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Biochar remediation of soil: linking biochar production with function in heavy metal contaminated soils

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Abstract: The focus of this study is on the soil physicochemical, biological, and microbiological processes altered by biochar application to heavy metal (HM) contaminated soils. The aim is to highlight agronomical and environmental issues by which the restorative capacity of biochar might be developed. Literature shows biochar can induce soil remediation, however, it is unclear how soil processes are linked mechanistically to biochar production and if these processes can be manipulated to enhance soil remediation. The literature often fails to contribute to an improved understanding of the mechanisms by which biochar alters soil function. It is clear that factors such as biochar feedstock, pyrolysis conditions, application rate, and soil type are determinants in biochar soil functionality. These factors are developed to enhance our insight into production routes and the benefits of biochar in HM soil remediation. Despite a large number of studies of biochar in soils, there is little understanding of long-term effects, this is particularly true with respect to the use and need for reapplication in soil remediation.

Keywords: adsorption; toxic element; soil contamination; soil amelioration; soil pollutants; biochar function

The overexploitation and inappropriate use of soil has, and unless addressed, will continue to negatively impact on achieving required food production levels and the sustaining of biodiversity. The contamination of soil by inorganic chemicals is a major issue, particularly with respect to heavy metals (HMs) and their toxic impact (Bolan et al. 2014, Wang et al. 2018a), on agricultural productivity and quality, and health (Adriano et al. 2004, Park et al. 2011, Bolan et al. 2014).

The deposition of HMs in soil occurs primarily, but not exclusively, due to leaching and volatilisation (Boening 2000, Porter et al. 2004, Lu et al. 2011). HM toxicity has critical health risks, often due to environmental bioaccumulation in food-chains (Abbas et al. 2018). The resistance of HMs to organic detoxification combined with bioaccumulation can yield long half-lives in soil (Bolan et al. 2014, Bastami et al. 2015). Some HMs, such as Pb, Cu, Cd and Zn exist at high concentrations in soils and sediments globally; affecting large areas of agricultural land. For example, in China, some 26 million ha (around 19% of the total agricultural land) is contaminated with HMs (O'Connor et al. 2018b). It should be remembered, however, that some of the HMs in the soil, such as Co, Cu, Fe, Mn, Mo, Ni and Zn are essential for the plants' normal growth and metabolism until their concentration becomes higher than the optimum values (Singh and Kalamdhad 2011); and that sorption capacity of biochar may be a disadvantage in such cases when micronutrients are already deficient in the soil.

Biochar is produced by pyrolysis (under oxygen-limited conditions) and is the result of thermochemical conversion of biomass to a carbon rich product. Global interest in soil applied biochar has focused on exploita-

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tion of its ability to provide long-term sequestration of atmospheric carbon, as pyrolysed carbon is recalcitrant and its potential for soil structural (physical and chemical) and functional (biological) remediation. The notion here is to promote, or reestablish soil health using biochar, particularly with respect to chemical remediation, and/or structurally and chemically altering the capacity of biochar to absorb pollutants (Abbas et al. 2018). Recent reports provide evidence that biochar reduces the impacts of a range of soil and water contaminants; including inorganic (Kim et al. 2015, Kończak and Oleszczuk 2018, Sui et al. 2018), organic (Song et al. 2016, Zhang et al. 2018c), and radioactive (Zhang et al. 2018a) contaminants. In particular, biochar is now being used to remediate HM mine soil contamination (Gwenzi et al. 2015). Biochars have been shown to immobilise HMs and reduce their phytoavailability (Kim et al. 2015, Shen et al. 2016, 2017), and uptake (Xu et al. 2016). A range of changes in physicochemical, biological and microbiological processes have been induced by biochar promoting revegetation (Coomes and Miltner 2017, Igalavithana et al. 2017). These changes are linked to reductions in HM transfer and accumulation in crops (Ahmad et al. 2014, Moreno-Barriga et al. 2017).

Less descriptive work has examined the physical and chemical nature of biochar to understand the mechanisms of amelioration and revegetation of HM contaminated soil. These studies, however, in general, provide little insight into the effects of biochar on factors such as soil biology, which could support a more critical and mechanistic informed use of biochar. Suggested remedial benefits from biochar often comes from short-term laboratory, or greenhouse experiments and fails to provide insight into long-term soil changes and biochar soil resilience. While some findings indicate adverse effects on native soil biological communities, e.g. a decrease of microbial diversity (Cheng et al. 2018a), increases in earthworm mortality (Pukalchik et al. 2018), and decreased arthropod reproduction (Kończak and Oleszczuk 2018). These impacts appear linked to high biochar application rates for which there is often limited financial justification.

Review aims

This review focuses on considering the factors which are key determinants of biochar functionality in soil and to develop a framework of understanding to provide insight into the links between production methods and impacts on HM soil remediation, *via* an understanding of their mode of action. Specific focus will be on soil-biochar processes and their HM remedial action through knowledge of soil physicochemical and biological changes. The implications of this research will be linked to biochar production and potential knowledge gaps identified. A focus has been given to articles published over the last ten years. To accomplish this part of the review compares referenced biochar production processes with remediation impacts, within a tabular format.

SOIL-BIOCHAR PRODUCTION PROCESSES AND REMEDIAL ACTION IN HEAVY METAL CONTAMINATED SOILS

Biochar, has a carbonaceous composition (Krull 2012) with an alkaline pH, high base cation content, cation exchange capacity (CEC), and surface area, along with an array of functional aromatic hydrocarbon groups (Rinklebe et al. 2016, Rizwan et al. 2016). The highly porous structure of many biochars contains extractable humic and fulvic acids in significant amounts (Trompowsky et al. 2005). It also has a high degree of chemical and microbiological stability (recalcitrance) which suggests long soil residence (10 s to 1 000 s of years), depending on the environment (Cheng et al. 2008). Biochars heterogeneous composition with hydrophilic, hydrophobic, basic and acidic surface moieties enables reactivity with a wide range of soil substances (Atkinson et al. 2010). This divergence of compositional chemistry makes biochar a potential adsorbent of a wide range of soil contaminants, including toxins such as HMs (Fellet et al. 2011, Abdelhafez et al. 2014, Ahmad et al. 2014, Brennan et al. 2014, Zhu et al. 2015, Zhao et al. 2016, Soudek et al. 2017, Bogusz et al. 2017, Cao et al. 2018).

