REVIEW



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Biochar mitigates allelopathy through regulating allelochemical generation from plants and accumulation in soil

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Abstract

Soil health is a key factor for the global food security. One of the main bottlenecks for the efficient utilization of farmland soil is the accumulation of allelochemicals excreted by plant roots, leached and volatilized from plants, and released during the decomposition of plant residues. The negative impacts derived from allelochemicals include: decreasing soil fertility, breaking the balance of soil microbial community, reducing seed germination rate and crop yield, and interfering with the growth of neighboring or successive plants. Recently, biochar, a soil amendment with low cost and high performance, is extensively studied on its functions and effects on farmland systems. In this review, we focused on the potential role of biochar in controlling the environmental fate and risk of allelochemicals in soils. The source and potential toxicity of allelochemicals in farmland and the deficiencies in current methods for alleviating the allelopathy were summarized first, followed by the discussion on their sorption and degradation by biochar as well as the impact of biochar properties on the performance and mechanism. At the same time, the generation of allelochemicals regulated by biochar through improving plant growth conditions was analyzed. The modification of biochar for alleviating the allelopathy and then overcoming the obstacles of agricultural production was also addressed. Future research directions, such as targeted biochar production, the interactions between different allelochemicals and biochar investigated by simulations and modeling, negative effects and stability of biochar or modified biochar, were then proposed. This review will provide useful theoretical and technical supports for targeted biochar applications in soils with allelopathy.

Highlights

1. Biochar can regulate the generation and release of allelochemicals by plant and be more effective after modification;

2. Biochar alleviates the allelopathy by adsorbing or degrading allelochemicals and buffering ecological stress;

3. Biochar promotes biodegradation of allelochemicals by activating soil organisms and providing habitats for them.

Keywords: Cultivated land, Environmental fate, Farming obstacles, Sustainable agriculture, Rhizosphere

Graphical abstract

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1 Introduction

With the fast increase of the global population, the demand for food is also growing explosively. To ensure the grain yield and quality, intensive soil cultivation is promoted globally, resulting in the deterioration of soil quality and reduction of crop yields (Lu and Huang 2010; Zhou et al. 2017). The obstacle of crop rotation has become the main bottleneck of efficient soil utilization, which will exacerbate the competition between plants for resources in the exhausted soil. Even worse, the plants may secrete many chemicals to ensure themselves in the leading position of the competition. These competitions may reduce the soil fertility, seed germination rate and crop yields, or disturb the balance of microbial communities (Li et al. 2021). Most of the above negative impacts are associated with the accumulation of allelochemicals (Bennett et al. 2012). These chemicals are secondary metabolites generated by plants. They enter soil through volatilization from plant stems and leaves, secretion from the root, decomposition of plant residues, and leaching from leaves by rain or dew (Inderjit 2005; Weston and Mathesius 2013). The content of allelochemicals generated by plants is highly determined by the nutrients, pH value, humidity, microbial community structure and activity, planting structure and density, as well as the chemical stresses of the soil environment (Duke 2010). On one hand, allelochemicals could induce the colonization of beneficial microorganisms, and thus plant can take advantage in competing for resources (Qu and Wang 2008). On the other hand, these chemicals can inhibit the growth of surrounding plants by interfering with their photosynthesis, interrupting the synthesis of adenosine triphosphate (ATP), and disrupting the normal metabolism of plants (Kumar and Bais 2010; Weir et al. 2004). In high-density crop planting, the accumulated allelochemicals may exceed the defense ability of its own kind (Yang et al. 2015). Therefore, understanding the release and regulation of allelochemicals is crucial for improving the utilization efficiency of cultivated soil.

Generally, allelochemical release is heavily associated with the environmental stresses of plants. Biochar has been proven to improve soil environment and microbial activity (Fig. 1). Biochar is a kind of carbon-rich particle produced by pyrolyzing waste biomass under hypoxic conditions. Biochar application can increase soil porosity and pH, retain water and nutrient (Yang et al. 2016),



and immobilize various contaminants (Kondrlova et al. 2018), which could reduce the environmental stress on crops and thus decrease the generation of allelochemicals. Moreover, biochar can provide habitats for microorganisms or establish a redox environment in soil, which promotes allelochemical degradation, biologically and/or chemically (Yan et al. 2021). Therefore, biochar displays a promising potential to alleviate the allelopathy as a soil amendment. However, some technical and practical barriers are reported in biochar application in large quantities, such as its relatively high cost, release of PAHs, and high redox potential (Das et al. 2020). Unfortunately, most previous work only focused on a certain process in soil, lacking attention on the comprehensive system with multiple processes, especially in complex environmental media.

In this review, the occurrence and implications of allelochemicals in soil systems were first introduced. The feasibility of biochar for regulating the release and environmental fate of allelochemicals were examined and summarized. The interactions between biochar and microorganisms, plants, or allelochemicals were systematically analyzed and discussed. The modification of biochar with enhanced functions for alleviating allelopathy was also proposed. This review concludes with emphasizing the future challenges and opportunities of biochar application in mitigating allelopathic effects and eventually contributing to sustainable agriculture.

