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REVIEW

Mitigation strategies for soil acidification based on optimal nitrogen management

Pengshun WANG[1,2], **Donghao XU** (✉)[1,2], **Prakash LAKSHMANAN**[3,4,5], **Yan DENG**[1,2], **Qichao ZHU** (✉)[1,2], **Fusuo ZHANG**[1,2]

1 College of Resources and Environmental Sciences, China Agricultural University, Beijing 100193, China.
2 National Academy of Agriculture Green Development, China Agricultural University, Beijing 100193, China.
3 Interdisciplinary Research Center for Agriculture Green Development in Yangtze River Basin, College of Resources and Environment, Southwest University, Chongqing 400716, China.
4 Sugarcane Research Institute, Guangxi Academy of Agricultural Sciences, Nanning 530007, China.
5 Queensland Alliance for Agriculture and Food Innovation, University of Queensland, St Lucia 4067, Brisbane, Queensland, Australia.

KEYWORDS

Soil acidification, nutrient management, nitrogen, soil buffering capacity

HIGHLIGHTS

- Soil acidification is determined by proton production and soil buffering capacity.
- Cropland acidification is mainly caused by anthropogenic activities.
- Nitrogen transformations dominate anthropogenic soil acidification processes.
- Acidification stage-specific strategies are needed for managing soil acidification.
- Optimizing N rate and N form is highly effective in mitigating soil acidification.

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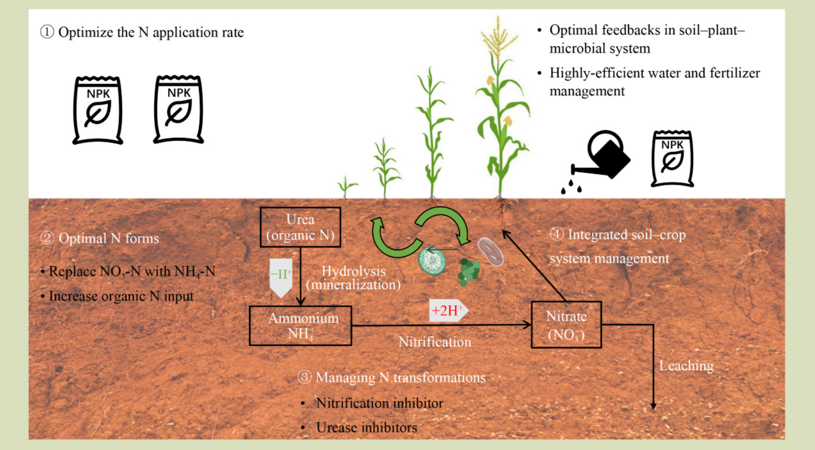
Correspondences: dominic.xu@foxmail.com,
qichaozhu@126.com

GRAPHICAL ABSTRACT



ABSTRACT

Soil acidification is a serious constraint to food production worldwide. This review explores its primary causes, with a focus on the role of nitrogen fertilizer, and suggests mitigation strategies based on optimal N management. Natural acidification is determined by the leaching of weak acid mainly caused by climate and soil conditions, whereas the use of ammonium-based fertilizers, nitrate leaching and removal of base cations (BCs) by crop harvesting mostly accounts for anthropogenic acidification. In addition, low soil acid buffering capacity, mainly determined by soil parent materials and soil organic matter content, also accelerates acidification. This study proposes targeted mitigation strategies for different stages of soil acidification, which include monitoring soil carbonate content and pH of soils with pH > 6.5 (e.g., calcareous soil), use of alkaline amendments for strongly acidic soils (pH < 5.5) with aluminum toxicity risk to pH between 5.5 and 6.5, and decreasing acidification rates and supplementing BCs to maintain this optimal pH range, especially for soils with low acid buffering capacity. Effective mitigation involves optimizing the rate and form of N fertilizers used, regulating N transformation processes, and establishing an integrated soil–crop management system that balances acid production and soil buffering capacity.

1 Introduction

Soils are vital for terrestrial ecosystem and support terrestrial life on the planet[1,2], and healthy soils are critical for global food production and human well-being[3]. However, around 33% of the global arable land is moderately to highly degraded, with soil acidification being one of the primary types[4]. It affects approximately 30% of the total ice-free land area and as much as 50% of potential arable land[5]. Soil acidification causes various conditions that restrict crop growth, which include decreased soil fertility with decreased availability of phosphorus and base cations (BCs) such as calcium, magnesium and potassium[6], and the release of toxic aluminum and manganese[7]. Also, soil acidification enhances the availability of toxic heavy metals, especially lead and cadmium, increasing the risk of their accumulation in crops and animals affecting food quality and human health through the food chain[8,9].

1.1 Definition of soil acidification

Soil pH is a measure of the H^+ concentration in the soil solution, and it serves as a key predictor of soil biology, chemistry, and physical processes, directly affecting plant growth and development[10]. The United States Department of Agricultural National Resources Conservation Service has categorized soil based on its pH as extremely acidic (< 4.5), very strongly acidic (4.5–5.0), strongly acidic (5.1–5.5), moderately acidic (5.6–6.0), slightly acidic (6.1–6.5), neutral (6.6–7.3), slightly alkaline (7.4–7.8), moderately to strongly alkaline (7.9–9.0), and very strongly alkaline (> 9.1)[11]. Most agricultural plants grow optimally when soil pH falls between 6 to 8, and the optimal availability for most of the nutrients is found in the slightly acid pH range[12]. Soil pH is calculated by the negative logarithm of the H^+ concentration in soil solution when the soil solid-liquid phase is in equilibrium. In this context, soil pH only represents the active protons in soils, neglecting the exchangeable acidity held near clay and humus surfaces[13], as well as the residual acidity, representing the H^+ and Al^{3+} bound on the clay and humus surfaces[14]. These latter forms of acidity constitute potential acidity, which is usually quantitatively greater than active acidity[14]. The amount of potential acidity is mostly determined by the soil pH and cation exchange capacity (CEC) and it increases substantially as pH drops below 5.5, mainly due to increased exchangeable acidity[15]. Consequently, total soil acidity hinges not only on soil pH but also on cation holding capacity, which encompasses exchangeable and residual acidity.

