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How application of agricultural waste can enhance soil health in soils acidified by tea cultivation? a review --Manuscript Draft--

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Abstract:	<p>Tea is one of the world's most consumed drinks and an important crop of many developing countries. As tea plants can retain their productive life span for decades, intensive tea cultivation has negative impacts on soil health properties and the environment. Globally, soil acidification in tea plantations has become a severe issue, threatening soil health, tea production and the environment. However, the ways in which soil acidification affects soil health, tea productivity and the environment, and suitable methods to control this issue have not been critically reviewed. Here, we review the mechanisms of tea soil acidification and its consequences; the potential of common agricultural wastes for ameliorating soil acidity and enhancing soil health and crop productivity, as well as reducing environmental pollution under tea cultivation. We show that intensive application of chemical nitrogen is the main cause of soil acidification in tea plantations, while tea plants also play a part in accelerating tea soil acidity. Agricultural waste and products derived from these resources have a great potential to correct soil acidity, enhance soil health and tea productivity and quality. These soil amendments also introduce risks such as heavy metals and/or pathogens as well as production and application costs that require consideration.</p>
Suggested Reviewers:	Wenyan Han, Dr Chinese Academy of Agricultural Sciences Tea Research Institute hanwy@tricaas.com Dr Han published several relevant papers on soil acidification in tea plantations L Wang, Dr

	<p>Nanjing Institute of Environmental Sciences pingfanshangren@126.com Dr Wang published relevant paper about the utilization of agricultural wastes such as biochar for increasing low soil pH in tea plantations</p> <p>Jianyun Ruan, Dr Institute of Genetics and Developmental Biology Center for Agricultural Resources Research jruan@tricaas.com Dr Ruan is very much interested on the effects of organic substitution for synthetic N fertilizer on soil bacterial diversity and community composition in tea plantations</p>
<p>Response to Reviewers:</p>	<p>Environmental Chemistry Letters ECLE- D- 21-00537 Responses to the Editor's comments</p> <p>We are sincerely indebted to the Editor for giving comments on our manuscript. We have revised our manuscript following all the comments and we hope that this revised version will fit with the editor's expectations.</p> <p>Editor's comments</p> <p>1. The introduction is too long and needs to be shortened down to 1-2 paragraphs Answer: Thanks for the Editor's comment. The introduction has now been rewritten and the length has been reduced by around a half.</p> <p>2. Possibility to discuss the ocean and other acidifications for attracting a wider range of readers. Answer: We have now added a new sub-section 2.1 to briefly discuss the status and mechanism of ocean and soil acidification and how they relate to agricultural activities.</p> <p>3. The figures need to be accurately presented (texts, legend, white space, Y and X axis...) Answer: We have now carefully revised all the figures attached in the manuscript. We hope that they will fit with the editor's expectations.</p> <p>4. Write at the end of each article section a conclusion of about 1-2 sentences to summarize the major points of the section and its significance. Answer: Thanks for the Editor's advice. We now reviewed the whole manuscript and added summary/ conclusion to many sections/subsections where we believe that they are accurate.</p> <p>5. Finding relevant articles published by Environmental Chemistry Letters (5-10 articles, using keywords and search from the journal home page) and consider citing them as the references. I believe we have published articles concerning soil and its degradation, tea as well as acidification. Answer: Thanks for the Editor's advice. We have been carefully checked and cited 8 articles published by ECL as references intext and highlighted these references in the bibliography. They are as follow:</p> <p>1. Akhil D, Lakshmi D, Kartik A, Vo D-VN, Arun J, Gopinath KP (2021) Production, characterization, activation and environmental applications of engineered biochar: a review. Environ Chem Lett 19: 2261-2297. http://doi.org/10.1007/s10311-020-01167-7 2. Gunarathne V, Ashiq A, Ramanayaka S, Wijekoon P, Vithanage M (2019) Biochar from municipal solid waste for resource recovery and pollution remediation. Environ Chem Lett 17: 1225-1235. https://doi.org/10.1007/s10311-019-00866-0 3. Ochedi FO, Yu J, Yu H, Liu Y, Hussain A (2021) Carbon dioxide capture using liquid absorption methods: a review. Environ Chem Lett 19: 77-109. https://doi.org/10.1007/s10311-020-01093-8 4. Patra BR, Mukherjee A, Nanda S, Dalai AK (2021) Biochar production, activation</p>

- and adsorptive applications: a review. *Environ Chem Lett* 19: 2237-2259. <https://doi.org/10.1007/s10311-020-01165-9>
5. Rana A, Rana S, Kumar S (2021) Phytotherapy with active tea constituents: a review. *Environ Chem Lett* 19: 2031- 2041. <https://doi.org/10.1007/s10311-020-01154-y>
6. Saliu T, Oladoja N (2021) Nutrient recovery from wastewater and reuse in agriculture: a review. *Environ Chem Lett* 19: 2299–2316. <https://doi.org/10.1007/s10311-020-01159-7>
7. Sánchez A, Artola A, Font X, Gea T, Barrera R, Gabriel D, Sánchez-Monedero MÁ, Roig A, Cayuela ML, Mondini C (2015) Greenhouse gas emissions from organic waste composting. *Environ Chem Lett* 13: 223-238. <https://doi.org/10.1007/s10311-015-0507-5>
8. Sharma H, Dhir A (2021) Capture of carbon dioxide using solid carbonaceous and non-carbonaceous adsorbents: A review. *Environ Chem Lett* 19: 851-873. <https://doi.org/10.1007/s10311-020-01118-2>

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4 **How application of agricultural waste can enhance soil health in soils acidified by tea cultivation: a**
5
6 **review**

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3 **Application of agricultural waste to enhance soil health in soils acidified by tea cultivation: a review**
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8 **Abstract**
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10 Tea is one of the world's most consumed beverages and an important crop of many developing countries.
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12 As tea plants can retain their productive life span for decades, intensive tea cultivation has negative
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14 impacts on soil health properties and the environment. While soil acidification in tea plantations is
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16 globally acknowledged to be a severe issue, threatening soil health, tea production and the environment,
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18 the ways in which soil acidification affects soil health, tea productivity and the environment, and suitable
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20 methods to control this issue have not been critically reviewed. Here, we review the mechanisms of tea
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22 soil acidification and its consequences; the potential of common agricultural wastes for ameliorating soil
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24 acidity and enhancing soil health and crop productivity, as well as reducing environmental pollution under
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26 tea cultivation. We show that intensive application of chemical nitrogen is the main cause of soil
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28 acidification in tea plantations, while tea plants also play a part in accelerating tea soil acidity.
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30 Agricultural waste and products derived from these resources have a great potential to correct soil acidity,
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42 **Keywords:** Agricultural waste · Soil acidification · Biochar · Organic manure · Soil health · Tea
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1. Introduction

Soil acidification has been a major threat to soil health and environmental sustainability in various agricultural systems and regions (Dai et al. 2017; Li et al. 2016; Yan et al. 2020), and occurs in many tea growing countries, such as China (Lin et al. 2019; Ni et al. 2018; Zou et al. 2014), India (Bandyopadhyay et al. 2014), Japan (Oh et al. 2006), Sri Lanka, Rwanda (Mupenzi et al. 2011), and Vietnam (Huu Chien et al. 2019). In China, the leading global tea producer and exporter, greater soil acidification occurred in tea plantations compared to other cash and cereal cropping systems, with 46% of tea plantations nationwide reporting soil pH below 4.5 (Yan et al. 2020). The reduction of soil pH in tea plantations will have impacts of soil characteristics by changing soil chemical processes, resulting in soil nutrient losses and imbalance, and increasing occurrence of Al and Mn toxicity (Alekseeva et al. 2011; Ni et al. 2018; Yan et al. 2018). In addition, soil acidification significantly degrades the diversity and functionality of soil organisms (Goswami et al. 2017; Li et al. 2017). While soil acidification occurs naturally in tea plantations and increases with increasing tea plant age and plant density, intensive application of mineral nitrogen (N) is the main cause of the issue (Li et al. 2016; Yan et al. 2018).

The use of agricultural organic waste products to ameliorate soil acidification has been recognized in Agriculture systems worldwide (Cai et al. 2015; Cornelissen et al. 2018; Dai et al. 2017). By definition, agricultural wastes or agriculture by-products are the unwanted residues generated from agriculture activities, such as crop residues, animal manure, forest waste, vegetable matter and weeds (Dai et al. 2018; Ramírez-García et al. 2019). Animal wastes, green manures and products derived from these wastes such as biochars and compost are generally alkaline in nature and have high pH buffering capacity which can neutralize soil acidification (Cai et al. 2021; Rayne and Aula 2020). Also, the presence of basic cations such as Mg^{2+} and Ca^{2+} , and organic anions in these materials contribute to increased soil pH (Cai et al. 2021; Tang et al. 2013). In addition to increasing soil pH, agricultural wastes have long been known to enhance soil health, including soil physical, chemical and biological properties (Bhatt et al. 2019; Cai et al. 2021; Rayne and Aula 2020). Globally, an estimated of 1 billion tons of agricultural wastes per year is generated, which China, USA and

India being the largest agricultural waste producing nations worldwide (Fig. 2) (Clauser et al. 2021; Obi et al. 2016), and this figure has been projected to increase rapidly because of the growing demand of agricultural products (Dai et al. 2018; Wei et al. 2020). Thus, the utilization of agricultural wastes as soil amendments could be a win-win strategy, which can benefit not only soil health but also reduce the pressure of using fossil fuels, mitigate serious environmental problems and human health threats (Bijarchiyan et al. 2020; Mpatani et al. 2021).

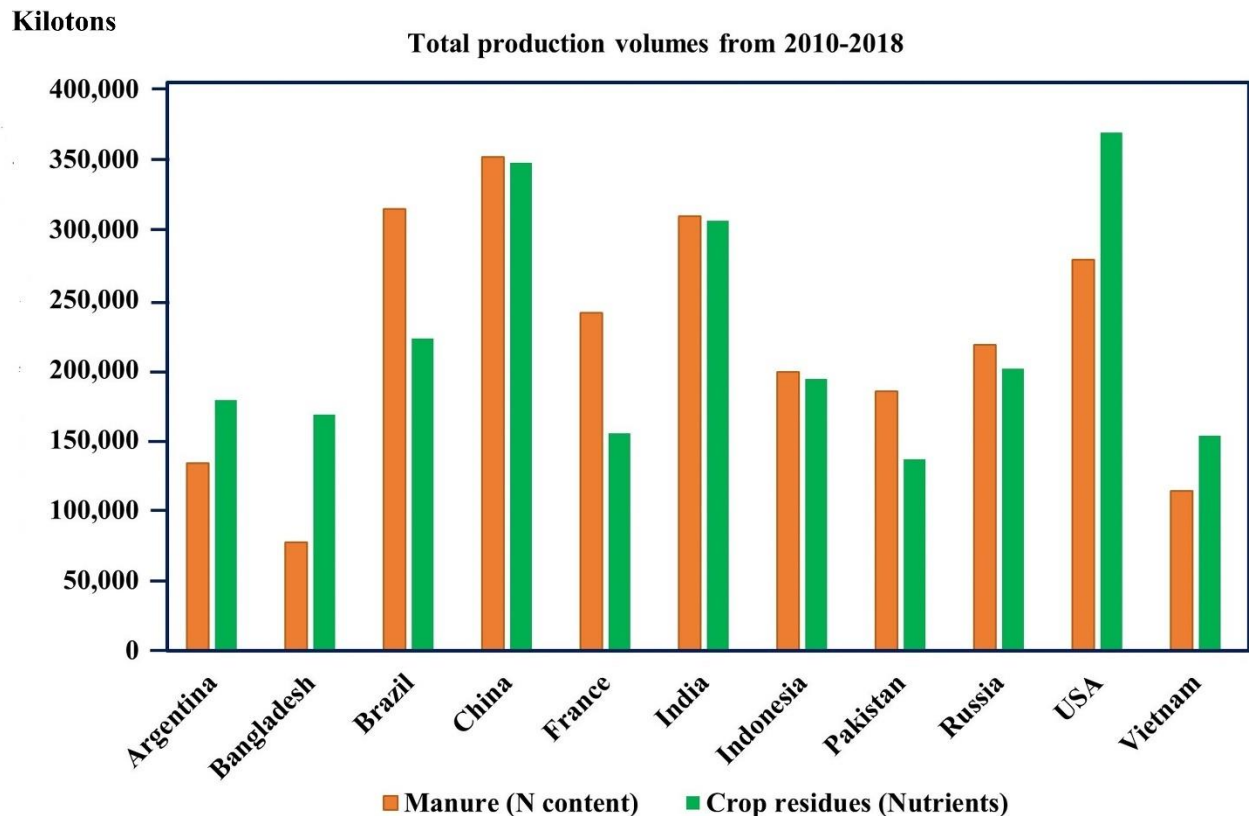


Fig. 1 Total production volumes of manures and crop residues in the world’s largest agricultural waste generating countries from 2010-2018. Manures and crop residues were measured by kilotons of N content and nutrients, respectively. Of these countries, China, India, Vietnam, Indonesia and Argentina have been also the top global tea producers in the same period. Data was based on FAO (2021).