Experimental studies of soil amelioration show a wide range of variable impacts, with much depending on the biochar production processes and soil type. Critical analyses of these studies have provided some clarity of the general underlying mechanisms (Atkinson et al. 2010). This work suggests that there is a strong potential, given a more mechanistic understanding of how biochar works, that the entire production process can be developed to achieve biochars with specific desired functions/performance – a "designer", or "smart" biochar. However, using biochar for ameliorating HM contaminated soil is still at an early stage (O'Connor et al. 2018b). Biochar can promote amelioration of degraded soils, particularly HM contaminated soils

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through, (1) abiotic: directly by altering soil physicochemical properties, including HM adsorption (2) biotic: by indirectly changing soil microbiological and biological diversity and function. The inherent characteristics of biochar and its capacity for adsorption and immobilisation of toxic substances in soil, from selected studies, are presented along with the additional benefits, for some soil types, *via* improved soil physical and chemical properties in Table 1.

Soil physicochemical properties

Differences in biochar chemistry and structure increase the variability of its behaviour in the soil. Changes are manifest through physicochemical properties (bulk density, aggregate stability and available water), which can increase plant growth and yield (Atkinson et al. 2010, Abdelhafez et al. 2014). Biochar also increases soil alkalisation, CEC and electrical conductivity (EC) values and nutritional status (Herath et al. 2014). Although biochar itself rarely contains high concentrations of available nutrients (with few exceptions, e.g. K). Biochar's value in plant nutrition is attributed to its an ability to retain soil solution plant nutrients (e.g. fertiliser additions) and thereby reduce leaching and volatilisation losses, resulting in increased nutrient availability and uptake, and higher crop yields (Chan and Xu 2009). For HM contaminated soils, however, the likely biochar positive impact is *via* its capacity to reduce HM mobility, bioavailability and dispersal (Zhao et al. 2016, Zhang et al. 2017, Cao et al. 2018). Improvements in soil physical properties depend on its physical nature (Fryda and Visser 2015). Differences in biochar pore size and porosity are important attributes in determining the adsorption of HMs. Biochar pyrolysis temperature determines its structural and surface properties (such as, pore size, pore volume and surface area, which generally increases with pyrolysis temperature) and these influence its different HM adsorption potential (Méndez et al. 2013, Yuan et al. 2013, Chen et al. 2014). Chen et al. (2014) in their study with biochar produced from sewage sludge found that 900 °C pyrolysis temperature was optimal for Cd adsorption by surface precipitation and ion-exchange; linking pyrolysis temperature with surface properties of biochar and their resulting HM adsorption behaviours.

Soil porosity is also improved directly by the biochar's micro-structural porosity along with its particulate macro-structural improvements of soil aggregate stability (Hardie et al. 2014). Clay soils are improved by biochar-induced increased aggregate stability, water-holding capacity (WHC) and pore-size distribution (Sun and Lu 2014), while sandy soils show improved physical properties such as bulk density and total porosity (Głąb et al. 2016) and plant available water (Atkinson 2018). The relationship between these factors is however complex due to their impact capacity often being more than additive.

Soil aggregate stability and water retention. Addition of biochar to soil can affect a wide range of soil physical characteristics, such as the bulk density, surface area, particle size distribution, particle density and pore size distribution and these all influence soil, texture, porosity and homogeneity; which in combination, determine soil aggregate stability and soil water holding capacity (Amonette and Joseph 2012, Abdelhafez et al. 2014, Atkinson 2018). In addition, biochar also effects soil quality by influencing its response to water and temperature changes, aggregation, permeability and swelling-shrinking dynamics (Lehmann and Joseph 2012). These factors can all influence plant growth *via* changes in ease of root penetration and nutrient availability and the oxygenation of the rooting zone. A number of soil chemical and biological properties, related to soil fertility and HM adsorption, such as soil pH, CEC and micro-habitats for microbes, are linked to changes in soil physical characteristics induced by biochar (Brady and Weil 2016). Soil physical conditions provide a key element in influencing multiple soil-biochar processes which influence amelioration and adsorption of HMs. HM contaminated soils treated with biochar were suggested to benefit through soil water conservation and increased plant available water during revegetating degraded sandy soils in arid regions (Atkinson et al. 2010).

Soil chemical properties and organic matter change. Addition of biochar to the HM contaminated soil frequently increases its pH (Fellet et al. 2011, Abdelhafez et al. 2014, Herath et al. 2014, Rees et al. 2014, Yang et al. 2016, Ali et al. 2017, Igalavithana et al. 2017, Seneviratne et al. 2017, Soudek et al. 2017). This is primarily due to the differences in feedstock which produce different amount of ash which is alkaline. The amount of ash produced is also influenced by the method of pyrolysis, where lower maximal production temperatures produce more acidic biochars. Biochars produced from feedstocks high in minerals, pyrolysed at high temperatures, with a high ash proportion, result in soil alkalisation due to (Cao and Harris 2010, Lehmann et al. 2011). Generally, by adding biochar and raising the soil pH, the competi-

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Table 1. The soil-biochar processes and their remedial outcomes in heavy metal contaminated soils derived from some selected recent studies