1.2 Buffering mechanism of soil acidification

Soil acidification is generally indicated by a decrease in soil pH. However, the change in soil pH is usually smaller than the amount of proton input from external sources, primarily due to the existence of acid-buffering substances within the soil. Therefore, soil acidification is defined as a decrease in acid neutralizing capacity of the solid phase of soil, which is conceptually defined as the sum of basic components minus the strongly acidic components at the reference pH of soils (generally pH 2.0–5.0)[16].

Acid buffering processes and the buffering capacity of soil solid phase substances differ as pH changes (Table 1)[6,17]. When the soil pH drops from 8 to around 3.5, it generally goes through a carbonate buffer system, a BC exchangeable buffer system, and a hydroxy aluminum and a hydroxy iron buffer system. When the soil is in the carbonate buffer system, it takes about 100 years to consume 1% calcium carbonate under natural conditions[18,19]. The buffering capacity is about 1500 $keq \cdot ha^{-1}$ H^+ in a calcareous soil per 1% $CaCO_3$ content in a soil with a weight of 15 kt (Table 1). Therefore, a calcareous soil can generally maintain a high soil pH of 7.0–8.5 for a long period. However, when the carbonate in the soil is exhausted, entering the BC exchange buffer system, the acid buffering capacity can be as low as 250 $keq \cdot ha^{-1}$ H^+. When the soil enters the buffer system of aluminum hydroxide and iron hydroxide (pH < 5.0), the buffering capacity increases to 1000–1500 $keq \cdot ha^{-1}$ H^+ per % clay. This indicates that the main acid buffering systems of both calcareous soil and very strongly acidic soil (pH < 5.0) generally have a strong buffering capacity, thereby the soil pH is insensitive to H^+ input, whereas the pH of soils in the BC exchange system (pH 5.0–7.0) is more sensitive to H^+ addition, as the dominated acid buffering element is BCs[20]. Therefore, the global soil pH at 0.5 m depth has two types of buffering systems approximately at pH 8.2 and 5.1 regulated by $CaCO_3$ and $Al(OH)_3$ buffers, respectively[21].

Table 1 Main buffering systems and their buffering capacity in different soil pH ranges

pH range	Main buffering system	Buffering capacity*
$6.2 < pH < 8.6$	Carbonate	1500 keq H^+ per % $CaCO_3$
$5.0 < pH < 6.2$	Base cation (BC) exchange#	250–750 keq H^+ per % (primary) silicate
$pH < 5.0$	Hydroxy-aluminum	1000–1500 keq H^+ per % clay
$pH < 3.8$	Hydroxy-iron	Lacking data

Note: *The acid buffer capacities were based on a soil weight of 15,000 t, calculated by assuming a bulk density of 1500 $kg \cdot m^{-3}$, expressed ha^{-1} and 1 m^{-1} soil depth[6]. #More specifically, the weathering of silicate minerals dominates the buffer reaction when soil $pH > 5.0$ as H^+ and Al^{3+} ions replace the BCs on the exchange complex[17].

2 Causes and processes of soil acidification

2.1 Causes of soil acidification

Causes of soil acidification can be categorized into the following aspects.

Natural soil acidification. Naturally, H^+ production occurs when a weak acidic anion is formed, such as the dissolution of CO_2 to form carbonic acid (H_2CO_3) and the dissociation of carboxylic acids (RCOOH) produced by plants and microorganisms[22]. Dissolution of H_2CO_3 and RCOOH releases H^+ exchanging with BCs, which is then leached to the subsoil accompanied with HCO_3^- and $RCOO^-$ by excess precipitation, being the main contributor to the natural forest soil and grassland, especially for the humid forested ecosystem. For example, Fujii et al.[23] found that $RCOO^-$ dominated the anions fluxes in the O horizon of the Japanese forest soil, being the most important driver of soil acidification. Natural acidification is the main driver in soils with high organic carbon content as in brown and dark brown soils in north-eastern China[24] and those with high pH or under high CO_2 pressure as in calcareous soils in the Netherlands; however, it has minor acidifying effects in non-calcareous soils[18].

Anthropogenic soil acidification. Without any human intervention, soil acidification is a naturally slow process. It was estimated that the pH of red soil declined by 1 unit after 2.3 million years without artificial disturbance[25]. However, the soil acidification rate has been greatly accelerated by human activities, such as burning fossil fuels causing atmospheric acid deposition, application of acidifying fertilizers (including neutral fertilizers like urea but with acidifying effects) and nutrient removal through harvested parts of crops.

Atmospheric acid deposition. From the beginning of the Industrial Revolution, emissions of acidifying compounds such as sulfur dioxide (SO_2) and nitrogen oxides (NO_x) greatly increased due to the rapid industrial development. Consequently, atmospheric acid deposition has become a serious global environmental problem, elevating the acidity of precipitation and acidification of soils and lakes in Europe and North America[26]. In forest soils in the UK, a 1.5-unit decline in topsoil pH (0–23 cm) was found during 1904–1964, mostly driven by the elevated atmospheric acid inputs caused by SO_2 and NO_x emissions (from 0.8 to 2.6 $kmol \cdot ha^{-1}$) and N transformations (from 0.6 to 2.9 $kmol \cdot ha^{-1}$)[27]. In the regions of southern Sweden (Europe) and the USA (North America) where SO_4-S deposition declined, there had been slow recovery of the soils from acidification[28,29]. However, in some areas in Asia, though strict regulations have declined the total S deposition, there was a delayed recovery of soil from acidification as N deposition-induced soil acidification has increased, either due to the increase in N deposition[30] or increase in NH_4^+/NO_3^- ratio in N deposition caused by intensive agricultural activities[31].

Acidifying fertilizers. In agricultural systems, the increase in N fertilizer application has increased food production but also increased acidification risks. Nearly all processes in the N cycle affect H^+ production or consumption. Overuse of N fertilizers, in particular ammonium-based (NH_4-N) fertilizers, caused significant soil acidification on major Chinese croplands[32]. The application of NH_4-N fertilizers greatly increased soil acidification due to the enhanced net H^+ production by nitrification. Also, leaching of NO_3^- results in permanent acidification by accompanying soil BCs (the decrease in soil buffering capacity). It was observed in the Park Grass Experiment at Rothamsted in Hertfordshire, England that ammonium sulfate application caused a rapid decrease in pH, starting in the surface soil but occurring throughout the profile to at least 1 m deep, compared to the treatment without any fertilizer application[33,34].