Studies on the utilization of agricultural wastes and its components to alleviate soil acidification caused by tea cultivation have been well-reported in China, but poorly implemented in other parts of the world. Among these soil amendments, biochar application is considered as the most effective way to counter low soil pH,

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3 resulting in subsequent benefits to soil health and tea productivity (Wang et al. 2018; Wang et al. 2014; Yan
4 et al. 2021). Several studies have also reported the positive impacts of organic manures on acidification of tea
5 soil (Lin et al. 2019; Qiu et al. 2014), while the benefit of plant residues varied significantly. Recent reviews
6 have highlighted the potentials of biochar in mitigating soil acidification (Dai et al. 2017), and the effects of
7 organic manure on soil health (Bhatt et al. 2019; Rayne and Aula 2020). However, to our best knowledge,
8 there has not been any reviews published that specifically focus on the mechanisms and consequences of
9 acidification in tea plantation soils, the advantages and drawbacks of using agricultural wastes and other
10 relevant options in alleviating soil acidification as a result of long-term tea cultivation. This review provides a
11 comprehensive overview of mechanisms and consequence of soil acidification by tea cultivation, the
12 utilization of agricultural wastes and its products on mitigating soil acidification and enhancing soil health
13 properties under tea plantations.
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30 **2. Soil acidification by tea cultivation and its consequences**

31 *2.1. Ocean and soil acidification*

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36 Ocean and soil acidification have been widely reported as the most critical issues, affecting the sustainability
37 of numerous ecosystems and regions around the world (Ochedi et al. 2021; Yan et al. 2020). Ocean acidity
38 has increased by ~25% since 1860s, and the soil pH values of 50% of total arable land worldwide are below
39 5.5 (Dai et al. 2017; Hall et al. 2020). Ocean acidification appears due to rising atmospheric carbon dioxide
40 (CO₂) concentrations and absorption by seawater, which subsequently leads to a fall of pH and carbonate ion
41 concentrations in surface seawater (Agostini et al. 2018; Sharma and Dhir 2021). Ocean takes up around 25%
42 of global anthropogenic CO₂, making it the largest atmospheric CO₂ absorbent on Earth (Hauck and Völker
43 2015). Among the CO₂ emission sources, agriculture directly contributes around 14% of the total amount
44 globally, and this proportion is likely to be exceeded in the future (Ayyildiz et al. 2021). Intensive agriculture
45 and land use practices have been also the main causes of global soil acidification, particularly inappropriate
46 uses of ammonium-based fertilizers (Cai et al. 2015; Dai et al. 2017). Additionally, soil nutrient leaching,
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product removal, acidic parent materials, acid deposition and host plants are all likely to be significant factors resulting in soil pH reduction (Tang et al. 2013, Yan et al. 2020).

2.2. Soil acidification in tea plantations

Tea plant

Tea (*Camellia sinensis* Kotze) is one of the oldest and most popular beverages in the world, and is an important crop being cultivated in around 50 countries (Gebrewold 2018). Global tea production in 2019 was more than 9.2 million tons, valued at approximately \$US55.3 billion (Fig. 2) (Allied Market Research 2020; Food and Agriculture Organization (FAO) 2021).

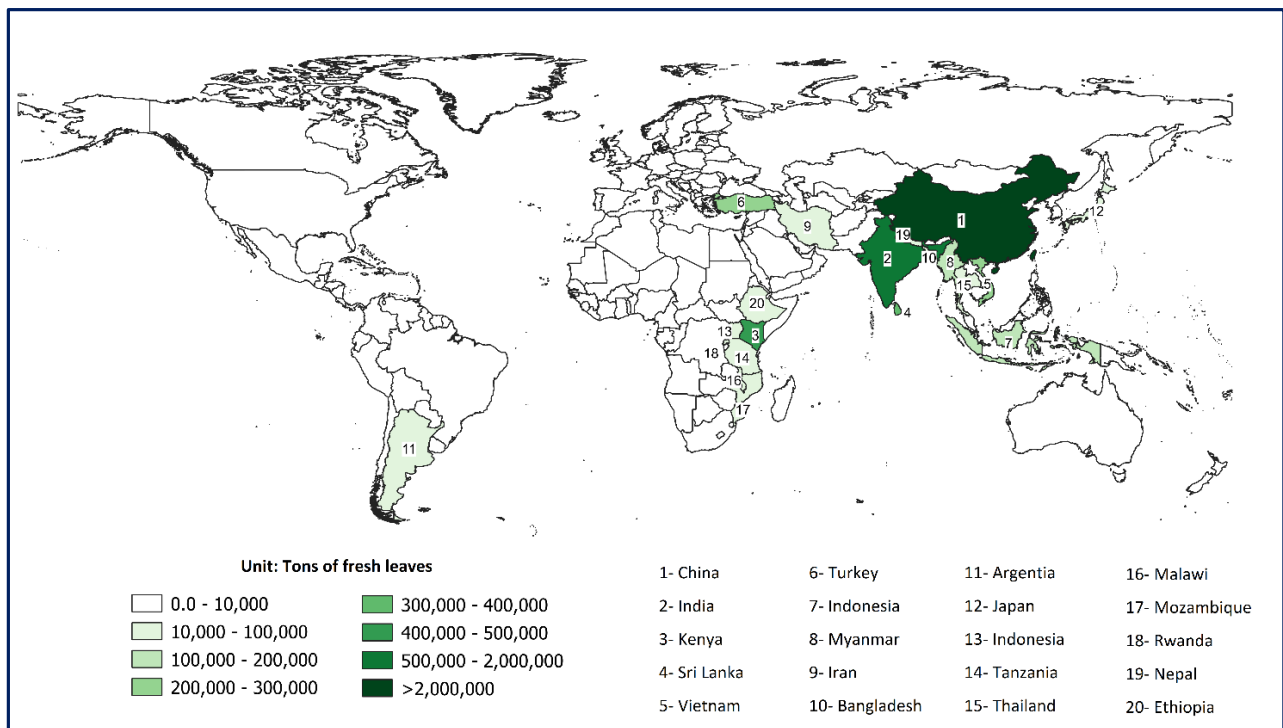


Fig. 2 Map of the 20 world's largest tea producing nations in 2019. China was the largest tea producer worldwide in 2019, followed by India, Kenya, Sri Lanka and Vietnam. Most of the global tea producers are in Asia and Africa continents. The top 20 global tea producing countries contributed to around 70% of total global tea production volume in the same year. Data was retrieved from FAO (2021).

Tea plants are native to the Asia continent, but they can adapt to a wide range of soil and climatic conditions (Rana et al. 2021; Yan et al. 2018; Yao et al. 2012). This perennial crop requires acidic soils for optimum

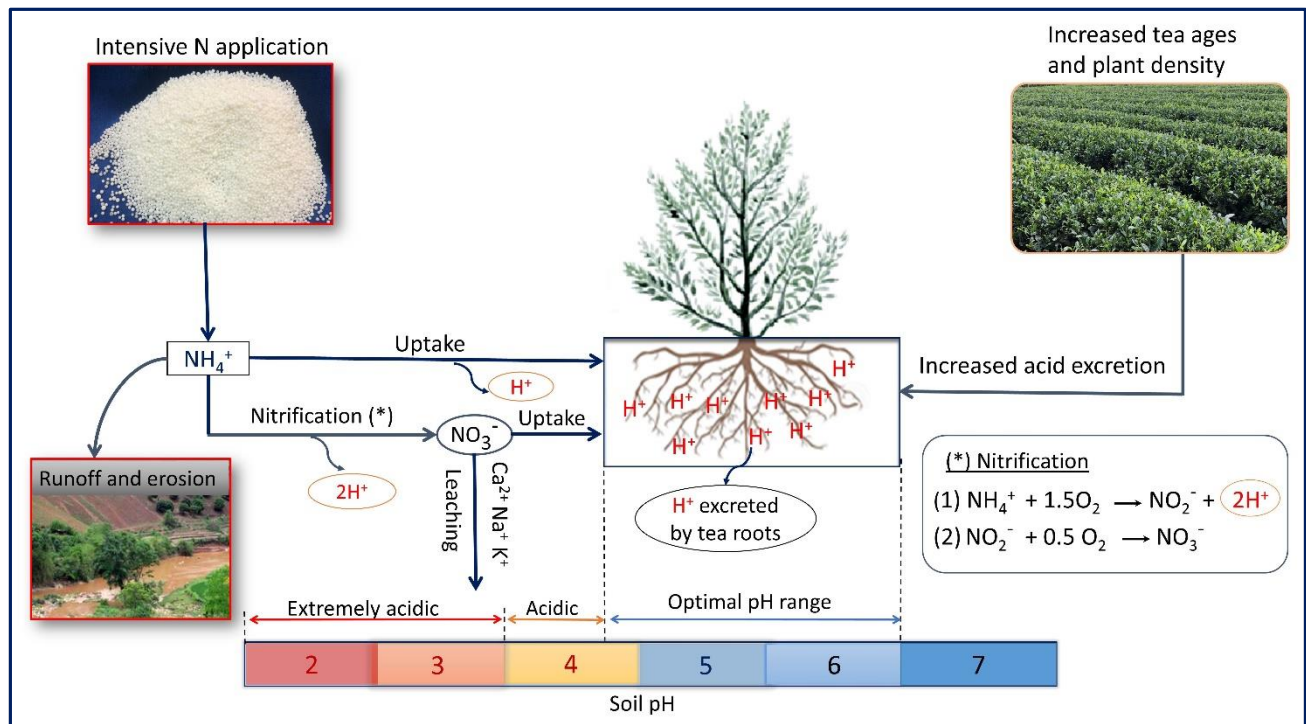
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3 growth and productivity, with the optimal soil pH for tea plants being between 4.5- 6, and the plant
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5 themselves are capable of acidifying soil (Fig. 3) (Gebrewold 2018; Li et al. 2016). Being a woody perennial,
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7 tea plants can remain their productivity for decades, and thus have long-term interactions with soil organisms
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9 and physicochemical processes, affecting soil health and plant productivity (Arafat et al. 2020; Yan et al.
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11 2020).
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13 14 15 *Soil acidity by tea cultivation practices*

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17 Soil acidification in tea plantations results predominantly from inappropriate management practices,
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19 particularly the intensive overuse of mineral N (Li et al. 2016; Yan et al. 2018). Tea growers apply N to
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21 ensure high tea productivity and as a replacement for soil nutrient loss. In Japan, tea fields are amended with
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23 more than 1000 kg/ha of N fertilizers per annum (Abe et al. 2015; Zou et al. 2014) and a majority of tea
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25 farmers in China apply a large amount of nitrogen to ensure high tea yield and maintain soil fertility (Yan et
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27 al. 2018). A recent study has shown that nitrogen fertilizer application rate can even reach 1200 kg/ha in
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29 Chinese tea plantations (Wu et al. 2016). Soil pH significantly reduces when N fertilizers such as ammonium
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31 nitrate and urea is applied above 50kg/ha/year, and increased N added rate will accelerate soil acidification
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33 (Tian and Niu 2015). Moreover, heavy N application results in greater decrease of subsoil pH compared with
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35 that of the topsoil (Ni et al. 2018). When fertilizers are applied at 2700 kg/ha, only 18,3% of applied nitrogen
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37 were absorbed by tea plants and of that, about 52% of nitrogen were stored in the soil, and 30% were lost
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39 through runoff, polluting surrounding watercourses and soils (Chen and Lin 2016; Xie et al. 2021).
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48 The main mechanisms of soil acidification resulting from inappropriate management practices in tea
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50 cultivation are shown in Fig. 3. When $\text{NH}_4^+\text{-N}$ fertilizer is applied, tea plants directly take up the nutrient and
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52 tea roots subsequently excrete an equivalent proton into the rhizosphere, causing the concentration of
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54 hydrogen ions to increase. NH_4^+ nitrification leads to a net production of 2 mol H^+ for each mol of NH_4^+
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56 applied, contributing to the decrease of soil pH (Hui et al. 2010; Li et al. 2016; Yan et al. 2020). Cai et al.
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58 (2015) estimated that an application rate of 300kg/ha/year of N fertilizers could produce 21.4 kmol
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60 H^+ /ha/year by the nitrification processes. N fertilizer application in the long term also promoted the
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3 accumulation of exchangeable Al^{3+} including hydrolysis, which further generated H^+ and aggravated the
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5 acidification of tea plantation soils (Zhang et al. 2020). Finally, increasing tea plant age and planting density
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7 also result in an increase of organic and carbonic acids induced by tea roots into the rhizosphere, which
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9 facilitate soil acidification (Hui et al. 2010). Tea plantation soil is not acidified at planting densities of 5000
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11 plants/ ha (Li et al. 2016).



