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REVIEW

Soil inorganic carbon sequestration through alkalinity regeneration using biologically induced weathering of rock powder and biochar

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HIGHLIGHTS

• Soil acidification caused severe losses of soil inorganic carbon stock worldwide.
• SIC losses could be mitigated via alkalinity regeneration approaches.
• Rock/mineral powder can supply substantial basic cations to soil to reduce acidification.
• Microorgnisms could be utilized to enhance weathering of rock/mineral powder.
• Biochar and bone biochar could reduce SIC losses via alkalinity regeneration.

GRAPHICAL ABSTRACT
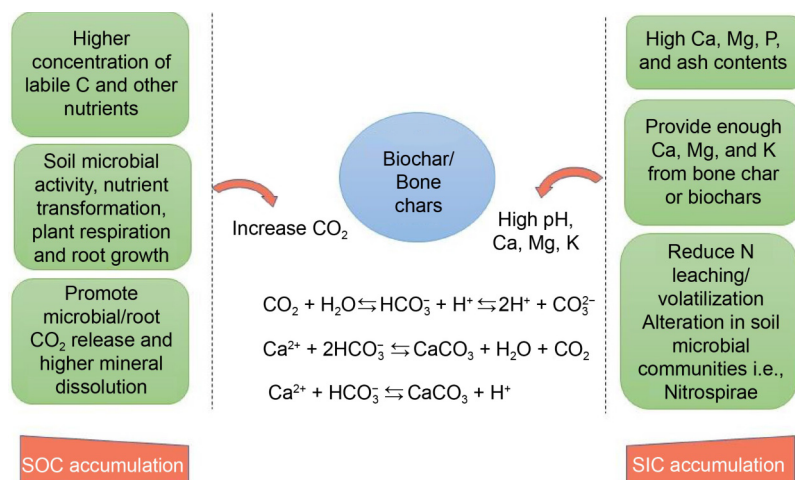
ABSTRACT

Soil inorganic carbon (SIC) accounts for about half of the C reserves worldwide and is considered more stable than soil organic carbon (SOC). However, soil acidification, driven mainly by nitrogen (N) fertilization can accelerate SIC losses, possibly leading to complete loss under continuous and intensive N fertilization. Carbonate-free soils are less fertile, productive, and more prone to erosion. Therefore, minimizing carbonate losses is essential for soil health and climate change mitigation. Rock/mineral residues or powder have been suggested as a cheaper source of amendments to increase soil alkalinity. However, slow mineral dissolution limits its efficient utilization. Soil microorganisms play a vital role in the weathering of rocks and their inoculation with mineral residues can enhance dissolution rates. Biochar is an alternative material for soil amendments, in particular, bone biochar (BBC) contains higher Ca and Mg that can induce even higher alkalinity. This review covers i) the contribution and mechanism of rock residues in alkalinity generation, ii) the role of biochar or BBC to soil alkalinity, and iii) the role of microbial inoculation for accelerating

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alkalinity generation through enhanced mineral dissolution. We conclude that using rock residues/BBC combined with microbial agents could mitigate soil acidification and SIC losses and also improve agricultural circularity.

1 Introduction

Soils are the largest carbon (C) sink (i.e., 2470 Pg C) in a terrestrial ecosystem (Eswaran et al., 2000) comprising soil inorganic C (SIC) and organic C (SOC) pools, thus playing a key role in the global C cycle and climate change (Lal et al., 2021a). Microbial biomass, rhizodeposits, and plant litter are considered to be the main sources contributing to the formation of SOC (1500 Pg C/100 cm depth, (IPCC, 2007)). Agroecosystems soils are a major source of CO_2 (Lal et al., 2021b) as the SOC is highly influenced by the agricultural practices (i.e., fertilization and tillage) and land-use change (Ruddiman, 2003; Chai et al., 2019). Due to the increasing level of CO_2 release to the atmosphere, previous studies have mainly focused on the management of SOC losses, while SIC has been studied less (Haddaway et al., 2017; Merante et al., 2017; Amundson and Biardeau, 2018; Lal, 2016, 2020). Consequently, more focus has been given to SOC as a climate mitigation strategy, and much less attention has been paid to SIC (Raza et al., 2021). Compared to SOC, SIC accounts for a dominant C share of 54% worldwide (exceeding 2300 Gt C in the top 200 cm) soil carbon, which is almost equivalent to the SOC reservoirs (2400 Gt C in the top 200 cm) (Zamanian et al., 2018). Globally, the SIC reservoir is mainly found in arid/semi-arid regions, together comprising 90% of the total SIC reserve (Filippi et al., 2020). Compared to SOC, SIC is generally considered as an inert C pool and assumed less sensitive to agricultural practices and more stable within the anthropogenic life timescale (Sanderman, 2012). Though, earlier studies have described those intensive agricultural activities (especially fertilization) have accelerated SIC losses worldwide (Raza et al., 2021).

Soil acidification (Raza et al., 2020), land-use change, water flow, and recharge cause a reduction in SIC reserves (Kim et al., 2020; Raza et al., 2020; Raza et al., 2021; Song et al., 2021). Even deep SIC reserves (up to 7 m belowground) that are generally considered to be stable for millennia are prone to losses via agricultural activities (Raza et al., 2021). These SIC losses predominantly take place as CO_2 emissions, mainly through N-fertilization-induced acidity causing $CaCO_3$ dissolution (Zamanian et al., 2018). For example, 12 years of urea application (i.e., 100 kg N ha^{-1} yr^{-1}) resulted in a depletion of $CaCO_3$ of 46–95 kg ha^{-1} yr^{-1} (Conyers et al., 1996). Therefore, N fertilization decreases SIC reserves (Dalal et al., 2005), increases atmospheric CO_2 emissions and eventually affects the global C cycle and global warming (Huang et al., 2015). Similarly, a study by Zamanian et al. (2018) also revealed that N fertilization-induced acidity (i.e., the release of H$^+$) causes dissolution of $CaCO_3$ and subsequently higher CO_2 emissions from the soil (i.e., 7.48 Tg C yr^{-1}). They also predicted that this higher continuous release of CO_2 will remain constant if N fertilization continues to be applied at a constant high rate. Apart from accelerating climate change, SIC losses decrease soil buffering capacity, nutrient availability, and microbial activity, eventually leading to the formation of acidic soils with reduced agricultural productivity (Rousk et al., 2010; Raza et al., 2020). $CaCO_3$ in the soil ensures a sustainable environment for biochemical processes, microbial functions, and ultimately plant growth.

