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Is the climate change mitigation effect of enhanced silicate weathering governed by biological processes?

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Abstract (max. 300 words)

A number of negative emission technologies (NETs) have been proposed to actively remove CO₂ from the atmosphere, with enhanced silicate weathering (ESW) as a relatively new NET with considerable climate change mitigation potential. Models calibrated to ESW rates in lab experiments estimate the global potential for inorganic carbon sequestration by ESW at about 0.5-5 Gt CO₂ y⁻¹, suggesting ESW could be an important component of the future NETs mix. In real soils, however, weathering rates may differ strongly from lab conditions. Research on natural weathering has shown that biota such as plants, microbes and macro-invertebrates can strongly affect weathering rates, but biotic effects were excluded from most ESW lab assessments. Moreover, ESW may alter soil organic carbon sequestration and greenhouse gas emissions by influencing physicochemical and biological processes, which holds potential to perpetuate in even larger negative emissions. Here, we argue that it is likely that the

climate change mitigation effect of ESW will be governed by biological processes, emphasizing the need to put these processes on the agenda of this emerging research field.

Introduction

Conventional climate change mitigation alone will not be able to stabilize atmospheric carbon dioxide (CO₂) concentrations at a level compatible with the “well below 2°C warming” limit of the United Nations’ Paris Agreement (UNFCCC, 2015). Safe and scalable negative emission technologies (NETs), which actively remove CO₂ from the atmosphere and ensure long-term carbon (C) sequestration, will be needed to meet this goal (Gasser et al., 2015). Depending on how fast greenhouse gas (GHG) emissions are reduced, 100-1000 Gt CO₂ will have to be removed from the atmosphere by 2100 (IPCC, 2018; Psarras et al., 2017; Rockström et al., 2017). Decarbonization roadmaps show that NETs must be deployed quickly and at large scale: CO₂ removal would need to reach about 5 Gt CO₂ y⁻¹ by 2050, and increase further to about 10 Gt CO₂ y⁻¹ between 2050 and 2100 (Obersteiner et al., 2018; Rockström et al., 2017). Fast progress in achieving cost-efficient NETs is needed if we are to meet the Paris Agreement’s ambitions (Hilaire et al., 2019).

Enhanced silicate weathering (ESW) is a relatively new, low-tech NET with considerable climate change mitigation potential (Beerling et al., 2020; Fuss et al., 2018; Strefler et al., 2018). The mechanism of CO₂ removal by ESW is based on speeding up the natural process of silicate weathering. The principle of ESW is the reaction of silicate grains with CO₂ and water to form bicarbonates which can either leach out of the soil into groundwater, rivers and eventually the ocean, or precipitate in the soil, forming pedogenic carbonates (Fig. 1).

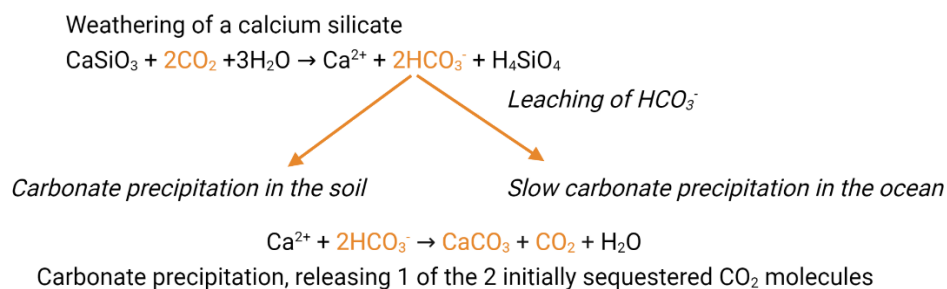


Figure 1: Simplified silicate weathering reaction indicating the two pathways: bicarbonate leaching out of the system and carbonate precipitation in the soil.