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41 **Fig. 3** The main mechanical causes of soil acidification by tea cultivation. Heavy addition of N fertilizers is
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43 the main reason causing soil acidification, and the accumulation of organic and carbonic acids released by tea
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45 roots also play a part in acidifying tea plantation soils.

50 *Soil acidification by tea plants*

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53 Acidification of soils may naturally occur in soils cultivated with tea – even without any imposed N proton
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55 additions, and this issue becomes more challenging with increasing tea plantations (Arafat et al. 2017; Han et
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57 al. 2007; Li et al. 2016). In tea plantations, soil pH in the topsoil naturally decreased by 0.071 units per
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59 annum, and the values following 13, 34 and 54 years of tea cultivation were 1,1; 1,62 and 2,07 units
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3 respectively (Hui et al. 2010; Ni et al. 2018). The acidification rate observed in the cultivated soil layers (0-
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5 10cm) could reach 4.40 kmol H⁺/ha/year during the 0-13 years of tea cultivation period (Hui et al. 2010).
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7 Organic acids secreted by tea roots such as malic acid, citric acid, and oxalic acid are the main proton source
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9 for soil acidification in the tea tree- soil systems (Fig. 3) (Yan et al. 2018). Tea roots also excrete carbonic
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11 acids and polyphenols which can aggravate soil acidification, affect soil nutrient release and subsequent
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13 element uptake (Ni et al. 2018; Wang et al. 2013). Additionally, the accumulation of chemical compounds
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15 such as epigallocatechin gallate, epigallocatechin, epicatechin gallate, catechin, and epicatechin, found in the
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17 tea residues also negatively affect soil pH and soil health properties (Arafat et al. 2020). Thus in summary,
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19 intensive application of N fertilizers is the main cause of soil acidity under tea plantations, and the
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21 accumulation of acid excreted by tea plants promotes the acidification.
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29 ***2.3. Consequences of acidification in tea plantation soils***

30 *Soil chemical parameters*

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34 Soil acidification negatively affect chemical processes and properties of tea plantation soils (Fig. 4). One of
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36 the most serious challenges of soil acidification under tea cultivation can be the reduction and imbalance of
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38 nutrient base cations, including Ca²⁺, Mg²⁺, Na⁺ and K⁺ (Alekseeva et al. 2011; Ni et al. 2018; Zhang et al.
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40 2020). Under heavy N application, released protons (H⁺) may replace the soil exchange base cations, which
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42 may have leached with the NO₃⁻ as accompanied cations due to the charge balance in soil solutions (Cusack et
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44 al. 2016; Ni et al. 2018). Moreover, a significant increase of Al³⁺ and Mn²⁺ has been widely recorded in acidic
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46 tea plantation soils, which could lead to Al and Mn toxicity (Alekseeva et al. 2011; Hui et al. 2010). Under
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48 acidic soil conditions, mineral Al solubilizes into trivalent Al³⁺, which is highly toxic to animals, plants and
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50 microorganisms (Zioła-Frankowska and Frankowski 2018). Gruba and Mulder (2015) indicated that the
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52 concentration of exchangeable Al maximizes in soils with a pH_{H2O} ≈ 4.2. Similarly, with decreasing soil pH,
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54 the amount of exchangeable Mn²⁺ increases in the soil solution (Millaleo et al. 2010). High concentration of
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56 Al³⁺ can inhibit the expansion, elongation, and division of root cells, reducing water and nutrient uptake by
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58 the root systems (Wang et al. 2015). Similarly, high levels of Mn²⁺ in soil is one of the main factors causing
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3 nutrient imbalances, especially with divalent cations such as Mg^{2+} , Zn^{2+} and Ca^{2+} (Venkatesan et al. 2010).
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5 Soil acidification can also promote the dissolution of minerals and movement of Fe in the profile, resulting in
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7 reduction of ferrimagnetic mineral content (Alekseeva et al. 2011). Increased Al and Mn toxicity have been
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9 considered as the most serious consequences of soil acidification by tea cultivation regarding soil chemical
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11 property.
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14 15 16 17 18 *Soil biological parameters*

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20 Soil pH is a crucial factor affecting soil organisms (Li et al. 2018; Neina 2019). Mulder et al. (2005) indicated
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22 that soil acidification has close inverse relationship with bacterial, fungal, nematode and arthropod
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24 abundance. Long-term soil acidification is responsible for reduction of soil microorganisms, which are
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26 regulating the reduction in soil pH by both ecological and evolutionary mechanisms because of the
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28 environmental changes (Zhang et al. 2015). In tea plantations, a low soil pH ($pH < 4$) could lead to a loss of up
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30 to 70% of important soil biota (Han et al. 2007). Likewise, soil fauna communities were significantly higher
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32 in the soil with pH 7.0 (21 classes) compared to acidic soil with pH 2.5 (11 classes) and pH 3.5 (14 classes).
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34 In this study, in terms of total individuals, the figures were 3710 (pH 7.0); 759 (pH 3.5) and 645 (pH 2.5)
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36 (Wei et al. 2017). Severe soil acidification also leads to significant decreases in soil enzymatic activities,
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38 microbial activities, and microbial biomass (Li et al. 2017; Zhang et al. 2015). Arafat et al. (2019) found a
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40 close association between the decline of some beneficial fungus such as *Mortierella elongatula* and
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42 *Mortierella alpina* and a low soil pH caused by long-term tea monoculture. Soil acidification also enhances
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44 the environment for growth of some soil-borne pathogen diseases. For instance, when soil pH reduced from
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46 5.07 to below 3.5 as a result of 35 years of continuous tea monoculture, the abundance of some pathogenic
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48 bacterial species including *Fusarium oxysporum*, *Fusarium solani*, and *Microidium phyllanthi*, which are
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50 responsible for diseases in tea plants such as root rot and die back, was significantly increased (Arafat et al.
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52 2019). Investigating the relationship between soil acidity and bacterial wilt disease, Li et al. (2017) found that
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54 the proportion of soil affected by bacterial wilt much higher when the soil pH lower than 5.5, and
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56 significantly less as the soil pH increases. Likewise, the highest population of *Xiphinema chambersi* was
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found in soil with a pH 4.5, and the figure decreased when soil pH increased from 4.5 to 6.4 (Chen et al. 2012). Thus, soil acidification by tea cultivation could not only impact soil beneficial microbial diversity, but also promote the development of some potentially pathogenic microbes (Fig. 4).

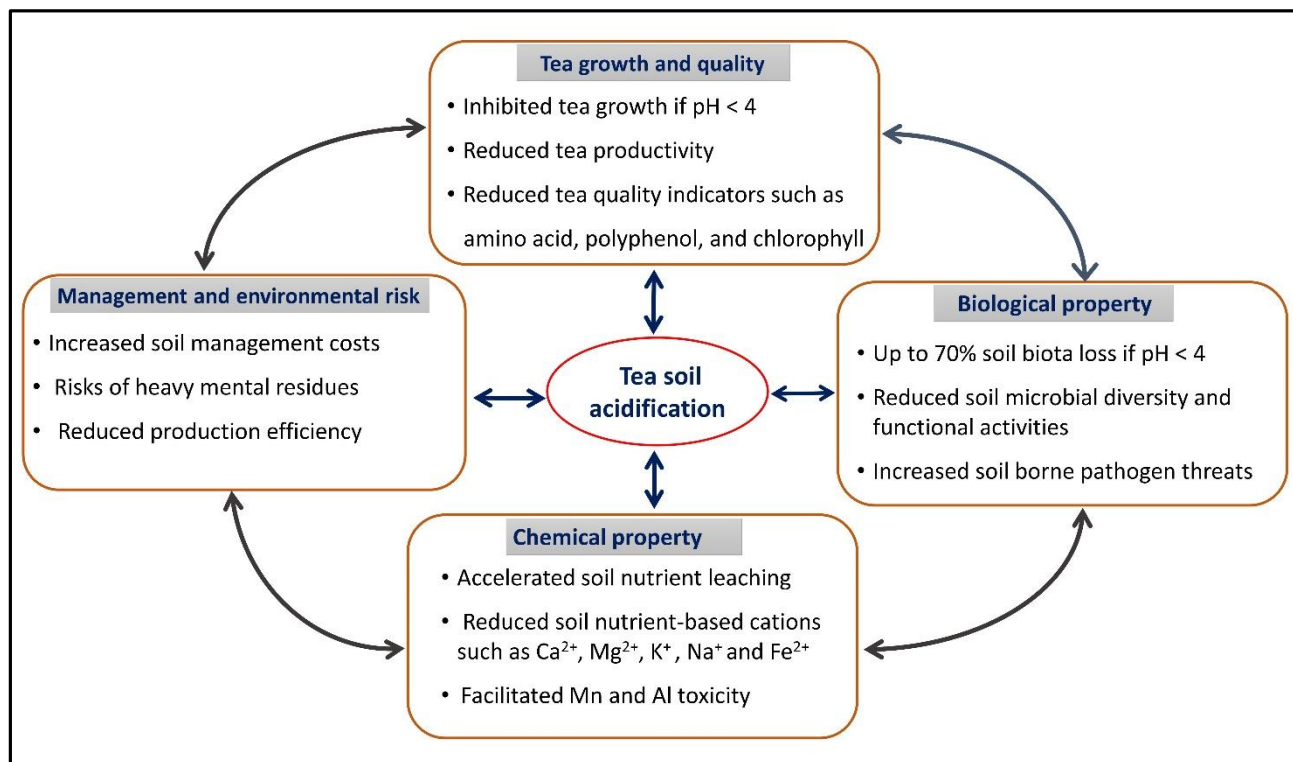


Fig. 4 A summary of the main consequences of soil acidification caused by tea cultivation in the aspects of soil chemical and biological properties, tea growth and quality, soil management cost and the environmental risks.

Tea productivity and quality

Although tea plants prefer acidic soil for optimal growth and productivity, severe soil acidity negatively effects plant performance and quality (Fig. 4). When the soil pH is lower than 4.0, tea plant growth is inhibited, affecting both the quality and quantity of tea production (Li et al. 2016; Yan et al. 2020). Heavy N addition also significantly decreases the Polyphenol/free amino acid ratio and affects other tea quality indicators by altering the relative content of chemical constituents (Qiao et al. 2018). High concentrations of Mn^{2+} negatively affects tea quality indicators such as amino acid composition and reduces the chlorophyll

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3 and carotenoid content of tea leaves (Venkatesan et al. 2010). Free Al³⁺ at a concentration of more than 1 mM
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5 retards tea growth, while the concentration at 10 mM leads to defoliation of tea plants (Fung et al. 2008).
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10 *Management cost and environmental risks*

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14 Despite the limited study on the management and other associated costs of soil acidification in the tea
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16 farming industry, research conducted on negative impacts of soil acidification on other agricultural sectors
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18 has highlighted the issues this causes. For instance, the annual loss of agricultural production due to soil
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20 acidification in New South Wales, Australia was around \$387 million (Li 2020). Likewise, soil acidification
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22 resulted in an estimated economic value decrease of \$US214,000 per hectare (ha) in the forest industry in
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24 America (Caputo et al. 2016). Lime has been considered as the most effective ameliorant to control acidic
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26 soils, but it is still too costly for farmers in many countries, due mainly to its transportation costs (Cai et al.
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28 2015; Tang et al. 2013). In tea plantation soils, acidification also occurs at the subsoil layers (100-120cm),
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30 thus deep incorporation of lime and other alternatives could be very expensive or even impractical due to the
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32 costs of suitable machinery (Li et al. 2016; Tang et al. 2013). Tea soil acidification can also promote the
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34 accumulation of chemical elements such as arsenic (As), mercury (Hg), lead (Pb), chromium (Cr), cadmium
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36 (Cd) and nickel (Ni) in the soil and tea leaves, increasing the human health and environmental risks of heavy
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38 metals (Bayraklı and Dengiz 2020; Zhang et al. 2020). It has been reported that more than 75% of soil Cd,
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40 Hg, Pb and Zn under acidic tea plantations exceeded uncultivated background concentrations, possibly due to
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42 the acidic environment promoted weathering pedogenic process releasing heavy metals (Tao et al. 2021).
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50 **3. Possible agricultural wastes for correcting tea soil acidification and enhancing soil health**

51 *3.1. Agricultural wastes for soil acidification and soil health*

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54 Agricultural wastes such as organic manures have been considered as a significant resource for agriculture for
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56 over hundred years (Rayne and Aula 2020), and since the downsides of agrochemical intensification on
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58 human beings and the ecosystem have become the global issue, the potential role of these alternate materials
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60 is being scrutinised increasingly closely (Chen et al. 2018; De Corato 2020). Most of agricultural wastes are
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widely available, cheap, biodegradable and rich in organic matter and nutrient and thus can be recycled as fertilizers or soil amendments (Kaur 2020; Onwosi et al. 2017; Saliu and Oladoja 2021). The nutrient compositions of agricultural wastes and products derived from these resources varies greatly and depend on multiple factors, such as their original sources, animal diets, waste storage and management, as well as production procedures (Amoah-Antwi et al. 2020; Dai et al. 2017; Rayne and Aula 2020). Common agricultural by-product and their components applied to agricultural soils as fertilizers and amendments are illustrated in Fig. 5.

