



Acidification of European croplands by nitrogen fertilization: Consequences for carbonate losses, and soil health

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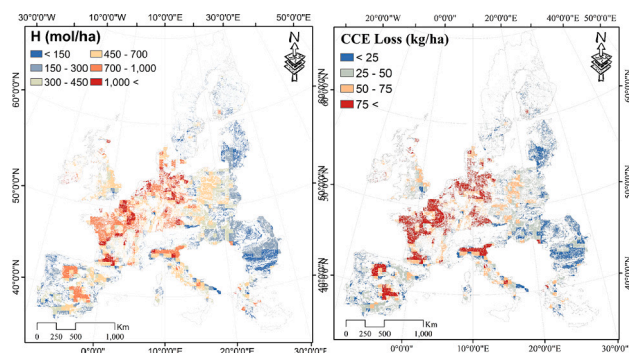
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HIGHLIGHTS

- Nitrogen fertilization annually adds 6.1×10^7 kmol H⁺ to European croplands.
- Strong acidification of 2.6×10^6 ha of European croplands within the next 50 years
- Lost soil carbonate stocks by acidification correspond to 1.5 % of EU budget for 2023.

GRAPHICAL ABSTRACT



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ABSTRACT

Soil acidification is an ongoing problem in intensively cultivated croplands due to inefficient and excessive nitrogen (N) fertilization. We collected high-resolution data comprising 19,969 topsoil (0–20 cm) samples from the Land Use and Coverage Area frame Survey (LUCAS) of the European commission in 2009 to assess the impact of N fertilization on buffering substances such as carbonates and base cations. We have only considered the impacts of mineral fertilizers from the total added N, and a N use efficiency of 60 %. Nitrogen fertilization adds annually 6.1×10^7 kmol H⁺ to European croplands, leading to annual loss of 6.1×10^9 kg CaCO₃. Assuming similar acidification during the next 50 years, soil carbonates will be completely removed from 3.4×10^6 ha of European croplands. In carbonate-free soils, annual loss of 2.1×10^7 kmol of basic cations will lead to strong acidification of at least 2.6 million ha of European croplands within the next 50 years. Inorganic carbon and basic cation losses at such rapid scale tremendously drop the nutrient status and production potential of croplands. Soil liming to ameliorate acidity increases pH only temporarily and with additional financial and environmental costs. Only the

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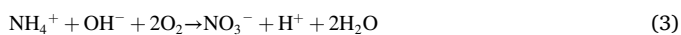
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direct loss of soil carbonate stocks and compensation of carbonate-related CO₂ correspond to about 1.5 % of the proposed budget of the European commission for 2023. Thus, controlling and decreasing soil acidification is crucial to avoid degradation of agricultural soils, which can be done by adopting best management practices and increasing nutrient use efficiency. Regular screening or monitoring of carbonate and base cations contents, especially for soils, where the carbonate stocks are at critical levels, are urgently necessary.

1. Introduction

Acidification of agricultural soils is an ongoing process, which happens mostly because of fertilization, in particular by inefficient utilization of ammonium-based nitrogen (N) fertilizers (Goulding, 2016; Tian and Niu, 2015; Wu et al., 2022). Annually, about 110 million tons of N mostly as ammonium-base fertilizers such as urea is added to the global agricultural soils (FAO, 2022), where through nitrification process, ammonium turns to nitrate, and concomitantly releases protons into the soil (Eq. 1–3). There will be no net soil acidity if plants take up all of the nitrate, because roots have to release equivalent amounts of hydroxyl ions to the soil solution to keep the plant's internal charge density balanced (Bolan et al., 1991; De Vries and Breeuwsma, 1987; Guan et al., 2022).



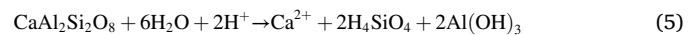
N use efficiency is on average about 50 % in the whole world and about 60 % in the European croplands (Zhang et al., 2015). This means 50 % of the unconsumed fertilizer N provides a strong source of soil acidity (Eq. 3) (Dal Molin et al., 2020; Hao et al., 2020; Zhang et al., 2021a). Even if crops take up all N in form of ammonium, it will still acidify the soils because roots will release the equivalent amount of protons to the rhizosphere (Custos et al., 2020). Despite very-well known consequences of acidification for vegetation growth and human nutrition, the extend of such effects on a large scale and on the stability of the whole ecosystem are missing. This is of crucial importance to see the drawbacks of soil acidification on continental or global scales instead of limiting to the farm scales.

Soils counteract acidification through various buffering substances, such a carbonate minerals (Bowman et al., 2008; Raza et al., 2020), which are dissolved and lost mainly as CO₂ during acidity neutralization (Eq. 4) (Sharififar et al., 2023). As a result, inorganic carbon stocks gradually decrease, and can be completely exhausted under continuous acidification (Tao et al., 2022; Wu et al., 2022). Besides being a strong buffering substance (de Vries et al., 1994; Li et al., 2023; Zhu et al., 2018), carbonates directly and indirectly (via adjusting pH or increasing concentration of base cations in solution) increase soil aggregation (Fernández-Ugalde et al., 2011; Safar and Whalen, 2023) and the stability of soil organic matter (Rowley et al., 2018, 2020), control the activity and structure of microorganisms (Lauber et al., 2009; Li et al., 2018; Schlatter et al., 2020) as well as their biodiversity (Guo et al., 2010; Pabian and Brittingham, 2012), and finally determine nutrients cycling and availability (De Vries and Breeuwsma, 1987; Kemmitt et al., 2006; Zheng et al., 2021). Therefore, carbonate losses due to acidification can adversely affect soil health and productivity (Zamanian et al., 2021; Zamanian and Kuzyakov, 2022; Raza et al., 2024).