2 The negative impacts of allelochemicals in soil systems and the current preventive methods

2.1 The types and negative impacts of allelochemicals generated by plants

Many plants can generate allelochemicals (Wang et al. 2005). For example, allelopathic wheat generated allelochemical (2,4-dihydroxy-7-methoxy-1,4-benzoxa-zin-3-one) in roots with the concentration of 300 to 900 μ g/g dry weight (Kong et al. 2018); three chemicals, veratric acid, maltol, and (–)-loliolide, were generated by crabgrass with the concentrations ranged from 0.16 to 8.10 μ g/g (Zhou et al. 2013). The generated allelochemicals enter the soil systems through volatilization from plant stems and leaves, secretion and drip washing from the plant root, decomposition of plant residues,

and leaching from leaves by rain or dew (Fig. 1). The allelochemical types and associated plant species, bioactive concentration range, and corresponding action modes are summarized in Table 1. Different allelochemicals may have different target physicochemical processes. For instance, ester allelochemicals can reduce the number of micronucleus in plant root tip cells (Duan et al. 2013); alkaloids can destroy electron transport and ATP synthesis (Veiga et al. 2008); quinones, benzoic acid derivatives, cinnamic acid derivatives, flavonoids, and terpenoids can inhibit the photosynthesis and respiration, nutrient accumulation, nutrient circulation, membrane permeability, enzyme activity, and hormone balance of plants (Jose and Gillespie 1998). Allelochemicals from plants can also cause diseases and pests to the competing species and enlarges their own territory (Yang et al. 2015). They may cause a substantial decrease in the quality and yield of other crops and even the death of other species in extreme cases (Aschehoug et al. 2016). The overall action mechanisms include the inhibition of cell differentiation by changing the cellular and submicroscopic structures, enhancement of cell membrane permeability, reduction of selective permeation, decrease of hormone and enzyme activity, inhibition of photosynthesis, and blockade of gene expression. Mostly, several mechanisms play a synergistic role in inhibiting the growth of plants, which is more pronounced when multiple allelochemicals coexist.

2.2 Deficiencies in current methods for alleviating the allelopathy

Recently, investigators have carried out various studies to alleviate the allelopathy in soil by regulating the biological conditions and environmental factors (Sujeeun and Thomas 2017; Zhang et al. 2018). For instance, Li et al. constructed a reductive system of soil disinfestation to effectively degrade the allelochemicals generated by Panax notoginseng and suppress the pathogens (Li et al. 2019b). However, the variety of allelochemicals produced by different plant species, combined with the complexity of soil systems, pose many difficulties in alleviating allelopathy (Yang et al. 2016). Cropping modes, such as crop rotation and intercropping, are helpful in alleviating allelopathy (Korai et al. 2018). They can maintain the crop yields for a short time and are economically preferred without additional financial investment. However, longterm crop rotation and intercropping may cause higher accumulation and diversity of allelochemicals in soil. Soil still faces the problems of fertility reduction, quality degradation, and microbial imbalance (Korai et al. 2018). The use of chemical fertilizer in soil can supplement nutrients in a short period of time, reduce the occurrence of pests and diseases, and thus alleviate the allelopathy (Ding et al. 2012). However, nutrients may be leached during rain events, and long-term use of chemical fertilizers will cause soil degradation and environmental pollution (Tong et al. 1997). Therefore, it is urgent to find a new method to regulate the generation and release of allelochemicals and release in soil systems and thus improve agricultural production.

3 Biochar application decreases the effective allelochemical concentration through sorption and degradation

Biochar may alleviate the negative impacts of allelochemicals in soil through sorption and degradation owing to its physicochemical properties. The sorption and degradation of allelochemicals by biochar released from plants are presented in Fig. 2.

3.1 Biochar properties affecting allelochemical sorption

The physical and chemical properties of biochar determining their sorption characteristics include specific surface area, particle size, porous structure, surface charge, and functional groups. Generally speaking, the higher the pyrolysis temperature, the larger the surface area of biochar, the more sorption sites for organic molecules, thus the greater sorption capacity to allelochemicals (Ahmad et al. 2014). The sorption irreversibility is another dimension that should be carefully evaluated in sorption studies. For example, micropores may play an important role in retaining pollutants (Ma et al. 2019). When allelopathic molecules enter pores with similar size, the pores will be expanded, accompanied by an exothermic process, which makes them difficult to desorb (Ma et al. 2019).

The particle size of biochar is associated with several other properties, such as the surface area and the abundance of surface functional groups, which generally shows a positive relationship with the sorption rate and capacity (Patel et al. 2021). Recently, studies have emphasized the importance of nano-sized biochar in the removal of organic contaminants and its superior performance compared to the bulk counterparts (Shen et al. 2020). The particles of nano-biochar share similar characteristics with engineered nanomaterials with great surface area and abundant functional groups, which have high sorption affinity to organics. It seems that the application of nano-sized biochar could be a more efficient technique to remove allelochemicals compared to bulk biochar (Rajput et al. 2022). However, it should be noted that the biochar particles of colloidal state tend to aggregate or move away from the original application spot, which likely reduces the effectiveness of the nanosize biochar. At the same time, a wide knowledge gap still exists between the laboratory tests of nano-biochar and its pilot application. Thus, the efficiency of nano-sized