Experiment type and heavy metal (HM) species	Biochar type and pyrolysis condition	Research findings: processes and remedial outcomes of biochar identified or observed or presumed	Reference
3-year field study of soil with biochar under wheat cropping system (Cd, Pb)	wheat straw (485 °C)	δ fluxes in soil pH caused by redox resulting from seasonal drought and flood cycles δ fluxes in soil organic carbon (OC) content and its subsequent stabilisation resulting from seasonal drought and flood cycles δ lesser effect of biochar on reducing HM uptake in wet (flooding) than the dry (draught) soil conditions	Sui et al. (2018)
18-month field study of soil with biochar (Cd, Cu, Zn, Cr, Pb, Ni, Co)	willow (700 °C)	+ reduction of sewage sludge toxicity + increase of bacteria luminescence (*Vibrio fischeri*) with an increased rate of biochar application δ reduction of reproduction stimulation of arthropods with an increasing rate of biochar	Kończak and Oleszczuk (2018)
5-month field study of soil with biochar under sunflower cropping system (Pb, Cd, As, Zn)	lychee branches (500 °C)	+ enhanced HM-extraction effect of sunflower + accumulation of Pb, Cd, As, and Zn in the leaf and receptacle + stimulation of sunflower plant growth	Jun et al. (2020)
8-day germination and 8-week pot cultivation of wheat in soil with biochar and bacterial strains (*Pseudomonas japonica* and *Bacillus cereus*) (Cr)	wood chips (525 °C)	+ positive physicochemical changes of the soil + reduction of phytotoxic influences of Cr – reduction of α and β amylase activities + improved height of plant, production of biomass, germination of seed, protein, carbohydrate, and chlorophyll content of wheat (*Triticum aestivum* L.) in the biochar with bacteria experiment	Arshad et al. (2017)
60-day pot culture of soil with biochar, wood ash and different humic substances (Zn, Cu, Cd, Pb)	wood chips (700–900 °C)	δ restored quality of the multi-contaminated soil at relatively low doses biochar δ high mortality of earthworm in increased (5%) biochar treatment	Pukalchik et al. (2018)
6-week greenhouse pot cultivation of maize and ryegrass in soil with biochar, rice straw, and wheat straw (Cd, Pb)	bamboo wood (750 °C)	+ increase of soil pH, electrical conductivity (EC) and OC content + increase of soil alkalinity – decrease of soil alkali-hydrolysable N content – reduction of N availability + lower plant uptake of Cd and Pb + reduced Cd concentration in maize and ryegrass shoots ± no improvement in plant growth	Xu et al. (2016)
7-week pot cultivation of *Brassica juncea* in mine-polluted soil with biochar and pig manure compost (Cd, Cu, Zn, Pb)	bamboo wood	+ increase of soil pH and EC + immobilisation of Zn, Cu, Pb, and Cd + decrease of Zn, Cu, Pb, and Cd bioavailability + transformation into the geochemically stable fraction of the readily available fraction of HMs + reduced root uptake of Zn, Cu, Pb, and Cd by mustard (*Brassica juncea*) + enhanced chlorophyll (*a* and *b*) and carotenoid in mustard (*Brassica juncea*)	Ali et al. (2017)

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Table 1 to be continued

Experiment type and heavy metal (HM) species	Biochar type and pyrolysis condition	Research findings: processes and remedial outcomes of biochar identified or observed or presumed	Reference
Laboratory study of soil with biochar and sewage sludge mixture (Cd, Cu, Ni, Zn)	willow (600 °C)	+ enhancement of HM immobilisation in the order of Zn < Cd < Cu < Ni + reduced risk of contamination of the environment + reduced risk of migration of contaminants in the plants, water, and living organisms + reduced risk of HM bioavailability and contamination of the food-chain	Bogusz et al. (2017)
4-month greenhouse plot cultivation of lettuce in soil with biochar (Cd)	rice straw (500 °C)	+ increase of soil pH and organic matter OM content + decrease of exchangeable Cd fractions in the soil δ reduced Cd concentration in lettuce (*Lactuca sativa* L.) shoots in lightly polluted soils δ no or prompted Cd concentration in lettuce (*Lactuca sativa* L.) shoots in highly polluted soils	Zhang et al. (2017)
50-day study of soil with biochar in a composting system (Cd, Cu, Zn, Cr)	rice straw (500 °C)	+ changes in soil OM and water-soluble C content + changes in the C/N ratio + reduction of HM toxicity to microorganisms + improvement of bacterial community structure + a weakened contribution of temperature to community succession of microbes	Chen et al. (2017)
90-day incubation study of river sediment with biochar (Cd, Cu, Zn, Pb)	rice straw (600 °C)	+ improvement of the soil physicochemical properties + improvement of the soil pH + increase of soil OM content δ improvement of soil microbial community composition (influenced by biochar intensity and incubation time) δ improvement of enzymes activity (influenced by biochar intensity and incubation time) + succession of bacterial community δ decrease of the relative intensity of dominant bacterial species with high concentrations of biochar – possibility of the indigenous microbial community to be affected by biochar	Huang et al. (2017)
112-day greenhouse pot cultivation of Italian ryegrass in soil with biochar (Cd, Cu, Zn, Pb)	wheat straw (350–550 °C)	+ decrease of Cd, Cu, Zn and Pb concentrations + increase of C and plant nutrient availability + stimulation of Proteobacteria and Bacteroides + increase of catalase, phosphatase, and urease activities	Liu et al. (2016)

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Table 1 to be continued

Experiment type and heavy metal (HM) species	Biochar type and pyrolysis condition	Research findings: processes and remedial outcomes of biochar identified or observed or presumed	Reference
15-day incubation of soil from an experimental farm in Huajiachi Campus, Zhejiang University, China (Cd)	commercially produced straw biochar	+ increased shoot (98.27%) and root (85.96%) dry biomass + increased net photosynthesis (45.52%) + increased transpiration rate (161.34%) + reduced bioavailable Cd	Kamran et al. (2019)
15-day pot culture of biochar and soil (contaminated by mine tailings) (Cd, Pb, Tl, Zn)	orchard pruning (500 °C)	+ increase of soil water holding capacity (WHC) and nutrient retention capacity (NRC) + increase of soil pH and cation exchange capacity (CEC) + reduction of Cd, Pb, Tl, and Zn bioavailability + favoured establishment of green cover on mine wastes in a phytostabilisation process	Fellet et al. (2011)
49-day incubation study of soil with biochar (Cd, Pb)	macadamia nutshell (465 °C)	+ reduction of HM toxicity of the soil + increase of biomass C + increase of microbial respiration and C use efficiency + mitigation of biotoxicity of the soil microorganisms	Xu et al. (2018)
90-day greenhouse pot study of soil with biochar and compost under cucumber plantation (Cu, Zn)	empty palm fruit bunches (600 °C)	+ increase of soil WHC + reduction of nutrient and HM leaching + increase of soil CEC + increase of microbial biomass C, nitrogen (N) and phosphorus (P) + improved plant growth + reduced environmental risks in excessively fertilised vegetable soils	Cao et al. (2018)
8-week incubation and 30-day pot cultivation of lettuce in soil with biochar (Cd, Pb, Cu, Zn)	rice hull (500 °C)	+ immobilisation of HMs + increase of soil pH + decline in the phytoavailable metal pool – decline in available nutrients, such as N + decreased metals concentrations in lettuce (*Lactuca sativa* L.) tissue except for Cu δ decreased lettuce growth with increased biochar application	Kim et al. (2015)
3-month pot cultivation of tobacco in calcareous soil with biochar (Cd, Pb)	tobacco stalks	+ increase of soil OM total C, N, P, and K contents, and C/N ratio + increase of available K content with increasing biochar application rates + decrease of bioavailable HM concentrations with the increasing rate of biochar + increase of tolerance of microbes to HMs + increase of bacterial richness and diversity + increase of operational taxonomic units (*Adhaeribacter, Rhodoplanes, Candidatus Xiphinematobacter* and *Pseudoxanthomonas*) – decrease of certain species of bacteria (*Kaistobacter*, *Lacibacter* and *Pirellula*)	Cheng et al. (2018a)