Liming and manure fertilization are important strategies to reduce soil acidification. Both offer short-term increments in soil pH (Ge et al., 2018; Mkhonza et al., 2020). Liming during acidity neutralization releases CO_2 emissions, and such emissions are estimated at 273 × Tg CO_2-C annually (Zamanian et al., 2018). Biochar is a promising strategy to reduce acidification due to its dynamic properties: high alkalinity, ash contents, surface functional groups (i.e., adsorption of NH_3 and NH_4^+), and the small particle size provides additional sites for the adsorption of NH_4^+ and NO_3^- (Dai et al., 2017; Shi et al., 2019; Azeem et al., 2020). A few studies have also documented that biochar can enhance SIC stock in field trials (Dong et al., 2019a; Shi et al., 2021). However, further long-term field experiments are necessary to determine the potential of biochar for enhancing SIC reserves, particularly in acid soils. In addition, SIC stock can be reversed while applying Ca^{2+} and Mg^{2+} and by utilizing those feedstocks that contain a higher proportion of these cations (Ca^{2+} and Mg^{2+}). For example, biochar production from bone feedstock (BBC) can provide higher Ca^{2+} and Mg^{2+} contents with an additional significant proportion of C (organic and inorganic). Previously, pyrolyzed bone has been amended to enhance soil pH and EC (Siebers 2013; Siebers et al., 2014; Azeem et al., 2021a,2021b). Though, to the best of our knowledge, no study has yet been conducted to elucidate the effect of BBC on the SIC content in the soil.

In addition, rock/mineral residues from local reserves are also a paramount source of fertilizers. Crushed rocks are abundant in nutrients (e.g., Ca, Mg, P, K, and Fe), are less vulnerable to nutrient leaching, and are inexpensive compared to conventional fertilizer (Ramos et al., 2022). These rock materials, in particular silicate minerals (e.g., basalt and peridotite) with high Ca^{2+} and Mg^{2+}, could be utilized to sequester SIC in acidic soils. However, the low dissolution rate of minerals limits their agronomic and alkalinity regeneration efficacy as potential fertilizers. Conversely, the solubility of these minerals in the soil relies on microbial or plant actions to accelerate the weathering process (Ribeiro et al., 2020; Ramos et al., 2022). The release of nutrients during the weathering process supplies essential nutrients to

plants after being released into the soil. The utilization of responsible microbes could be a cost-effective method, not only to enhance the biological weathering of added rock/mineral powders, but also to provide indigenous nutrients to the plants.

In this review, we discuss the status of SIC stocks, the role of soil acidification in the losses of SIC reservoirs, and potential strategies to overcome these losses using biochar, BBC, and rock residues. The objective of this review is to determine the potential of rock residues and biochar, in particular BBC with enhanced biological weathering using soil microbes. The possible solutions to reduce soil acidification with enhanced biological weathering by supplying Ca^{2+}, Mg^{2+}, K^+, and P and other nutrients from crushed rocks or BBC are discussed regarding the mitigation of soil acidification and carbon sequestration.

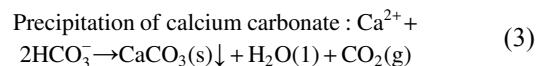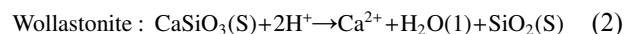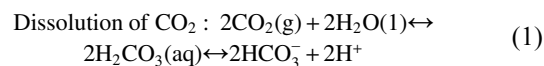## 2 Global acceleration of inorganic carbon losses

During the last six decades, anthropogenic N additions to soils via N-based fertilization (i.e., 12 to 108 Tg yr⁻¹), manure, and biological N fixation (i.e., 6 to 10 Tg N yr⁻¹) have increased twofold compared to pre-industrial natural N amounts (Guo et al., 2010; Zamanian et al., 2018). More than half of this added N is not utilized by plants and eventually causes soil acidification and other environmental degradation worldwide (Zamanian et al., 2018; Raza et al., 2021). Zamanian et al. (2021) estimated an irrecoverable loss of CO_2 to the atmosphere of 0.41 Gt C over the past 50 years and predicted that a further 0.72 Gt C will be liberated until 2050. Similarly, continuous N fertilization for 80−90 years has resulted in the complete removal of carbonate stocks in one of the world's longest fields trials conducted at Rothamsted research station UK (started in 1843). Likewise, a continuous overapplication of N addition to Chinese croplands over a period of 40 years also generated carbonate-free soils on about 7 million hectares, and this area is expected to expand to 30 million ha in 2100 (Raza et al., 2020; Song et al., 2021). This extensive depletion of soil carbonates globally leads to additional ecological problems, largely associated with the drop of soil functions such as lower soil organic matter (SOM) stability and reduced net primary production (Rowley et al., 2020). The loss of $CaCO_3$ decreases the soil's buffering capacity and promotes the basic cations leaching including Ca^{2+}, Mg^{2+}, K^+, and Na^+ (Lucas et al., 2011). Loss of $CaCO_3$ results in a reduction of soil pH that affects nutrient availability to plants, soil microbial function/activity, community composition, and structure (Rousk et al., 2010). Lower carbonates in soil and associated nutrients show detrimental effects on plant functions, such as lower foliar Ca^{2+} and Mg^{2+} contents resulting in a decrease in the photosynthetic pigment contents, capacity, and rates (Bethers et al., 2009). This limits the ability of plants to survive under various abiotic stresses including oxidative stress (Guo et al., 2016), high temperatures (Fahad et al., 2017), mechanical injury (Vega-Muñoz et al., 2020), and drought (Hosseini et al., 2019). This, in turn, leads to the depletion of SOC, accelerated erosion, and thus rapid losses of other irretrievable SOC stocks. $CaCO_3$ dissolution has several consequences for soil physicochemical and biological properties, and productivity.