Table 1 The negative in	mpacts of the allelocher	nicals on the plant growth	
Allelochemical types	Bioactivity range (mg/ kg) (Macias 1994)	Plant species	Action models
Simple esters	0.631–15.9	Wheat (Wang et al. 2006), rice (<i>Oryza sativa</i> L.)(Kato-Noguchi et al. 2002), <i>Mirabilis jalapa</i> L. (<i>Nyctaginaceae</i>) (Anaya and Pelayo-Benavides 1997),Welsh onion (<i>Allium fistulosum</i> L.) (Xu et al. 2013), asparagus (Yeasmin et al. 2013)	Reducing the number of micronucleus in plant root tip cells (Duan et al. 2013).
Alkaloids	79.4–794	Hortia superba (<i>Rutaceae</i>) (Veiga et al. 2008), <i>Peganum harmala</i> L. (Shao et al. 2013), Lupin (Tyski et al. 1988)	Destroying electron transport and ATP synthesis, and causing damage to root tip cell walls and organelles (Liu and Lovett 1993).
Polyacetylenes	0.794-50.1	Conyza canadensis L. Cronquist syn. (horseweed) (Queiroz et al. 2012)	1
Sulphur compounds	1.26–10, 31.6–126	Wheat (Dinelli et al. 2007), ryegrass (Dinelli et al. 2007), wild cabbage (Chen and Yan 2007)	1
Fatty acids	0.199-0.631	Wheat, cucumber (Yuansen et al. 2007), peanut (Liu et al. 2012)	1
Quinones	19.9–158	Juglans regia L. (Willis 2000)	Causing the wilting and browning of the vascular tissue and plant death. Inhibition of photosynthesis and respiration (Kocacaliskan et al. 2009).
Benzoic acid derivative	15.8-158	Wheat (Carlsen et al. 2009), Welsh onion (<i>Allium fistulosum L</i> .) (Xu et al. 2013), eggplant (Wang et al. 2005), peanut (Huang et al. 2013), tobacco (Ren et al. 2015), rice (<i>Oryza sativa</i> L.) (Chung et al. 2018), cucumber (Kato-Noguchi et al. 2012), soybean (Porter et al. 1986)	Inhibition of plant nutrient accumulation, nutrient cycling, photosyn- thesis, respiration, membrane permeability, enzyme activity, hormone balance (Li et al. 2010).
Cinnamic acid derivatives	1.58–1584	Wheat (Carlsen et al. 2009), eggplant (Wang et al. 2005), peanut (Huang et al. 2013), rice (<i>Oryza sativa</i> L.) (Chung et al. 2018), watermelon (Abu-Reidah et al. 2013)	Inhibition of plant nutrient accumulation, nutrient cycling, photosyn- thesis, respiration, membrane permeability, enzyme activity, hormone balance (Abrahim et al. 2003).
Coumarins	3.98–79.4	Eggplant (Wang et al. 2005), arabidopsis thaliana (Jrg et al. 2016), sweet vernalgrass (<i>Anthoxanthum odoratum L</i>) (Yamamoto 2009)	Reducing photosynthetic pigment content and inhibiting nitrogen metabolism (Chen et al. 2011).
Flavonoids	1.58–19.9	Soybean (Porter et al. 1986), alfalfa (<i>Medicago sativa L</i>) (Hartwig et al. 1991), <i>Trifolium repens</i> L. (Carlsen et al. 2014), <i>Trifolium pratense</i> L., Stellera chamaejasme L. (<i>Thymelaeaceae</i>) (Zhiqiang et al. 2015), <i>Typha</i> <i>angustifolia</i> (Yongvanich et al. 2002)	Inhibition of photosynthesis, respiration, enzyme activity.
Monoterpenes	0.100-158	Ambrosia trifida L. (Peng et al. 2008), Pinus ponderosa (Madronich et al. 2011)	Inhibition of respiration. Induction of oxidative stress (Zunino and Zygadlo 2004).
Sesquiterpenes	0.0001-0.001, 0.199-158	Helianthus annuus (Suo and Yang 2006)	Inhibition of photosynthesis (Bennett et al. 2012).
Sesquiterpene lactone	0.0001-10.0	Juncu effusus (Ervin and Robert 2000), <i>Centaurea repens</i> (Robles et al. 2015)	Induction of oxidative stress (Robles et al. 2015).
Diterpenes	2.51–19.9	<i>Cistus ladanife</i> r L. (Natividad et al. 2019), <i>Chrysoma pauciflosculosa</i> (1992)	Destroying electron transfer (Ding et al. 2008).
Triterpene	0.0010-31.6	American ginseng (Nicol et al. 2003), <i>panax pseudoginseng</i> (Ding et al. 2018), ginseng (Ding et al. 2018), alfalfa (<i>Medicago sativa</i> L.) (Jennings and Nelson 1998)	Causing the decrease of mitochondria and the rupture of the outer membrane of organelles. Destroying root tip cell division. Induction of oxidative stress (Yang et al. 2015).



biochar for organic contaminant removal in soil should be systematically evaluated in pilot-scale studies.