In carbonate-free soils or following complete exhaustion of carbonate minerals by acid neutralization, silicates (e.g. Eq. 5) and exchangeable base cations on surface of clay minerals and organic compounds (Eq. 6) are dominant buffering substances. Over the time while buffering pH value between 6.5 down to about 4.5 (Li et al., 2023), the percentage of basic cations such as calcium (Ca²⁺), magnesium (Mg²⁺) and

potassium (K⁺) on the exchangeable sites, defined as base saturation (Eq. 7) declines and the easily weatherable minerals disappear (Haynes and Swift, 1986; Lucas et al., 2011; Tian and Niu, 2015; Yu et al., 2020). Note that the lost cations cannot generally be compensated via cations released by silicates weathering over decadal to centennial periods as weathering of silicates, in contrast to carbonates is kinetically too slow (Cecchini et al., 2019; Fang et al., 2023; Slessarev et al., 2016). Since these cations are nutrients, the crop growth and the yield, especially when the pH value falls below 5.5 will considerably decrease (Haynes and Swift, 1986; Pan et al., 2020). At pH values about 5.5 or less, aluminum (Al) toxicity is a common problem (Eq. 8) (Schroder et al., 2011; Zhu et al., 2018). Presence of free Al³⁺ ions in soil solution prevents root elongation (Haling et al., 2011; Matzner et al., 1986; Yang et al., 2013), where a weak rooting system declines nutrients and water uptake and consequently the crop growth. Furthermore, the concentration of Al³⁺ and protons increases on exchangeable sites, decreases microbial activity, soil respiration and nutrients cycling (Blake et al., 1999; De Vries and Breeuwsma, 1987; Kemmitt et al., 2006; Li et al., 2018; Liu et al., 2022a; Xing et al., 2022). In addition, the toxicity of various ions like Fe³⁺ (Fageria et al., 2008) as well as heavy metals released at low pH such as cadmium and lead (Vanstraelen and Bergema, 1995; Zhang et al., 2021b; Zhao et al., 2020) may decrease crops yield and quality.



(X is the exchanger site, and M is an exchangeable cation with valence n)

Base saturation (BS, milli equivalent in 100 g soil)

$$= \text{Sum of base cations/potential soil cation exchange capacity} \quad (7)$$



Liming, i.e. addition of crushed limestone or other materials with liming effect such as biochar are usually recommended to increase soil pH (Dai et al., 2017; Wang et al., 2021). However, the financial capability to provide lime and labor to distribute that on the field may still prevent proper liming management (Goulding, 2016). Furthermore, neutralization of lime in soil is a considerable CO₂ source with consequences for global warming (Kuzyakov et al., 2021; Tao et al., 2022; Wang et al., 2021; Zamanian et al., 2018, 2021).

Soil acidification is thus a considerable global threat for sustainable crop production, which directly endangers food security (Zhu et al., 2020). Therefore, quantifying the acidification rate, and subsequent losses of carbonates and base cations are vital for devising policies and control measures. The main goals of this study are (i) to determine the rate of losses in acid neutralization capacity of soils at large-scale, i.e. within the European countries, (ii) to predict and locate the areas and soils, which are susceptible to complete decalcification or decreasing BS to less than 50 % in the next 50 years upon current N fertilization rates, and (iii) to determine the potential amount of CO₂ emission from the lost carbonates or the applied lime. The soils in countries of the European Union (EU) are chosen as the study area, because long-term intensive land-use with high N fertilization rates have already imposed severe degradation to the soils (Serebrennikov et al., 2020), which are of highly importance for the global nutrition. In addition, a large number of point measurements over the continent, including all the data needed for this

research, are available by Land Use and Coverage Area frame Survey (LUCAS) of the European commission (Orgiazzi et al., 2018).

2. Materials and methods

We employed geospatial analysis techniques to assess the impact of nitrogen fertilization on agricultural croplands across the European Union (EU) countries. Our investigation comprised several significant stages (Fig. 1). Initially, we identified and delimited the EU's cropland areas, which allowed us to focus our analysis on the most relevant regions for our research questions. Secondly, we classified the soils within the study area into two categories based on their calcium carbonate content: carbonate-containing and carbonate-free soils. Thirdly, by considering N fertilization rates to both soil types, we evaluated various consequences of losses of calcium carbonate (i.e., soil inorganic carbon or SIC), changes in the area of carbonate-containing and carbonate-free soils, and reduction rates of base saturation, which can result in the development of acid soils with a pH value below 5.5. Moreover, to anticipate the long-term consequences of N fertilization on these soils, we conducted simulations spanning 50 years, assuming a constant fertilization rate as present. These simulations allowed us to assess the potential cumulative effects of N fertilization on soil acidification in the EU.

2.1. Study area

The 27 countries of the European Union (EU) and Great Britain cover an area of approximately 4.4 million square kilometers. The climate in EU is mainly temperate with mild to cold winters and warm summers, where the mean annual precipitation is around 300 mm in some parts of the Mediterranean region, to over 2000 mm in certain areas of the Alps and Scandinavia. The soils in EU exhibit remarkable variability, spanning from sandy soils in northern Europe to loamy soils in central and

eastern Europe. Moreover, the EU soils have various physical, chemical, and biological properties, highlighting the importance of assessing the effects of agricultural practices, such as nitrogen fertilization, on soil quality and sustainability. The dominant land cover types across the EU countries consist of forests, croplands, grasslands, wetlands, urban areas, and water bodies (Büttner, 2014). The agricultural lands in the EU are essential for producing a wide range of crops, including cereals, vegetables, and fruits, and have been subjected to intensive nitrogen fertilization (van Egmond et al., 2002).

It is worth noting that our study area was limited to cropland area covering 135 million ha in 27 EU Member States and Great Britain and was based on the available maps of topsoil (0–20 cm) properties at the European scale (Ballabio et al., 2019) and the spatial patterns of global fertilizer application (Potter et al., 2010). As shown in Fig. 2, the agricultural lands were later classified into two categories based on their calcium carbonate content in the upper 20 cm: carbonate-containing and carbonate-free soils.