The proof of principle that silicate weathering draws down atmospheric CO₂ can be found in the geological record, where a negative temperature-weathering feedback is believed to have stabilized Earth's climate (Berner, 2004; Walker et al., 1981). Increasing CO₂ concentrations raise temperatures and increase rainfall, thereby accelerating silicate weathering rates and atmospheric CO₂ removal, hence slightly mitigating the warming trend by about 0.04 W m⁻² K⁻¹ (Goll et al., 2014). The idea of ESW is to increase C sequestration through mineral weathering by actively amending soils with finely

ground, fast-weathering silicates such as basalt (Hartmann et al., 2013; Schuiling & Krijgsman, 2006). Soil amendment with basalt, an abundant rock rich in calcium (Ca) and magnesium (Mg), is particularly promising in agriculture, due to the potential for co-delivery of multiple ecosystem services, including increased crop yield (Beerling et al., 2018; Van Straaten, 2006). In fact, the positive effects on soil and crops are the primary current reason for the use of basalt and other silicates in agriculture (Haque et al., 2020b; Leonardos et al., 1987; Van Straaten, 2006; Wang et al., 2018a; Zhang et al., 2018). Another potential application that is gaining interest is the use of silicates for nature restoration, as this would help to abate soil acidification and replenish soil calcium (Taylor et al., 2021).

Early lab experiments and modelling indicate the highest potential for ESW on acid soils in humid and warm environments and estimate the global C sequestration potential of ESW between 0.5 and 5 Gt CO₂ y⁻¹ (depending on cost assumptions, among others; Beerling et al., 2020; Fuss et al., 2018). This emphasizes the clear potential of ESW to provide a substantial part of the required decarbonization. However, the uncertainty on current estimates derived from lab experiments and modelling is large and the largest uncertainty concerns the *in natura* weathering rate and associated C sequestration (Fuss et al., 2018). The first field assessments of inorganic C sequestration by ESW indicate large variability, even between sites with similar climate, soil, silicate material, and rate of application (Haque et al., 2020b). Moreover, in the real world, processes such as secondary mineral formation, soil pore water saturation, and low water-silicate contact rates can substantially slow down weathering rates (Zhang et al., 2018) – as was the case in one of the first ESW mesocosm experiments (Amann et al., 2020). In addition, ESW will almost certainly impact primary production, soil organic carbon (SOC) sequestration and soil GHG emissions. These impacts will affect the climate change mitigation potential of ESW, but have not yet been considered in current calculations.

Biota stimulating silicate weathering

We postulate that biota are key to understanding the effect of ESW on atmospheric GHG concentrations and anticipate that an explicit consideration of biotic context is necessary to unlock ESW's full climate change mitigation potential. Much of our ESW knowledge is derived from lab experiments that excluded biota such as plants and soil fauna, although it is known that natural weathering is strongly influenced by biota (Berner, 2004). Many biota have evolved mechanisms to enhance the weathering of minerals and access the nutrients contained in them (Dontsova et al., 2020; Zaharescu et al., 2020). During Earth's history, this biotic stimulation of mineral weathering has substantially altered the mobilization of multiple macro- and micro-elements (Bergman et al., 2004; Zaharescu et al., 2020), inducing for example global shifts in the provision of dissolved silicates to aquatic and marine ecosystems (Derry et al., 2005; Falkowski et al., 2004; Kidder & Gierlowski-Kordesch, 2005). Without such biological influences on weathering, the Earth would be considerably warmer than today due to comparably low abiotic weathering rates (Schwartzman & Volk, 1989). Despite the profound effect of biota on the weathering process, surprisingly little attention has been paid to their role in optimizing ESW efficiency, and to their role in ESW in general. Below, we first discuss the potential effects of plants, microbes and macro-invertebrates on ESW, which can in part be derived from knowledge on natural (geological) weathering. In the following section, we evaluate the expected responses of biota to the implementation of ESW. Then we discuss how ESW may

interact with SOC stocks, and GHG emissions in general, and lastly we provide a way forward in addressing the most important questions that arise.