3 Rock residues and biochar: potential ways to enhance SIC

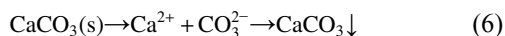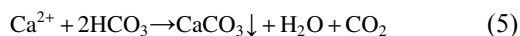### 3.1 Regeneration of alkalinity through rocks/minerals addition

Mineral carbonation using silicate rocks weathering can transform atmospheric CO_2 into carbonate minerals (Benson and Cole, 2008). Mineral carbonation is considered the most stable method of sequestering CO_2, which refers to the formation of pedogenic carbonates upon the reaction of atmospheric CO_2 with silicate minerals containing higher Ca^{2+}/Mg^{2+} contents (Benson and Cole, 2008; Snæbjörnsdóttir et al., 2020). Atmospheric CO_2 incorporation into the soil is regulated by the two main natural pathways: (i) via photosynthetic activity and SOM cycling, and (ii) weathering or mineralization of alkaline minerals. The first pathway mainly affects the SOC pool, while the second pathway controls the SIC stock. Possible inorganic rock materials for enhanced biological weathering include silicates of Ca^{2+} and Mg^{2+} (such as olivine, serpentine, and pyroxene minerals) with global availability and higher reactivity with CO_2 (Haque et al., 2019). Addition of fine crushed/powdered silicate rocks to the soil upon weathering releases alkaline metal cations, which can increase the pH and buffering capacity of soils (Ng et al., 2022). With the passage of time and the action of biotic/abiotic factors, these cations can gradually react with carbonate anions to form pedogenic carbonates (Equation (Eq.) (1)). CO_2 sequestration via wollastonite weathering occurs following the pathway shown below (Equations (1)−(3); Hangx and Spiers, 2009):

$$\text{Dissolution of } CO_2 : 2CO_2(g) + 2H_2O(1) \leftrightarrow 2H_2CO_3(aq) \leftrightarrow 2HCO_3^- + 2H^+ \tag{1}$$

$$\text{Wollastonite}: CaSiO_3(S) + 2H^+ \rightarrow Ca^{2+} + H_2O(1) + SiO_2(S) \tag{2}$$

$$\text{Precipitation of calcium carbonate}: Ca^{2+} + 2HCO_3^- \rightarrow CaCO_3(s) \downarrow + H_2O(1) + CO_2(g) \tag{3}$$

Although the rock powders are mostly studied as a potential rock fertilizer, their role in the formation of associated carbonates is rarely studied (Jones et al., 2020). In fact, silicate minerals are rich in Ca^{2+} and Mg^{2+} contents but their presence hinders the disintegration of minerals and release of nutrients due to the common-ion effect, for example higher contents of Ca^{2+} or Mg^{2+} in the soil solution

limits the further dissolution of Ca^{2+}/Mg^{2+} containing minerals. In a batch experiment, Haque et al. (2020) found that an optimum wollastonite dosage (1−10 wt.%) can potentially sequester 0.08 kg of CO_2 m^{-2} month^{-1} and made an assumption that wollastonite mineral application (1 mm-equivalent wollastonite layer) at the rate of 29 tons per hectare (up to 100-m depth) to agricultural fields could sequester stoichiometrically 11 tCO_2 ha^{-1} yr^{-1} (Haque, 2019).

The carbonation reaction is dependent on the presence of divalent cations such as Ca^{2+}, or Mg^{2+} as found in mafic (such as peridotite) and ultramafic (like basalt) rock, which are favorable for mineral-CO_2 sequestration (Benson and Cole, 2008; Kelemen and Matter, 2008; Matter et al., 2016). CO_2 release from soil microbial/root respiration or SOM mineralization on reaction with water initiates the generation of a proton and carbonates/bicarbonates (Eq. (4)). The dissolution of mineral cations upon reaction with bicarbonates makes pedogenic carbonates (Eq. (5)). However, this is a reversible reaction and higher CO_2 production always needs cations (Eq. (6)) to balance the system, or an even higher quantity is needed to sequester pedogenic carbonates in soils,

$$CO_2 + H_2O \rightarrow H_2CO_3 + HCO_3^- + H^+ \tag{4}$$

$$Ca^{2+} + 2HCO_3 \rightarrow CaCO_3\downarrow + H_2O + CO_2 \tag{5}$$

$$CaCO_3(s) \rightarrow Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3\downarrow \tag{6}$$
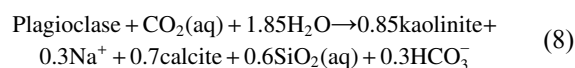
Silicate minerals are abundant on the earth's surface; some are exposed in continental settings and others lie beneath the earth's ocean. In particular, basaltic rocks are considered as the most favorable cation-rich silicates with their porous and permeable structure making them favorable for effectively capturing CO_2 (Snæbjörnsdóttir et al., 2020). The dissolution of basalt is enhanced under high CO_2 concentration and the availability of water. During basalt-CO_2-water interactions, the dissolved CO_2 will lead to the formation of plagioclase (e.g., anorthite; $CaAl_2Si_2O_8$ and anorthite; $Ca_{0.7}Na_{0.3}A_{l1}.7Si_{2.3}O_8$) and other primary minerals such as olivine and clinopyroxene, and their dissolution will help to release Ca^{2+} which helps to regenerate alkalinity in acid soils following equation (Eq. (7)) (Tutolo et al., 2021):

$$\text{Anorthite (An70)} + 6.8H^+ \rightarrow + 1.7Al^{3+} + 0.3Na0.7Ca^{2+} + \\ 2.3SiO(aq) + 3.4H_2O \tag{7}$$
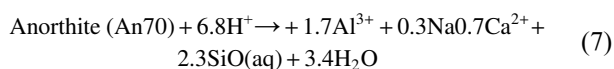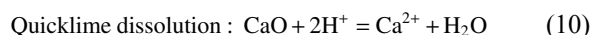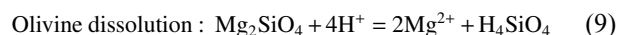
The weathering of primary silicate minerals and rate of dissolution is dependent on various environmental factors including moisture, temperature, size of mineral or grain (if applied in powder), and mineral composition (Harley and Gilkes, 2000). After the disintegration of primary minerals, the rate of mineral dissolution and alkalinity generation is also dependent on the dissolution of secondary minerals, in particular poor aluminosilicates comprising kaolinite (containing cations of Ca^{2+}, Mg^{2+}, Na^+, or Fe^{2+}). Also, minerals like phyllosilicates (dolomites, siderite, ankerites; (Ca, Mg, Fe(CO$_3$)$_2$)), carbonates of aragonite/calcite ($CaCO_3$), and

phyllosilicates secondary including phyllosilicates saponite and analcime are important secondary silicates minerals that are formed from the dissolution of primary silicates. However, their supersaturation in the soil solution may limit their alkalinity generation capacity and will cause them to precipitate. Thus, the solubility of secondary silicates, clays and carbonates is the rate-limiting step in weathering, which is highly dependent on the removal of base cations from the solution (Tutolo et al., 2021); Eq. (8)),

$$\text{Plagioclase} + CO_2(aq) + 1.85H_2O \rightarrow 0.85\text{kaolinite} + \\ 0.3Na^+ + 0.7\text{calcite} + 0.6SiO_2(aq) + 0.3HCO_3^- \tag{8}$$