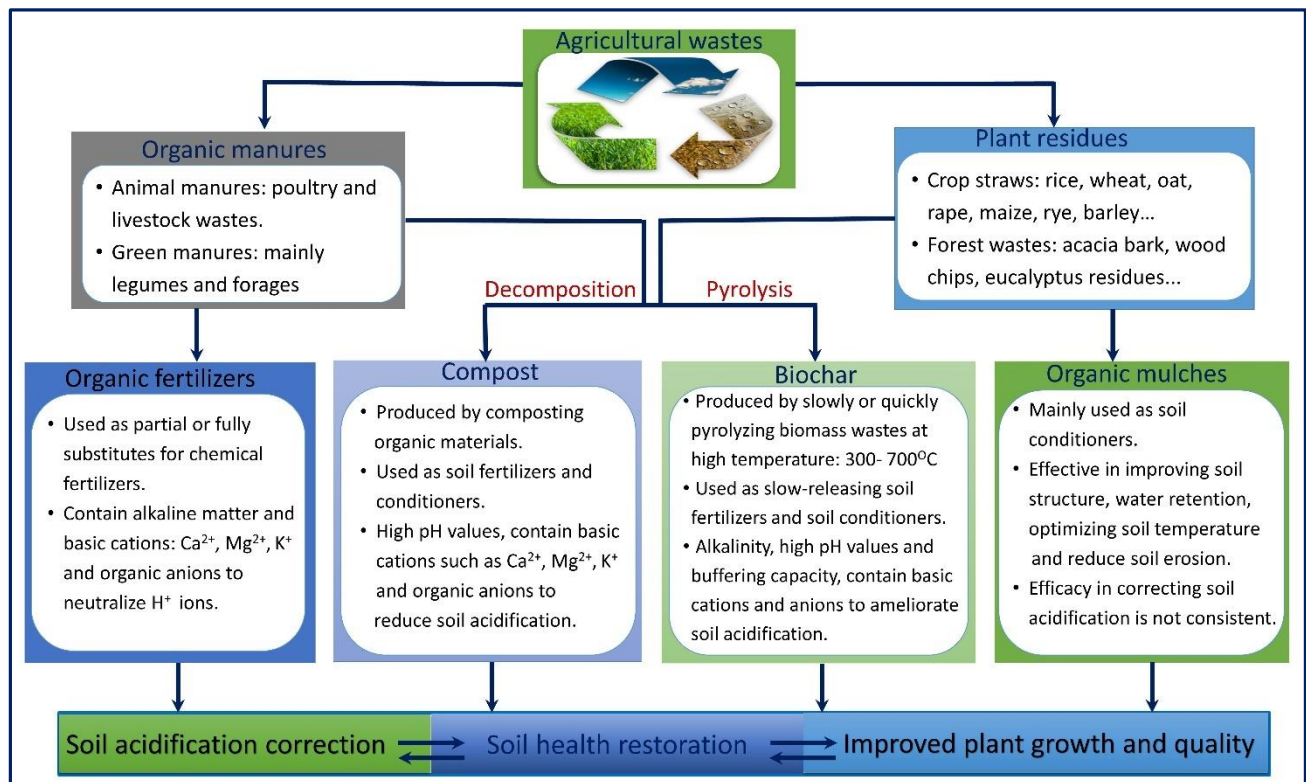


Fig. 5 A simplified illustration demonstrated the common types of agricultural wastes and products using these wastes as main feedstocks, how they could be produced and used to mitigate soil acidification and improve soil health, crop growth and quality.

There are various types of agricultural organic wastes applied to croplands, but they can be divided into two different groups based on their origins and common uses (Fig. 5). Organic manures include animal wastes from livestock and poultry industries, and green manures are mainly leguminous and forage crops (Maitra et al. 2018; Rayne and Aula 2020). Globally, animal waste has been predominantly attributed to manures from

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2
3 livestock and in 2018, contributed around 35 million tons of N applied to croplands globally, compared to
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5 more than 13 million tons from poultry (FAO 2021). Organic manures can be applied to soils or used as main
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7 materials for compost production, the natural biological processes of decomposing organic wastes involving
8
9 numerous microbial species (Azim et al. 2018; Bhatt et al. 2019; Sánchez et al. 2015). Compared to manures
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11 and compost, plant straws and other organic biomass such as wood chips and tree pruning residues are not
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13 often applied directly to soils as fertilizers, but can also be incorporated as mulches, mainly for enhancing soil
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15 structure and water retention (Amoah-Antwi et al. 2020; Siedt et al. 2020). Alternatively, using agricultural
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17 by-products to produce biochar has been also an increasingly accepted way of recycling wastes. Biochar
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19 could be best described as a “soil conditioner”, a rich carbon product produced by thermochemical
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21 decomposition of organic matter under low oxygen environment and high temperature, normally from 300-
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23 700°C (Peng et al. 2018; Verheijen et al. 2010). Feedstocks for biochar production consist of various biomass
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25 types, including municipal wastes and agro-industrial residues, and the feedstock types are important factors
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27 affecting biochar properties (Amoah-Antwi et al. 2020; Gunarathne et al. 2019; Guo et al. 2020). Details of
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29 elemental properties of some common agricultural wastes, compost and biochar are summarised in Table 1.
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36 The various agricultural wastes have differing effects on alleviating soil acidification. Organic compost and
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38 biochar produced from organic manures and plant residues are naturally alkaline and have a higher pH value
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40 compared to that in the acid soils, so the addition of these organic amendments can increase soil pH to some
41
42 extent (Cornelissen et al. 2018; Shi et al. 2019). Additionally, organic manure and its components naturally
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44 contain some basic cations such as Mg^{2+} , Ca^{2+} , Na^{2+} and K^{+} , which can form carbonates or oxides and then
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46 subsequently react with the H^{+} in the acidic soils and lead to the acid neutralization (Dai et al. 2017; Rayne
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48 and Aula 2020). In contrast, some studies showed that the decomposition of some mulching materials such as
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50 woody chips, crop straw and pine bark could generate organic and carbonic acids, which facilitate soil acidity
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52 (Arafat et al. 2020; Zhao et al. 2018). Nevertheless, numerous studies have reported the neutral to positive
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54 effects of mulching practices on soil acidification (Cu and Thu 2014b; Ni et al. 2016; Sadek et al. 2019; Vijay
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7 With regards to soil physical aspects, plant residues, organic fertilizers and biochar applications can benefit
8 soil hydrothermal environment, soil structure and water holding capacity (Kader et al. 2017; Siedt et al. 2020;
9 Wang et al. 2020). In terms of soil chemical properties, adding organic fertilizers and biochar significantly
10 improve soil organic matter, soil macronutrients and micronutrients, reduce Al and Mn toxicity risks and
11 nutrient leaching (Ding et al. 2020; Gong et al. 2020; Patra et al. 2021; Siedt et al. 2020; Zhongqi et al. 2016).
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14 Recently, a number of studies have reported the positive impacts of agricultural residue practices on soil
15 organism abundance and functional diversity, such as the applications of organic mulches (Xiang et al. 2021;
16 Zhang et al. 2020b), biochar and compost (Amoah-Antwi et al. 2020; Liu et al. 2021) and organic manures
17 (Rayne and Aula 2020; Su et al. 2021). Despite the preference in using synthetic fertilizers, agricultural
18 wastes and products derived from these resources are being used intensively as soil amendments and
19 fertilizers, to partially or fully substitute for chemical fertilizers (Amoah-Antwi et al. 2020; Lin et al. 2019;
20 Shaji et al. 2021). However, since the nutrient compositions and efficacy of agricultural wastes and its
21 products varied significantly (Table 1), they cannot be applied in a homogenous manner (Dai et al. 2017;
22 Rayne and Aula 2020). Therefore, having a good understanding of characters of agricultural wastes and its
23 components would be important to increase their application efficiency and reduce the pollutant risks to
24 ecosystems (Amoah-Antwi et al. 2020; Ayilara et al. 2020; Cai et al. 2021).
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Table 1 Nutrient composition of some main types of agricultural wastes and its based products used as soil amendments in tea cultivation and croplands.

Type of waste	Nutrient composition									Reference
	N	P	K	Na	Fe	Cu	Mn	Zn	Total C	
1. Animal manure										
Horse	20.7	7.6	41.4	7.58	729	22	110	167	43.3	Moreno-Caselles et al. (2002); Chong et al. (2019)
Cow	18.6	7.89	17.6	5.38	3527	20	111	79	43.88	Mendonça Costa et al. (2015); Moreno-Caselles et al. (2002)
Calf	17.5	9.6	35.1	24.6	2839	40	225	233	-	Moreno-Caselles et al. (2002)
Pig	21.7	14.4	8.9	2.34	1559	170	328	427	-	Moreno-Caselles et al. (2002)
Sheep	18.7	5.67	34.3	6.94	3786	21	137	159	41.84	Mendonça Costa et al. (2015); Moreno-Caselles et al. (2002)
Goat	22.2	8.1	59.2	16.9	1729	31	170	202	-	Moreno-Caselles et al. (2002)
Rabbit	17.9	9.2	18.2	5.07	2623	61	225	453	-	Moreno-Caselles et al. (2002)
Chicken	31.4	13.2	24.7	4.85	154	40	237	304	34	Moreno-Caselles et al. (2002); Ravindran and Mnkeni (2016)
Turkey	39.7	10.9	24.5	3.97	172	45	327	336	39.7	Moreno-Caselles et al. (2002) ; Calbrix et al. (2007)
Ostrich	16.5	7.7	10.7	4.64	1303	56	257	200	-	Moreno-Caselles et al. (2002)
Earthworm	17.3	11.9	7.8	2.34	6503	78	335	348	-	Moreno-Caselles et al. (2002)
<i>Note:</i> N, P, K (g/kg, dry weight); Na, Fe, Cu, Mn, Zn (mg/kg, dry matter); Total C (% , dry weight).										
2. Plant residues	N	P	K	C	Ca	Mg	pH	C:N ratio	Ash content	
Wheat straw	55	9	42	43.9	22.61	2.88	5.1	124.4	23.2	Jalali and Ranjbar (2009); Torma et al. (2018); Wang et al. (2009)
Potatoes	59	6	61	-	-	-	6.1	22.0	20.4	
Maize straw	39	3	19	42.14	6.40	4.60	-	-	48.8	
Oat straw	55	8	58	36.35	-	-	-	54.25		Torma et al. (2018); Zhao et al. (2018)
Rye	45	8	24	-	-	-	-	-	-	Torma et al. (2018)
Barley	43	7	40	-	-	-	-	-	7.14	Torma et al. (2018); Plazonić et al. (2016)
Triticale	54	8	28	-	-	-	-	-	5.27	
Pea straw	112	14	74	43.56	17.32	6.51	-	-	61.6	Torma et al. (2018); Wang et al. (2009)

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Soybean straw	132	14	72	44.06	18.24	17.86		44.06	72.0	
Sugar beet	20	2	13	-	-	-	-	-	-	Torma et al. (2018)
Mustard	91	21	127	-	-	-	-	-	-	
Sunflower	108	15	218	-			5.3	81.4	10.4	Jalali and Ranjbar (2009); Torma et al. (2018)
Rape	107	15	218	-	-	-	5.1	65.5	5.4	
Rice straw	0.5- 0.8 ^a	0.07- 0.12 ^a	1.16- 1.66 ^a	41.25	7.03	3.96	-	-	33.6	Ayinla et al. (2016); Chivenge et al. (2020)

Note: N content, P, K (kg/ ha); OM, C (%); Ca, Mg (cmol (+)/kg); Ash content: (%; dry weight); ^a (%).