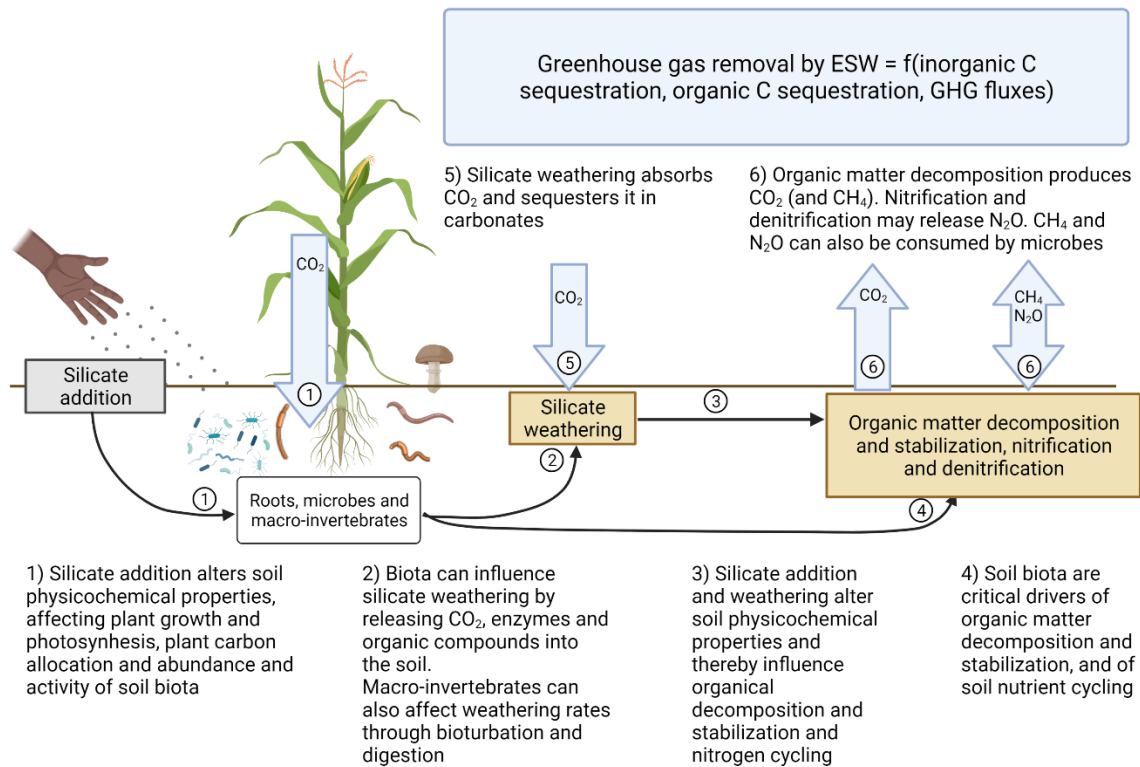


Figure 2: Overview of the biota/silicate-weathering interactions and their influence on the greenhouse gas (GHG) removal potential of ESW. Blue arrows show major GHG fluxes that can be positively or negatively influenced directly or indirectly by ESW. GHG removal through ESW includes not only inorganic C sequestration through the weathering reaction, but also covers the effect of silicate addition on soil organic C sequestration and soil GHG emissions.

Plants - Plant roots can create physicochemical conditions that accelerate the dissolution of silicate minerals (Burgelea et al., 2015; Drever, 1994; Hinsinger, 1998; Hinsinger et al., 2001). They also improve soil structure and hydrology (Angers & Caron, 1998), possibly stimulating weathering rates. A recent microplot study found up to 10-fold higher inorganic C sequestration in planted compared to unplanted soils amended with silicates (Haque et al., 2020c). Roots take up elements such as Si, Mg, Ca, and Fe that are released during weathering, and thereby avoid pore water saturation with reaction products to slow down weathering rates (Harley & Gilkes, 2000; Hinsinger, 1998). By releasing protons and CO_2 , roots reduce soil pH and increase the CO_2 concentration in the rhizosphere (Lenzowski et al., 2018), both of which stimulate mineral weathering (Harley & Gilkes, 2000). Plant roots also exude organic compounds such as organic acids that stimulate weathering by chelating reaction products and dissolving silicate minerals (Dontsova et al., 2020; Drever, 1994). Moreover, organic acids can dissolve silicate minerals at near-neutral pH, where abiotic dissolution rates are limited (Harley &

Gilkes, 2000). The latter compounds may be particularly relevant for ESW applications in soils that are not acidic.

Most leguminous plants such as beans and soybean live in symbiosis with nitrogen (N) fixing bacteria. During N fixation, atmospheric nitrogen (N_2) is converted to ammonia (NH_3), which is used to form amino acids. The latter dissociate to form organic acids such as malate and citrate, also releasing protons (Bolan et al., 1991). These acids help to solubilize silicate minerals and Akter and Akagi (2005) indeed found weathering of basalt to be higher in the presence of soybean than with corn or rice (non-legumes). Also Haque et al. (2019b) reported that in soils treated with wollastonite, weathering rates were higher with beans than with corn or for bare soil without plants.