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Table 1 to be continued

Experiment type and heavy metal (HM) species	Biochar type and pyrolysis condition	Research findings: processes and remedial outcomes of biochar identified or observed or presumed	Reference
30-day incubation study of soil with biochar (Pb)	sugarcane bagasse, orange peel (500 °C)	+ increase of soil aggregate stability and soil WHC + increase of soil pH, CEC, OM content, and N status + decrease of availability of labile Pb fractions + decrease of solubility of Pb to values lower than the toxic regulatory level	Abdelhafez et al. (2014)
45-day incubation study of soil with biochar (Pb)	vegetable waste, pine-cone (200 °C, 500 °C)	+ immobilisation of HMs + increase of soil pH and EC + increase of existing organic C degradation and supply of the readily available C substrates + improvement of microbial community composition and structure + increase of dehydrogenase activity	Igalavithana et al. (2017)
Greenhouse pot cultivation of rice in soil with biochar (Cd, Pb, Cu, Ni, Zn, Cr)	wine lees cereal waste (600 °C)	+ decrease of exchangeable HM contents of soil (Transformation of HM into a residual fraction) + increase of soil pH + decreased migration and reduced accumulation of HMs to the aboveground part of the paddy plant + reduced contents of HMs in rice grains	Zhu et al. (2015)
60-day incubation study of soil from a Cu-mining area of Riotinto, Spain (As, Cu, Co, Cr, Se, Pb)	manure waste (450 °C and 600 °C)	+ increased biomass of *Brassica napus* + reduced accumulation of heavy metals by *Brassica napus* + decreased amount of As, Cu, Co, Cr, Se and Pb in the soil	Gascó et al. (2019)
9-week greenhouse pot cultivation of tomato in soil with biochar (Ni, Cr, Mn)	bioenergy waste (900 °C)	+ decrease of bioavailable concentrations of Cr, Ni, and Mn in soil (surface sorption) + increase of soil pH, CEC, EC and total organic C (TOC) with increasing concentration of biochar δ maximum decreased bioaccumulation of Cr, Ni, and Mn in the tomato plants at 5% biochar rate δ highly favourable microbial growth at 2.5% and reduced growth at 5% biochar rate	Herath et al. (2014)
3-month greenhouse pot cultivation of mung bean in soil with biochar and bacterial strains (*Bradyrhizobium japonicum*) (Ni, Mn, Cr, Co)	bioenergy waste	+ increase of soil pH + increase of concentration of soil nutrients (N, P) + increase of microbial biomass C + gradual reduction of bioavailable fractions of HMs with the increased rate of biochar application + reduction of Cr and Mn mobility with biochar + reduction of bioavailable HM in the presence of bacteria δ reduced plant uptake of Ni with an increase of biochar application rate from 1% to 2.5% δ enhanced plant growth of mung bean (*Vigna radiata*) at 2.5% while retarded plant growth at 5% biochar application rate	Seneviratne et al. (2017)

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Table 1 to be continued

Experiment type and heavy metal (HM) species	Biochar type and pyrolysis condition	Research findings: processes and remedial outcomes of biochar identified or observed or presumed	Reference
Laboratory study of HM stress on sorghum seed germination with biochar (Cd, Pb, Cu)	ash tree, beech tree, rice husk, bamboo wood	+ increase of soil pH + reduction of the mobility of Cd, Cu, and Pb in the soil + reduction of Cd, Cu, and Pb toxicity in the soil + reduced abiotic stress on plants and seeds of sorghum (*Sorghum bicolor* L.)	Soudek et al. (2017)
3-week laboratory study of biochar for HM stabilisation in soil (Cd, Pb, Cu)	pine tree sawdust, switchgrass (Co-pyrolysis with phosphate at 500 °C)	+ stabilisation of HMs + reduction of HM bioavailability + greater C retention + slower P release + improved soil fertility	Zhao et al. (2016)
1-year incubation study of soil with biochar (Cd, Pb, Cu, Zn)	bamboo, rice straw (750 °C, 500 °C)	+ improvement of soil physicochemical properties + increase of soil pH and electrical conductivity + reduction of HM availability δ increase of enzyme (urease and catalase) activity (influenced by biochar type, rate, and particle size)	Yang et al. (2016)
21-day rhizobox cultivation of maize in soil with biochar (Cu)	pine woodchip, olive tree pruning (450 °C)	δ reduction of HM mobility and availability in the soil influenced by biochar type + reduced uptake of HM by plants + improved root traits of maize	Brennan et al. (2014)
1-week laboratory study of HM sorption kinetics in soil with biochar (Cd, Cu, Zn, Pb, Ni)	coniferous chips, hardwood chips (450 °C)	+ increase of soil pH + immobilisation of HM influenced by biochar particle size	Rees et al. (2014)
4-week greenhouse experiment of soil from a Zn mining area located in Vazante, State of Minas Gerais, Brazil (Cd, Pb, Zn)	sewage sludge (500 °C) wood (*Eucalyptus* sp.) (350 °C)	+ increased leachate and soil pH + reduced the concentration of bioavailable Cd, Pb and Zn concentration	Penido et al. (2019)
12-week incubation study of soil from an anonymous contaminated site in urban Victoria, Australia (Pb)	poultry litter and biosolids (300, 400 and 500 °C)	+ significantly reduced the concentration of bioavailable Pb + biochars were able to outperform phosphate amendments for Pb immobilization	Netherway et al. (2019)
24-month incubation of soil from a fallow field of the Federal University of Sergipe experimental station, Northeast Brazil	coconut husk, orange bagasse and sewage sludge (500 °C)	+ reduced the most available fractions of Cu – increased Cu associated with OM	Gonzaga et al. (2020)