The sorption strength is also highly dependent on the functional groups on biochar (Zhu et al. 2005). For example, oxygen-containing functional groups (OFGs) on biochar could adsorb organics through the formation of electron-donor-acceptor (EDA) complexes in binary and ternary sorption systems (Mohammad et al. 2018). The structures of allelochemicals and biochar may contain aromatic rings, carboxyl groups, hydroxyl groups, ester groups, and quinone groups. These functional groups can be paired to form binary complexes. The binding energies of the interactions can be calculated based on the density functional theory (DFT) calculations and they can be used for comparing the sorption strength (Chen et al. 2021). In these EDA interactions, the donors and acceptors are dependent on the actual sorption systems, which can be elucidated based on the frontier orbital theory in quantum chemical calculations (Chen et al. 2019). The molecular dynamics simulation is also an efficient approach to study the sorption of allelochemicals, by which the sorption process can be visualized and the sorption energy and mechanisms can be elucidated in statistics (Zuo et al. 2022). At the same time, hydrogen bonds including charge-assisted hydrogen bonds (Ahmad et al. 2014), hydrophobic effects, and partitioning could all contribute to allelochemical sorption on biochar (Dai et al. 2019). Notably, the ternary sorption, such as cation bridging formed between the negatively charged allelochemicals and biochar, could also increase the sorption (Xu et al. 2021b).

After their application in soil, biochar particles might be covered by soil organic matter or microbes forming an eco-corona (Wang et al. 2021). In this case, the sorption or degradation may be mainly mediated by the coated organic matter and soil microbes. If this situation is not properly considered, the sorption or degradation of allelochemicals by biochar may be misestimated, most likely overestimated. The contribution of biochar in these conditions may be quantitatively analyzed by measuring the effective surface area.

3.2 Biochar properties affecting allelochemical degradation

Recent studies pointed out that biochar could mediate the degradation of organic pollutants. The degradation

of organic pollutants mediated by biochar is recognized as free radical pathway and non-radical pathway (Ruan et al. 2019; Yuan et al. 2017). The main contributors in the free radical pathway are the environmental persistent free radicals (EPFRs), hydroxyl radical (•OH), and superoxide radical (•O $_2^-)$ (Fang et al. 2015b). The EPFRs on biochar may degrade organic pollutants after their direct contact, or induce $\cdot OH$ or $\cdot O_2^-$ generation from water or oxygen to degrade organic pollutants (Qin et al. 2018). In addition, it is noted that redox-active structures of biochar might exchange electrons with organic pollutants to promote degradation (Van der Zee and Cervantes 2009), which is known as a non-radical pathway. Especially, the phenolic and quinones functional groups on biochar are electron-donating functional groups and electron-accepting functional groups, respectively (Yuan et al. 2017). Many allelochemicals possess abundant phenolic hydroxyl groups, which are readily participating in redox reactions with biochar. In these cases, the organics were easily attacked by electrons, which promoted both transformation and biotransformation of the chemicals (Yuan et al. 2017). In addition, the strongly oxidized biochar will generate ${}^{1}O_{2}$ through the energy transfer of the quinone structure (Zhang et al. 2021a). It has strong oxidizing properties and can degrade allelochemicals, which is also a non-radical degradation pathway.

Additionally, when biochar is added to soil, biodegradation of allelochemicals can be accelerated. For example, adding biochar to soil increased the diversity of fungi and reduced the diversity of bacteria, which accelerated the biodegradation of allelochemicals (Li et al. 2013). The addition of biochar can increase the biodegradation of phthalates by 36% (Ren et al. 2021). At the same time, the effective activity concentration or even the activityrelated structures of the extracellular enzymes may be altered after they interacted with biochar. Therefore, the biodegradation of allelochemicals can be promoted or inhibited but it is heavily associated with the type of enzymes (Fang et al. 2021). Therefore, biochar could effectively alleviate allelopathy in the soil if the chemical or biological degradation could be properly mediated.

3.3 Allelochemical types affecting biochar-allelochemical interactions

The hydrophobic properties, polarity, and molecular size of allelochemicals regulate their interactions with biochar. For example, hydrophilic allelochemicals (p-hydroxybenzoic acid) are more easily adsorbed by biochar (Teixido et al. 2011). The hydrophobic allelochemicals (such as phthalates) could be adsorbed through the hydrophobic effect or van der Waals forces (Dickenson and Inyang 2015). The allelochemicals with larger molecules (such as ginsenosides) are retained in mesopores, while allelochemicals of smaller molecules (such as citric acid) are more easily protected by the micropores of biochar (Ma et al. 2019). It is thus a comprehensive task to alleviate allelopathy using biochar, and it is also enlightening to specifically design biochar to remove a class of targeted allelochemicals. Therefore, how to specifically alleviate the allelopathy produced by different allelochemicals requires more in-depth exploration. In addition, if biochar-allelochemicals interaction mechanisms could be revealed on how allelochemicals complementarily occupy different sorption sites, a biochar designed for a broad-spectrum sorption and degradation ability might be a promising way, which will be discussed in the subsequent section.

4 Biochar application decreases allelochemical generation through improving plant growth condition

The generation of allelochemicals is dependent on the physiological requirement of the plant. Generally, physical or chemical stresses will promote the generation of allelochemicals, while a low amount of allelochemicals will be excreted if the plant is in a preferred condition. The application of biochar may improve soil fertility or productivity, but at the same time, may pose some additional stress to plants. As presented in Fig. 3, the apparent impact of biochar on allelochemical generation may be a combination of various processes, which will be individually discussed in the following subsections.