2.2. Datasets

The objective of this study was to evaluate the impact of N fertilizers on agricultural lands within the EU by utilizing a variety of freely available geospatial data sources. Table 1 and Fig. 3 present the datasets used in the study, along with a brief description of each. Soil data were extracted from the pan-European Land Use/Land Cover Area Frame Survey (LUCAS) topsoil database, which collected 19,969 topsoil (0–20 cm depth) samples from all EU countries in 2009 (Orgiazzi et al., 2018). The LUCAS project aims to monitor land use and land cover changes to support policymaking across the EU. The spatial distribution of soil properties, including cation exchange capacity (CEC), calcium carbonate equivalent (CCE), potential of hydrogen (pH), and bulk density (BD) were originally computed using machine learning methods based on the LUCAS data. Furthermore, the base saturation (BS) map was obtained

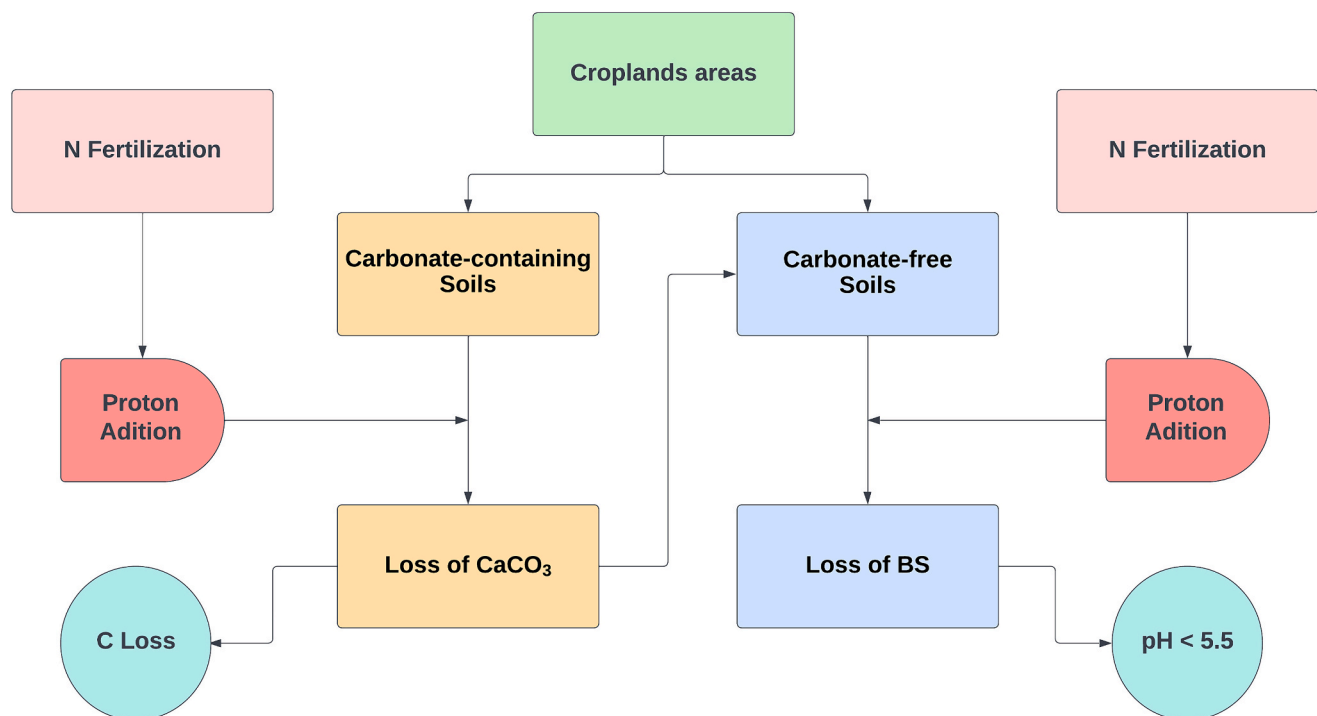


Fig. 1. Flowchart to show the work processes. The EU croplands were divided based on presence or absence of carbonate minerals into carbonate-containing and carbonate-free soils as the mechanism of acid neutralization differs among these groups. In carbonate-containing soils, acidification leads to loss of carbonate stocks, and the soils become carbonate-free over the time. In carbonate-free soils, acidification leads to the loss of base cations i.e. decreasing the base saturation percentage (BS). When the soil BS drops to about 50 %, the pH value will be about 5.5 or less and the soil generally becomes unsuitable for most of crops because of low nutrients availability and Al^{3+} toxicity.

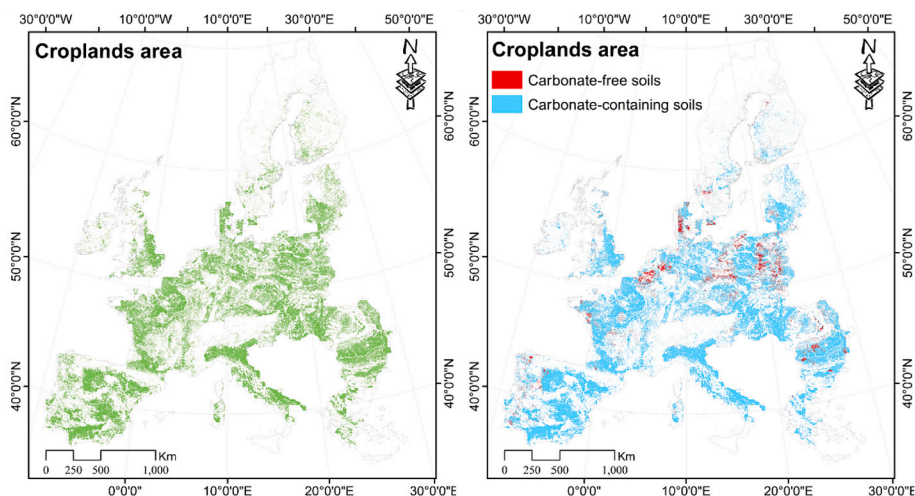


Fig. 2. (left) Croplands area (ca. 135 million ha) in the European Union (source: CORINE Land Cover) and (right) distribution of carbonate-containing (ca. 126 million ha) and carbonate-free soils (ca. 8.72 million ha) in 2019. Note that the calculated area of carbonate-containing and carbonate-free soils were based on the LUCAS dataset, where could also include the limed soils. Note that about 64 % of the carbonate-containing area has a carbonate content below 5 %.

Table 1

List of geospatial data used in this study.

Datasets	Unit	Resolution (m)	Source
CEC*	cmol kg ⁻¹	500	(Ballabio et al., 2019)
CCE	g kg ⁻¹	500	(Ballabio et al., 2019)
pH	–	500	(Ballabio et al., 2019)
BD	T m ⁻³	500	(Ballabio et al., 2016)
BS	%	1000	(Panagos et al., 2012)
N fertilizer	kg ha ⁻¹	1000	(Potter et al., 2010)

* CEC: cation exchange capacity, CCE: calcium carbonate equivalent, BD: bulk density, BS: base saturation.

from the European Soil Database Maps (Panagos et al., 2012). The N fertilizer application map was obtained from the spatial patterns of global fertilizer application. A detailed and sophisticated methodology for computing fertilizer application rates that involved a rigorous and multi-stage process, integrating data from various sources to ensure comprehensive and consistent estimates is well described by (Potter et al., 2010).