Bullet description: + – positive outcome; – – negative outcome; ± – no outcome; δ – dependent outcome

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tion for sorption sites among H^+ and metal cations (M^{n+}) is reduced, which decreases the soil mobility and availability of HMs (Gomez-Eyles et al. 2013). However, a biochar reduction in HM mobilisation at high pH cannot be explained entirely by HM biochar surface sorption, as soil surface sorption also increases (Gomez-Eyles et al. 2013). The increase in soil pH is explained by a greater pH-dependent CEC with organic matter, oxyhydroxides and soil clay minerals. Precipitation of HMs as insoluble hydroxides, phosphates, carbonates, and other mineral species result from a raised biochar soil pH and is a common explanation for reductions in HM mobilisation rates (Gomez-Eyles et al. 2013). The relationship between soil pH and HM mobility is dependent on soil properties, quantity of applied biochar and the duration of its application (Fellet et al. 2011, Herath et al. 2014, Rees et al. 2014, Igalavithana et al. 2017, Seneviratne et al. 2017).

Biochar amendments frequently increase soil CEC (Fellet et al. 2011, Abdelhafez et al. 2014, Herath et al. 2014, Cao et al. 2018). The reason for this are the inherently high CEC of biochar. Increased surface area for adsorption of cations, or increased charge density per unit surface, or a combination of both leads to elevated biochar CECs. Post-production of biochar results in its oxygenation upon exposure to air (Cheng et al. 2006), creating oxygen-containing negatively charged functional groups (phenol, hydroxyl, carboxyl, and carbonyl groups) over the biochar's surface (Liang et al. 2006, Lee et al. 2010, Uchimiya et al. 2010, 2011). Biochar adsorption of HM ions induces the liberation of H ions (Uchimiya et al. 2010), and of Na, K, Ca, Mg, and S (Uchimiya et al. 2011) into the soil solution. The organic carbonaceous nature of biochar results in increased soil OM content, with evidence of a simple biochar organic matter dose-response (O'Connor et al. 2018b). Biochar induced increased soil OM aids the retention of available nutrients (Herath et al. 2014, Liu et al. 2016, Huang et al. 2017, Seneviratne et al. 2017, Cheng et al. 2018a). Biochar application can increase soil OM content and is rate dependent (Sui et al. 2018). The decomposition rate of soil OM, derived from biochar, is dependent on microbial activity and has been shown to be lower in HM contaminated soil relative non-contaminated soil. Cheng et al. (2018a) showed that an increase in soil microbial abundance and diversity, induced by a biochar increase in soil pH, brings about favourable changes in soil physicochemical properties with an increased supply of available nutrients.

Immobilisation of heavy metals. Biochar has been shown to immobilise and reduce the phytoavailability (Kim et al. 2015), and plant uptake of HMs (Xu et al. 2016), including Pb, Al, Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, and Tl (Fellet et al. 2011, Brennan et al. 2014, Herath et al. 2014, Zhu et al. 2015, Yang et al. 2016, Bogusz et al. 2017, Yu et al. 2019). Multiple mechanisms have been proposed to understand biochar's potential for soil remediation. These include surface sorption (Egene et al. 2018), electrostatic interaction, ion exchange (Kumarathilaka and Vithanage 2017), precipitation and chemical complexion (Abbas et al. 2018). For example, peat moss biochar reduces mobility and bioavailability of Cu, Cd, and Pb by the co-ordination of metal electrons to C=C (π-electron) bonds (Park et al. 2016). The presence of biochar functional groups, along with a binding affinity with HMs, reduces their bioavailability. For example, an oak wood biochar resulted in a > 66% decrease in Cd and Zn availability (Egene et al. 2018). Immobilisation was attributed to the higher oak biochar pH and lower DOC concentrations (due to biochar surface sorption). Similarly, bioenergy waste biochar reduced bioavailability of Ni (68–92%) and Mn (76–93%) (Kumarathilaka and Vithanage 2017). The immobilisation of these HMs was due to surface diffusion and electrostatic attractions. However not all biochars, derived from different feedstock show high reductions of HM mobility, for example Pb availability using coconut fibre biochar, only declined by around 20%, despite high biochar application rates (Li et al. 2019). Many of these experiments also recorded reduced mobility of HMs in the leachate which has important implications for reducing soil losses to aquatic ecosystem (Zhou et al. 2017). The reduced mobility mechanism was due to a biochar induced decline in the acid-soluble HM fraction. Biochar has also been used in soil phytoremediation, to compliment living plants acting as HMs absorption sinks. Alfalfa phytoremediation of contaminated soil was shown to decrease soil Cd at a rate around 90 g Cd/ha and was due to root exudates complexing Cd and reducing root Cd absorption (Zhang et al. 2019).

These experiments do not, however, show that biochar can be used to totally remediate a soil HM issues. This may be due, at least in part, to an inappropriate stoichiometric ratio between soil HM load and biochar dosage being achieved (Abdelhafez et al. 2014, Zhang et al. 2017). It is generally true, however, that higher rates of application provide greater surface area and more HM bonding sites (Seneviratne et al. 2017). There may also be a complex interaction and stability of HMs with soil particles and biochar and their impact on HM ion mobility and chemistry. There are suggestions that non-specific sorptions are reversible and HMs can be released to the soil solution (Campillo-Cora et al. 2020). Experiments with Cu, Pb, Ni and Zn show these elements have a higher retention when present singly compared to when present in combination, highlighting the importance of synergistic effects in HM mobility. It can, however, be concluded that biochar application alone does not determine soil HM mobility and bioavailability.