Nutrient removal by crops. To maintain the inherent charge balance, crops release H^+ to soil solution when absorbing cations more than anions and vice versa[35]. The removal of cations through harvested parts permanently decreases soil buffering capacity (BCs losses) and releases H^+ when the uptake of anions is less. The acidifying effects vary between crop types due to different amount and composition of nutrients in the harvested component. For cereal crops, crop-induced acidity can vary from 0.25 to 0.76 $mol·kg^{-1}$[35], whereas fruits and upland crops generally have higher ratios of BCs/anions uptake than, for example, paddy rice[36]. For grain legumes, apart from the fixed N and enhanced N-induced acidification, crop-induced acidity can be as high as 1.77 $mol·kg^{-1}$[35], thereby appearing highest acidifying potential. Bolan and Hedley reported that, where legumes had been grown continuously in Australia for > 30 years, soil pH declined by 1 unit[22].

2.2 Main processes causing soil acidification

In the soil–plant system, nutrient transformations and element cycles generate proton or hydroxyl ion (equal to H^+ consumption). The most important elements include C, N, cations (BCs and Al^{3+}) and anions ($H_2PO_4^-$, SO_4^{2-}, and Cl^-)[37], other elements with minor effects are not discussed here; the processes are listed in Table 2.

3 Main factors affecting soil acidification in cropland

Soil acidification of cropland has gained wide attention since Guo et al.[32] reported the significant acidification in Chinese croplands. Characters of cropland acidification are greatly different from natural ecosystems because of intensive disturbance of nutrient cycles by fertilizer and water management[38]. Overall, the main factors affecting soil acidification rates in cropland are: (1) fertilizer application (in particular N), (2) crop harvest, and (3) soil acid buffering capacity.

3.1 Fertilizer application

N fertilizer application is one of the most important contributors to soil acidification. China is the largest synthetic N fertilizer consumer in the world, where the N use efficiency (N removal by crop harvests divided by total N input) is lower than 50%, indicating that over half of N input was lost[39]. The overuse of N fertilizers caused soil pH to decline by 0.5 units during the 1980s–2000s in major Chinese croplands[31]. This acidification rate was much faster than previous findings in the forest soils of Europe caused by acidic deposition in the 1980s[27,34] but was quite comparable with intensely managed agricultural and grassland soils of Australia[40]. Globally, N

Table 2 Some reaction equations of H^+ production and consumption processes in the carbon, nitrogen, cation, and anion cycles

Process	Reaction equation
Carbon cycle	
- Dissociation of CO_2	$CO_2 + H_2O \leftrightarrow HCO_3^- + H^+$
- Dissociation of organic acids (ROOH)	$RCOOH \leftrightarrow RCOO^- + H^+$
Nitrogen cycle	
- N fixation	$2N_2 + 3CH_2O + 4ROH \rightarrow 4RNH_2 + 3CO_2 + H_2O$
- Mineralization of organic N	$RNH_2 + H^+ + H_2O \leftrightarrow ROH + NH_4^+$
- Volatilization of NH_3	$NH_4^+ \leftrightarrow NH_3 + 2H^+$
- Nitrification	$NH_4^+ + 2O_2 \rightarrow NO_3^- + 2H^+ + H_2O$
- Uptake of NO_3^-	$ROH + NO_3^- + H^+ + 2CH_2O \rightarrow RNH_2 + 2CO_2 + 2H_2O$
- Denitrification	$5CH_2O + 4NO_3^- + 4H^+ \rightarrow 2N_2 + 5CO_2 + 7H_2O$
Anion cycle	
- Plant uptake of anions (An^-, e.g., SO_4^{2-}, $H_2PO_4^-$, Cl^-)	$ROH + An^- + H^+ + CH_2O \rightarrow RH-An + CO_2 + H_2O$
- Soil absorption of An^-	$(OH)_{ex} + An^- + H^+ \rightarrow An_{ex}^- + H_2O$
Cations cycle	
- Plant uptake of cations (M^+, e.g., Ca^{2+}, Mg^{2+}, K^+)	$RCOOH + M^+ \leftrightarrow RCOOM + H^+$
- Soil adsorption of M^+	$M^+ + H_{ex}^+ \leftrightarrow M_{ex}^+ + H^+$

addition significantly decreased soil pH by 0.26 on average[41]. More recent studies also found that N fertilizer input can lead to extremely serious soil acidification in tea plantations and grassland systems[42,43].

N fertilizer application significantly enhances soil acidification by affecting the cycling of H^+ in the soil–plant system. N form is important in the degree acidification caused. Common mineral N fertilizer types include urea, liquid ammonia, ammonium bicarbonate and ammonium sulfate. These fertilizers can be categorized into three types based on the N form of their main substances: non-charged N (such as urea and liquid ammonia), ammonium-based N and nitrate-based N. In addition to the different amounts of H^+ contributed by N transformations, the selective uptake of NH_4^+ and NO_3^- also affects the soil acidification process (Fig. 1).

Ammonium-based fertilizers pose the greatest acidification risk due to the nitrification process (conversion of NH_4^+ to NO_3^-) and subsequent NO_3^- leaching. Theoretically, the nitrification of 1 mol of NH_4^+ to form 1 mol of NO_3^- produces 2 mol of H^+. If the produced NO_3^- is taken up by plants, it releases 1 mol of OH^-, and overall 1 mol of NH_4^+ applied to soils produces at least 1 mol of H^+, regardless of whether the NH_4^+ is taken up by plant (either in the form of NH_4^+ or NO_3^- after nitrification) or absorbed by soils or even lost to air as NH_3. However, if the NO_3^- is fully leached, 1 mol of NH_4^+ would produce 2 mol of H^+. Consequently, in well-ventilated sandy soil, the nitrification of NH_4^+ and the subsequent leaching of NO_3^- have the greatest impact on soil acidification[44].