2.3. GIS operations

After obtaining the geospatial data presented in Table 1, we converted the N fertilizer map (kg ha⁻¹ y⁻¹) into mol H⁺ ha⁻¹ y⁻¹ added to the soils using Eq. 9.

$$\begin{aligned} \text{N fertilization (kg N ha}^{-1}\text{y}^{-1}) \times 58/100 \times 71.43 \times 40/100 \\ = \text{mol H}^+ \text{ ha}^{-1}\text{y}^{-1} \end{aligned} \quad (9)$$

It is assumed that (1) the total added N via applying mineral fertilizers only was in form of urea, which is the main type of applied mineral N fertilizer (Chao et al., 2011; Rice and Herman, 2012). Note that application of all other mineral fertilizers, which are also generally ammonium-based such as ammonium nitrate, ammonium sulfate or ammonium phosphate is comparatively small. Nevertheless, based on our recent studies (unpublished data), application of the other mentioned ammonium-based fertilizers may even add more acidity to the soil than urea. (2) about 58 % of the total added N to the EU croplands came from mineral fertilizers (Leip et al., 2011) and the probable contribution of organic fertilizers such as manure and crop residues were not considered in the calculations. Further, (3) we used a value of 71.43 to account for the release of 1 mol H⁺ for each mole of N in urea-form fertilizers. The assumption was made based on the fact that 1 mol N

weighs 14 g. Therefore, 1 kg N (1000 g N) corresponds to approximately 71.43 mol H⁺. Last, (4) we considered a N use efficiency of approximately 60 %, which is typical of European countries (Zhang et al., 2015). This means that approximately 40 % of the added N contributed to soil acidification. Note that it is assumed that the released nitrate and accompanying protons via mineralization of 60 % of the added fertilizer was instantaneously taken up by the roots and had no contribution to soil acidification. Hence, our calculations involved several assumptions related to the origin and form of the added N, as well as the efficiency of its use. These factors were considered in the conversion of N fertilizer values to mol protons, which provided a more precise measure of the impact of N on soil acidity and changes in base saturation. After converting the N fertilizer map to mol protons, we were able to estimate the number of protons added to the soils in the current year i.e. . 2023. Assuming a constant rate of fertilizer application in future, we can then calculate the number of protons that will be added to the soils in the next 50 years (Fig. 4).

In carbonate-containing soils, the addition of the protons leads to the loss of carbonates, which can be determined by equating the added H⁺ (mol ha⁻¹ y⁻¹) to the mol CCE loss ha⁻¹ y⁻¹. To convert this value to mass units, we multiply by 100 (g CCE loss ha⁻¹ y⁻¹) and 1/1000 (kg CCE loss ha⁻¹ y⁻¹) to obtain the amount of CCE lost per unit area. It is important to note that we assumed that only about 60 % of the lost CCE contributed in the emitted CO₂ from carbonate-containing soils, while the other 40 % may stay as bicarbonate ions in soil solution and reach groundwater (West and McBride, 2005). To calculate the amount of CO₂ (mol ha⁻¹ y⁻¹) from carbonates, we multiply the added H⁺ (mol ha⁻¹ y⁻¹) by 60/100 and convert to mass units by multiplying by 44 (g CO₂ mol⁻¹) and 12 (g C mol⁻¹). This calculation was performed for N fertilizer addition over 1 and 50 years. Furthermore, we accounted annual changes for the conversion of some parts of the carbonate-containing into carbonate-free soils.

In carbonate-free soils, the addition of protons leads to a reduction in base saturation (BS), which can be determined by equating the added H⁺ (mol ha⁻¹ y⁻¹) to the mol positive charges, i.e. base cations lost per hectare per year. This generates a map showing the annual loss of basic cations (mol positive charge ha⁻¹ y⁻¹). To account the given wide range of BS on the original map and considering variability in the BS, we used three maps with minimum, maximum, and mean values for each pixel of the map, and all calculations for declining BS were computed three times. We also computed the loss of BS up to 50 years with a constant rate of N fertilization. By decreasing the BS, we determined the area of soils, where the pH value will go below 5.5 in the EU croplands in the