Similarly, a change in the carbonate chemical equilibrium to HCO_3^- and CO_3^{2-} (Eq. (7)) would accord with a decrease in the CO_2 flux, so that additional CO_2 from the system could be sequestered and stored permanently (Bach et al., 2019). Such a change toward HCO^- and CO_3^{2-} can occur via the solubility of minerals such as quicklime (CaO) or olivine (Mg_2SiO_4) (Eqs. (9)−(10)). During the mineral's dissolution, H^+ is consumed and substituted by conservative ions with cations like Ca^{2+} and Mg^{2+} (Wolf-Gladrow et al., 2007; Oelkers et al., 2018). Thus, utilization of the rock/minerals, and induced biological weathering approaches, could supply divalent cations to buffer the soil pH and the associated risk of SIC losses in acid soils,

$$\text{Olivine dissolution}: Mg_2SiO_4 + 4H^+ = 2Mg^{2+} + H_4SiO_4 \tag{9}$$

$$\text{Quicklime dissolution}: CaO + 2H^+ = Ca^{2+} + H_2O \tag{10}$$

3.2 Use of biochar and bone derived bio (chars)

Application of various organic amendments including manure, crop residues, or compost help in the short-term decreases of acidification, but at a cost of higher CO_2 release. For instance, manure addition to soil can generate alkalinity via the addition of base cations, organic anions decarboxylation, labile organic N ammonification, and by forming the aluminum-SOM complexes (Xu et al., 2006; Xiao et al., 2013; Dippold et al., 2014; Cai et al., 2015; Hagvall et al., 2015). In addition, the addition of these amendments and their mineralization releases N to the soil which further leads to soil acidification (Chen et al., 2014). In contrast, biochar application to soil causes a long-term reduction in soil acidification and induces a liming effect as it contains a significant proportion of alkaline ash or base cations (Dai et al., 2017; Shi et al., 2019). Biochar supplies Ca^{2+} to soil (Wang et al., 2014), which can further facilitate carbonate precipitation (Monger et al., 2015; Dong et al., 2019a). Furthermore, the sequestration of CO_2 in biochar-amended soil could be mainly associated with the adsorptive/reactive capture of organic and/or inorganic carbon (IC). The adsorptive removal mechanisms for organic CO_2 by biochar comprise H-bonds interaction electrostatic force, π interaction (i.e., electron donors/acceptors), hydrophobic sorption, π, and pore filling (Woolf et al., 2010). However, various factors

control the dominant adsorption mechanisms such as the physicochemical and biological biochar characteristics, surface functional groups, and carbon species in soils (Pan et al., 2021).

Results of various incubation experiments revealed that biochar amendment resulted in higher dissolved and total IC contents in soil (Table 1) (Fernández et al., 2014; Luo et al., 2017; Dong et al., 2019a, 2019b; Shi et al., 2021). Biochar addition may increase SIC content, especially by promoting pedogenic IC precipitation. For example, in a long-term field trial, Dong et al. (2019a) tested the impact of different levels of biochar (i.e., 0, 30, 60, and 90 kg ha^{-1}) on the SIC contents in the soil profile (0–100 cm). The results revealed an increment in the SIC by 18.8%, 42.4%, and 62.3%, respectively (0–20 cm soil depth) suggesting the higher ash content of biochar (36%) increased the soil pH (from 7.6 to 7.7−7.93), respectively.

However, the SIC concentration can be affected by Ca^{2+} contents, and CO$_2$ concentration in soil. Hence, the mechanisms that drive biochar-induced SIC sequestration could be: i) higher ash, CEC, water-soluble salt ion (such as Ca^{2+}) contents in biochar serves a key factor for the formation and retention of associated carbonates (Sommer et al., 2006; Monger et al., 2015; Dong et al., 2019a), ii) high base cations in the biochar enhances soil pH which helps to accelerate SIC formation (Zhao et al., 2016b; Dong et al., 2019a). Higher pH favors the dissolution of CO$_2$ into the alkaline soil solution-phase resulting from the formation of dissolved IC, and iii) higher soil microbial activity promotes more SIC formation by releasing CO$_2$ in response to the decomposition of SOC, resulting in higher SIC precipitation (Fauriel and Laloui 2012; Zhao et al., 2016a; Dong et al., 2019b).

The small particle size of biochar helps to enhance soil porosity, water, and nutrient holding capacity, and ultimately soil microbial activity (Lehmann et al., 2011; Azeem et al., 2021c). In long-term biochar amended field trials, a significant improvement in soil moisture contents and the abundance of microbial biomass suggests that biochar application promotes soil microbial activity (Azeem et al., 2019; Shi et al., 2021). Higher soil respiration, possibly due to stimulated microbial activities with biochar addition, provides enough dissolved CO$_2$ involved in the formation of the secondary carbonates (Shi et al., 2021). In addition, after biochar amendment to soil, the IC component of carbonates (HCO$_3^-$ and CO$_3^{2-}$) in biochar itself, linked with base cations in the ash, also contribute to the SIC accumulation, as reported in a long-term field experiment (Yuan et al., 2011; Shi et al., 2021). However, limited studies have reported only the occurrence of higher SIC in biochar-amended soils; thus, further studies are required to better understand the associated mechanism of SIC accumulation/depletion in particular in biochar-amended acid soils.

It is widely accepted that dissolved CO$_2$ could be possibly sequestered to form stable carbonates in the soil liquid phase provided with enough water-soluble Ca^{2+} and Mg^{2+}. This could be true while using BBC, as it contains a significant proportion of Ca^{2+} and Mg^{2+} (Siebers and Leinweber, 2013; Azeem et al., 2021a,2021b) (Table 2), and

Table 1 Effect of biochar application on SOC and SIC in the agricultural soils.