Compared to ammonium-N fertilizers, liquid ammonia and urea have relatively lower acidification capacity, as their conversion to NH_4^+ consumes the same amount of H^+. In the case of organic manure, the acidification ability of N is the same as that of urea because of the same form of N; organic manure, however, can alleviate soil acidification due to high BCs content. Generally, nitrate-N fertilizers do not produce H^+[45] and when plants uptake 1 mol of NO_3^-, equal amount of H^+ is released, resulting in an increase in soil pH in the rhizosphere[46]. The same happens with the denitrification of NO_3^- to N gases (N_2, NO, and N_2O). In summary, N fertilizer types have different impacts on soil acidification, with the ranking as follows: ammonium-N fertilizer > non-charge fertilizer > nitrate-N fertilizer. It is worth noting that when urea and NO_3^- are used, theoretically soil pH hardly changes or may even increase following NO_3^- uptake.

Numerous long-term field experiments have consistently demonstrated the impacts of N fertilizers on soil acidification. In the Park Grass experiment, $(NH_4)_2SO_4$ application for 150 years decreased soil pH by 1.7 units compared to unfertilized

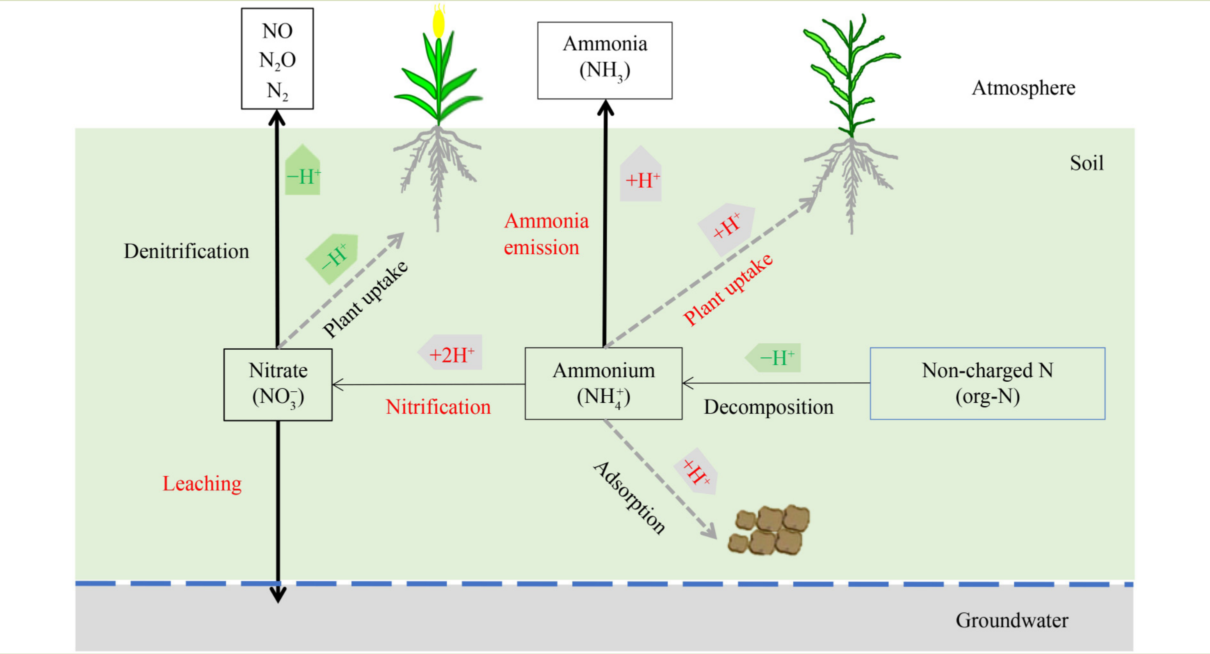


Fig. 1 Processes of N transformations associated with soil acidification. The process in red label indicates that it produces H^+ and in black consumes H^+.

treatment. In contrast, the application of sodium nitrate increased soil pH by 0.7 units[34]. Khonje et al.[47] found that the soil acidification rates associated with nine annual applications of the N fertilizers at 300 kg·ha^{-1} N was of the order: $(NH_4)_2SO_4$ > NH_4Cl > anhydrous NH_3 = NH_4NO_3 > urea > $Ca(NO_3)_2$ = $NaNO_3$. Many long-term trial sites in China also showed that N fertilizer application increases soil acidification[44,48]. For example, the pH of red soil treated with urea for 18 years decreased by more than 1 unit compared to the control (unfertilized) plot located in southern China[49]. Yang et al.[42] found marked soil pH decline in both topsoil (0–40 cm) and subsoil (40–90 cm) at a relatively high N application rate (569 kg·ha^{-1}), being from 4.16 to 3.15 and from 3.67 to 3.35, respectively, over 8 years compared to the control without N application[42].

A recent study revealed that N fertilizer application contributed more than 55.1% of H$^+$ production between the 1980s and the 2010s in Chinese croplands[50]. The high contribution of N cycling to soil acidification in China can be explained for two reasons. First, the increasing application of N fertilizer has greatly exceeded crop demand over the past 20 years[51] (see also Fig. 2). The average N surplus (total N input minus crop uptake) of major crops (including cereal crops and cash crops) varied from 133 to 429 kg·ha^{-1} according to a national scientific fertilizer application survey data set in 2019. Secondly, the changing fertilizer type also contributed to the N-induced acidification. Urea and compounded N (often in urea form) are the main forms of N fertilizer in China, accounting for over 87% in 2020, and nitrate-based nitrogen fertilizer only accounted for less than 3.5%, according to International Fertilizer Association. The structure of the N fertilizer form has significantly changed from the 1980s when the ammonia bicarbonate dominated in N fertilizer. In other words, the N fertilizers applied are still dominated by acid-type N fertilizers.