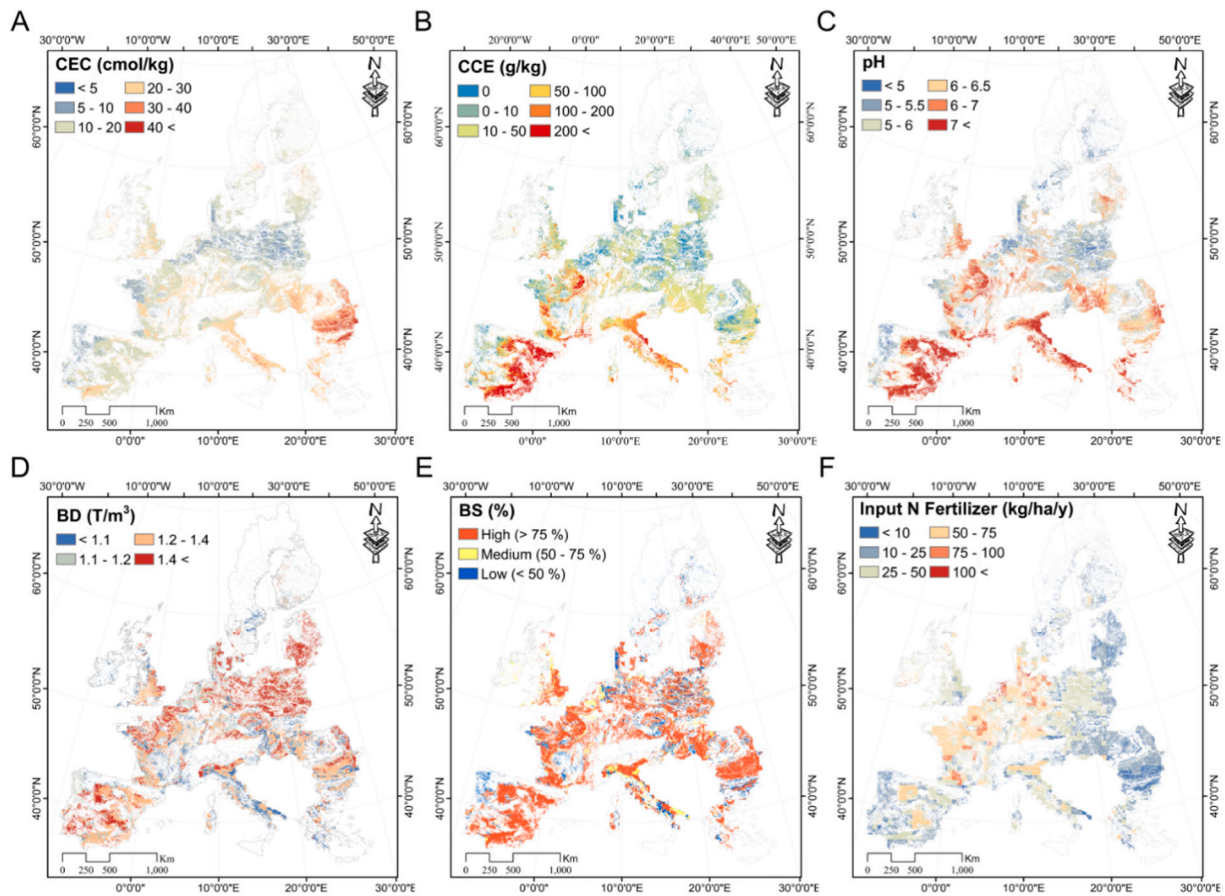


Fig. 3. Spatial distribution of parameters used in this study. (A-D) Soil properties, including cation exchange capacity (CEC), calcium carbonate equivalent (CCE), pH values, and bulk density (BD) are from the LUCAS dataset. (E) Soil base saturation (BS) map was obtained from (Panagos et al., 2012). (F) Nitrogen fertilization map is from (Potter et al., 2010).

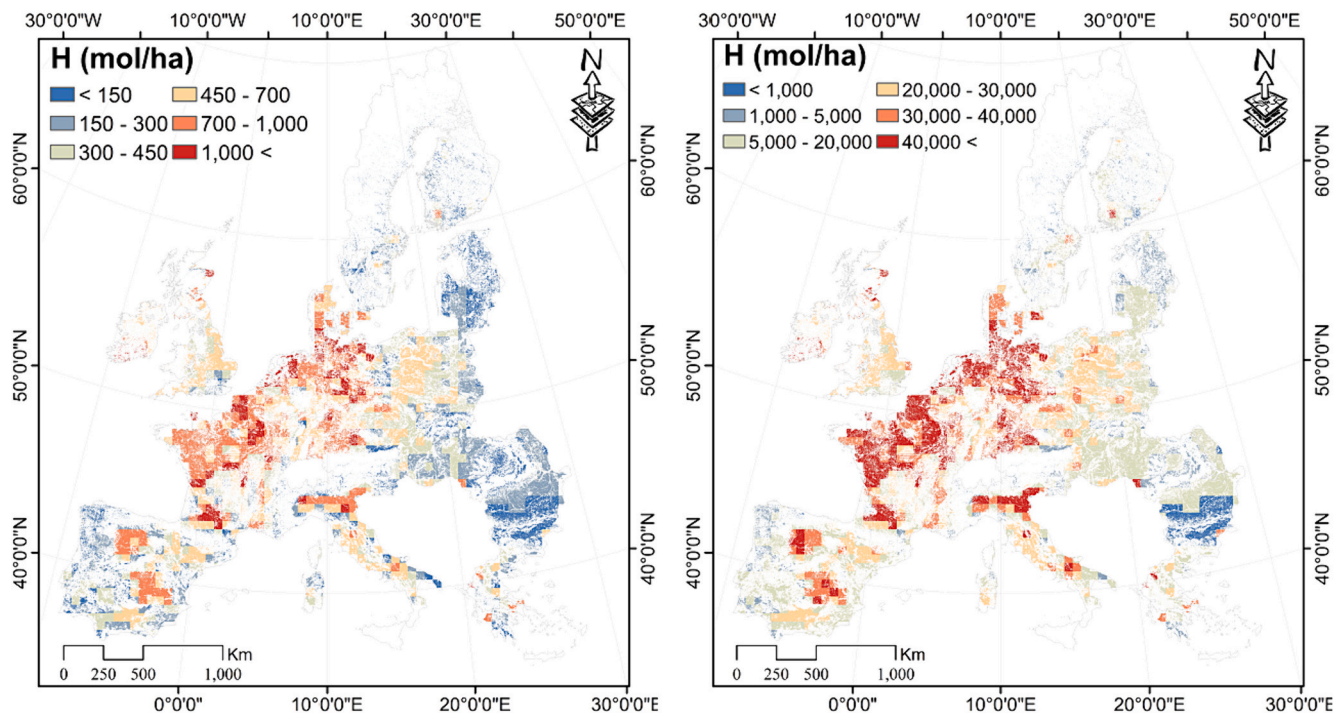


Fig. 4. Spatial distribution of protons added into soils over time, with maps for 1 (left) and 50 (right) years of N fertilization. Note different scales for classes of acid input on the maps as well as the expansion of the red area over time, which spreads throughout of the Europe.

next 50 years. We considered that soils with pH values below 5.5 and a BS smaller than 50 % are unsuitable for economic cultivation of most crops (Stahr et al., 2016).

2.4. Uncertainties in data generation

Various factors potentially introduce degrees of uncertainty into our results, meriting careful consideration: Firstly, our study is restricted to agricultural areas within the European Union, covering approximately 1.3 million hectares. This scope is primarily dictated by the limitations inherent in the available maps.

Secondly, spatial data input introduces several uncertainties. CEC, CCE, pH, and BD maps, though likely less uncertain due to machine learning and ground truth validation, still have inherent imperfections. The vector-based BS map, with its three-level categorization, can lose significant information compared to raster maps. Additionally, the N fertilizer map, at a 1000-m resolution, displays the highest uncertainty due to its global scale, limited ground data, and coarse resolution inadequately representing many agricultural areas.