Microbes - About 90% of land plant species live in symbiosis with mycorrhizal fungi (Brundrett & Tedersoo, 2018). Mycorrhizal fungi are thought to have significantly increased mineral dissolution rates at evolutionary timescales and experiments have shown that they indeed stimulate rock weathering (Bonneville et al., 2011; Burghelca et al., 2015; Burghelca et al., 2018; Zaharescu et al., 2020). Given that mycorrhizal fungi depend on their host for C, their influence on ESW is likely to depend strongly on plant activity and C allocation. Depending on soil conditions, plants can allocate substantial amounts of C to mycorrhizal fungi (Ven et al., 2020), and thereby stimulate their weathering activity, increasing the release of P and other mineral elements from the silicate minerals (Verbruggen et al., 2021).

Other fungi can also accelerate weathering; mineral dissolution rates can be 10 times higher underneath individual fungal filaments compared to areas where fungi are absent (Wild et al., 2021). Fungi accelerate weathering by exuding protons, organic acids, chelators, and by creating gradients through channeling elements away from mineral surfaces (van Hees et al., 2006). Because fungal hyphae are very thin, they can also interact with surfaces much more tightly than plant roots can (Howard et al., 1991; Wild et al., 2021). Moreover, specific genetic pathways that stimulate conversion of CO_2 into carbonates and thus accelerate weathering can be upregulated in response to exposure to minerals (Xiao et al., 2012). This suggests specific fungal adaptations towards dissolution of minerals. The effect of fungi on ESW will likely depend on fungal species and on the extent to which elements contained in the applied silicates (e.g. Mg, Ca, Fe, K) are limiting their growth.

Also other microorganisms such as bacteria can stimulate weathering of rocks and minerals (Gouda et al., 2018). One of the key processes underlying microbially enhanced weathering is the lowering of pH by releasing acids, such as low molecular mass organic acids and dissolved CO_2 . Some bacteria can lower pH to values as low as 2.3 (Ahmed & Holmström, 2014). Basak & Biswas (2009) found that *Bacillus mucilaginosus* significantly enhanced the K release of muscovite mica, which is among the most weathering resistant silicate minerals (Palandri & Kharaka, 2004). In addition, both bacteria and fungi can produce chelates and enzymes that can enhance mineral dissolution rates up to 100 times (Buss et al., 2007; Sun et al., 2013; Xiao et al., 2015). Chelates like siderophores are usually specific to a single element, and their production depends on the type of geological material and soil fertility, again emphasizing high variation among microbial taxa and dependence on environmental context.

Soil enzymes -The enzymes and proteins that play an important role in weathering of silicates are often excreted by microbes experiencing nutritional deficiency. The extracellular excretions are biologically activated both by nutrient limitation and the proximity to the nutrient-carrying mineral (Xiao et al., 2015; Zaharescu et al., 2020). Some enzymes, such as carbonic anhydrase (CA), have a combined effect of both increasing silicate weathering and carbonate precipitation. CA catalyzes the equilibrium reaction between CO₂ and bicarbonate ions, which in contact with free metal ions combine to form solid carbonate precipitates, such as magnesite or siderite. The abiotic process of carbonate precipitation is slow and the addition of CA accelerates this reaction considerably (Bose & Satyanarayana, 2021). In fact, CA is one of the fastest enzymes, performing up to 10⁶ CO₂ conversion reactions per second. CA is most efficient at high pH and may thus be especially important for ESW in alkaline soils.

Urease is another enzyme used by prokaryotes and eukaryotes for efficient biomineralization. As with CA, urease increases pH locally, inducing precipitation of carbonates, and is inhibited by low pH. One of the more efficient inhibitors of urease are humic acids, that irreversibly inhibit the hydrolytic decomposition of urea and concomitantly the biomineralization of carbonates (Liu et al., 2019). Nonetheless, the humic acid concentrations in soils may be too low to have a profound impact on the precipitation capacity of urease (Al-Taweel & Abo-Tabikh, 2019; Moghal et al., 2020). Moghal et al. (2020) tested the retention of heavy metals in soils by inducing carbonate precipitation using urease. They found that urease efficiently precipitated carbonates which had the coupled effect of also decreasing heavy metal concentrations in the soils. Enhanced weathering of ultramafic silicate minerals such as olivine can release heavy metals such as Ni and Cr, but with addition of urease, the toxic effect of those metals may be diminished. In other words, urease may not only increase weathering rates, but may also help in overcoming potential heavy metal contamination upon addition of some silicate materials. This would be particularly interesting to further investigate for fast-weathering minerals such as olivine that contain high amounts of Ni and Cr.