Tea and wood residues	N	P	K	Dry matter	C	Ca	Mg	C:N ratio	Ash content	
Tea pruned foliage	252	30	72	7.2	2.9	-	-	11	-	Kamau (2008)
Tea pruned twigs	85	10	21	3.6	1.4	-	-	17	-	
Primary wood	101	28	2	4.2	1.8	-	-	42	-	
Secondary wood	44	13	13	4.2	1.8	-	-	40	-	
Acacia bark	133.4	2.6	8.4	8.9	-	76.5	1.2	-	2.1	Taflick et al. (2015); Van Bich et al. (2018)
Eucalyptus biomass	307.5	28.8	249.3	-	-	-	455.7	131.7	15.4	Reina et al. (2016); Resquin et al. (2020)

Note: N, P, K, Ca, Mg (kg/ ha, dry weight); C (t/ ha).

3. Biochar	N	P	K	Ca	Mg	Total C	pH	C:N ratio	Ash content	
Rice straw biochar at 400 °C	19.8	2.0	24	8.8	5.7	56	8.7	-	39	Naeem et al. (2017)
Wheat straw biochar at 400 °C	19.4	3.8	33	10.3	9.6	62	7.8	-	36	
Pine woodchip biochar at 500 °C	0.7	<0.001	2.1	10.1	2.7	244.5 ^c	8.7	366	-	Brantley et al. (2015)
Rice biochar at 500 °C	0.92 ^a	3.23 ^a	2.48 ^a	875.2	578.9	46.4	11.0	-	34.6	Yan et al. (2021)
Bamboo biochar at 750-800 °C	0.58 ^a	1.85 ^a	1.01 ^a	560.3	320.6	77.3	11.3	-	5.8	

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Peanut biochar at 300 °C	2.6 ^a	-	22.0 ^b	47.4 ^b	45.6 ^b	55.1	9.2	21.5	228.4 ^b	Wang et al. (2014a)
Vermicompost	8.7	<0.1	1.3	26.3	-	181 ^c	8.09	20.9	8.09	Adhikary (2012)

Note: Total N, P, K Ca, Mg, (g/kg); Total C (%); Ash content (%); ^a (%), ^b (cmol (+)/kg), ^c (g/kg).

4. Compost	N	P	K	Ca	OC	pH	C:N ratio	OM	Moisture	
Chicken manure compost	13.19	12.5	20.00	-	325.3	7.92	26.06	72.56	29.9	Li et al. (2021)
Pig manure compost	29.82	15.13	8.16	-	-	8.37	-	73.01	78.89	Li et al. (2012)
Buffalo manure compost	1.3	-	-	-	-	7.3	14	-	-	Doan et al. (2014); Ngo et al. (2011)
Cow manure compost	21.3	10,4	21.7	23.7	-	9.6	-	56.96	29.1	Gil et al. (2008)

Note: N, P, K, Ca (g/kg); OC, OM and moisture (%).

3.2. *Organic fertilizer and organic tea management practices*

Applying animal manure to tea plantation soils could be an effective solution not only for ameliorating soil acidification, improve soil health of tea plantations but also as a waste management tool. Manures from various animals such as sheep, pig, cow and chicken used as organic fertilizers or compost for tea gardens significantly increased pH of acid soils, compared to their chemical nutrient counterparts (Cai et al. 2015; Gu et al. 2019; Ji et al. 2018; Lin et al. 2019; Qiu et al. 2014). For example, Gu et al. (2019) indicated that long-term applications of animal manure resulted in a significant increase of soil pH (5.36), compared to that in non- fertilizer (4.71) and chemical fertilizer practices (4.31). Likewise, application of pig manure over 18 years increased soil pH by 1.1 units (Cai et al. 2015). Additionally, the replacement of chemical fertilizer by organic fertilizer in organic and agroecological tea cultivation has also had positive impacts on soil pH and other soil health indicators (Li et al. 2014; Viet San et al. 2021; Yan et al. 2020). Analyzing more than 2000 tea soil samples collected from conventional and organic tea plantations, Yan et al. (2020) concluded that conventional tea cultivation which employ heavy application of synthetic fertilizers caused severe soil acidification, while organic tea management approach did not result in significant soil acidification. Similarly, our recent study shown that agroecological tea management practices with chicken and buffalo manures as main nutrient supplies significantly improved soil pH compared to conventional tea cultivation which employs intensive chemical NPK (unpublished data). As outlined above, the mitigation of acidification of tea plantation soils by organic substance addition could be by alkaline matter and basic cations from added organic fertilizers, which can neutralize the soil acidity (Ji et al. 2018). Moreover, other chemical processes involving manure supplementation such as organic anion decarboxylation and organic N ammonification may play a part in reducing soil acidity (Xiao et al. 2013; Xu et al. 2006). Organic fertilizer can also support soil buffering action, thus reducing soil acidification (Chen et al. 2009). More examples of positive effects of organic manure and compost usage on soil acidification are indicated in Fig. 6 and Table 2.

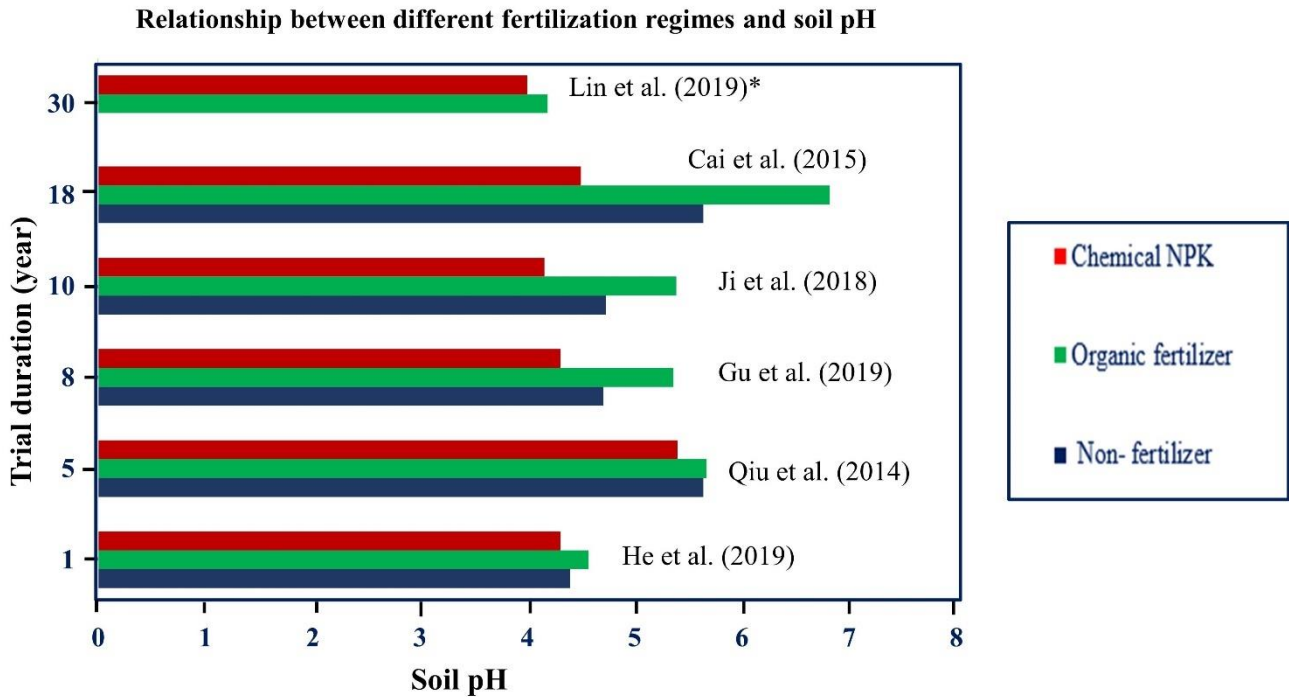


Fig. 6 Effects of different fertilizer type applications on soil pH under tea cultivation. Organic fertilization consistently resulted in greater soil pH in comparison with chemical fertilizer and non-fertilizer practices. Heavy uses of synthetic fertilizers also led to highest reduction of soil pH, compared to other fertilization approaches. Adapted from Lin et al. (2019); Cai et al. (2015); Ji et al. (2018); Gu et al. (2019); Qiu et al. (2014); He et al. (2019). (*) the data for non-fertilizer management practice not available.

Apart from ameliorating soil acidification, recycling organic amendments as the partial or full substitutes for chemical fertilizers can bring about a range of benefits for other aspects of tea plantation soil health and the environment. Organic fertilizer applications consistently improved soil OM, soil OC, soil exchangeable cations such as Ca^{2+} , Mg^{2+} , Na^+ and K^+ , and nutrient availability, while reducing risks of Al toxicity, heavy metal accumulation, greenhouse gas emissions and nutrient run off such as N and P (Table 2) (Cai et al. 2015; He et al. 2019; Ji et al. 2018; Lin et al. 2019; Qiu et al. 2014). Sustainable effects of adopting organic soil amendments in tea plantation soils on biological soil health has been also clearly indicated. Organic materials such as sheep, cow, chicken manures or compost significantly improved soil fauna communities, soil microbial diversity and functional structures (Gui et al. 2021; Li et al. 2014; Lin et al. 2019; Zhang et al. 2020a). Organic fertilizers are

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3 naturally rich in nutrients contain more organic matter compared to chemical compound, thus the replacement of
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5 organic amendments provide more organic matter in the soils (Wu et al. 2020; Xie et al. 2019). Richer soil
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7 organic contents will attract soil fauna and facilitate the activities of soil microbial communities in converting
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9 soil nutrients, which ultimately increase soil nutrient of tea plantation soils (Fan et al. 2017; Xie et al. 2019; Xie
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11 et al. 2021). These positive changes in turn, will result in increasing soil organism diversity and community
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13 structure (Gu et al. 2019; Wu et al. 2020).
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20 There do exist some concerns for recycling animal manures and organic compost which need further
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22 consideration. Firstly, organic fertilizer such as rapeseed cake had inconsistent effect on soil pH (Xie et al. 2019;
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24 Xie et al. 2021). This discrepancy may result from the dissimilarity of chemical composition of the product and
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26 other conditions such as soil type, application rate and management practices (Gu et al. 2019; Wu et al. 2020).
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28 Secondly, it has been reported that organic manure cannot ameliorate deep-soil acidification in tea plantations
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30 (Li et al. 2016). In this case, biochar or a combined utilization of manure and biochar may be an effective
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32 solution to not only mitigate soil acidification but also enhance soil health and tea productivity (Dai et al. 2017;
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34 He et al. 2019). Thirdly, long- term application of animal manure and compost to manage acidic tea soils and
35
36 restore soil health could led to the risks of heavy metal accumulation and manure- borne pathogen contamination
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38 (Cai et al. 2021; Li et al. 2020). For heavy metal contamination, Ji et al. (2018) indicated that 10 - year
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40 application of pig manure did not result in increase of most heavy metals, and Lin et al. (2019) found that sheep
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42 manure and rape cake application reduced levels of Cd, Pb and As in soils as well as in tea leaves. To date
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44 however, the relationship between animal manure, compost and pathogenic diseases of tea plants has been
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46 poorly understood. Thus, an integrated approach including appropriate application rates, reducing chemical
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48 inputs and concentrations of heavy metals in animal feed could be all necessary to minimize the environmental
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50 risks from using these organic materials as soil amendments and increase their efficacy (Cai et al. 2021; Ji et al.
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52 2018).
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3.3. Biochar amendment

Among the ameliorants of soil acidification, biochars could be one of the most effective options as it can also improve soil quality, plant productivity and contribute to a reduction in greenhouse gas emissions (Akhil et al. 2021; Siedt et al. 2020; Zhang et al. 2018). In tea farming, biochars produced from plant residue such as rice, wheat straw and bamboo residues have been commonly incorporated as soil amendment (Chen et al. 2021; Ji et al. 2020b; Wang et al. 2018). Depending on biochar types and application rates, soil condition, tea management practices and the application duration, the liming effect of biochars varied significantly, (Wang et al. 2014a; Yan et al. 2021). As demonstrating in Fig. 7, applying biochars at rates of from 1% to 5% of soil dry weight can significantly increase soil pH from 0.2 to more than 1 units within a few months (Ji et al. 2020a; Oo et al. 2018; Wang et al. 2018; Zheng et al. 2019). Studies conducted in tea plantations also demonstrated the positive outcomes of biochar utilization for correcting soil acidification caused by tea cultivation (Table 2) (He et al. 2019; Ji et al. 2020b; Yang et al. 2021).