3.2 Crop harvest and residue management

The plant-induced soil acidification mainly includes two aspects: (1) net H$^+$ production by the plant due to excess uptake of non-N cations over non-N anions[35]; and (2) removal of BCs (e.g., Ca, Mg, and K) in harvest parts such as grain, straw and hay decreasing soil buffering capacity. In

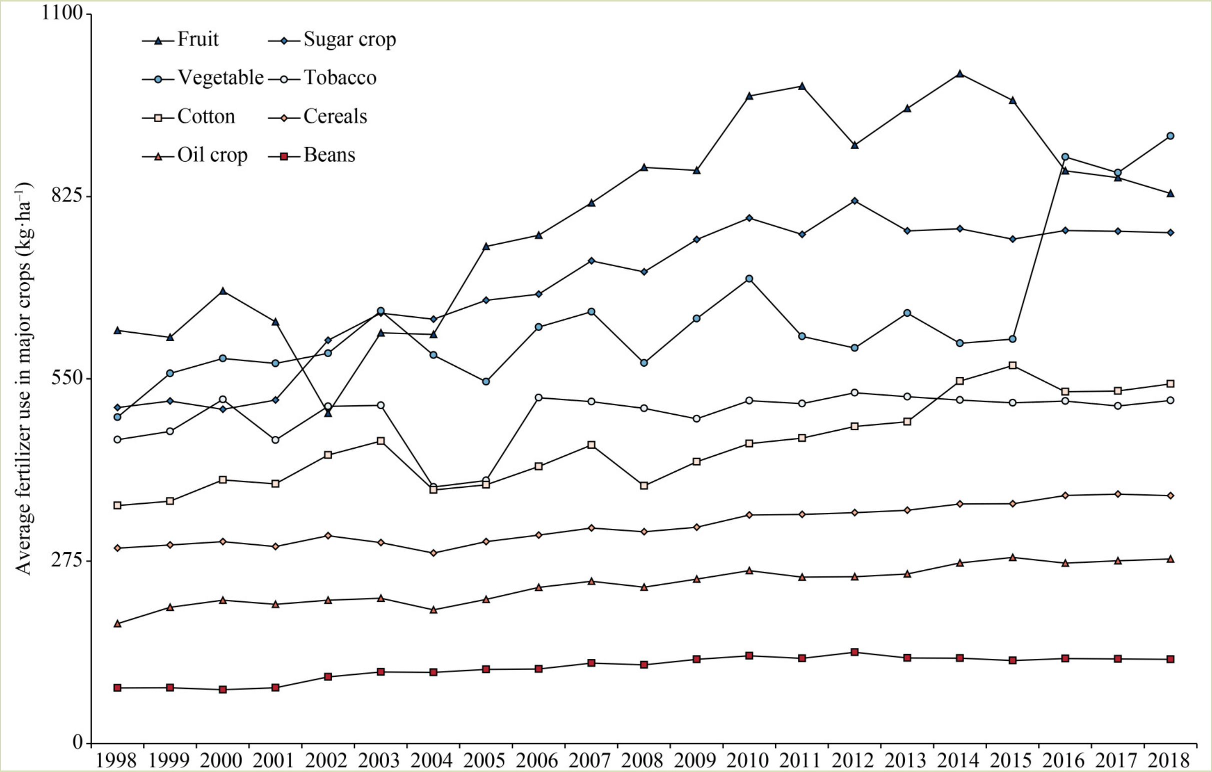


Fig. 2 Fertilizer use of major crops in China during 1998–2018. The amount indicates the sum of N, P_2O_5, and K_2O (kg·ha^{-1}). The sugar crop includes sugarcane and sugar beet and oil crop includes peanuts, rapeseed, sesame, soybeans, and sunflower. Values are means for each crop.

agriculture systems, biomass harvesting is unavoidable, but returning crop residues, as much as possible, is potentially beneficial for minimizing soil acidification induced by crop removal[52,53]. The uptake of the BCs presents in the form of companied cations and organic anions in plants (Table 2), and the localized net H^+ production is balanced by the release of hydroxyl ions during subsequent plant decomposition. In other words, the contribution of plants to soil acidification is close to zero in the case of whole plant biomass returned to the soil, since no BCs removal occurs but net carbon input from the biomass. The returned carbon potentially increases soil organic carbon content and CEC, therefore increasing soil buffering capacity. Therefore, continuous removal of harvested parts (for crops) from the field causes permanent soil acidification, and the net impact keeps increasing as the aboveground biomass increases. It is noteworthy that excess uptake of cations over anions often occurs more in leaves or stems than grains. For this reason, soil acidification is faster under continuous removal of both grain and straw than when grain only is harvested[54].

3.3 Soil acid buffering capacity

Apart from external factors (e.g., acid deposition and fertilizers), soil acid buffering capacity also affects the soil acidification progress, which is naturally determined by parent materials and the amount of soil organic matter (SOM) presents. Different parent materials lead to distinct soil physical, chemical and biological properties, which in turn alter soil buffering capacity and acidification processes[55]. When the soil contains free carbonate (e.g., $CaCO_3$), every mole of carbonate can neutralize 2 mol of H^+, therefore the buffering capacity of calcareous soils is determined by the content of carbonate within the soil. As soils move to the BC buffering system, soil CEC controls the absorption and supply of exchangeable cations in the soil, which is mostly contributed by SOM and clay[56]. Higher CEC indicates that the soil has more exchangeable sites to absorb H^+ and Al^{3+} from the soil solution avoiding the decrease in soil pH, and the soil is capable of providing more BCs to buffer excess H^+ input. Soils that developed from granitic parent materials are likely to be more acidic than soils derived from alluvial sediments partly due to their low SOM content and thus decreased exchange capacity for BCs[57]. Also, the composition of clay minerals is closely related to soil buffering capacity. The 2:1 type phyllosilicate mineral has more isomorphous substitution, resulting in permanent negative charges in soils that can adsorb BCs and thus have a large acid buffering capacity than the 1:1 type phyllosilicate with little or no isomorphous substitution[58]. Additionally, soil texture is also an important factor in controlling soil acidification rates. When the soil sand content is higher, the potential leaching of mobile anions such as NO_3^-, Cl^-, and SO_4^{2-} is also higher, causing greater soil acidification under similar fertilizer management[59].