Macro-invertebrates - Earthworms are important ecosystem engineers (Blouin et al., 2013). It is long known that, through their burrowing and feeding, earthworms strongly affect soil physicochemical as well as biological parameters. Through ingestion of fresh residue and soil particles they can increase mineralization and mineral dissolution, leading to large local increases in nutrient availability (Van Groenigen et al., 2019). Recent research has also shown that availability of nutrients such as P can greatly increase during earthworm gut passage due to competitive desorption reactions with dissolved organic C (Ros et al., 2017). To test the effects of earthworms on mineral dissolution, de Souza et al. (2018; 2013) added gneiss and steatite rock powder to vermicompost containing the earthworm species *Eisenia andrei*. They found that earthworms increased rock weathering and nutrient release, indicated by higher maize yields, albeit only statistically significantly for steatite (de Souza et al., 2013).

Interestingly, several common earthworm species sequester significant amounts of inorganic C by producing calcium carbonate in their specialized calciferous glands (Briones et al., 2008; Darwin, 1892; Lambkin et al., 2011; Versteegh et al., 2014). Although the purpose of these glands remains a topic for debate, they may contribute to increasing weathering rates. The worm digestive system can also promote mineral weathering by inoculating mineral surfaces with microbes and stimulating microbial activity, albeit dependent on the minerals that are used (Carpenter et al., 2007; Liu et al., 2011). Hu et

al. (2018) isolated various silicate dissolving bacteria from the gut of earthworms and found that they increased quartz and feldspar weathering. Furthermore, inoculating potting soils with the isolated bacteria significantly increased soluble Si contents, and thereby enhanced Si uptake and growth of maize seedlings. Last, the positive effects of earthworms on soil structure and drainage (Blouin et al., 2013) can potentially help to distribute silicate grains to deeper soil layers and accelerate the infiltration of water in soils, decreasing the risk for saturation of soil pore water with reaction products.

Ants too might enhance weathering rates (Dorn, 2014). They are abundant in most terrestrial ecosystems, where they influence biogeochemical cycling and mineral weathering (Viles et al., 2021). Ants alter soils in various ways, including effects on soil pH, water infiltration, organic matter accumulation and mineral weathering. Several ant species produce organic acids such as formic acid, which can stimulate rock weathering (Viles et al., 2021). In a 25-year long experiment, Dorn (2014) placed grains of plagioclase and olivine in ant nests and found dissolution rates that were 60 to 330 times higher than in the control plots. On the one hand, ants are thus potentially powerful biotic weathering agents, while on the other hand their area of influence is likely diminishing with distance from the nest. More research is still needed on the role that ants play in natural and enhanced rock weathering, to quantify their potential effect on ESW and to explore the interaction between ants and silicates applied in the field.

Impact of ESW on biota

If biota are important in steering weathering rates, their response to silicate addition will be critical for the climate change mitigation effect of ESW. Biotic responses to silicate addition will co-determine their influence on the weathering rates. Moreover, side-effects on biodiversity associated with changes in the trophic status of ecosystems induced by ESW could occur and both positive and negative effects on plants and soil biota may have economic and/or health consequences. These will influence desirability and societal acceptance of ESW and will thus co-determine the feasibility of ESW in agriculture and in more natural settings.

Plants - Many silicates that can be used for ESW contain mineral nutrients that plants need to grow, including P, Mg, Ca, K, Fe, Zn and Si. As a result, ESW can stimulate plant growth and increase crop yield (Battles et al., 2014; Haque et al., 2019a; Haque et al., 2020c; Taylor et al., 2021; Van Straaten, 2006), although this is not always the case (Haque et al., 2020c; Wang et al., 2018a). Of particular importance might be the widely neglected supply of Si, which is considered a beneficial rather than an essential nutrient, although there is wide agreement and accumulating evidence that Si can induce a broad range of plant biotic and abiotic stress resistances (Epstein, 1999; Guntzer et al., 2012; Haynes, 2014). Besides improved plant growth, ESW has been suggested to increase crop resistance to pests and drought, mainly due to increased Si uptake (Guntzer et al., 2012; Van Bockhaven et al., 2013). Furthermore, seven of the ten most important crops are considered to be Si-accumulators (FAOSTAT, 2018; Fig. 3), and yield increases in response to Si fertilization have been frequently demonstrated for e.g. wheat, rice and sugarcane (Korndörfer & Lepsch, 2001; Liang et al., 2015; Neu et al., 2017). The latter two tropical crops are typically grown on highly weathered and desilicated soils with Si concentrations usually 5-10 times lower than for temperate soils. The demand of Si in agriculture is therefore expected to increase in the future (Haynes, 2014).