Soil biology

Microorganisms. In general terms, the interactions between microbes, HM and plants can be supported by considerable biochemical and molecular understanding (Ma et al. 2016). While soil applied biochar can further alter microbial community composition (Igalavithana et al. 2017), and increase diversity (Cheng et al. 2018a) and thus stimulate specific microbial processes, enhancing soil biochemical cycles through rhizospheric plant-bacterial interactions to increase nutrient uptake and crop productivity (Hayat et al. 2010). Biochar induced changes in plant growth regulating, or promoting, rhizobacteria (bacteria belonging to the groups; *Azospirillum*, *Enterobacter*, *Klebsiella* and *Pseudomonas*) enable direct changes in microbial ecology and function within the rhizosphere.

There is good evidence that biochar is colonised by arbuscular mycorrhizal fungi (AMF) which subsequently increased plant growth (Steiner et al. 2007). Similarly to AMF, biochar colonisation by other soil microbes is known to increase; in response to the porous physical structure of the biochar increasing microbial habitat niches (pores) (Atkinson et al. 2010, Brady and Weil 2016). A relatively short incubation study, with biochars from different feedstocks, showed those produced at higher temperatures (600 °C compared with 400 °C) had greater microbial abundance (bacteria and fungi). The reasons for this were due to the greater proportion of micro- and meso-pores providing a favourble micro-habitat (Zhang et al. 2018b).

Amending HM contaminated soils with biochar has shown increased microbial abundance which suggests increased HMs tolerance (Liu et al. 2016, Chen et al. 2017, Cheng et al. 2018a), with changes in microbial population size, composition and activity (Liu et al. 2016, Yang et al. 2016, Chen et al. 2017, Huang et al. 2017, Xu et al. 2018). A number of studies have shown biochar dosage influences soil microbial population and activity, at low concentrations of biochar (1%) increased the relative abundance of bacterial and fungal species, while at higher application rates (5%) abundance declined (Huang et al. 2017). Generally, high concentrations of soil HMs lead to detrimental impacts on microbiological function, most importantly, enzyme function (Yang et al. 2016, Huang et al. 2017). This is despite the fact that HMs, such as Fe, Mn, Ni, Co, Cu and Zn at low (non-toxic) concentrations, play crucial roles in cell production and regulation of particular enzymes (e.g. soil-borne pathogen and antibiotic resistance) (Bååth 1989, Azarbad et al. 2015).

Fauna. Earthworms and arthropods maintain different soil functions which influence plant growth (*via* changes in soil structure (increasing soil OM), aeration, water infiltration and nutrient cycling). Earthworm activity is important in soil turnover and mineralisation of soil OM and N (Bhadauria and Saxena 2009). Arthropods account for some 85% of the soil fauna species, and functional plant litter transformers and soil ecosystem engineers; modifying soil structure, mineral and OM and hydrology (Culliney 2013).

Generally, studies show the benefits of biochar, but there is evidence that at high dosages, despite immobilising HM, increase biochar application rates induce greater earthworm mortality (Pukalchik et al. 2018). Similarly, an increase arthropod reproduction (*Folsomia candida*) occurred at low biochar rates with a decline at higher rates in HM contaminated soil (Kończak and Oleszczuk 2018). The reasoning behind this was due to intestinal surface accumulation of the nutrient-absorbing biochar leading inhibited arthropod growth. Earthworms have been shown to improve biochar properties considerably through soil enrichment with extracellular enzymes involved in biogeochemical and bioremediation enzyme pathways (e.g. alkaline phosphatase, β-glucosidase, arylsulfatase, and carboxylesterase) (Sanchez-Hernandez 2018). The latter enzyme is known to inactivate several agrochemicals (e.g. organophosphorus and methyl carbamate pesticides) when using enriched biochar. Earthworms are also known to promote the abundance HM degrading soil microorganisms (Rodriguez-Campos et al. 2014, Morillo and Villaverde 2017). Generally, these studies are confined to organic soil pollutants (Castracani et al. 2015, Sanchez-Hernandez et al. 2019, Silvani et al. 2019). Despite very a limited number of studies recent Chinese

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patents have described remediation technologies for HM contaminated soils in combination with biochar and earthworms (Ma 2015, Cheng et al. 2018b).

Flora. When incorporated into soils, biochar can contribute to improved soil and crop nutrition, particularly in nutrient-deficient soils which HM soils are often (Chan and Xu 2009, Baronti et al. 2010, Nigram et al. 2019, Wang et al. 2020). Enhancements of plant growth, on HM contaminated soils, also provides the means by which HM contamination can be directly reduced (Kumar et al. 2018, 2020, Zhang et al. 2020b). The use of "remediating plants" provides a green technology which has a low-cost and potential for a more sustainable approach to soil remediation (Saxena et al. 2020, Wang et al. 2020). Extensive mechanistic knowledge exists regarding the utilisation of specific HM phytoremediation combinations which includes the contaminated media type (e.g. soil or water), the contaminant in question (the toxic element) and an appropriate remediating plant species (Kumar et al. 2018, Bian et al. 2020, Wang et al. 2020). The appropriateness of remediating plant species has also been determined mechanistically and links directly to process of phytoremediation mentioned above. These studies also include the development of a molecular understanding of plant HM tolerance mechanisms (Yang et al. 2015, Fischer et al. 2017, Peng et al. 2017).

Successful soil remediation depends on plant growth, development and subsequent successful reproduction, to minimise the effects of HMs, particularly with respect to annual plants (Wenzel 2009, Karami et al. 2011). For example, biochar application has been shown to reduce the phytotoxic effects of Cr, improve seed germination, and the amount of biomass produced and grain quality (Arshad et al. 2017). These improvements are frequently linked to measureable reductions in tissue HM concentrations, e.g. Cd in lettuce leaves (Zhang et al. 2017). The mode of action of the biochar was *via* an increase in the fraction of Cd bound to soil OM and the presence of its oxides and carbonates, thereby reducing Cd availability and potential plant uptake. More recent work shows that biochar production temperature can be used as factor in the development of biochars to promote phytoremediation (Zhang et al. 2020a). However, despite an understanding of the mechanisms of biochar action on soil HM and the many positive experimental effects of biochar application on plant growth in non-contaminated soils (Lehmann et al. 2011, Prendergast-Miller et al. 2014), we have very little insight into the longer-term effects of biochar on HM remediation of plant performance or soil HM availability.