In conclusion, there are three main factors that cause soil acidification in modern agriculture. Firstly, the excessive application of N fertilizers, especially ammonium-N fertilizers, increases NO_3^- leaching from the soil accompanied by BCs, resulting in large amounts of H^+ remaining in the soil. This also occurs with the overuse of S fertilizers. Secondly, the continuous removal of harvested crops depletes soil BCs, exacerbating the loss of soil acid buffering capacity. Additionally, cultivation of certain crops can increase soil acidification. For example, legumes produce and accumulate most of its N through N_2 fixation and the large input of N from legumes into soils increases NO_3^- leaching. Lastly, the soil acid buffering capacity, largely affected by factors such as the content of SOM and clay minerals, determines soil sensitivity to acidification processes. Soils with low levels of SOM and clay minerals tend to have a lower acid buffering capacity, and are more prone to acidification at a faster rate.

4 Mitigation strategies of soil acidification based on optimal nitrogen management

Soil acidity limits food production in many major agricultural production regions globally[4]. As N fertilizer is one of the most important factors exacerbating soil acidification, we propose effective and sustainable mitigation strategies for soil acidification based on optimal N management which can be achieved by carefully designed, region-specific optimization of N management, as discussed below.

4.1 Classified management scheme for soil acidification

Soil acidification accompanies gradual consumption of soil carbonate until it is consumed fully before the soil moves to the next buffering stage of BC exchange system and then to hydroxy aluminum/iron buffer system. This is reflected by the carbonate and exchangeable BCs losses, decrease in soil pH and increase in exchangeable Al to toxic level, thereby stunting growth or even crop failure[60–62]. Considering the ultimate target of sustainable crop production and sustainable land use, we propose a crop management scheme including three major soil buffering stages, that focuses on decreasing soil acidification rates, enhancing soil acid buffering capacity and alleviating the damage caused by acidification (Fig. 3).

The application of one or multiple mitigation principles and strategies is based on the nature and extend of the acidification problem, soil buffering capacity and the intrinsic characteristics of the soil buffering system in different ranges of soil pH. Giving attention to all these aspects is critical to make the solution effective. Note that while the soil buffering stage classification is arbitrary, they are based on sound knowledge of soil acidification and its management practices. We also assumed that, in most cases, the slightly acid soils (pH ranging from 5.5 to 6.5) are optimal for crop production. Also, it is important to note that care should be taken when defining critical/target soil pH for crop yields, as the measure soil pH will be dictated by the soil extractant, for example, soil pH of soil water extract is higher than that of $CaCl_2$ soil extract[63].

In soils with a pH above 6.5, generally categorized as non-acidic and slightly alkaline, the pH remains relatively stable due to the high acid buffering capacity of carbonate. In such soils, soil acidification has negligible or positive effects on crop growth. However, in the long run, continued acidification poses a risk to even such soils when the buffering materials (mainly carbonate) are exhausted. For example, a study has revealed that long-term N-induced acidification led to 7 Mha of cropland becoming carbonate-free from 1980 to 2020 in China[61]. Therefore, regular monitoring of soil quality, especially the carbonate content and soil pH by establishing fixed observation points is crucial in this stage of acidification to avoid potential acidification risks. However, considering that acidification may deplete soil inorganic carbon stock, lowering soil acidification rates by optimizing N form and input rate will be valuable even in this context.

In slightly acidic soil with a pH ranging from 5.5 to 6.5, soil acidification accelerates the losses of BCs causing substantial decline in soil fertility, which negatively affects crop growth. Hence, the primary objectives of measures at this stage aim to lower the soil acidification rate and supply the lost BCs, increasing the acid buffering capacity. These key measures include: (1) optimizing the form and amount of N fertilizers to minimize soil acidification caused by N fertilizer[64]; and (2) increasing the input of alkaline materials, such as crop straw, manure and biochar, rich in BCs, preventing soil acidification and enhancing soil buffering capacity[65]. The lower threshold pH for this stage is around 5.5, slightly higher than the turning point pH (~5.0) between the BC exchange and aluminum buffering systems because most field crops fail to obtain the maximum yield when soil pH is 5.0 or lower[63].

When the soil pH is below 5.5, soil acidification generally starts to decrease crop yield because of poor soil fertility (e.g., low Ca and Mg content due to large loss of BCs) and the toxicity caused by increasing levels of Al^{3+} and Mn^{2+}[60]. Priority measures at this stage should focus on raising soil pH quickly by alkaline amendment, for example, the application of lime or other alkaline substances to increase soil pH to ensure optimal crop production[66], or crop selection which increases the tolerance to elevated Al^{3+}. However, minimizing soil acidification rates is still essential, since it is not practical to apply high rates of alkaline substances considering the cost[67], as well as potential re-acidification after liming[68]. Therefore, decreasing soil acidification rates through optimize N fertilizer

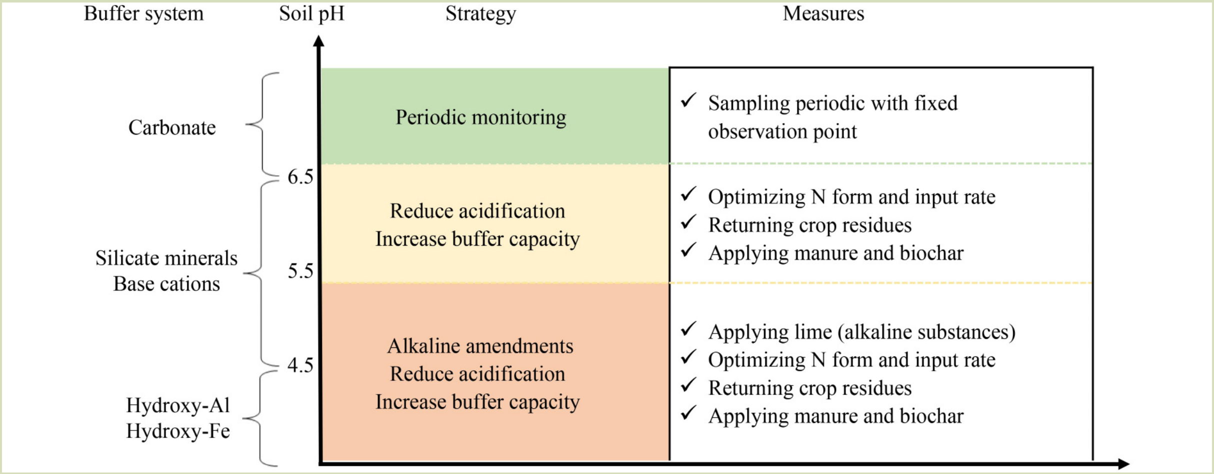


Fig. 3 Mitigation strategies for sustainable management of cropland acidification.

form and rate, and improving acid buffering capacity by increasing the input of organic manure or straw are also required to guarantee sustainable crop production and soil quality.