Biochar feedstock and pyrolysis temperature	Application rate	Soil and experiment type	Effect on SOC	Effect on SIC	Reference
Corn stover (500°C) and wood mixture (600°C)	-	Silt loam, maize filed, incubation (140 days)	Wood mixture enhanced SOC from 0.001–0.0069 mg C kg^{-1} of soil.	Wood mixture enhanced (SIC) up to 0.045 mg C kg^{-1} of soil.	Fidel et al., 2017
Holm oak wood, 400°C for 8 h	5% mixed with 95% compost	Copper mine spoil planted with *Brassica juncea*, pot experiment (90 days)	Increased 16–180 g C kg^{-1}	3.8–37 g C kg^{-1} increment in SIC	Rodríguez-Vila et al., 2016
Calotropis procera, 450°C for 1h	30 and 60 tons ha^{-1}	8 years old reclaimed mine spoil and forest soil. 6 months incubation in the field, natural conditions	Total SOC increased by 36–40 g C kg^{-1} compared to forest soil (21 g C kg^{-1})	The SIC enhanced form 1.9 g C kg^{-1} to 3.5 g C kg^{-1} and 4.5 g C kg^{-1} at BC30 and BC60, respectively.	Ghosh and Maiti, 2021
Rice husks (70%) and cotton seedhulls (30%), 400°C for 4 h	0, 30, 60, and 90 Mg ha^{-1}	Wheat and maize rotation	TOC contents were enhanced by 32%–104% with 30, 60, and 90 Mg ha^{-1} of biochar application	SIC accumulated by 18.8%, 42.4% and 62.3%, respectively, in the 0–20 cm soil layer.	Dong et al., 2018, 2019ab
Corn straw, 360°C	4.5 Mg (ha^{-1} yr^{-1} B4.5) and 9.0 Mg ha^{-1} yr^{-1}	Wheat and maize rotation	After 10 years of biochar incorporation, 62–81% of biochar-C (was transformed to SOC under 0–100 cm soil layer)	Straw incorporation (3.2–3.7 g kg^{-1}) revealed higher SIC accumulation compared to biochar (1.0–2.0 g kg^{-1}) and control (1.0–2.0 g kg^{-1}), under 0–40 cm soil layer	Lu et al., 2021
Corncobs, 360°C	0, 4.5 and 9.0 Mg ha^{-1} year^{-1}	Silt loam, wheat–maize rotation	Total SOC contents increased up to 16%–82%	Enhanced SIC content (3.2%–24.3%),	Shi et al., 2021

*SOC=Soil organic carbon; TOC=Total organic carbon, SIC=Soil inorganic carbon.

thus can facilitate associated carbonate precipitation under higher CO_2 respiration. The BBC is made by pyrolyzing animal bones following the similar process of plant-derived biochar preparation i.e., under deficit / no oxygen conditions. Following this procedure, the BBC retains more C, recalcitrant C, and a larger surface area (Azeem et al., 2021a, 2021b). In addition, BBC contains macrospores with carbon (10%−24%), and minerals contents (75%−90%), and the P_2O_5 fertilizer content is around 10%−30% (Azeem et al., 2022). These features make BBC a suitable soil amendment as a natural organic fertilizer (Koron et al., 2017). BBC can provide enough C, Ca^{2+}, and Mg^{2+} with organic C, which helps sequester both SOC and SIC (Azeem et al., 2021a, 2021b). This helps to improve soil microbial activity and will provide enough CO_2 to make carbonates with Ca^{2+} and Mg^{2+} present in the added BBC (Fig. 1). This could be a potential strategy for sequestering C acidic soils with the

extra benefit of being a source of organic P. Table 2 summarizes the physiochemical composition of biochar and BBC derived from various feedstocks.

Biochar addition to the soil also affects the nitrogen cycle by reducing ammonia volatilization and nitrate leaching (Van Zwieten et al., 2015). The high pH of biochar, ash content, CEC with abundant positive and negative surface functional groups, helps to retain both NH_4^+ and NO_3^-, as described in the previous studies (Hale et al., 2013; Sun et al., 2017; Yuan et al., 2017; Palviainen et al., 2018; Wu et al., 2018). Biochar amendment as a compound fertilizer (biochar chemical fertilizers, and bentonite) at the rate of 1 t biochar ha^{-1} into a paddy field, significantly helped to reduce CH_4 (25%–50%) and N_2O (7%–39%) emissions with an increment int the grain yields of 10%–31% (Qian et al., 2014). This suggests a substantial potential for lowering C and N losses under rice crop production, mainly in acidic soils

Table 2 Physicochemical composition of biochars derived from various feedstocks (The table represents the mean average of the results given in Table S1).

Feedstock	EC(dS m⁻¹)	pH	CEC(cmol kg⁻¹)	Surface area (m² g⁻¹)	Chemical composition (%)								
					C	H	O	N	P	Ca	Mg	SIC	Ash
Wood	7.46±6.1	9.05±1.2	18.2±8.1	106±300	63.71±16	3.08±1.3	19.23±8.8	0.95±1.1	0.40±0.04	26.3±2.60	0.52±0.08	3.92±3.11	5.3±7.0
Manure	0.46±0.5	9.94±1.3	66.1±8.00	22±36	30.80±10	1.39±0.6	34.01±33	4.44±6.2	2.57±0.14	52.7±3.64	5.78±2.15	-	44.6±0.97
Crop residues	2.52±3.3	8.81±1.6	56.3±3.92	111±112	59.24±12	2.54±1.1	17.34±8.5	1.22±1.4	0.80±0.09	20.7±2.16	0.61±0.07	-	21.1±0.54
Bone char	3.12±0.01	7.94±0.5	-	88±61	8.86±3.5	0.75±0.5	27.52±5.4	1.59±1.2	17.13±2.3	39.53±7.3	-	-	80.53±11.2
Bone biochar	0.88±0.5	9.37±1.3	-	115±99	24.34±19	2.02±1.3	16.74±8.6	2.49±1.3	11.77±5.8	35.04±50.4	12.6±0.11	-	51.79±5.33
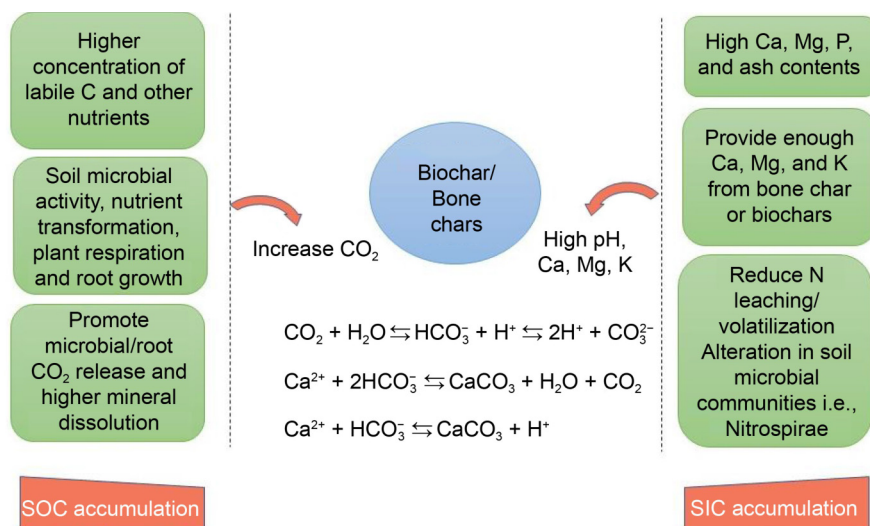
Fig. 1 Bio (bone) char role in SIC accumulation in soil. Mechanism responsible for the higher accumulation of SIC under biochar amendment includes; i) higher concentration of labile C and another nutrient such as N, P, K, Ca, Mg, K and improved soil properties will lead to enhance soil microbial activity, root proliferation, and higher microbial/plant respiration, ii) The higher soil microbial activity promotes higher SIC formation via releasing CO_2 in response to the decomposition of SOC, resulting in higher SIC precipitation, iii) biochar or bone biochar will provide enough cations to react with CO_2 and form pedogenic carbonates. The soil acidification induced with N addition will be balanced by biochar (via adsorption of NH_3^- and NH_4^+ with functional groups on biochar surface) and alteration in the microbial communities responsible for nitrification.