4.1 Nutrients released from or retained by biochar

When crops are densely grown, they compete for the limited nutrients (Aschehoug et al. 2016). According to the statistics of intraspecies and interspecies interaction studies in recent years, 67% of these studies showed negative outcomes because of intraspecific and interspecific competitions for the nutrients (Adler et al. 2018). Biochar application may greatly alleviate these competitions, and thus reduce the generation of allelochemicals.

Biochar itself can release a variety of nutrients, such as C, N, P, and K, which can alleviate the nutrient stress, and thus reduce the release of allelochemicals (Lou et al. 2016). The elemental composition and content of biochar largely depend on the raw materials and pyrolysis temperatures. Hair and chicken feather-derived biochar has a high content of N, while animal manure-derived biochar has high contents of P and K (Domingues et al. 2017; Hass et al. 2012). When the pyrolysis temperature increased between 300 and 600 °C, the content of total P and total K in wood and herbal biochar increased accordingly (Zheng et al. 2013). However, it should be noted that not all these nutrients are available to plants. Only soluble components can be readily used for plant growth.



As a matter of fact, available P and N decreased with the increasing temperature (300–600 °C) (Zheng et al. 2013). The reason is that P was converted to less soluble minerals at high temperatures, and N was lost as gas products at high temperatures or converted into heterocyclic-N (Zheng et al. 2013). Soil chemistry conditions also play an important role in the nutrient availability of biochar. Taken P as an example, P release in 24h increased from 8.9 to 4.5, which was comparable to conventional P fertilizers (Silber et al. 2010). Therefore, adding biochar to soil can effectively reduce the nutrient limits to plants and decrease the generation and release of allelochemicals.

Biochar can also adsorb nutrients in the soil and release them slowly to function as a slow-release fertilizer. The nutrient detaining by biochar is closely related to the feedstock. For example, rice husk biochar could strongly retain more nutrients than camellia oil shell biochar, because the pores of rice husk biochar were well developed (Zhao et al. 2021). In wheat cultivation experiments, corn stalk biochar was a better slow-release fertilizer matrix than wheat stalk biochar, because of its larger cation exchange capacity (CEC) and higher sorption to nutrients (Zhao et al. 2021).

The intensive tillage may result in both nutrient shortage and allelochemical accumulation (Dong et al. 2018). The application of biochar could introduce some additional nutrients and partly alleviate the nutrient shortage (Rajkovich et al. 2012). At the same time, biochar could adsorb or degrade allelochemicals (Gámiz et al. 2021). Both processes may be associated with the apparent decrease of the detected allelochemicals. Clearly, previous studies did not carefully identify these two processes. How to distinguish these processes will need a specifically designed experiment. The investigators suggested to add a reference experiment with the addition of biochar supernatant to quantify the impact of biochar nutrients on allelochemical release.

4.2 Improvement of soil physical and chemical properties by biochar

The addition of biochar to soil can improve soil effective water content, increase soil porosity, promote soil aggregates, enhance soil CEC, and thus develop a more suitable environment for plant growth, and reduce the generation of allelochemicals (Abel et al. 2013). Studies have shown that effective water content was increased by about 22% after the application of biochar, which can reduce the drought stress on plants (Peake et al. 2014). Soil porosity was also increased by 20% after biochar application, which alleviated the pressure caused by soil compaction on plant growth (Laird et al. 2010). The addition of biochar can increase soil CEC by up to 35.4% due to the lower original content of exchangeable Ca²⁺ compared with biochar (Hailegnaw et al. 2019). The addition of biochar can also increase soil aggregation by 8-36% because of the increased organic matter contents (Oguntunde et al. 2004). Moreover, biochar is effective to increase soil pH value of acidic soils, thus alleviating soil acidification stress to plants (Wu et al. 2018). Considering the highly heterogeneous structures of biochar, biochar with different properties may have different advantages to alleviate one of the stresses. If a systematic comparison could be conducted on the strength of the individual stresses to trigger the generation and release of allelochemicals, the

biochar could be site-specifically designed to overcome the environmental stresses of a certain remediation site.

4.3 Pollutants immobilized or removed by biochar

Soil pollution, such as heavy metals, organic pollutants, has a major negative effect on plant growth. When the pollution exceeds plant tolerance, their self-protective system will be activated and allelochemicals will be secreted to detoxicate the pollution. For example, the secretion of citric acid, malic acid, and succinic acid in the wheat root system increased significantly under Al stress (de Andrade et al. 2011). These allelochemicals could form relatively stable complexes with heavy metals, which reduce the bioavailability and stress of heavy metals on plants (Riaz et al. 2018). Under the stress of organic pollutants, such as phenanthrene, the secretion of organic acids in ryegrass was significantly stimulated (Xie et al. 2008). These allelochemicals can promote the growth of bacteria (Sphingobacteriia and Actinobacteria) that degrade phenanthrene in the rhizosphere, and alleviate the stress of organic pollutants on plants (Li et al. 2019a).