IMPROVEMENT OF BIOCHAR'S EFFICACY FOR HEAVY METAL REMEDIATION

Recent biochar studies describe primarily two methods of adsorption by biochar (He et al. 2019a, b), firstly, by direct adsorption and secondly, by improving the soil's physicochemical properties (such as pH, CEC, mineral, and OM content) (Wang et al. 2021). The mechanisms involved in controlling the removal of HMs from contaminated soils by direct adsorption of biochar includes physical sorption, ion exchange, electrostatic interaction, precipitation, and complexation (Inyang et al. 2016). The surface of biochar possesses various functional groups, including hydroxyls, carbonyls, and carboxyls (Tan et al. 2015), and their abundance, as indicated by CEC, is the most important factor for regulating the sorption based HM stabilisation (Guo et al. 2020). Biochar exhibits its electrical charges depending on the pH and dissociation (protonation) of these functional groups. Many biochars having negatively charged surfaces, through electrostatic attractions, can sorb the HM cations (Inyang et al. 2016). The introduction of additional alkalinity followed by an elevation of soil pH by biochar provides another key mechanism for the precipitation of soil HMs. The hydroxyls react with the HM cations and precipitate as metal hydroxides (Guo et al. 2020). Additionally, the mineral components serving as supplementary adsorption sites, provide biochar with another property for controlling HM adsorption processes in the soil (Tan et al. 2015). The biochar properties controlling these mechanisms predominantly include ash and mineral content, aromaticity, surface structure, functional groups, and pH (Wang et al. 2018b). Moreover, the soil type, biochar amendment rate and its placement in the soil controls the overall efficacy of biochar for HM remediation (O'Connor et al. 2018b, Guo et al. 2020). Biochar's preparation condition also influence its physicochemical properties, which indirectly also control its HM immobilisation effects (Wang et al. 2021). The production parameters that were found to control these biochar properties include, pyrolysis temperature, heating rates, vapour residence time, biomass type and particle size (Sakhiya et al. 2020). Hence, recent biochar research directed at HM remediation highlights routes to improvement and modification of biochar's HM absorption efficacy through altering its production processes (Wang et al. 2019).

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Biochar modification

In recent years the most widely studied modification technologies of biochar include physical modification, chemical modification, impregnation with mineral oxides, and magnetic modification (Rajapaksha et al. 2016). Although the physical modification processes of biochar are in general simple and economically feasible. A conventional method of physical activation of biochar is "steam activation" conducted during the initial pyrolysis reactions. In this process, pyrolysis is conducted in two stages where during the second stage, the biochar in the pyrolysis chamber is subjected to limited gasification with steam. The resulting biochar is characterised by high surface area and improved carbonaceous structures (Rajapaksha et al. 2016). Gas purging of biochar (another physical modification process) with CO_2 at high temperature was found to increase biochar surface area and pore volume relative to unmodified ones (Xiong et al. 2013). Chemical modification, in general, is a heat treatment process (450–900 °C) of biochar with chemical activating reagents (Sakhiya et al. 2020). Studies show that chemical modification creates opportunities for biochar to chemically react with HMs more efficiently through (1) increased surface area and sorption sites; (2) more conducive surface to electrostatic attraction, surface complexation, and/or precipitation, and (3) specific surface functional groups for greater sorption affinity and stronger interactions (Rajapaksha et al. 2016). Multiple ways of chemical modification have been studied to improve biochar's efficacy for HM remediation in soil, including acid/base treatment and chemical oxidation, organic solvents treatment, functional groups modification, surfactant modification, and biochar coating (Rajapaksha et al. 2016). Corn straw biochar modified by Na_2S and KOH when applied in a $Hg^{(II)}$ contaminated soil showed increased adsorption capacity of $Hg^{(II)}$ by 77% and 32%, respectively (Tan et al. 2016a). Similarly, dairy manure biochar modified by NaOH when applied to a Pb and Cd contaminated soil showed increased adsorption capacity of Pb and Cd. For the two HMs the highest adsorption capacity was 176 and 68 mg/g, respectively (Chen et al. 2019). In another study, rice husk biochar modified by sulfur when applied to a Hg contaminated soil increased the adsorptive capacity of biochar by 73% (O'Connor et al. 2018a).

Biochar nano-composites

Combining/loading nano-material(s) with biochar to form biochar nano-composites is another innovative way of achieving higher biochar efficacy for HM removal (Tan et al. 2016b, Mandal et al. 2020, Pan et al. 2021). These biochar nano-composites exhibit improved physicochemical properties relative to standard biochar, such as pore properties, surface sites, functional groups (Tan et al. 2016b), and greater stability (Pan et al. 2021). "Smart" biochar nano-composites can be synthesised through selecting an appropriate feedstock and the nano-material(s). Depending on the loading/doping method of the nano-material in biochar, the technique of producing biochar nano-composites is either as a pre-treatment or as post-treatment (Pan et al. 2021). Different studies were carried out with biochar nano-composites to remediate HM (including As, Cd, Pb, and Hg) contaminated soils. A recent study of Fe-Mn modified biochar, in an As contaminated soil, showed significant changes in terms of soil pH, redox potential, and a reduction of As contamination (Lin et al. 2019). Although there have been many studies using biochar nano-composites for remediation of HM contaminated water, there are comparatively few studies for remediation of HM contaminated soils.

Use of biochar-microorganism synergism

Metal-immobilising bacteria are known to reduce metal uptake of plants (Cheng et al. 2020). Additionally, certain soil bacteria interact with HMs and reduce metal bioavailability and toxicity (Chen et al. 2016, Rizvi and Khan 2017). This has generated studies into their use of biochar with bacteria to enhance metal immobilisation in HM contaminated soils (Tu et al. 2020). Two strains of metal(loid)-resistant bacteria, *Ralstonia eutropha* Q2–8 and *Exiguobacterium aurantiacum* Q3–11 were shown to reduce the uptake of Cd and As in wheat (Wang et al. 2018c). In another recent study biochar +*Serratia liquefaciens* CL-1 reduced the Cd and Pb content in wheat grain and the soil rhizosphere compared to that of the biochar and the bacteria alone (Cheng et al. 2020). Elsewhere, biochar inoculated with *Pseudomonas* sp. NT-2 (5%) reduced soil Cd and Cu bioavailability (Tu et al. 2020).