Soil pH change

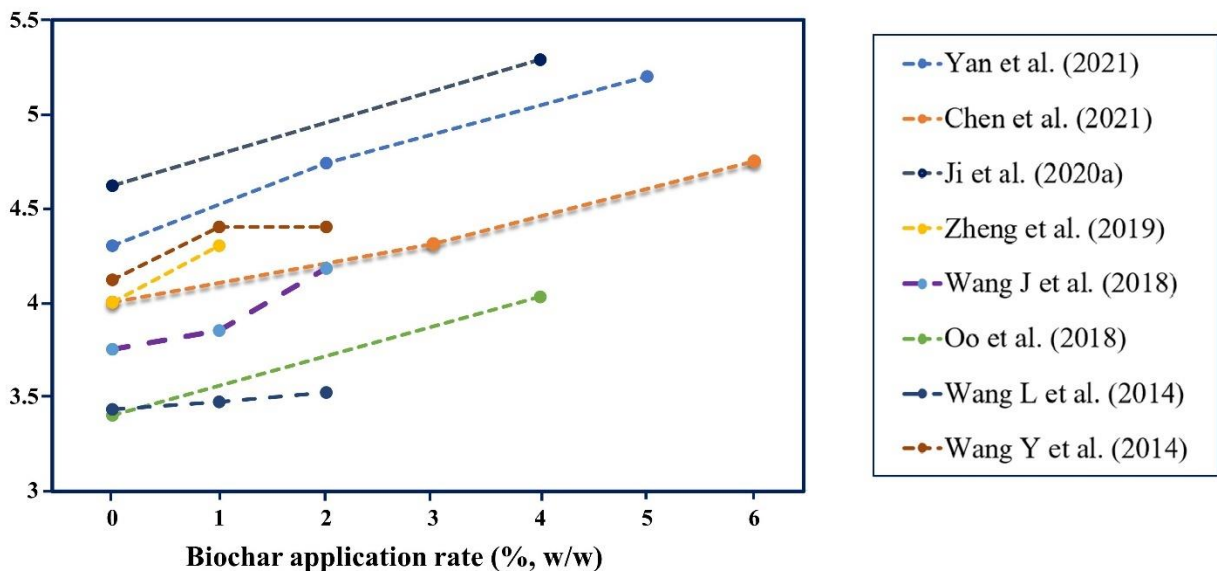


Fig. 7 Effects of biochar application rate on pH of tea plantation soils. Data collated from recent publications: Chen et al. (2021); Ji et al. (2020a); Oo et al. (2018); Wang J et al. (2018); Wang L et al. (2014); Wang Y et al. (2014) and Zheng et al. (2019).

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3 Biochar ameliorates soil acidification by its natural alkalinity, high pH value and pH buffering capacity. Biochar
4 generally has an alkaline pH value, thus soil amended with this product can become less acidic (Table 1). For
5 instance, a meta- analysis by Dai et al. (2017) indicated that biochar applications significantly increased soil pH
6 by up to 2 units, and in most cases, the pH of biochars is greater than 7.0, which is at least 1.5 units higher than
7 the pH in acid soils. Moreover, mineral constituents of biochar including basic cations such as Ca, Mg, K, Na
8 and alkaline oxides that originated from feedstocks can mitigate soil exchangeable acidity (mainly H⁺ and Al³⁺)
9 in the soil and ultimately increase soil pH (Dai et al. 2017; Patra et al. 2021; Yuan et al. 2011). In addition, soil
10 pH buffering capacity is an important factor contributing to biochar amelioration of soil. Shi et al. (2019)
11 illustrated that rice straw and peanut straw biochar application increased pH buffering capacity by 22% and 32%
12 respectively. It has been verified that the increases in CEC of the soil by biochar incorporation, driven by
13 protonation- deprotonation processes, was the main mechanism of increasing soil pH buffering capacity (Shi et
14 al. 2017; Xu et al. 2012). Biochar application also suppressed soil nitrification by limiting the availability of NH₃
15 or NH₄⁺ for oxidation because of the surface adsorption or increased emissions of NH₃ due to enhanced soil pH
16 (Wang et al. 2018; Yang et al. 2015). This in turn generally reduces the proton (H⁺) released into soil and
17 ultimately increase soil pH (Shi et al. 2019).
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41 Biochar addition also enhanced soil quality indicators, tea growth and productivity, as well as reduced the
42 environmental risks from pollution by heavy metals and greenhouse gases such as CO₂, N₂O and NO (Chen et al.
43 2021; Ji et al. 2020a; Yan et al. 2021). Consistently, biochar incorporation in soil improved soil OC, soil nutrient
44 availability including Ca, Na, Mg, P and K contents, soil total N and C (Yan et al. 2018; Wang 2014; Zheng et
45 al. 2019). While the impact of biochar on soil fauna has been poorly investigated, this carbon-rich material has
46 significant effects on enhancing soil microbial diversity and community structure (Table 2) (Ji et al. 2020a; Yang
47 et al. 2021; Zheng et al. 2019). Biochar itself is a source of nutrients, including microminerals, trace elements,
48 ash and so on, so its application also supplies essential agronomic benefits to farmers (Rawat et al. 2019). More
49 importantly, biochar can absorb fertilizers and slowly release these into the soil, which helps to not only retain
50 the nutrient availability in the soil but also reduce fertilizer leaching and drainage, which then contribute to
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3 environmental pollution (Rawat et al. 2019). Since soil pH and nutrient status has a close correlation with soil
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5 microorganism, the changes in soil chemical and physical properties as a result of biochar application could be
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7 the key driven factor for the alteration of soil biological properties (Cheng et al. 2019; Yang et al. 2021).
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14 Several downsides of biochar incorporation need to be considered to improve its effectiveness and reduce the
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16 detrimental effects on the environment. Biochar has been considered as the most expensive soil management
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18 solution, particularly for large-scale use in agriculture (Siedt et al. 2020). Since the application rate of biochar
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20 normally ranges from 10 to 150 tons/ha and controlling strongly acid soils may require large quantity of biochar,
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22 which leads to an increased costs for energy inputs, feedstocks, transportation and incorporation (Dai et al.
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24 2017). Furthermore, most studies on biochar application for managing soil acidification in tea farming to date
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26 have been conducted in controlled conditions in China, suggesting that further research either in long-term field
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28 conditions or in other tea producing areas would be needed. Overall, biochars indicate a great potential in
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30 ameliorating soil acidification and improving tea plantation soil health, however, more comprehensive and
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32 reliable evidence should be provided to validate these advantages.
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40 **3.4. Plant residues as organic mulching practices**

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42 Organic mulching practices employing plant residues and other agricultural wastes have received limited
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44 attention to date. Some studies conducted on tea fields indicated that mulching materials such as Fern
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46 (*Gleichenia linearis*) and tea pruning materials can alleviate soil acidity (Cu and Thu 2014a; b). Other materials
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48 such as crop straws and legume residues also had positive effects on increasing pH of tea plantation soils, either
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50 in field or laboratory trial conditions (Table 2) (Wang et al. 2009; Xianchen et al. 2020). In contrast, there have
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52 been a number of investigations revealing the negative impacts of organic mulching on soil pH from other
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54 cropping systems. Otero-Jiménez et al. (2021) found that rice straw mulch and rice straw burning significantly
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56 reduced soil pH by 0.55 and 0.19 units respectively, and the application of wheat straw mulching reduced soil
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58 pH by 0.11 units (Mehmood et al. 2014). Finally, some studies have demonstrated that plant residues have no
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3 significant effects on soil pH (Iqbal et al. 2020; Ni et al. 2016). Positive effects of crop residues in increasing soil
4 pH could be mainly due to the decarboxylation of organic anions, which can neutralize soil exchangeable H^+ and
5 Al^{3+} , and also reduce the toxicity of Al species to plant roots (Dai et al. 2017). Declines in soil pH following
6 application plant residue mulches could be attributed to the release of H^+ from nitrification of NH_4^+ , which is
7 produced during the mineralization of organic N in the residues (Dai et al. 2017). Decomposition of crop
8 residues may also produce some organic and carbonic acids, potentially causing soil acidity (Arafat et al. 2020).
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10 The potential of crop residue mulching in enhancing other soil health indicators have been widely recognized.
11 Plant residues improve soil moisture content, soil structure and regulate soil temperature, support soil microbial
12 activities and improve soil nutrient availability, as well as suppress weeds and reduce soil erosion, all of which
13 contribute to enhance soil health and crop productivity (Chatterjee et al. 2017; Kader et al. 2017; Ngosong et al.
14 2019). These benefits have also been demonstrated in tea cultivation systems. Covering the surface of tea
15 plantation soils with rice straw and tea pruning residues significantly reduced soil temperature variation, soil
16 compactness and soil bulk density, while increasing soil water retention and soil moisture (Cu and Thu 2014b;
17 Xianchen et al. 2020). Organic mulches can also enhance soil nutrient availability (Ca^{2+} and Mg^{2+} , available N,
18 P, K) soil OM content but reduce soil Al^+ concentration (Cu and Thu 2014a; Wang et al. 2009; Xianchen et al.
19 2020). Enrichment of soil microbial diversity and community structure as a result of mulching material addition
20 have been reported in these studies (Cu and Thu 2014a; b) (Table 2). Organic mulch cover creates favorable
21 moisture and thermo regimes in soils by controlling surface evaporation rates and alter soil temperatures, by
22 reducing temperature in the summer and raising it in the winter (Kader et al. 2017). Under appropriate soil
23 microclimatic conditions, plant litter can decompose and add nutrients to soils. Plant residues and other organic
24 mulch materials generally contain higher level of nutrients compared with inorganic mulch materials, but the
25 influence of organic mulching application on soil nutrients has been also determined by other factors such as soil
26 characteristics, climatic conditions (Iqbal et al. 2020; Kader et al. 2017). In addition, soil physicochemical
27 conditions including soil moisture, soil temperature and soil nutrients play a crucial part in governing soil
28 organisms (Kader et al. 2017; Onwuka and Mang 2018; Tan et al. 2018). For example, Brockett et al. (2012)
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concluded that soil moisture is the major factor affecting the community structure of soil microbes as well as enzyme activities. Examples of plant residue mulching and the summary of beneficial impacts of organic mulching, organic fertilizer and biochar applications in tea plantation soils are shown in Fig.8.

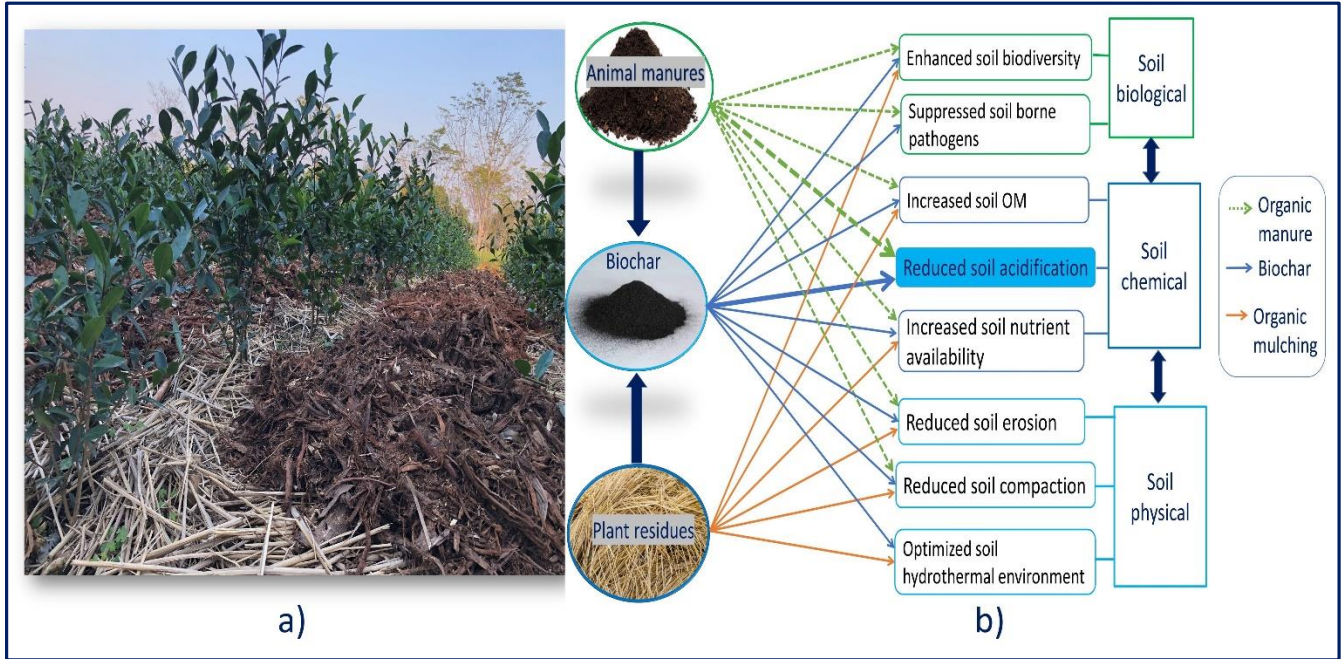


Fig. 8 Plant residues (rice straw, Acacia bark and woodchips) and organic manure (poultry manures) applications in tea plantations (a) and summaries of the beneficial effects of some soil amendments derived from agricultural wastes on soil properties of tea plantations (b). Photo was taken in Thai Nguyen province, Northern Vietnam by the author.

