that liming-induced increases in soil pH resulted in little or no changes in CH_4 oxidation rates (Klemedtsson & Klemedtsson, 1997; Saari et al., 2004). In our meta-analysis, liming showed an insignificant effect on soil CH_4 uptake by upland ecosystems, which reflects the controversy on pH effects on soil methanotrophic activity outlined above.

In contrast to the insignificant effects of liming on upland CH_4 fluxes, our study reveals that liming of lowland paddy soils significantly reduces net CH_4 emissions from these systems by 19.0% (Figure 5a). These reductions in CH_4 emissions go along with increasing rice yields (Figure 3). While neither hydrogenotrophic nor aceticlastic methanogenesis has been found to be specifically pH sensitive (Conrad, 2020), the reductive effect of liming on CH_4 emissions is likely linked to increased CH_4 oxidation. Increased plant growth not only stimulates root C exudation and consequently methanogenesis (Zheng et al., 2006), but may also stimulate oxygen transport into the rhizosphere via the aerenchym (Butterbach-Bahl et al., 1997), thereby increasing the net consumption of CH_4 in the rhizosphere (Conrad, 2007; Jiang et al., 2018). We assume that the latter effect prevails and that this explains the observation that liming reduces CH_4 emissions from acidic paddy soils. Also, as previously discussed, liming has the potential to improve soil structure and to increase the abundance of soil macropores. This is particularly important at times of paddy soil drainage, as soils with a high abundance of macropores drain quicker and, thus, turn aerobic faster as compared to soils with a lower number of macropores. Increasing soil O_2 availability hampers CH_4 production and turns soils from a net source into a net sink for atmospheric CH_4 (Borken & Brumme, 1997; Butterbach-Bahl & Papen, 2002; Page et al., 2009).

While liming greatly enhanced soil respiration (CO_2 emission), the overall effect on soil C stocks in paired studies comparing limed and unlimed soils for >5 years was positive (4.51%; Figure 2). Theoretically, changes in SOC are closely associated with the balance between C inputs (e.g., through plant biomass) and C losses (e.g., via organic matter decomposition), with both processes being strongly affected by changes in soil pH. As discussed above, liming significantly increases plant productivity. This stimulates C inputs into the soil profile and soil respiration, but may as well lead to increasing SOC stocks (Egan et al., 2018; Kunhikrishnan et al., 2016). However, while liming stimulated soil respiration from upland soils, for example, due to increased microbial growth and plant litter inputs (Neale et al., 1997; Paradelo et al., 2015), we also found that soil respiration from limed rice paddy soils had the tendency to be reduced (Figure 6a). In agreement with the argumentation by Hamilton et al. (2007), we assume that the negative effect of liming on paddy soil respiration is due to (a) the higher pH buffering capacity of lowland as compared to upland soils, which reduces the positive effect of lime application on soil pH, and (b) generally lower rates of organic matter decomposition under anaerobic conditions. Besides, with regard to the positive effect of liming on SOC stocks, many studies also argue that liming promotes incorporation of C in soil clay-organic complexes, which protects C from decomposition, and is a direct link to the liming-induced amelioration of soil structure and increase in soil aggregate stability (Fornara et al., 2011; Kunhikrishnan et al., 2016; Paradelo et al., 2015). However, increases in SOC stocks will only last for 10–20 years, that is, until a new equilibrium between soil C input an soil respiration is reached (Stewart et al., 2007; Tian et al., 2015).