4.2 Optimizing nutrient management to mitigate soil acidification

China is a major contributor to the global food systems; however, nearly 20 Mha (47%) of croplands are acidic ($pH < 6.5$), mostly in southern China[69]. A recent study made an alarming observation that if the current nutrient management practices continue, future production losses due to soil acidification will be about 16% or more by 2050[70] even if the "Action Plan for Zero Growth of Fertilizer Use by 2020" remains in force. Since N cycling is the main factor exacerbating soil acidification, nutrient management measures based on N regulation are crucial for acidification management. The effective measures include optimizing the N application rate, managing N forms, optimizing N transformation processes and practicing an integrated soil–crop system management as shown in Fig. 4.

Optimize the N application rate. The optimal N application rate based on crop N demand should be considered first. Currently, crop N use efficiency in China is only about 40%, which could be increased to 60%–70% to maintain soil fertility and decrease environmental costs[71,72], implying a great operational space for decreasing N application. It was estimated that the N fertilizer use in China could be decreased from 27 Mt in 2020 to 21 Mt in 2060 toward an ambitious environmentally sustainable land-use transformation pathway[73]. Different crops have different potential to decrease N application in China. For example, the N input can be decreased by 21% to 28% in cereal crops (maize, rice, and wheat)[74], and a projected 38% decrease in the N application rate can be achieved for vegetables based on root zone N management without compromising the vegetable yield compared to conventional N fertilizer use[75]. Fruit systems have higher N inputs with lower N use efficiency, in which over 60% of N inputs can be eliminated if N management based on crop demand is followed[76]. In general, optimizing N application rate is an effective measure with great potential to decrease soil acidification. A recent meta-analysis showed that decreasing N input in grasslands from 300 to 50 $kg \cdot ha^{-1} \cdot yr^{-1}$ effectively slowed down soil acidification, with pH declining by 0.76 units in 300 $kg \cdot ha^{-1} \cdot yr^{-1}$ compared to 0.30 units in 50 $kg \cdot ha^{-1} \cdot yr^{-1}$ over a 5-year period[77]. In a field experiment, the acidification rate was decreased by 24% through optimization of urea application, and by 44% through optimization of ammonium-N fertilizer application in a wheat-maize system[71].

Optimize nitrogen forms. Nitrification is an important process producing H^+, which can be inhibited to mitigate soil acidification by adjusting fertilizer forms. In general,

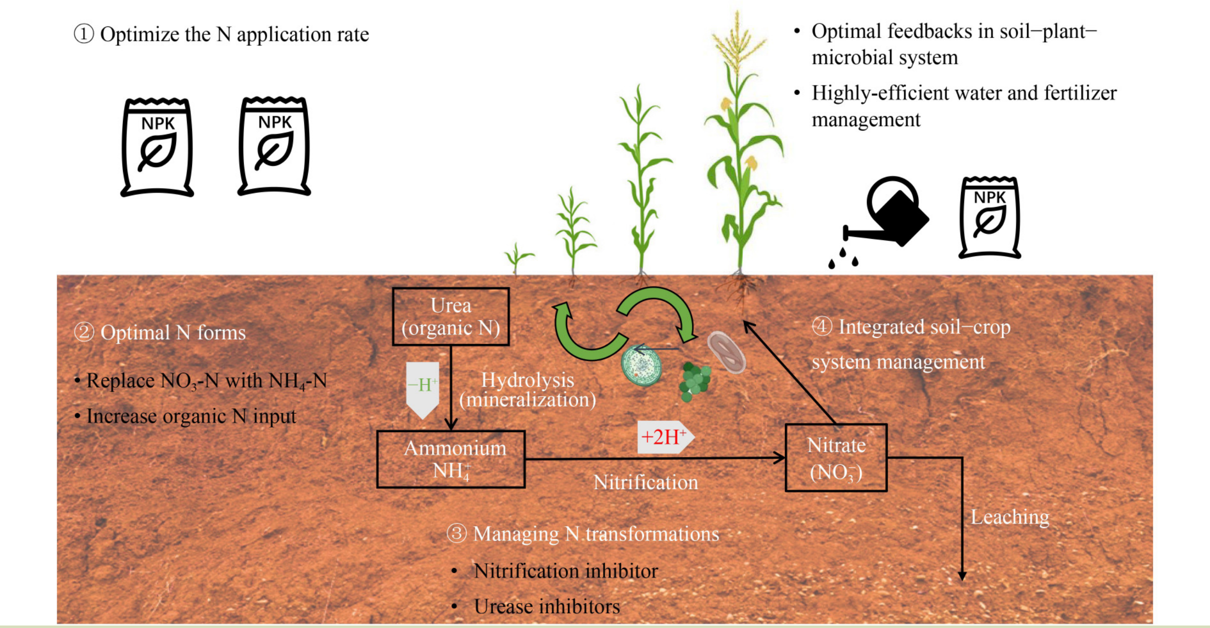


Fig. 4 Mitigation strategies for sustainable N management of cropland acidification.

ammonium-based fertilizers (e.g., $(NH_4)_2SO_4$ and NH_4Cl) acidify soils greater than urea[71,77,78]. Hao et al.[71] showed that the average acidification rate was 10 keq·ha^{-1}·yr^{-1} H^+ when urea was applied and 41 keq·ha^{-1}·yr^{-1} when NH_4Cl was applied in a wheat-maize double cropping system with a silty loam soil (initial pH was 5.1 in topsoil of 0–20 cm). In contrast, rhizosphere soil pH increased with the application of nitrate-N fertilizers[46]. Therefore, transforming ammonium-based fertilizers to nitrate-based fertilizers can decrease soil acidification given that most upland crops, such as maize, tobacco, vegetables and wheat, are more effectively grown with nitrate-based fertilizers[79,80].