Thirdly, there is an issue of temporal relevance as the maps utilized were created in different years. As a result, the values for nitrogen fertilization rates, applied fertilizer amounts, and soil chemical properties may not correspond precisely to the actual conditions in 2023.

Lastly, our analysis identified discrepancies in regions where carbonate-containing and carbonate-free soils intersect, especially in terms of their pH values. Contrary to expectations, some carbonate-containing soils demonstrated lower base saturation levels than carbonate-free soils, despite an anticipated higher pH value in the former.

In conclusion, our study emphasizes significant uncertainties in spatial data analysis for EU agricultural areas, primarily due to the limitations of existing maps and varying uncertainties in CEC, CCE, pH, BD, and BS map data. Notably, the N fertilizer map, with its coarse resolution and limited data, introduces considerable uncertainty. Temporal variances and discrepancies in soil properties, especially in areas with varying soil types, further complicate the accuracy of our analysis. These challenges highlight the need for improved data collection and

processing methods to enhance the reliability of agricultural assessments in the EU.

3. Results

Carbonate-containing (intrinsically or limed) and carbonate-free soils, respectively comprise about 93.5 % and 6.5 % of the total cropland area in the countries of the EU (Fig. 2). Note that the calculated area of carbonate-containing and carbonate-free soils were based on the LUCAS dataset, where could also include the limed soils. From whole area of carbonate-containing soils, about 26 % and 64 % have respectively carbonate contents below 1 %, and below 5 %. The EU croplands annually receive about 6.1×10^7 kmol H^+ due to N fertilization. If a similar fertilization trend continuous for the next 50 years, the total acidity added to the EU croplands will reach 2.9×10^9 kmol. Hence, in carbonate-containing soils, every year 6.1×10^9 kg $CaCO_3$ will be lost via neutralization of the added acidity, where the total loss of carbonates over the next 50 years may reach 2.9×10^{11} kg (Fig. 5). Correspondingly, about 3.4×10^6 ha of EU croplands will be decalcified over the next 50 years. The greatest loss of carbonates occurs primarily in central and southern Europe, including France and Spain. Nevertheless, the areas becoming carbonate-free spread throughout much of the EU in the next 50 years (Fig. 5).

The dissolved bicarbonate ions following carbonate neutralization may not necessarily remain in soil solution or in the leachate but may release to the atmosphere as CO_2 (Kuzyakov et al., 2021; Li et al., 2023; Zamanian et al., 2018, 2021), directly from the field or by degassing as soon as soil drainage reaches the surface (Wallin et al., 2013). By a conservative assumption that about 40 % of bicarbonate ions (Eq. 4) may remain as dissolved ions (West and McBride, 2005), neutralization of carbonate minerals in the EU croplands because of N fertilization annually releases about 19 Tg C (19×10^{12}) to the atmosphere. This means emission of about 0.9 Pg C (0.9×10^{15}) over the next 50 years.

In carbonate-free soils, the neutralization of fertilization-induced acidity leads on average to an annual loss of about 2.1×10^7 (1.6×10^7 – 2.7×10^7) kmol of basic cations. The loss of basic cations may amount to 7.5×10^8 kmol after 50 years of fertilization. These rates of

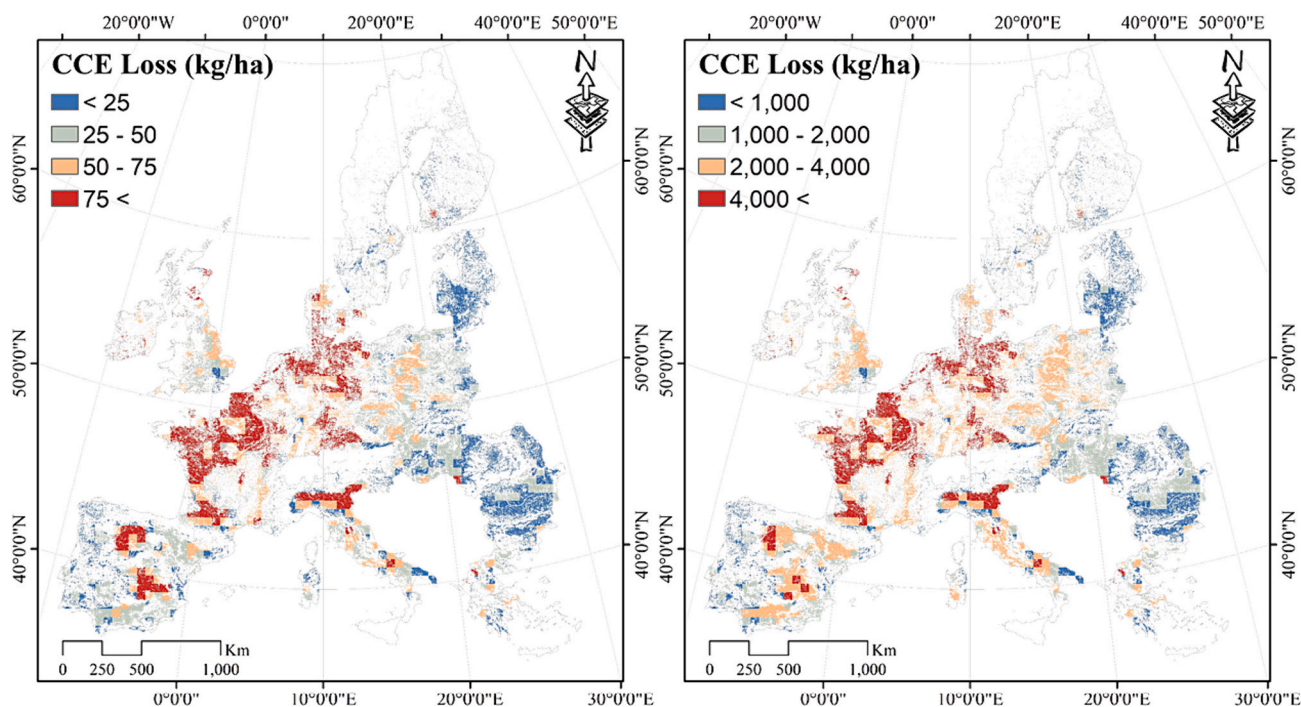


Fig. 5. Loss of carbonates over 1 and 50 years of N fertilization. Note different scales for classes of CCE loss on the maps. Carbonate losses will cause complete decalcification of the top 20 cm of about 3.4 million ha of European croplands within the next 50 years.