4.3 | Global GHG and crop production benefits due to liming acid soils

To meet the Paris Agreement's goal of limiting the increase in average global temperature to 1.5 or 2°C above preindustrial levels and the second Sustainable Development Goal (SDG2) of achieving zero hunger by 2030, mankind is facing substantial challenges in reducing GHG emissions from the agricultural sector while meeting basic nutritional needs. Our synthesis shows that liming of acid soils reduces soil N_2O and paddy CH_4 emissions, while supporting atmospheric CH_4 uptake by upland soils and stabilization and increases in SOC stocks (Figure 2). To assess the global effect of liming on soil GHG fluxes from agricultural soils and crop and biomass yields, information on the magnitude of soil liming is required. Surprisingly, we were not able to find data about the fraction of acid agricultural soils getting limed periodically. For some countries, such as Germany, we found evidence that at least 50% of all agricultural soils in the state of North-Rhine Westphalia show a significant deficiency of lime application (Jacobs, 2012). Also for the United Kingdom, Goulding (2016) reported that 40% of all arable soils and 57% of grassland soils are in urgent need for lime application as their pH is below the critical threshold value of 6.5 (arable soils) or 6.0 (grassland soils), respectively. Given the difficulty to find tangible information, we used the rather broad assumption that all acid agricultural soils will be limed in the future. Based on the two calculation methods (one further stratifying acidic soils in different pH categories, while for the other we broadly scaled effects to all agricultural soils with a pH < 6.5), we conclude that if all acid agricultural soils would be limed, soil N_2O and CH_4 emissions would decrease by about 0.60–0.67 Tg N_2O-N year^{-1} and 1.75–2.21 Tg CH_4 year^{-1}, respectively (Table S4; Figure 7). Furthermore, this would lead to an increase in soil CH_4 uptake and SOC stocks by about 0.06–0.13 Tg CH_4 year^{-1} and 65.7–110 Tg C year^{-1}, respectively. Recently, Tian et al. (2020) estimated N_2O emissions from global agricultural soils at an average of 2.3 (ranging from 1.4 to 3.8) Tg N_2O-N year^{-1}. Accordingly, liming of all acid agricultural soils would reduce the reported soil N_2O emissions by 26.1%–29.1%. Similarly, appropriate liming of acid paddy soils would reduce the global source strength of rice paddies for CH_4 (36 Tg CH_4 year^{-1}, IPCC, 2013) by 4.9%–6.1%. Aggregating all liming effects on the soil GHG balance, that is, the sum of achievable reductions in CH_4 and N_2O emissions plus the positive effects on upland CH_4 uptake and SOC stocks, results in a total GHG balance benefit of up to 633–749 Tg CO_2-eq year^{-1} (Table S4; Figure 7). On basis of a modeling study, Zhang et al. (2014) estimated the overall soil GHG balance of Chinese agricultural soils to be 296 Tg CO_2-eq year^{-1} between 2005 and 2009. By using both bottom-up and top-down approaches, Tian et al. (2016) estimated the overall biogenic GHG balance of global terrestrial biosphere between 2001 and 2010, and concluded that the terrestrial biosphere is a net source, with the net GHG emission ranging from 3900 to 5400 Tg CO_2-eq year^{-1} (mean: 4650 Tg CO_2-eq year^{-1}). Hence, our upscaled liming-induced mitigation of net GHG fluxes from all acid agricultural soils globally has the potential to completely negate the GHG source strength of Chinese croplands, and to counteract 13.6%–16.1% of the GHG source strength from global terrestrial biosphere. However, our synthesis focused solely on the changes in net GHG fluxes via biological pathways following liming of acid agricultural soils, while neglecting liming-derived CO_2 release from chemical processes such as lime dissolution. Several studies found that CO_2 emissions originating from the chemical dissolution of applied lime are significant at global scale (e.g., Page et al., 2009; Zamanian et al., 2018). However, part of the H_2CO_3 in the soil solution as derived from lime dissolution may finally end in recalcitrant soil C pools in deeper soil layers (Hamilton et al., 2007) as liming combined with fertilization has been found to favor the storage of C recalcitrant pools and even promotes the transformation of stored C from more labile to more recalcitrant pools (Manna et al., 2007). Additionally, part of the dissoluted H_2CO_3 may finally end up as lime due to the calcination of limestone or will be transported to the ocean where it may precipitate and dissolute (West & McBride, 2005).