However, the interactive mechanisms between microbes and biochar remain unclear (Tu et al. 2020). Applied soil microbiological studies using biochars need to understand the synergisms that could be exploited to use species specific cocktails and application rates alongside their likely effective field duration. Understanding the diversity and function-

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Table 2. A summary of the processes and outcomes of biochar use

Processes		Outcomes
Abiotic: Physicochemical changes of the soil	**Biotic:** Microbiological and biological changes of the soil	effects on plants, food-chain, and the environment
Positive changes	**Positive changes**	**Positive outcomes**
1. Stabilisation, and reduction of HM mobility, bioavailability, and decreased solubility of HMs to values lower than the toxic regulatory level.	1. Mitigation and increase of tolerance of HM biotoxicity of the soil microorganisms.	1. Reduced abiotic stress, improved soil fertility, seed germination, root traits, plant growth, and biomass production.
2. Increase of soil pH, CEC and EC values.	2. Improvement of microbial community composition and structure with an increase of operational taxonomic units, and a weakened influence of temperature to community succession of microbes.	2. Increased chlorophyll, protein, and carbohydrate in crops.
3. Increase of availability of plant nutrients, soil OM content, the supply of readily available C substrates, soil TOC, and N status.	3. Increase of bacterial richness and diversity.	3. Reduced root uptake (migration) of HMs to the aboveground parts of plants (i.e., shoots, leaves) and reduced concentrations of HMs (phytotoxicity) in the edible parts of the plant (i.e., fruits and grains).
4. Increase of the soil aggregate stability.	4. Increase of microbial respiration, microbial C use efficiency, microbial biomass C, N, and P.	4. Reduced HM contamination of the food-chain.
5. Increase of soil WHC, and reduction of nutrient and HM leaching.	5. Increase of enzymic activity.	5. Reduced leaching and risk of migration of HMs to the aquatic, biotic and abiotic environment.
Negative changes	**Negative changes**	**No or negative outcomes**
1. Increased mobility of certain HM in particular biochar type (indicating that albeit biochar application reduces HM mobility, some biochars may also increase it).	1. Decrease of the relative concentration certain species of bacteria (including the dominant species) at high biochar rates with the possibility of the indigenous microbial community to be affected by biochar application.	1. No improvement of plant growth, decreased plant growth with increased biochar application.
2. Declined available nutrients in increased biochar dosage condition (indicating that increased biochar dosage may sometimes be the reason for reduction of nutrient availability in the soil).	2. Increased mortality of earthworms at high biochar rate.	2. Increased HM uptake in high pollution condition.
3. Reduced availability of soil N in increased biochar dosage condition.	3. Decreased reproduction stimulation of arthropods with the increasing rate of biochar.	3. Lesser effect of biochar on reducing HM uptake in wet and flooding soil conditions than the dry and draught soil conditions.

HM – heavy metal; CEC – cation exchange capacity; EC – electrical conductivity; OM – organic matter; TOC – total organic matter; WHC – water-holding capacity

ing of these microbiological components in biochar applied soils, remains very crucial. The complementary role of soil microbes and biochar in improving plant growth in HMC soils, is little understood, and requires development using molecular screening tools to determine their diversity, abundance and functionality (Seneviratne et al. 2017).

SUMMARY

Biochar application induces changes in many soil factors (Atkinson 2010, 2017), this work however generally lacks mechanistic understanding. This limits predictive outcomes of biochar-soil interactions, particularly and importantly, over the long-term and this limits biochar use in HMC soil remediation. However, lab-based short-term studies do help in the design, direction and application of longer-term field experiments (Table 2).

There is an urgent necessity to acquire such data to develop efficacy and biochar production specificity before any likely commercialisation (Janus et al. 2015). The influence of biochar production processes and feedstocks provides the means to prediction

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and optimisation of biochar HM detoxification. The goal would be to exploit such knowledge to provide biochars designed to tackle specific HM immobilisation issues. The emphasis given to understanding mechanisms will enable biochar capacity to be developed, which is both specific, as well, an innovative in regard to biochar purpose, i.e. "smart biochars" and biochar plus soil conditioners (with microorganisms). However, a precautionary approach is needed to understand the trade-offs in the biochar supply chain, i.e. production, feedstock selection and soil performance over time. This should also include a biochar's potentially unwanted contaminants (e.g. polycyclic aromatic hydrocarbons, polychlorinated dioxins and furans). Contaminants such as these may explain the negative effects of biochar on soil biota. Attention to the study of soil biota population dynamics and functionality, particularly in regard to specific HMs and their bioavailability's is required. The recalcitrant nature of biochar is a primary benefit to its use in soil improvement, but the changes to soil structure and function, both short- and long-term, need to be understood in relation to the amount of biochar applied and the requirement for reapplication. This is particularly important with respect to phytoremediation where initial revegetation requires sustaining over longer periods and where soil HM pollutants are required to remain immobilised. Understanding biochar field performance requires knowledge of HM adsorption/bioavailability and retention capacity in relation to changes in soil environmental factors over time. The impacts of environmental change and their potential to alter the capacity of biochar to influence soil remediation need to be incorporated into the development of a biochar.

CONCLUSIONS

Incorporation of biochar into HM contaminated soils is a relatively novel concept for remediation, restoration and revegetation and has yet to be carried out long-term. Biochar adsorbs a wide range of soil HMs and can provide an environmentally-friendly solution for remediation, with a low risk of causing short-term ecological hazards. Biochar application to contaminated soils has its greatest impact when it does not alter the inherent features of the soil, such as the biotic environment and its nutrient status. The descriptive literature shows the importance of taking into consideration a multi-functional ecosystem approach when assessing HM soil remediation impacts. Biochar benefits to contaminated soils are dependent on soil type, biochar feedstock and dosage, and the environment. The extent of remediation, combined with the dependencies above, implicates the need for engineering of "designer/smart biochar" exploiting existing knowledge of feedstock, pyrolysis conditions and application rate. Despite this there remains a considerable gap regarding the longer-term effects of field applied biochar in contaminated soils, particularly with respect to application rate and frequency, along with more complex issues of restoring and maintaining ecosystem functioning.

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