However, some of mulching materials such as crop straws generally decompose quickly, thus need to be frequently incorporated for long-term use. This may require extra labour and investments, preventing farmers from adopting them in the long run (Amoah-Antwi et al. 2020; Dai et al. 2017). Extensive use of plant residues such as tea pruned litters to mulch tea soils could also lead to a decrease of soil pH and the accumulation of active allelochemicals, which can cause soil sickness and tea growth deterioration (Arafat et al. 2020). Too much organic mulch could also result in other issues such as excess moisture and nitrogen, pests and anaerobic conditions, damaging the plant root and negatively affecting its growth and productivity (Iqbal et al. 2020; Kader

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3 et al. 2017). Overall, organic mulching employing plant residues is an effective soil management tool to improve
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5 soil physicochemical properties, but its role in controlling tea soil acidity needs further investigations.
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10 11 **3.5. Intercropping and agroforestry**

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14 Tea plants intercropped with loquat, waxberry and citrus significantly improves soil pH, organic matter, N, P and
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16 K availability, tea quality indicators, and reduces soil heavy metal concentrations compared with monoculture
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18 tea gardens, regardless of sampling seasons (Wen et al. 2019). Similarly, Xianchen et al. (2020) found that inter-
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20 planting of *Vulpia myuros* at the density of 22.5kg/seeds/ha in tea plantations significantly increased soil
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22 nutrients (OM, available N, P, K), soil water holding capacity while reducing soil temperature fluctuations and
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24 soil compactness at all observed soil depths (0-10 and 10-20cm). In terms of soil organism, intercropping
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26 adoption in tea cultivation enriched soil enzyme activity and regulated tea pests (Xianchen et al. 2020; Zhang et
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28 al. 2017) (Table 2). In addition, tea – Ginkgo tree (*Ginkgo biloba* L.) agroforestry significantly increased soil pH
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30 (5.86 vs 5.21), soil organic carbon (17.92 vs 16.38 and total N (1.91 vs 1.79) compared with single tea
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32 plantations (Tian et al. 2013). The increase of soil pH in the Ginkgo – tea agroforestry is likely due to the
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34 alkaline matter formed during the decomposition of Ginkgo tree residues which neutralizes soil acidity (Tian et
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36 al. 2013). Intercropping and agroforestry might increase overall ecosystem productivity and nutrient retention by
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38 increasing species diversity, increase soil organic matter by plant residues, attribute to the decomposition of fine
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40 roots in the deep mineral layers and surface leaves of trees (Brooker et al. 2015; Cong et al. 2015; Dollinger and
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42 Jose 2018). Among these impacts, organic matter enrichment could play a key role, containing basic cations and
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44 contributing to increasing the supply of important nutrients (Cardinael et al. 2020; Dollinger and Jose 2018).
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Table 2 Summaries of current studies of organic fertilizers, biochar, plant residues and other relevant options on mitigating soil acidification and improving soil health, tea plant growth, and reducing environment risks.

Material/ Practice	Soil type Location	Experiment type Application rate/time	Soil pH effect	Other positive and/or negative impacts on soil, tea plants and the environment	Reference
Sheep manure + rape cake	Red soil China	- Field experiment - Trial time: 30 years	- Organic fertilizers resulted in an increase by 0.2 units (4.2 vs 4.0) compared to chemical fertilizers.	- Significant increased soil bacterial abundance, total K, while decreased the contents of Cd, As and Pb in rhizosphere and tea leaves. - Reduced soil total N (0.23 g/kg); total P (1.24 g/kg).	Lin et al. (2019)
Pig manure	Red soil (Ferralic Cambisol) China	- Field experiment - Trial time: 18 years	- Increased by 1.1 units after 18 years of pig manure application.	- Pig manure application reduced exchangeable Al ³⁺ and significantly increased soil exchangeable Ca ²⁺ , Mg ²⁺ , Na ⁺ and K ⁺ .	Cai et al. (2015)
Cow manure + Pig manure	Haplic Acrisol Chia	- Field experiment - Manure: 1000-2.000kg/ha - Trial time: 1 year	- Soil pH value with chicken and pig manure practices were 5.36 and 5.09 respectively, compared to 4.71 of non- fertilization and 4.31 of mineral compound (NPK) application.	- Organic fertilizer application increased soil microbial diversity by 8.59–33.14% and resulted in an improvement of potential ecosystem function compared with synthesized fertilizer. - Increased total P but decreased total N.	Gu et al. (2019)
Pig manure	Red soil China	- Field experiment - Substitution of 25%, 50%, 75% and 100% N by organic manure - Trial time: 10 years	- 0.66 unit increased by application of 100% N substitute compared to the non- fertilizer plots - 1.23 units higher compared to the pH value of synthetic fertilizer use.	- Significantly increased soil OC, total N, NH ₄ ⁺ -N contents, available P and K. - Soil microbial biomass carbon (MBC) and microbial biomass nitrogen (MBN), soil bacterial diversity and community structure were improved significantly.	Ji et al. (2018)
Cattle manure	Planosols (Clay loam) China	- Field experiment - Manure + biochar, 20.000 kg/ha - Trial time: 2 years	- Organic fertilizer and biochar application resulted in greater soil pH compared to chemical fertilizer.	- Cattle manure and biochar applications reduced NO emission. - Adding cattle manure as a partial substitute for biochar reduced NO emission, and solely biochar application reduced N ₂ O emission by 14%.	Han et al. (2021)

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Chicken manure	China	- Field experiment - 11.400kg/ha - Trial time: 5 years	- Chicken manure application resulted in the highest soil pH (5.67), compared to non-fertilization (5.64) and mineral compound (NPK) (5.40).	- Significantly increased soil OM, total N and P; available N, P and K. - Organic manure uses promoted bacterial diversity, while that was reduced by chemical fertilizer application.	Qiu et al. (2014)
Rapeseed cake	Yellow brown China	- Field experiment - 1.904, 3.928, 6.207 kg/ha - Trial time: 1 year	- Rape seed cake (6.207 kg/ha) decreased soil pH by 0.19 units while with chemical fertilizer was 0.33 units.	- Soil OM, available P and K increased by 31.4%, 26.2%, and 21.7%, respectively - Increased restoration of NH ₄ - N, NO ₃ -N, total P and K contents in soil while reduced the substances in runoff water.	Xie et al. (2019)
Cow manure	Brown loamy China	-Field experiment - 20 tons/ha - Trial time: 6 months	- Data not provided	- Significantly increased the relative abundance of <i>Proteobacteria</i> and <i>Bacteroidetes</i> species and enhanced the diversity of bacterial communities.	Zhang et al. (2020a)
Rapeseed cake	Acid yellow brown China	- Field experiment - 1.708, 4.270, 6.831 and 8.539 kg/ha/year - 8 months	- Significantly increased soil pH by 2.19 – 4.29% compared to chemical compound treatments.	- Increased total OM and preserved soil C and N pools of the tea plantations - Reduced the nitrogen inputs (NH ₄ - N and NO ₃ -N) in the tea plantation runoff.	Xie et al. (2021)
Pig, chicken and cattle manure compost	Alfisol China	- Field trial - Trial time: 1 year	- Soil pH for pig, chicken and cattle manure compost uses were 4.56, 4.48 and 4.57 respectively, compared to 4.44 of non-fertilizer and 4.31 of chemical fertilizer practices.	- Increased soil OC, total N while reducing N ₂ O and NO emissions. - Organic fertilizer has no influence on tea yield, but that was increased by chicken manure and biochar combined application.	He et al. (2019)
Organic management (Chinese Pennisetum, rape cake and farmyard manure)	Ferralsol China	- Field trial - Chinese Pennisetum: 4.000kg/ha; rape cake: 3.000kg/ha; farmyard: 2.000kg/ha/year - Trial time: 6 years	- Organic tea management with organic fertilizer uses resulted in greater soil pH compared to conventional tea management; but lower compared to natural tea plantations.	- Increased soil OM, soil N and C/N ratio. - Enhanced species diversity, species richness and trophic diversity of nematodes in the soil.	Li et al. (2014)
Organic management (rape cake, compost and commercial organic fertilizers)	Ultisols China	- Field experiment - 4.500- 9.000 kg/ha/year - Trial time: around 10 years	- Soil pH has an inconsistent correlation with tea management methods.	- Increased soil microbial C by 164.4% and soil microbial N by 482.9% on average. - Total OC, N and available P increased significantly in organically managed tea plantation soils, but Ca and Mg availability decreased in comparison with conventional management.	Gui et al. (2021)

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Agroecological management (chicken and cow manure as main nutrient supplies)	Ferralsols Vietnam	- Field experiment - 6.000- 8.000 kg/ha/year - Trial time: 5-10 years	- Increased soil pH by 0.35 units on average, compared to conventional tea plantations.	- Significantly improved soil OM, colonization and intensity of arbuscular mycorrhizal fungi (AMF). - Reduced soil total N.	Unpublished data
Organic management (cow and pig manure, commercial organic fertilizer)	Red soil China	- Field experiment - Management duration: 14 years	- Soil pH increased by 0.91 units compared to conventional tea plantations, and 0.06 units compared with the tea plantations employed a combined application of organic and chemical fertilizers (non-polluted management practices).	- Increased total OC, available P, NH ₄ -N and NO ₃ -N but total P and N were lower than that in the non-polluted tea management). - Improved soil microbial diversity, increased the abundances of beneficial soil microbes, and altered the interaction network structure compared with conventional and pollution-free management practices.	Tan et al. (2019)
Organic management	Bangladesh	- Field research	- Soil pH of organically managed tea plantation was 5.1, compared to 4.2 of conventionally managed tea plantation.	- Increased total OM and nutrient availability (K, Ca, Mg, P, Zn and S) - Significantly increased tea yield and economic efficiency.	Sultana et al. (2014)
Organic management (Sheep manure)	Laterites China	- Field research - 6.000kg/ha/year, dry matter - Management time: 3 years	- Soil pH was significantly lower compared to that in longan orchard, both in the surface (5.05 vs 5.32) and 10-20cm depth (5.04 vs 5.24). - No significant difference compared to conventional tea management plantations.	- Organic tea management increased soil P availability, enhance soil microbial communities (bacteria, fungi, actinomycetes and AMF) compared to conventional tea management. - Conversion of longan to tea plantation significantly reduced soil fertility.	Wu et al. (2020)
Rice straw biochar	Oxisols China	- Laboratory incubation - 1%, 2% and 5% of the dry soil weight (w/w) - Trial time: 21 days	- Soil pH was 4.4; 4.2 and 3.9 for 5%, 2% and 1% of biochar applications respectively) - Soil pH significantly increased by biochar application, but that was lower compared to lime (CaO) application.	- Nitrification would be detrimental to the N uptake of tea, while NO ₃ -N produced from nitrification could be lost by leaching, runoff and denitrification. - Tea soil pH should be maintained at higher value than the optimum pH for nitrification (~5.1)	Wang et al. (2018)
Rice husk biochar at 550 °C	China	- Laboratory incubation - 0.5%, 1%, 2% (w/w) - 60 days	- Application of biochar at 2 and 4% significantly increased soil pH (3.52 and 3.63 respectively).	- The incorporation of fast pyrolysis rice husk led to a significant increase of soil total C, N, extractable Ca, Na, Mg and K contents, while available Al and Pb were reduced.	Wang et al. (2014)
Rice, wheat and peanut residue biochar at	Ultisol China	- Laboratory incubation - 1%, 2% (w/w)	- Soil pH increased in all biochar application treatments, and the highest soil pH value	- Significantly increased soil exchangeable cations but reducing soil exchangeable Al and acidity	Wang et al. (2014)