(Jeffery et al., 2016). Biochar addition also alters the soil microbial communities' structure, abundance, and activity (Azeem et al., 2019, 2021c; Palansooriya et al., 2019). Biochar limits the availability of N which reduces the abundance of nitrifying communities. A significant reduction in the *Nitrospira* community has been noticed in various agricultural and turfgrass soil systems (Wang et al., 2015; Azeem et al., 2020; Liu et al., 2021). This will decrease nitrification and associated H^+ release and thus can help to reduce acidification, C and N losses, enhance N use efficiency, and sequester both SOC and SIC in low C soils.

4 Role of microbes in enhancing mineral/BBC solubility

Rock/mineral powders are low-cost potential sources of most of the nutrients essential needed by higher plants for optimum growth and development. However, slow mineral dissolution/solubility rates are the main hindrance to its widespread utilization of rock residues/powders in agricultural soils (Ribeiro et al., 2020). Biological weathering and rhizosphere processes may accelerate the dissolution of minerals (Haldar, 2020). Plants and soil microbes act directly on the mineral surface and play a key role in mineral dissolution, sustaining a wide variety of weathering microbes in the root environment (Fig. 2; Table 3). Previous studies revealed that the rate of microbial-induced weathering of silicate minerals or rock powder is higher compared to pure chemical weathering (Perez et al., 2015; Hu et al., 2020). Also, surface bacterial/fungal attachment promotes higher mineral dissolution (Ahmed and Holmström, 2015). The

mechanism of microbial-mineral interaction and mineral dissolution depends on various factors: i) production of metabolic agents, i.e., extracellular secretions, enzymes, organic acids, siderophore, or other chelation agents, ii) disintegration of the mineral lattice via redox reactions, and iii) exchange of cations charge balance for protons at the surface of the mineral i.e., Ca^{2+}, Na^+, and/or K^+ (Hu et al., 2020).

Microbially induced mineral weathering involves a chain of actions including biofilm formation, acid production (e.g., carbonic, sulfuric, acetic, citric, formic, gluconic, lactic, oxalic, pyruvic, and succinic acids), chelating agents (e.g., siderophore production), redox reactions, and disintegration with microbial filaments. Biofilm formation (mostly bacteria) and filament-induced cracking (by fungi) are the first key steps in microbial mineral dissolution via the release of extracellular polysaccharides. Biofilms wrap microbes and adhere to the mineral surface via extracellular polysaccharides (Ribeiro et al., 2020). Fungal communities also play an important role in the biological disintegration of rocks/minerals and the production of secondary minerals (Burford et al., 2006; Hu et al., 2020). Mycorrhizae develop a symbiotic relationship with plants thereby contributing mineral uptake, transformation, and/or redistribution of inorganic elements (Akhtar and Siddiqui, 2008). In addition, some fungi can withstand higher pH levels over a wider redox range, faster and more efficiently dissolve minerals and mobilizing metals, compared to bacterial communities (Castro et al., 2000; Burford et al., 2006). The action of fungi on the rocks makes visible dissolution channels as seen for *Aspergillus niger, Aspergillus* sp. FS-4, *Talaromyces flavus,* and *Suillus tomentosus*, on the muscovite, basalt, lizardite,
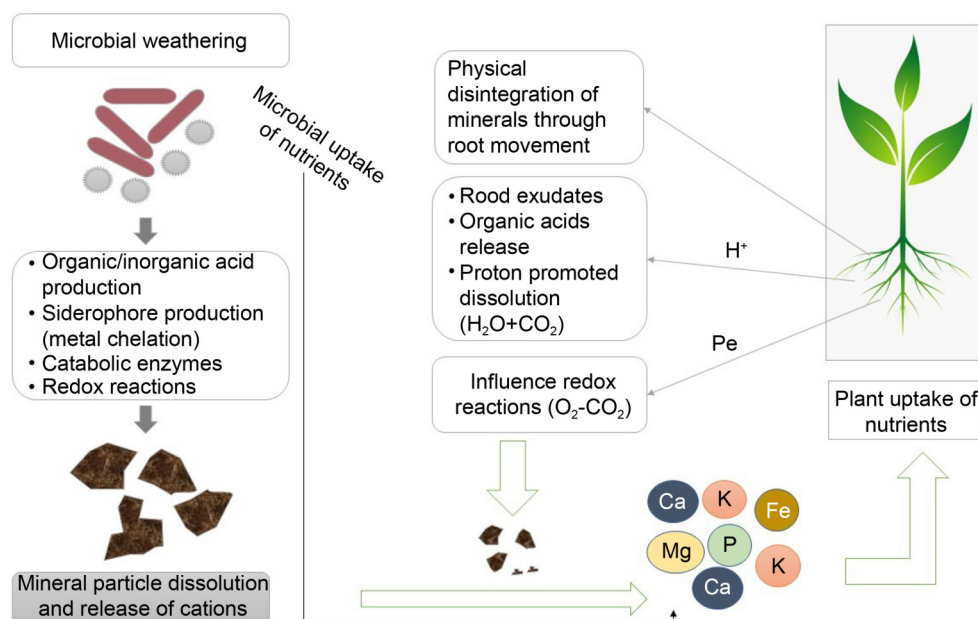
Fig. 2 Biological mineral weathering induced by soil microbes and plant root-mineral interaction. Plants induce mineral weathering via physical disintegration, the release of root exudates, organic acids, redox reactions, and proton-promoted dissolution at the mineral surface. The microbial released by-products (i.e., organic/inorganic acids, siderophore production, catabolic enzymes, and redox reactions) accelerate the dissolution of soil minerals.