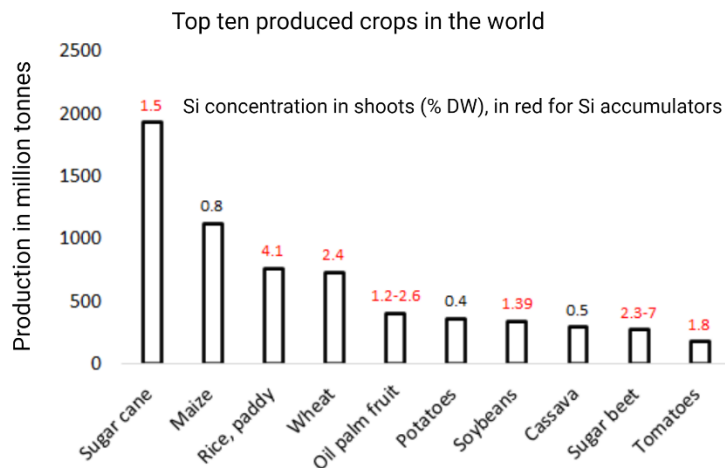


Figure 3: Top ten produced crops in the world in 2018 (FAOSTAT, 2018). Seven of these crops are classified as Si accumulators (>1.0% Si of dry weight (DW)). The values above the bar are average shoot Si concentrations reported by Hodson et al. (2005). Data for average Si shoot contents of oil palm fruit are from Munevar & Romero (2015) and for sugar beet from Draycott (2008).

A positive effect of silicate addition on plant growth and defense can create a positive feedback with ESW, especially if root production and belowground inputs increase. Moreover, positive growth responses can increase C sequestration in plant biomass if silicates are applied in (semi-)natural ecosystems where biomass can accumulate (Goll et al., 2021). On the other hand, it might be concerning that ESW is accompanied by the release of heavy metals like Ni and Cr (Beerling et al., 2018; Haque et al., 2020a; Hartmann et al., 2013). Nonetheless, the application of Ni is not necessarily problematic and below a certain threshold, Ni may even be beneficial for plants (Ahmad et al., 2011; Kumar et al., 2018). In one experiment, barley growth and yield increased with Ni additions of up to 10 mg Ni kg⁻¹ soil (Kumar et al., 2018). When the concentration of Ni exceeded those thresholds, growth and yield declined, while the uptake of Ni continued to increase with increasing Ni application to soil. This suggests that Ni accumulation in the food chain is proportional to the Ni addition. The application rate and choice of silicate minerals can be adjusted to control the heavy metal release (Haque et al., 2020a). In addition, phytoremediation may in some cases pose a way to mitigate the concentration of contaminants such as Ni in soil. As for urease, the potential of phytoremediation to reduce heavy metal availability following e.g. olivine application requires further investigation.

Microbes - Large shifts in soil microbial communities have been associated with the addition of silicates (Carson et al., 2007; Das et al., 2019a; Zhou et al., 2018). For example, Zhou et al. (2018) observed changes in bacterial and fungal community composition and reported a decrease in the abundance of microbial plant pathogens with silicate addition, likely related to improved crop defense (Zhou et al., 2018). Soil pH is one of the main determinants of microbial community composition (Fierer, 2017), and pH changes following silicate addition will thus directly influence which microbial taxa flourish (Das et al., 2019a; Fierer, 2017). Accordingly, silicate rock powder addition had contrasting effects on soil microbes in three Austrian forest soils with varying pH (Mersi et al., 1992). The rock powder additions increased the pH of all soils, but the most significant effects on microbial processes were

found for a Calcaric Regosol and Cambisol (pH 5.8), where the rock powder additions increased nitrification, microbial biomass and respiration, xylanase, and protease activity. Intermediate effects were found for a Stagno-Mollic Gleysol (pH 3.8), where protease activity increased but phosphatase activity decreased, whereas no effects were found on a highly acidic Stagno-Dystric Gleysol (pH 2.8). This illustrates that the effect of ESW on microbial communities depends on the soil properties and hence also the feedback to ESW is likely to vary depending on environmental conditions.