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300 °C		- Trial time: 65 days	was observed in peanut biochar, followed by wheat and rice residue biochar.	- Increasing biochar application rate has no further effect on soil pH. - Reduced acidity produced from N cycle.	
Rice straw biochar at 550 °C; Bamboo straw biochar at 750-800°C	Loamy clay China	- Glasshouse trial - 2% and 5% (w/w) - Trial time: 1 year	- pH increased by 0.9 units by bamboo biochar application, 1 unit (from 4.30- 5.30) by rice biochar use at the rate of 5%. - Increasing biochar additional rate resulted in greater soil pH increase.	- Increased plant nutrients (P, K and Mg concentrations), while reducing Mn and Cu concentrations. - Significantly improved tea growth characters compared to conventional tea management without biochar. - Rice and bamboo biochar has no significantly different effect on tea growth and tea soil nutrients.	Yan et al. (2021)
Tea pruning residue biochar at 500- 600°C	Red- yellow Japan	- Laboratory incubation - 4% (w/w) - Trial time: 90 days	- Biochar amendment significantly increased soil pH at the surface (0-5 cm, 0.23 units) and 5- 10 cm soil layer (0.73 units).	- Tea pruning residue use as mulch significantly increased soil total N, C, and also N ₂ O and CO ₂ emissions. - Converting tea pruning residue to biochar amendment and its incorporation significantly mitigate N ₂ O emission by up to 74.2%, but increased CO ₂ emission.	Oo et al. (2018)
Bamboo residue biochar at 500 °C	Inceptisols	- Glasshouse trial - 3% and 6% (w/w) - Trial time: 180 days	- Soil pH increased by 0.31 units with application rate of 3%, 0.75 units with incorporation rate at 6%.	- Reduced NH ₄ ⁺ -N leaching by up to 91.9%; NO ₃ ⁻ -N by a maximum of 66.9% and total N by up to 72.8%. - Enhanced soil nutrient retention (N by up to 23.9%). - Improved soil microbial biomass and enzyme activity.	Chen et al. (2021)
Wheat straw biochar at 450 °C	Plinthosols China	- Laboratory incubation - 4% (w/w) - Trial time: 35 days	- Soil pH increased 1.09 units compared to non-fertilizer practices, but lower compared to the combined application of biochar and N fertilizer (5.2 vs 5.4).	- Biochar amendment increased the abundance of ammonia oxidizing bacteria and Nitrous oxide reductase genes. - Increased soil C/N ratio and decreased N ₂ O emission in acidic soil. - Biochar could increase N ₂ O emission in alkaline soils	Ji et al. (2020a)
Legume and non-legume biomass at 500 °C	Utisols China	- Laboratory incubation - 1% (w/w) - Trial time: 30 days	- Soil pH immediately increased by around 0.4 units after biochar addition, then remained stably. - Legume biochar has greater impact on increasing soil pH compared to that of non-legume biochar.	- Increased soil dissolved OC but reduced inorganic N. - Suppressed N ₂ O emission by around 40% - Significantly altered fungal community structure, relative abundance of Ascomycota community, but has no significant effect on bacterial community.	Zheng et al. (2019)

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Wheat straw biochar at 450 °C	Plinthosols China	- Field experiment - 20.000kg/ha - Trial time: 2 years	- Significantly increased soil pH by 0.2 units.	- Biochar application decreased N ₂ O and NO emissions from acidic tea soils. - Denitrification was mainly responsible for producing N ₂ O in acidic soil. - Nitrification and denitrification processes were both facilitated by biochar addition.	Ji et al. (2020b)
Wheat straw biochar at 450 °C	Alfisol China	- Field experiment - 7.500 kg/ha - Trial time: 1 year	- Increased soil pH by 0.68 units compared to conventional chemical N, and by 0.55 units compared with non-fertilizer treatment.	- Biochar applications reduced N ₂ O and NO emission factor by 1.82 and 1.38 respectively, compared to chemical N use. - Biochar combined with manure chicken applied to tea soils could mitigate N gas emissions and increase tea productivity.	He et al. (2019)
Mushroom residue biochar at 500 °C	Ultisols China	- Field experiment - 1.350 kg/ha and 2.390kg/ha - Trial time: 1 year	- Biochar application at a rate of 1.350 kg/ha increased soil pH by 0.1 units after one year, while the figure for the higher rate (2.390kg/ha, biochar + based chemical fertilizer) was 0.27 units.	- Biochar application enhanced plant beneficial fungal genera such as <i>Chloridium</i> , <i>Clavulina</i> , <i>Amylocorticium</i> , <i>Rhodosporidiobolus</i> and bacterial genera such as, <i>Mizugakiibacter</i> , <i>Rhodanobacter</i> and <i>Pedobacter</i> . - Increased tea yield and yield components, tea quality indicators such as amino acids and water extract contents.	Yang et al. (2021)
Rice straw	- China	- Field experiment - 7cm thick - Trial time: 8 months	- Increased soil pH by 0.13 units compared to non- mulching practice.	- Reduced soil temperature variation and having a significant cooling effect in the deep soil layer - Significantly improved soil water retention while reducing soil compactness. - Significantly increased soil OM, available N, P, K and total N.	Xianchen et al. (2020)
Plant residue ash (canola, wheat rice, corn, soybean peanut...)	Alfisol China	- Laboratory incubation - 20g ash/ 350g soil - Trial time: 60 days	- Plant residue ash significantly increased soil pH (by 0.3 units on average). - Leguminous residues had more significant effects in raising soil pH than the non-legumes.	- Reduced soil Al exchangeable concentrations.	Wang et al. (2009)
Fern (<i>Gleichenia linearis</i>)	Acrisols Vietnam	- Field experiment - 0, 15, 25, 35 and 45 tons/ha (fresh weight) - Trial time: 3 years	- Application rate of 15 and 25 tons/ ha significantly increased soil pH at the 3 years of experiment, while the rates of 35 and 45 tons/ha had inconsistent effect on soil pH.	- Significantly increased soil basic cations (Ca ²⁺ and Mg ²⁺) while reducing soil Al ³⁺ - Improved soil moisture, soil bulk density and humus substances, and enhanced soil microbial activities. - Application rate at 25tons/ha of fern is recommended.	Cu and Thu (2014a)

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Tea pruned residues	Acrisols Vietnam	- Field experiment - 30 tons/ha - Trial time: 3 years	- Tea residue mulches significantly increased soil pH (by 0.3 units after 1 year; 1.1 units after 3 years) compared to no- mulching practice.	- Increased soil moisture, soil OM content and reduced soil bulk density. - Significantly increased total number of soil bacterial, fungi and actinomyces. - The influences of tea pruned residues on soil properties reduced rapidly after 3 application years.	Cu and Thu (2014b)
Peanut hull	Brown soil China	- Field experiment - 10cm thick	- Soil pH slightly increased (0.04 units) compared to non-mulch treatments.	- Significantly increased soil moisture contents, OM, total N and K, available N but reduced total P, available P and K. - Increased fungal community diversity in 0–20 cm soils and that of bacterial communities in 20–40 cm soils.	Zhang et al. (2020b)
Intercropping with <i>Vulpia myuros</i>	China	- Field experiment - 7cm thick - Trial time: 8 months	- Increased soil pH by 0.06 units compared to tea monoculture.	- Significantly increased soil OM, soil available N, P, K and total N, and soil enzyme activity. - Optimized topsoil temperature, increased soil water holding capacity while reducing soil compactness.	Xianchen et al. (2020)
Intercropping with aromatic plants (<i>Cassia tora</i> , <i>Medicago sativa</i> , <i>Leonurus artemisia</i> , and <i>Mentha haplocalyx</i>)	Acidic Histosols China	- Greenhouse trial - Trial time: 2 years	- Data not provided	- Decreased the population of tea green leafhoppers while increasing the natural enemies of tea pests such as spiders, lacewings, and parasitoids.	Zhang et al. (2017)
Intercropping with fruit trees (loquat, waxberry and citrus)	Yellow soil China	- Field experiment - Trial time: 30 years	- Soil pH at three soil depths (0-10, 10-20 and 20-30cm) significantly increased by intercropping practices, compared to that in mono tea plantations.	-Increased soil OM, available P and K while reducing heavy metal (Cr, Cd, As, Hg, and Pb) - Improved tea quality indicators such as amino acid and catechin.	Wen et al. (2019)
Agroforestry (tea-Ginkgo tree (<i>Ginkgo biloba</i> L)).	China	- Field experiment - Growing distance: 10 x 10m and 6 x 6m - Trial time: 11 years	- Increased soil pH at all observed soil depths (by 0.65 units at 0-10cm layer, 0.15 at 10- 20cm layer and 0.35 at 20-30 cm layer).	- Significantly increased soil OC, OM and total N contents, soil microbial biomass, and enzyme activity. - Enhanced soil productivity and sustainability.	Tian et al. (2013)

4. Conclusion and perspectives

Soil acidification is becoming an increasingly severe problem in many tea growing countries, resulting in serious impacts on soil chemical properties, tea productivity and quality and the environment. To date however, how low pH affects tea soil biological and physical properties as well as its management cost have been poorly explored. Agriculture wastes and products have demonstrated a great potential to mitigate soil acidification by tea cultivation and improve tea soil health. Being naturally alkaline with high pH value and buffering capacity, these materials could supply alkaline matter and essential elements to neutralize soil acidity and alter soil properties, positively influencing soil nutrient availability, enrich soil organisms and ultimately improve tea yield and quality indicators. While promising, their expanded uses would need further understanding to improve their application efficacy while reducing any potential negative consequences on the environment. In addition, the risks of introduction of heavy metal and pathogens from animal manures, compost and biochar applications have been widely reported (Alegbeleye and Sant'Ana 2020; Dai et al. 2017), but how they could affect soil and tea plants have not been clearly understood. Moreover, most of reports on effective impacts of biochar for correcting soil acidification have been the outcomes of laboratory or glasshouse studies, thus the results need to be validated in field conditions (Dai et al. 2017). Finally, the majority of studies on utilizing agricultural wastes in tea cultivation to date have been implemented in China, with specific but limited soil characteristics, climate conditions and tea management practices. It has been clearly indicated that differences in such conditions could significantly affect the effectiveness of these soil acidification ameliorants (Gu et al. 2019; Siedt et al. 2020; Wu et al. 2020). This research gap highlights the need and opportunities for further investigations in other systems to provide comprehensive knowledge and reliability in recycling these soil amendments.

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Conflicts of Interest

The authors declare no conflict of interest.

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Responses to the Editor's comments

We are sincerely indebted to the Editor for giving comments on our manuscript. We have revised our manuscript following all the comments and we hope that this revised version will fit with the editor's expectations.

Editor's comments

1. The introduction is too long and needs to be shortened down to 1-2 paragraphs

Answer:

Thanks for the Editor's comment. The introduction has now been rewritten and the length has been reduced by around a half.

2. Possibility to discuss the ocean and other acidifications for attracting a wider range of readers.

Answer:

We have now added a new sub-section 2.1 to briefly discuss the status and mechanism of ocean and soil acidification and how they relate to agricultural activities.

3. The figures need to be accurately presented (texts, legend, white space, Y and X axis...)

Answer:

We have now carefully revised all the figures attached in the manuscript. We hope that they will fit with the editor's expectations.

4. Write at the end of each article section a conclusion of about 1-2 sentences to summarize the major points of the section and its significance.

Answer:

Thanks for the Editor's advice. We now reviewed the whole manuscript and added summary/conclusion to many sections/subsections where we believe that they are accurate.

5. Finding relevant articles published by Environmental Chemistry Letters (5-10 articles, using keywords and search from the journal home page) and consider citing them as the references. I believe we have published articles concerning soil and its degradation, tea as well as acidification.

Answer:

Thanks for the Editor's advice. We have been carefully checked and cited 8 articles published by ECL as references in text and highlighted these references in the bibliography. They are as follow:

1. Akhil D, Lakshmi D, Kartik A, Vo D-VN, Arun J, Gopinath KP (2021) Production, characterization, activation and environmental applications of engineered biochar: a review. *Environ Chem Lett* 19: 2261-2297. <http://doi.org/10.1007/s10311-020-01167-7>
2. Gunarathne V, Ashiq A, Ramanayaka S, Wijekoon P, Vithanage M (2019) Biochar from municipal solid waste for resource recovery and pollution remediation. *Environ Chem Lett* 17: 1225-1235. <https://doi.org/10.1007/s10311-019-00866-0>
3. Ochedi FO, Yu J, Yu H, Liu Y, Hussain A (2021) Carbon dioxide capture using liquid absorption methods: a review. *Environ Chem Lett* 19: 77-109. <https://doi.org/10.1007/s10311-020-01093-8>
4. Patra BR, Mukherjee A, Nanda S, Dalai AK (2021) Biochar production, activation and adsorptive applications: a review. *Environ Chem Lett* 19: 2237-2259. <https://doi.org/10.1007/s10311-020-01165-9>
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