After biochar addition, the bioavailability of heavy metals in the soil will be reduced through sorption, complexation, precipitation, or even redox reactions. For example, some anions in biochar, such as OH^- , CO_3^{2-} , and SiO_3^{2-} , could form insoluble salts and reduce the content of exchangeable heavy metals (Zhu et al. 2015). Moreover, with the increase of soil pH, the negative charge on the surface of clay minerals also increased, which promoted the sorption of heavy metals by soil particles (Chao et al. 2018). The investigators summarized that the addition of biochar in the soil will reduce the mobility of Cu, Cd, Pb, and Zn by 36.4%, 25.6%, 26.1%, and 16.5%, respectively (Liu et al. 2021). Similarly, organic pollutants can be immobilized through surface sorption, matrix partitioning, sequestration, or degradation (Ruan et al. 2019; Van der Zee and Cervantes 2009). In addition, biochar may accelerate organic pollutant degradation, mediated by biochar redox potential or PFRs as discussed earlier.

It is generally accepted that the generation of allelochemicals stimulated by pollution could be greatly decreased after biochar application. However, the following points need to be carefully evaluated when assessing pollutant-biochar interactions. (1) It is unknown how to estimate the release of the adsorbed pollutants from biochar, especially with the change of soil chemistry conditions. It is easy to understand that the primary process for pollutant detoxification is retention or degradation, not the apparent sorption. Therefore, the batch sorption experiments provide only limited information to understand biochar function. (2) Biochar properties should be considered dynamically in soil systems. The active sites may easily interact with microorganisms or soil chemical compositions, likely being deactivated. Thus, biocharpollutant interactions are also changed. Only limited studies were carried out to investigate biochar aging and its implications. (3) The sorption of chemicals by biochar may reduce the apparent bioavailability of allelochemicals. However, if the allelochemicals were just adsorbed on biochar without degradation or decomposition, their allelopathy was not affected largely. Even worse, the adsorbed allelochemicals will be accumulated to increase their contact probabilities with soil organisms, including microorganisms and soil animals on the surface of biochar, resulting in the deterioration of soil health.

4.4 Improvement of soil biological/microbial compositions by biochar

The impacts of biochar on soil biological compositions or activities could be understood from enzymes, microorganisms, and animals. The activity of soil enzyme is one of the important indicators for soil quality and determines the decomposition of organic matter and nutrient cycling. Some enzymes are directly related to allelochemical excretion. For example, the activities of rhizosphere enzymes (such as protease, urease, phosphodiesterase, arylsulfatase, and β -glucosidase) are closely associated with the secretion of small molecular organic acids by regulating the C-, N-, S-, and N-cycling (Das et al. 2017). Biochar application could improve enzyme activities in soils through stimulating soil microbial biomass (Khadem and Raiesi 2017) or protect enzymes from the attack of organisms and reactive chemicals (Joseph et al. 2021).

It is widely reported that biochar could alter the microbial communities of soils, and it is also well-known that allelochemical generation could determine or be determined by plant-microorganism communications (Scavo et al. 2019). The porous structure and carbon-based properties of biochar as well as the nutrients and water retained by biochar are all beneficial for microorganism growth, especially in soil with low organic matters (Joseph et al. 2013). Studies have shown that biochar applied to the soil increased the microbial diversity by 1.25 times and the colonization rate of arbuscular mycorrhiza (AM) by 20% (Matsubara et al. 2008). The increase in AM colonization rate is beneficial to increasing the absorption capacity of nutrients and water by plant roots, thereby reducing the secretion of plant allelochemicals.

It should be noted that biochar application does not change only one microorganism species. Investigators generally agreed that biochar application enhanced the abundance of microorganisms, but reported different results on the diversity of the microbial community as affected by biochar addition (Xu et al. 2021a). It is still unknown how biochar-motivated microbial community



may generate a lower amount of allelochemicals. At the same time, biochar may adsorb or degrade allelochemicals to decrease their effective concentrations. Biochar can be specifically modified with improved properties to decrease allelochemical generation or regulate their behavior and activity

change is related to allelochemical release by plants because of the complex soil conditions and biochar properties. In addition, biochar not only promotes the growth of beneficial microorganisms but also promotes the growth of harmful microorganisms. It is hardly investigated how to balance the different species for a long term.

The activity of animals in the soil is also important for soil productivity. However, the interactions among soil animals, allelochemicals, and biochar application have not been studied yet. It was reported that allelochemicals could pose significant toxicity to soil animals, including earthworms (Liu et al. 2020) and nematodes (Hamaguchi et al. 2019). Biochar may have dual roles in benefiting soil animal activity. For example, biochar could adsorb or degrade allelochemicals to alleviate their toxic impacts. At the same time, biochar may maintain preferential habitats for soil animals by improving the soil quality (Sanchez-Hernandez et al. 2019).

4.5 The flip side of applying biochar to regulate allelopathy

Volatile organic compounds (VOCs), EPFRs, heavy metals and polycyclic aromatic hydrocarbons (PAHs) contained in biochar may induce oxidative stress in plant roots, destroy the integrity of root tip cell membranes, and stimulate the generation of allelochemicals (Gupta et al. 2011). Fortunately, the contents of these endogenous pollutants in biochar are normally low. The negative effects of biochar may be limited, especially when considering that biochar is generally applied below 5%.