losses in base cations mean that about 2.6 million ha of EU croplands will get acidic, i.e. will have a base saturation below 50 % in the next 50 years. Loss of base saturation by 50 % will dictate necessity of liming or addition of other pH increasing measures to the soil (Tang et al., 2013; Zamanian and Kuzuyakov, 2019). If we assume that (i) lime is the main acid neutralizing amendment, (ii) each hectare of acid soils receives at least 1 ton lime per year, and (iii) the rate of expansion in acid area is constant and about $52,000 \text{ ha y}^{-1}$, then annually about $6.2 \cdot 10^9 \text{ g}$ additional C will be released to the atmosphere from lime neutralization.

4. Discussion

Presence of carbonates either intrinsically or via liming in about 93 % of the EU croplands (Fig. 2), already points to the importance of carbonates for the agricultural industry. Hence, soil carbonate stocks should be seen as an asset (Groshans et al., 2018, 2019). By assuming about 0.2 € (0.22 US \$) as the mean price for 1 kg of agricultural lime (i.e. CaCO_3) (“EU’s Calcium Carbonate Market Report,” 2022), the EU croplands face loss of annually about 1.2 milliard € (1.34 milliard US \$) in their carbonate asset. This loss will reach to about 58 milliards € (65 milliard US \$) over the next 50 years. At the same time, if we consider a CO_2 price of about 80 € (89 US \$) for each ton of CO_2 (“EU Carbon Price Tracker,” 2022), which can potentially be emitted because of CaCO_3 neutralization, about 1.5 milliard € (1.7 milliard US \$) are needed for compensation of C footprint from the lost carbonates. These annual losses, both direct loss of soil carbonate stock and compensation of carbonates-related CO_2 , correspond to about 1.5 % of the proposed budget of the European commission for 2023, i.e. about 186 milliard € (207 US \$ milliard) (“EU budget 2023,” 2022).

All these estimations however, should be seen as conservative values. Because various other sources of soil acidification in agroecosystems such as acid input by cultivation of legumes (Bolam et al., 1991; Galloway et al., 2004), or via harvesting (Alves et al., 2019; Fujii et al., 2022; Hao et al., 2020, 2022; Zeng et al., 2017) and through acid and N deposition (Galloway et al., 2004; Gao et al., 2013; Johnson et al., 2008; Yu et al., 2020) are totally ignored in these calculations. Note that harvesting alone may add comparable acidity to soils as N fertilization (Dong et al., 2022; Hao et al., 2019, 2022). If we consider addition of about 0.10 and 0.36 kmol H^+ for each ton of the harvested grains and straw of cereals, respectively (Wang et al., 2024) and a total cereal harvest of about 271 million tons in EU (Eurostat, 2023), harvesting of cereals would add about 2.7×10^7 to $9.8 \times 10^7 \text{ kmol H}^+$ to the EU croplands, which is comparable to the annual addition of protons ($6.1 \times 10^7 \text{ kmol H}^+$) via N fertilization. In any case, by loss of soil carbonate stocks, all the corresponding ecosystem services such as aggregate stability, C sequestration as organic matter, resistance to erosion, organisms’ activity and biodiversity will also be lost (Raza et al., 2021, 2024; Shariffar et al., 2023; Zamanian et al., 2021), which then ultimately endangers crop production and food security (Zhu et al., 2020). In consequence there will be various other, yet completely unconsidered costs upon decline in soil carbonate stocks than the estimated values and the real social costs are much higher (Rennert et al., 2022).

Organic fertilization such as manuring (Jia et al., 2022; Liu et al., 2023; Niu et al., 2023; Tian et al., 2022; Wang et al., 2014) or straw addition (Bughio et al., 2016; Jia et al., 2022; Tian et al., 2022; Wang et al., 2014) as well as treating soils with biochar (Azeem et al., 2022; Dong et al., 2019; Ghorbani et al., 2021, 2022; Shi et al., 2021; Wang et al., 2023) have been frequently proposed as a preventive strategy against soil acidification due to slow mineralization of organic N and returning base cations via decomposition. However, if the straw, to be used as green fertilizer or as feed for livestock or for pyrolysis to biochar has to be transported to a new location, this will cause loss of base cations and acidification of the soil where the straw was produced (Zamanian and Kuzuyakov, 2022). Furthermore, mineralization of the added organic fertilizers (straw, manure), though with comparatively slower rates can also acidify soils because of nitrification of organic N

and release of organic acids during decomposition (Jia et al., 2022). In addition, to have a reliable and global perspective about the efficiency of organic fertilization and biochar application for alleviating soil acidification, the transportation, labor costs, N flow, and greenhouse gas emissions among the sectors should also be considered (Liu et al., 2022b).

The “enhanced weathering” and distribution of ground mafic and ultramafic silicates as fertilizer is also suggested as an effective strategy to overcome the negative consequences of acidification (Azeem et al., 2022; Beerling et al., 2020). Despite considerable acid neutralization potential and capability to replenish lost cations on exchangeable sites (Eq. 5), the rates of silicates weathering are some orders of magnitude slower than carbonates-acid reaction. Therefore, the enhanced weathering is applicable to soils which are already decalcified and acid. One may argue that enhanced weathering leads to production of carbonates in soils, where the release of base cations like Ca^{2+} ions as in Eq. 5 precipitates as CaCO_3 (Haque et al., 2020a, 2020b). Although, this is completely true under natural ecosystems, where carbonic acid is the main source of protons, CaCO_3 precipitation following silicates weathering might not necessarily happen in agroecosystems (Batool et al., 2024). In a fertilized soil, the presence of nitrate ions (Eq. 3) prevents CaCO_3 precipitation (Zamanian et al., 2018; Zamanian and Kuzyakov, 2022) and nitrate and calcium ions are simply leached into groundwater (Rengel, 2011; Tao et al., 2022).