The calculation of global rates of CO_2 release from lime application is further complicated by assumptions on lime application rates, which do depend on the acid-neutralizing capacity of the liming material and the pH buffering capacity of the soil (Kunhikrishnan et al., 2016). Also, information on global amounts of lime applied to agricultural soils is not available. Therefore, we assumed the following: (a) the total area of agricultural soils with a pH < 6.5 is 17.6 million km^2 (Tables S4 and S5; Figure 1), and (b) that the average amount of lime application to acid agricultural soils needed to combat detrimental low pH effects on crop growth is 1 Mg ha^{-1} year^{-1} on average (or 5 Mg per hectare every 5 years; West & McBride, 2005; Zamanian et al., 2018). The production, transport, and spreading of 1 Mg lime to agricultural soils entails about 38 kg CO_2-C (West & McBride, 2005). The mass fraction of C in limestone and dolomite is 0.12–0.13% (or 120–130 kg C per Mg crushed rock), and the study of West and McBride (2005) suggests that the net CO_2 release due to dissolution of applied lime is about 49%. This means that an additional CO_2 emission of 59–64 kg C per Mg of lime applied needs to be considered, totaling to 97–102 kg CO_2-C emissions associated with 1 Mg of lime application to acid agricultural land. Scaling this with the global area covered with acidic agricultural soils (i.e., 17.6 million km^2) results in a total global emission of 170–179 Tg CO_2-C year^{-1} (or 624–656 Tg CO_2 year^{-1}) due to lime production and application. Thus, the CO_2 release associated with the application of lime to acidic agricultural soils approximately equals the net GHG savings due to the positive effects of lime applications with regard to reductions in soil N_2O and CH_4 emissions and increases in SOC stocks, which were within the range of 500–774 Tg CO_2-eq year^{-1} (Table S4).

While according to our assumptions and calculations there might be no net GHG benefit due to the liming of agricultural soils, there is still a significant net benefit regarding agricultural production. Our calculations show that liming of agricultural acid soils would result in a global increase of upland crop yields by 5.21 × 10^8–7.70 × 10^8 Mg year^{-1}, rice yields by 0.48 × 10^8–0.56 × 10^8 Mg year^{-1}, and grass production by 0.87 × 10^8–5.90 × 10^8 Mg year^{-1} (Table S5; Figure S7). Based on FAOstats data (FAO, 2013), the average food demand per capita over the world is about 528 kg cereal and 133 kg meat (Crusciol et al., 2019) annually. Thus, our estimated liming-induced increment in crop and biomass production has the potential to feed nearly 876–1261 million people in the future (Table S5; Figure 7). Overall, our meta-analysis indicates that liming of acid soils (especially soils with coarse and medium texture) is a feasible measure, which allows to address the challenge of land degradation and food security, thereby being overall climate neutral. However, such measures should be carried out with caution, as for example, for fine-textured soils with high pH buffering capacity resulting from high SOC, liming had no significant or even reverse effect on plant productivity (Higgins et al., 2012), and over-liming could decrease the nutrient availability for plant metabolic processes and increase the susceptibility to crop diseases (Holland et al., 2018).

4.4 | Uncertainties and implications for future studies

While using a systematic synthesis approach to simultaneously examine liming effects on soil GHG fluxes and crop yield, the calculated response factors include uncertainties originating from limitations of field experiments, but also from statistical methods used. First, some field studies reporting on soil GHG fluxes in response to liming were short term and only covered one or two growing seasons. This will result in an unquantified uncertainty of calculated annual GHG fluxes (Liu et al., 2014). Second, uncertainties are also associated with simple extrapolation approach as we only use the global area of the acid soils (Van Groenigen et al., 2011), but not consider, for example, the severeness of acidification or other factors which may limit the plant growth (e.g., climate conditions). Moreover, most reported liming studies were carried out in Europe, East Asia, and North America, while for important agricultural production regions in South America and Africa, where 33.8% of all global acid soils are located (Zamanian et al., 2018), no or little information is available (Figure S1). Although we do know that liming effects on the composition and functioning of the soil microbial community remains severely understudied, new tools and techniques (e.g., isotopes, metagenomics) are available to allow to gain an in-depth understanding of the underlying mechanisms and to develop targeted liming strategies. Finally, it is noteworthy that liming effects are generally not included in biogeochemical and agroecosystems models, which hamper the applicability of these models for scenario studies to predict terrestrial ecosystem feedback to agricultural management changes and to identify innovative climate change-related mitigation and adaption strategies.

ACKNOWLEDGMENTS

The authors sincerely acknowledge the scientists for their various contributions to the dataset used in this meta-analysis. This work was financially supported by the National Natural Science Foundation of China (41977282, 41675144, and 41807327) and the Chinese Academy of Sciences (ZDBS-LY-DQCOO7).

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available in the supplementary material of this article.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

How to cite this article: Wang Y, Yao Z, Zhan Y, et al. Potential benefits of liming to acid soils on climate change mitigation and food security. *Glob Change Biol*. 2021;00:1–15. https://doi.org/10.1111/gcb.15607