Additionally, it may not be wise to over-inhibit allelochemical release. Some allelochemicals have specific ecological functions and are effective in pollutant detoxification or beneficial microorganism colonization. The concentration range of allelochemicals for the beneficial impacts is still unknown because the literature mostly focused on the implications of allelochemicals, let alone the biochar addition strategies to ensure this beneficial concentration range. Extensive studies of the beneficial concentration ranges of allelochemicals will be useful for synthesizing or producing herbicides based on natural structures (Zhao et al. 2019).

5 Biochar modification to mitigate allelopathy in agriculture

Because of the diversity of biochar feedstock and high heterogeneity of its structures, biochar may not always be selective or effective enough to its targeted application goals. A modification can be designed for a targeted biochar application. Fortunately, biochar modification has been widely studied. Researchers have established a variety of biochar modification technologies, including physical, chemical, biological, and combined methods (Zhang et al. 2021b). For example, the surface area of biochar could be enlarged by acid modification, alkali, or CO_2 activations; the intensity of PFRs in biochar can be enhanced by loading metals or metal oxides, N-doping, or incorporation of functional groups; the magnetism can be imparted to biochar by loading magnetic metal oxides on biochar through co-precipitation or hydrothermal method; the relative content of surface functional groups can be manipulated by using oxidizers or reductants (Zhang et al. 2021b). As shown in Fig. 4, biochar can be modified for different applications, all aiming at alleviating allelopathy from different aspects.

5.1 Biochar modification to increase allelochemical immobilization or removal

Allelochemical removal includes both sorption and degradation. The enhanced sorption generally correlated with the increased specific surface area and pore volume. The targeted modification methods include steam activation and acid treatment. When the biochar is purged by high-temperature CO₂, it will be thermally altered with CO generation, which will increase the micropores. This method is cheap with high feasibility (Rajapaksha et al. 2016). Phosphoric acid is a commonly used modifier. Phosphoric acid is dehydrated during activation of biochar and forms polyphosphates or phosphates cross-linkers. They stay in the porous structure inside the biochar, preventing the pores from shrinking during the pyrolysis (Fierro et al. 2010). When the ash content of the biochar is washed, their internal micropores increase greatly. The increased surface area and porosity all contribute to the improved sorption capacity to allelochemicals.

As discussed earlier, the apparent sorption may not completely inhibit the bioavailability of allelochemicals because of the desorption. Therefore, the strong interaction between biochar and allelochemicals is essential for allelochemical immobilization. A modification method to introduce specifical functional groups, such as oxygencontaining functional groups, especially the carboxyl group, will help to enhance the formation of stable chemical bonds between the biochar and allelochemicals (such as covalent bonds and hydrogen bonds) (Xue et al. 2012).

The doping of biochar is also a promising way to improve their sorption and degradation for allelochemicals. For example, the N-doped biochar exhibited a high catalytic degradation ability for organics owing to the abundant unpaired electrons on N atoms (Chen et al. 2019; Wan et al. 2020); N-doped biochar also can enhance the sorption strength and affinity for sulfamethazine because it could provide both π electrons and unpaired electrons for sorption. Biochar loaded with positively charged inorganic nanoparticles can increase the surface charge of biochar and promote the electrostatic interactions and surface complexations between biochar and allelochemicals (Zhang et al. 2013).

Recent studies have also suggested that biochar is reactive and could mediate the degradation of organics. Some modifications could increase the redox potential of biochar. For example, the modification by borohydride could increase the phenolic hydroxyl group of biochar, and the loading of metals or phenolic compounds on biomass could increase the concentration of PFRs in the biochar (Fang et al. 2015a; Zhong et al. 2019). These modifications could accelerate the degradation of allelochemicals by biochar. However, this degradation of allelochemicals by biochar may not be complete mineralization. It is known that incomplete degradation of organic contaminants could be accompanied by the generation of even more toxic byproducts (Sun et al. 2020). This type of degradation evaluation is still absent in allelochemicalbiochar interaction studies.

If biochar is modified to benefit microbial colonization, the degradation of allelochemicals by microorganisms could also be enhanced. The biochar coated with multi-layer conductive structure MXene served as the cathode of microbial electrosynthesis (MES). This modification not only enhanced the conductivity of biochar but also enriched microorganisms (such as *Proteobacteria* and *Firmicutes*), which enhanced the chemical and biodegradation of allelochemicals (Tahir et al. 2021). More effectively, some specific allelochemical-degrading microorganisms could be inoculated and protected in biochar for field application.

5.2 Biochar modification to decrease allelochemical generation and release

The decreased allelochemical generation is generally related to favorable growth conditions for plants, such as decreased pollutant stress, increased nutrients, and a balanced microorganism system. According to the previous discussion, it will be much easier to develop modification methods if the purpose is certain. It would be more preferred to modify biochar with one method but with multiple advantages to improve plant growth conditions. For example, acid modification (phosphoric acid, nitric acid, or sulfuric acid) can mostly decompose aromatic substances and lignocellulose, and increase the specific surface area and porosity of biochar (Fierro et al. 2010). In order to achieve the purpose of reducing soil bulk density, increasing soil moisture content, enhancing soil porosity, and reducing the pressure caused by drought compaction on crop growth, phosphoric acid and nitric acid modification can increase P and N contents in biochar and provide a food source for microorganisms (Abd El-Mageed et al. 2021). Sulfuric acid modification can also improve the sorption capacity to Na⁺, reduce the content of soluble Na⁺ and salt in the soil, and alleviate the salt stress in soil (Duan et al. 2021). Acidic methanol modified biochar can appropriately increase the specific surface area of biochar, and increase the number of esters and hydroxyl groups, as well as the electron density of oxygen atoms and pH value (Jing et al. 2014). Therefore, the sorption performance of biochar for pollutants is significantly improved.