Atmospheric deposition of base cations as dissolved ions in rain drops or present in dusts and aerosols can also be considered as another mechanism for replenishing of exchangeable sites (Cecchini et al., 2019; Draaijers et al., 1997a, 1997b; Mikhailova et al., 2013). The significance of atmospheric deposition is however, highly dependent on climatic conditions, distance to a source area like the Sahara desert and the land use type (Draaijers et al., 1997b; Goddard et al., 2007). For example, the potential of acid neutralization decreases from about 50 % in southern parts of Europe to about 25 % in central and northwestern Europe (Draaijers et al., 1997a). Regionally, as in central Italy, atmospheric deposition may annually add about $1.11 \text{ g Ca}^{2+} \text{ m}^{-2}$ and about $0.08 \text{ g K}^+ \text{ m}^{-2}$ to the soils, which can considerably replenish the lost cations and inactivate the acidification process in natural forest ecosystems (Cecchini et al., 2019). Nevertheless, in European agroecosystems, which on average annually receive about $0.45 \text{ kmol H}^+ \text{ ha}^{-1}$, atmospheric deposition, even at rates reported for regional hot spots such as Italy or eastern Europe should be considered insignificant in neutralization of fertilization-induced acidity.

Irrigation by using groundwater is a common management in carbonate-rich soils (Alexandratos and Bruinsma, 2012; de Soto et al., 2017, 2022), where dissolved calcium and bicarbonate ions may precipitate as pedogenic carbonates (de Soto et al., 2017; Entry et al., 2004; Ortiz et al., 2022). The amount of precipitated carbonates highly depends on concentrations of dissolved calcium and bicarbonate ions (Hannam et al., 2016), soil fertility (Bughio et al., 2016) as well as the aridity index (Tao et al., 2022). Therefore, contradictory results have been reported on the effects of irrigation on the carbonate content of soils (Sanderman, 2012; Wu et al., 2009). Generally, increasing soil carbonates content because of irrigation can be significant, where leaching is limited i.e. less than 30 % of the applied water (Naorem et al., 2022) and in arid areas with an aridity index smaller than 0.2 (Tao et al., 2022). The distribution of such arid areas in EU is negligible (Estrela et al., 1996). In addition, the area of irrigated soils in EU comprises about 6 % of the total croplands area (Eurostat, 2024). Hence, increasing soil carbonates content due to irrigation, if any, especially in the first 20 cm of the EU croplands should be insignificant. Besides this, it should be considered that the use of groundwater can lead to massive CO_2 release to the atmosphere through degassing (Hannam et al., 2016; Huo et al., 2022; Rajan et al., 2020), which is questioning the suitability of irrigation as a measure to sequester C in the form of soil carbonates.

After all, soil acidification is a natural process, which cannot be totally stopped. Furthermore, agricultural production still highly

depends on N fertilizers (Einarsson et al., 2021) and most probably the fertilization demand will increase along with the world population (Tian et al., 2021). Therefore, probably the best strategy to slow down soil acidification is increasing the N use efficiency (Hua et al., 2020; Zhang et al., 2021a; Zhu et al., 2023) and keeping the protons input to the agroecosystems ideally in a comparable range as in natural ecosystems. In this regard, excessive fertilization should be avoided and nutrient use efficiency should be increased through best management practices and enhanced efficiency fertilizers (Herrera et al., 2016; Raza et al., 2019; Shao et al., 2022). Replacing the ammonium bearing fertilizers with nitrate fertilizers could also be an option. Decreasing just 5 % in the annual application rate of N fertilizers will prevent decalcification of about 150,000 ha of EU croplands in the next 50 years (annually about 3900 ha). The use of detoxified municipal wastes (de Soto et al., 2022, 2023) as fertilizers along with addition of manure and straw to the soil should be promoted (Chen et al., 2020; Niu et al., 2023; Tian et al., 2022). Planting deep-rooted crops (Chen and Liao, 2017; Yu et al., 2015; Zamanian and Kuzyakov, 2022) and activating oxalotrophic microorganisms (Guan et al., 2022) to assimilate the excess of nitrate in soil solution from various depths, and managing soil water content to avoid N losses by leaching (Gentsch et al., 2022; Hess et al., 2020; Xu et al., 2020) can be most promising approaches to control soil acidification. Moreover, considering irreversible changes that decalcification and subsequent acidification cause for microorganisms (Ma et al., 2018; Ste-Marie and Paré, 1999; Zamanian and Kuzyakov, 2022), clay minerals (Matocha et al., 2016; Oort et al., 2022), and their functions, liming, as it is traditionally done in some parts of Europe like Germany is suggested to be applied before that soil carbonate stock reaches a critical threshold (Li et al., 2023) or lost completely.

5. Conclusions

This study quantifies proton addition to the European agricultural soils and subsequent effects of acidification on losses of carbonates and base cations by utilizing the comprehensive and high-resolution dataset of LUCAS. Annual addition of about 6.1×10^7 kmol H^+ leads to loss of ca. 6.1×10^9 kg $CaCO_3$ from European agricultural soils, and development of about 3.4 million hectares of carbonate-free cropland area over the next 50 years. Considering the actual price for $CaCO_3$ in the European market, direct loss of soil carbonate stocks and compensation of carbonates-related CO_2 correspond to about 1.5 % of the proposed budget of the European commission for 2023. In addition, carbonate losses at such a large continental scale, which were previously presumed to be very stable, has serious consequences on food production, soil-plant-water interactions, and environmental sustainability. As a result, the real social and environmental costs of $CaCO_3$ losses are much higher than this estimation. Therefore, devising policies on monitoring soil carbonate stocks, especially for soils with low or trace carbonate contents, and adopting best management practices that have less acidification potential can save soil deterioration from acidification.

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CRediT authorship contribution statement

Kazem Zamanian: Writing – review & editing, Writing – original draft, Validation, Supervision, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Ruhollah Taghizadeh-Mehrjardi:** Writing – review & editing, Visualization, Formal analysis, Data curation. **Jingjing Tao:** Writing – review & editing, Formal analysis. **Lichao**

Fan: Writing – review & editing, Formal analysis, Data curation. **Sajjad Raza:** Writing – review & editing, Funding acquisition, Formal analysis. **Georg Guggenberger:** Writing – review & editing, Validation, Formal analysis. **Yakov Kuzyakov:** Writing – review & editing, Validation, Supervision, Funding acquisition, Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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