In addition, replacing part of N fertilizers with organic N fertilizers is also effective in mitigating soil acidification, as most organic fertilizers are alkaline, directly neutralizing soil acidity and supplying BCs[81,82]. Also, organic fertilizers raise SOM content, which can effectively improve acid buffering capacity. Research has shown that replacing 50% of mineral N fertilizers by organic N increased the pH buffering capacity of Alfisol by 60% to 81%[82]. This can also effectively decrease the acidification by inhibiting nitrification of NH_4^+[82,83]. Additionally, unlike mineral fertilizers, nutrients in organic fertilizers are mostly slow-releasing, which lessens nutrient runoffs, help balances crop demand-based nutrient supply, slows the rate of soil acidification and improves soil and environmental quality[84]. The results of Cai et al.[64] showed that 40%, or more, of total N from manure can prevent or reverse acidification of red soil, and provide all the P crops required. Apart from organic manure, the combined application of urea and biochar can decrease the nitrification process, thus mitigating soil acidification[85]. Also, some slow-release or smart fertilizers (e.g., chitosan nanohybrid) can also decrease the N fertilizer application as a compensatory substitute[86].

Managing nitrogen transformation processes. The nitrification of 1 mol of NH_4^+ to form 1 mol of NO_3^- produces 2 mol of H^+. Therefore, it is plausible and effective to control soil acidification by regulating the nitrification process using nitrification inhibitors. Research has shown that adding the nitrification inhibitor DMPP along with N fertilizers increased soil pH, decreased N input by 37% without yield loss, and decreased NO_3^- leaching, promoting sustainable N management in tea plantations[87]. Also, using nitrification inhibitors can decrease the emissions of N_2O due to the less NO_3 produced[88]. The combined application of N and P can change N:P stoichiometry and negatively affect the abundance of ammonia-oxidizing bacteria and ammonia-oxidizing archaea, thereby inhibiting the nitrification process of urea in soil[89]. In addition, incorporating urease inhibitors into fertilizers is also an important technology to decrease N loss. Urease inhibitors can lower the activity of soil urease, slow down the hydrolysis of urea in soils, and decrease the loss of NH_3 volatilization. Individual application or co-application of urease inhibitors, chemical nitrification inhibitors and biochar could mitigate NH_3 volatilization by 12.5%–26.5%, N_2O emission by 62.7%–73.5%, and N leaching loss by 17.5%–49.0% in a wheat growth pot experiment with a calcareous soil developed from purplish shale[90].

Integrated soil–crop system management. Integrated soil-crop system management often recommends precise fertilizer application combined with optimized field management, which improves the utilization rate of fertilizer and slows down cropland acidification[91]. For example, optimizing a fertigation system to improve fertilizer application accuracy can decrease N input by 11% on average compared with the traditional irrigation-fertilizer system[92], slowing the rate of soil acidification. In addition, fertilizer placement close to seeds or plant roots (deep fertilizer placement) ensures high nutrient availability and nutrient use efficiency. A systematic review showed that deep fertilizer placement led to a 3.7% higher yield and 11.9% higher nutrient content in aboveground parts than fertilizer broadcast[93]. An integrated land-use management system for pasture and agriculture with precise fertilizer application achieve soil nutrient balance and significant decrease in fertilizer use[94]. Therefore, a combination of precise fertilizer application and optimized field management can improve fertilizer utilization efficiency and decrease fertilizer loss.

5 Conclusions

Soil acidification is one of the most serious threats to global food production and human health. Natural acidification occurs due to the net leaching of bicarbonate and organic acid accompanied by BCs, which is slowly driven by excess precipitation, surpassing soil water loss through evaporation. However, human activities have substantially increased soil acidification in croplands. Application of acidifying fertilizers, especially ammonium-based fertilizers which increases acid production via N transformations, excessive N application causing leaching of nitrate along with BCs, and continuous BCs removal by crops all lead to soil acidification.

Considering the distinct soil acidification status, processes and the rate of progress, different acidification mitigation strategies were proposed for soils with pH of > 6.5, 5.5–6.5, and < 5.5 accordingly. Generally, the soil with a pH > 6.5 have a strong acid buffering capacity, e.g., calcareous soils with free carbonate, which therefore only requires frequent monitoring of soil carbonate content and pH to avoid acidification risks; strongly-acid soils are potentially at aluminum toxicity risks and are required to raise soil pH to 5.5–6.5, an optimal pH range that ensures adequate nutrient supply for most crops. However, the soil with this range of pH is the most acid-sensitive, since soil exchangeable BCs is the main buffering substance. Decreasing the soil acidification rate and enhancing soil buffering capacity are then required in the stage.

Optimal N management can be regarded as the core of the mitigation strategies to maintain the optimal pH for crop production. The effective strategies include: (1) optimizing the N application rate, (2) managing N forms, (3) optimizing N transformation processes, and (4) integrated soil–crop system management. Optimizing N input based on crop demand can decrease nitrate losses via leaching, thereby reducing the loss of BCs. By adjusting N forms from ammonium-based to urea-based or nitrate-based, the acid production from nitrification of ammonium to nitrate can be significantly decreased; replacing synthetic fertilizer with organic fertilizers (crop residues, manure, and biochar) rich in BCs has the same effects and they also supplement soil BCs pool, enhancing soil buffering capacity. Also, use of nitrification inhibitors and urease inhibitors can decrease acidification from N transformations by minimizing N losses and increasing N use efficiency. The same effects occur with the integrated soil–crop system management by combining precision fertilizer application and optimal field management. Implementing these strategies can simultaneously decrease soil acidification and N losses, therefore optimizing crop production, minimizing environmental pollution, and promoting agricultural sustainability.

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Compliance with ethics guidelines

Pengshun Wang, Donghao Xu, Prakash Lakshmanan, Yan Deng, Qichao Zhu, and Fusuo Zhang declare that they have no conflicts of interest or financial conflicts to disclose. This article does not contain any studies with human or animal subjects performed by any of the authors.

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