However, it should be noted that sometimes the modification may not always be beneficial to plant growth. For example, biochar is always washed after modification, and the content of dissolvable organic matter in the biochar will be reduced. The nutrient value of the biochar could be also reduced. In addition, it is possible that some modifying reagents are retained in the pores, which may pose negative effects on the growth of plants. Loading metal oxides on biochar can also cause certain environmental risks. Although the content of the loaded metal oxides is low, the release or dissolution of the metals may impart heavy metal pollutants into soil systems.

6 Conclusions and future perspectives

The accumulation of allelochemicals in soil may inhibit the germination and growth of plants, even reduce crop yields. Biochar could regulate the behavior and fate of allelochemicals according to the following three aspects: 1) it partly overcomes the allelopathy in soil by adsorbing and degrading allelochemicals; 2) it reduces the generation of allelochemicals by buffering environmental or ecological stress for plants; and 3) it stimulates the activity of soil organisms and promotes biodegradation of allelochemicals. Specifically modified biochar could be more effective in alleviating allelopathy because of their enhanced properties in sorption or degradation of allelochemicals, decreasing the generation of allelochemical by plants, and improving the soil conditions targeted, which are all beneficial in decreasing the accumulation of allelochemicals in the soil.

A thorough evaluation should be carefully carried out before the mass application of biochar for allelopathy control. For example, biochar provides habitats or refuges for both beneficial and harmful microorganisms. How to balance the effects on microorganisms in a long term? Another example is that sorption of allelochemicals by biochar leads to their enrichment on the biochar particles. Does this process increase the contact between allelochemicals and microorganisms? How to mitigate the by the application of modified biochar? The regulation of the secretion of plant allelochemicals is a complicated process. If biochar is to be used in solving allelopathy problems, the following studies need to be considered:

- a. Because allelochemical generation is determined by various factors, it is important to identify the contribution of different functions of biochar to alleviate allelopathy. The above-mentioned three pathways may be related to different biochar properties. Thus, targeted biochar production is highly dependent on our understanding of the relationship between biochar properties and its functions.
- b. Both allelochemicals and biochar are diverse in their types and properties. It may not be easy to establish a general standard to optimize their interactions. It is thus necessary to explore the interactions between different allelochemicals and biochar, summarizing the best combinations to immobilize or degrade alle-

lochemicals by biochar for a certain plant species. It should be noted that it is not possible to test all the combinations. Hence, the integrated investigations of computational simulations, quantitative structure–activity relationship (QSAR) modeling, and meta-analysis might be an efficient way to reduce the experimental workload.

- c. The application of modified biochar can be an efficient way to mitigate the generation and release of allelochemicals. The targeted design and preparation of modified biochar should be considered to effectively regulate the concentration range of allelochemicals for beneficial impacts. Nanosization could be a promising technique for biochar modification, but the interactions between nano-biochar and soil animals, allelochemicals, soil microbes, and soil organic matter should be explored. The balance between cost and efficiency of field application of nano-biochar must be accounted.
- d. The possible negative effects of biochar or modified biochar should be taken into full consideration before and during application. The amount and type of biochar, the growth phase of plants, and soil characteristics should all be considered to establish a sustainable strategy for biochar application.
- e. The stability of biochar or modified biochar in soil should be paid more attention. The decomposition mediated by soil microorganisms and physiochemical aging may change the surface properties and thus the function of biochar, but the pros and cons of plant growth and allelochemical generation are not fully understood.

In summary, the application of biochar in alleviating allelopathy may have significant environmental and ecological benefits. Considering that biochar could be produced using the residual biomass from farmland, a sustainable agroecosystem could be established if biochar functions in relation to the behavior and activity of allelochemicals in soil are fully understood.

Abbreviations

BC: Biochar; ATP: Adenosine triphosphate; OFGs: Oxygen functional groups; π - π EDA: π - π electron-donor-acceptor; DFT: Density function theory; EPFRs: Environmental persistent free radicals; CEC: Cation exchange capacity; AM: Arbuscular mycorrhiza; VOCs: Volatile organic compounds; PAHs: Polycyclic aromatic hydrocarbons; MES: Microbial electrosynthesis; QSAR: Quantitative structure–activity relationship.

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Authors' contributions

Quan Chen: Investigation, writing-original draft, writing-review & editing, funding acquisition; Pinyao Lan: Investigation, writing-original draft; Min Wu:

Investigation, writing-original draft, funding acquisition; Meng Lu: Investigation; Bo Pan: writing-original draft, writing-review & editing, funding acquisition; Baoshan Xing: writing-original draft, writing-review & editing, funding acquisition. All authors read and approved the final manuscript.

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Declarations

Ethics approval and consent to participate Agree.

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Agree.

Competing interests

Bo Pan and Baoshan Xing are the editorial board member for Carbon Research and was not involved in the editorial review, or the decision to publish, this article. All authors declare that there are no competing interests.

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