Table 3 Microbial communities involved in the mineral dissolution.

Bacteria	Mineral	Crop	Type of experiment	Results	Reference
Bacillus pseudomycoides O-5	Waste mica	*Camellia sinensis* L.	Pot experiment	B. *pseudomycoides* solubilized the mica waste of 33.32 ± 2.40 µg K mL^{-1} in culture medium (7 days) while 47.0 ± 7.1 mg kg^{-1} in microcosm soil after 107 days of addition	Pramanik et al., 2019
Bacillus cereus	K-feldspar	Potato	Field experiment	42% increase in soil available K using co-use of B. *cereus* and K-feldspar with reduced soil pH via release of organic acids	Ali et al., 2021
Bacillus subtilis ANctcri3; *B. megaterium* ANctcri7	Feldspar rock powder (K 3.9%)	Elephant foot yam	Field experiment	475–522 mg kg^{-1} of K release was notices using both strains with lowering pH and attachment of bacteria on mineral surface	Anjanadevi et al., 2016
Bacillus mucilaginosus, Azotobacter chroococcum, and *Rhizobium* sp.	Waste mica	Maize and wheat	Hydroponic experiment	Higher plant uptake (4%-22%) and biomass accumulation of K (29%-86%)	Singh et al., 2010
Pseudomonas sp.	Muscovite	Tomato	Pot experiment	188 and 127% of K release from biotite and muscovite, respectively. Ability of K solubilization was in the range of 19–49 mg L^{-1}	Sarikhani et al., 2018
Bacillus edaphicus	Illite	Cotton and rape	Pot experiment	Oxalic acid production was the key in releasing the K	Sheng and He, 2006
Bacillus mucilaginosus	Waste mica	Sudan grass	Pot experiment	Mineral K release to water soluble and exchangeable K with bacterial inoculation. 13%–36% higher uptake of K in plants	Basak and Biswas, 2009
Enterobacter sp. GL7, *Klebsiella* sp. JM3, XF4, and XF11	K-feldspar	Tobacco	Pot experiment	0.5–4.4 mg L^{-1} of K release in a liquid medium	Zhang and Kong, 2014
Klebsiella oxytoca KSB-17	Waste mica	Maize	Pot experiment	Increased uptake of K (154%) in plants	Imran et al., 2020
Aspergillus sp. FS-4	27 vol% (Basalt-T) and 18 vol% (Basalt-F)	Glass composition	-	Higher release of Mg, Al, Fe, and K with *Aspergillus* with a reduction in pH and higher siderophores production	Hu et al., 2020
Verticillium sp.	olivine	Mg-deficient Forest soil	Incubation (9 months)	16% weathering flux in 9 months of soil incubation with (2.2 ± 1.2) × 10^{-10} moles of olivine per square meter per second (mol m^{-2} s^{-}) of dissolution rate	Wild, 2021

and biotite, respectively (Balogh-Brunstad et al., 2008; Wei et al., 2013; Li et al., 2016; Hu et al., 2020).

Furthermore, fungal hyphae/mycelia penetration results in the increment of turgor pressure at the mineral interface, which releases substrates and increases access to nutrients (Adeyemi and Gadd, 2005; Gadd, 2017). For example, the growth of microbial filaments into the cracks on the surface of pyrite results in bioerosion of pyrite via biomechanical penetration (Samuels et al., 2019). Penetration of mycorrhizal fungi was also reported for other minerals like hornblende, shale, and feldspars through channels/tunneling to access minerals nutrients, thereby suggesting the role of microbial filaments in the formation of dissolution channels resulting in the bio-weathering of minerals (Smits, 2006; Samuels et al., 2019).

After surface attachment, the microbial release of organic acids and chelating agents including siderophores, initiates a synergic process of biological mineral dissolution (Finlay et al., 2020). In this case, the decrease in pH around the soil particles, protonation (H$^+$)-promoted dissolution, chelation with minerals, soil matric and redox reactions are the key mechanisms of bacterial-rock weathering (Ribeiro et al., 2020; Samuels et al., 2020). The acidification process driven by microbes is frequently related to organic or inorganic acid production, such as the production of sulfuric and nitric acid. Sulfur-oxidizing bacteria, i.e., *Thiobacillus* genus, play a vital

role in the production of sulfuric acid (via oxidation of element sulfur) while nitrifying microbes are responsible for nitric acid production through the process of nitrification that leads to acidification and ultimately causes the dissolution of minerals (Ranalli et al., 2009). These changes in pH around the mineral surface enhance microbial activity that affects the ionic concentration equilibrium and ion solubility around the mineral-solution interface resulting from higher rates rock/mineral weathering. For example, *Bacillus pseudomycoides* isolated from soil planted with tea were reported to induce higher K$^+$ release from mica waste (i.e., muscovite type-mineral). The *B. pseudomycoides* solubilized 33.32 ± 2.40 µg K mL^{-1} of mica waste in 7 days of incubation (30 ± 1°C), while in a soil microcosm, *B. pseudomycoides* strain O–5 enhanced K$^+$ availability (47.0 ± 7.1 mg kg^{-1}) during 105 days of incubation, while the strain released 104.9 ± 15.3 mg K kg^{-1} in MW-treated soil (Pramanik et al., 2019). Parmar et al. (2016) mentioned that *B. licheniformis* and *Bacillus subtilis* had K-dissolution potentials of up to 42.37 and 46.37 µg mL^{-1}, respectively. These studies reported the acidolysis (i.e., organic/inorganic acid production), polysaccharides, chelation, complex lysis, and exchange reactions as the key mechanism for enhanced mineral dissolution (Etesami et al., 2017).

In addition, the conversion of aerobically respired CO$_2$ into carbonic acid could be enhanced by the enzyme carbonic

anhydrase, that is involved in the catalysis of the reversible hydration of CO_2 (Eq. (8)). Carbonic anhydrase is widely released by various bacterial strains involved in mineral weathering (e.g., calcite dissolution) (Xiao et al., 2015). Various studies reported that the introduction of bacterial inoculation of i.e., P solubilizing bacteria (*Pseudomonas* and *Bacillus* spp.) in a growth medium enhanced the weathering of added basalt, muscovite via the production of organic and inorganic acids, lowering the pH around rock particles, and ultimately released Ca^{2+}, Mg^{2+}, P, and Fe (Gupta et al., 2011; Olsson-Francis et al., 2015; Pramanik et al., 2019; de Oliveira et al., 2020). Olsson-Francis et al. (2015) reported higher basalt dissolution ($p < 0.05$) with the addition of *Arthrobacter Bacillus*, *Chromobacterium*, *Burkholderia, Paenibacillus, and Janthinobacterium*, which utilizes basalt as a nutrient source. In addition, they noticed a positive correlation of acidification (i.e., oxalic acid production) with the higher rate of basalt dissolution.

Gupta et al. (2011) studied the dissolution potential of muscovite rock phosphate using various phosphate solubilizing bacteria (PSB) (i.e., *Burkholderia gladioli* MTCC-10217, *B. gladioli* MTCC-10216, *Serratia marcescens* MTCC-10238, and *Enterobacter aerogenes* MTCC 10208). The PSB strain-induced dissolution of muscovite was in the range of 1.4 to 15.2 μg mL^{-1}; however, the highest level of dissolution was found with *Enterobacter aerogenes* 10208. In addition, the PSB-inoculated plants also had a higher P content (64%–273%) in comparison to uninoculated plants, while in another study, *Bacillus cereus* enhanced K-feldspar weathering in a potato field. The results revealed higher availability of N, P, and K in the potato plant. In particular, the soil available K was enhanced with co-use of K-feldspar and *Bacillus cereus* (42%), while, K uptake and potato yield were improved by 62% and 21%, respectively, in comparison to unamended plants (Ali et al., 2021).

Siderophores are the key chelating agents released by soil microbes under iron deficit conditions and play an important role in microbial weathering (Behnsen and Raffatellu, 2016). These substances are mostly involved in the dissolution/solubilization of Fe-bearing silicates (Ahmed and Holmström, 2015; Parrello et al., 2016). Oxidation/reduction processes occurring on the mineral particle surface can destabilize the crystalline structure, resulting in mineral dissolution. For example, *Desulfuromonas*, *Shewanella*, and *Geoalkalibacter ferrihydriticus* (Z-0531T) species utilized Fe (III), using oxides of Fe and clay minerals as a terminal electron acceptor, during anaerobic respiration (Ribeiro et al., 2020). Also, Zavarzina et al. (2016) reported a bacterial strain *Geoalkalibacter ferrihydriticus* Z-0531T, as a potential candidate involved in the phyllosilicate weathering (i.e., glauconite and biotite) through anaerobic oxidation of carbonate and utilizing Fe_2C as an electron acceptor. Increased rock/mineral dissolution rate is essential to supply enough cations to balance the soil pH and reduce the loss of Ca^{2+}/ Mg^{2+} in acid soils. Thus, potential microbial candidates for accelerating the biological weathering of silicate minerals can be applied with powdered rock/mineral or BBC dissolution, as an enhanced weathering approach, particularly in acid soils. However, the microbial agents along with rock/mineral powder need a carrier that supports microbial growth and activity. In this case, biochar can be utilized as a potential inoculant carrier for microbial agents as reported in earlier studies (Hale et al., 2015; Azeem et al., 2021c). Biochar from feedstock other than bone biochar can be utilized as an inoculation carrier for microbial agents for enhanced weathering of silicate minerals. Biochar will provide an appropriate habitat for microbes, bioavailable nutrients, and water contents, which will help to improve microbial activity and release of acids, siderophores, and other chelating agents. The composites will not only help to reduce soil acidity but also will help to supply other essential nutrients for plant growth, increase crop productivity, reduce organic fertilizer demand and lower the negative consequences on soil and human health. The comprehensive review of the potential mechanisms calls for further studies to determine the true potential of these approaches (biochar, rocks, microbes) with respect to specific mineral dissolution, various soil types, enhanced SIC precipitation, and economic benefits for crop production.

5 Conclusion

Lower N use efficiency and higher nitrogen addition accelerate soil acidification, which has exacerbated acidification and depletion of SIC stocks. Strategies are required to decrease SIC losses and maintain soil pH, mainly by enhancing fertilizer use efficiency. Biochar application (in particular derived from manure and high Ca^{2+}, Mg^{2+}, and ash sources such as bone wastes) possesses huge potential to overcome acidification-induced SIC losses. BBC is rich in Ca^{2+}, Mg^{2+}, P, and with an additional C source could serve as a nutrition source for soil microbes which could lead to enhanced biological weathering or soil mineralization. Very few previously published studies have shown enhanced SIC accumulation in biochar amended long-term field trials. There is a pressing need to determine the long-term impacts of biochar on SIC stocks, particularly in acidic soils. Furthermore, to our best knowledge, there is no field trial regarding BBC addition to acidic soil and its effect on SOC and SIC stocks. Adding to these, rock powder, particularly basalt, could be utilized for alkalinity generation in acidic soils to overcome acidification. The lower dissolution rate of rock residues limits their use, due to slow weathering and release of Ca^{2+} and Mg^{2+}. The dissolution of rock powder can be enhanced using a microbial inoculation of appropriate microbes belonging to the genus *Bacillus*, *Pseudomonas*, and *Aspergillus species*. In addition, the screening of potential microbial communities is necessary to determine which bacteria or fungi are specific to which mineral type. In addition, microbial inoculation needs a

suitable carrier that can supply essential nutrients (C, N, P, K, and S) and a habitat for microbes at the time of application. This could be achieved with the co-use of biochar as an inoculant carrier and rock powder or BBC.

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