In general, we can expect shifts toward microbial taxa that are better able to occupy new niches on mineral surfaces or those that profit from the released nutrients (Barker et al., 1998; Gleeson et al., 2006; Reith et al., 2015). Also the tolerance to toxic trace elements such as Ni or Cu, which can negatively impact microbes (Silva et al., 2012), can play a role. The various interactions between microbes and added silicate minerals can be expected to lead to a dynamic equilibrium between microbial community composition and mineral weathering. This may impact various soil processes relevant for soil C sequestration and GHG emissions, as illustrated by the observed increases in the abundance of functional genes involved in the degradation of labile C, fixation of C and N and CH₄ oxidation (Das et al., 2019a).

Macro-invertebrates – Few experiments have tested the effect of silicate additions on macro-invertebrates and to the best of our knowledge, these experiments have yet been limited to earthworms and rock powders mixed into vermicompost and manure. Divergent responses were reported, with earthworm growth increasing in some cases and decreasing in others, depending on the rock type and amount that was applied (de Souza et al., 2019; Zhu et al., 2013).

We propose three main pathways through which applying silicate minerals might affect earthworm functioning. First, the increase in pH and basic cations upon silicate addition may positively affect earthworm communities, especially in highly weathered, low pH soils. It is well known that earthworms are absent in soils with a pH lower than 3.5, and very scarce at pH lower than 4.5. Optimal pH ranges differ per species, but are generally within the range 5.0-7.4 (Curry, 2004). In addition, increased availability of basic cations such as Ca and Mg has been shown to increase earthworm populations (Fragoso & Lavelle, 1992) and a recent study showed a clear increase in the earthworm biomass after prolonged liming of forest soil (Persson et al., 2021).

Second, there may be physical interactions between earthworms and added minerals. It has only recently been established that the thickness of the body wall of earthworms varies between species and may affect their functioning in the soil (Briones & Álvarez-Otero, 2018). Although this is so far mostly related to susceptibility to desiccation and burrowing behavior, earthworms with thicker body walls might be better fit to function in systems where sharp mineral particles are added. This and the possibility of mechanical damage upon ingestion remain to be investigated.

Finally, as with plants and microorganisms, the release of toxic trace elements might be detrimental to earthworms. Earthworms can be affected by increased concentrations of e.g. Cu and Ni, especially under conditions of low pH when more cations are desorbed (Ma, 1988; Wang et al., 2020), although in general they are fairly tolerant to most heavy metals (Ireland, 1983). Accordingly, de Souza et al.

(2019) found that the high concentrations of Ni and Cr released during dissolution of steatite did not hinder earthworm growth.

Impact of ESW on soil organic carbon storage

In order to forecast the net effect of silicate addition on the C balance of an ecosystem, the impact of ESW on the largest pool of ecosystem C, i.e. SOC, must be taken into account. Here too, we expect biota/silicate-weathering interactions to play a critical role. Empirical data on the effects of silicate addition are still scarce, but Anda et al. (2013) applied basalt powder to an oxisol and observed significantly increased cacao plant growth but no significant change in SOC stocks was detected within 24 months after application. On the other hand, mineral weathering has previously been identified as the main driver of SOC sequestration across a natural weathering chronosequence (Doetterl et al., 2018). Doetterl et al. (2018) showed that primary mineral weathering was associated with increases in nutrient availability and higher potential of soils to stabilize carbon. Hence, similar to liming and fertilization, silicate addition can be expected to impact SOC sequestration by affecting the quantity of plant belowground C inputs, as well as the stabilization of these inputs in soil organic matter (SOM) (Paradelo et al., 2015; Van Sundert et al., 2020). Depending on soil heterogeneity and magnitude of the effect, it may take several years though before such changes in SOC stocks are detectable.

Plant belowground C inputs depend on plant productivity and C allocation patterns. Plants allocate substantial amounts of C belowground in the form of roots and exudates and through symbiosis with mycorrhizal fungi (Ven et al., 2019; Verlinden et al., 2018). Nutrient availability is a key driver of plant C allocation and plant C inputs to the soil are likely to be affected by silicate addition, although the magnitude and direction of the effect is expected to depend on environmental conditions (Litton et al., 2007; Poorter et al., 2012; Ven et al., 2020; Vicca et al., 2012). Especially soil nutrient status and plant growth responses to the silicate additions are expected to be important in this regard.

Stable SOM can be formed via two major pathways: modification of organic matter present in the soil and turnover of new C inputs. Modification of SOM is strongly driven by activity of extracellular enzymes, which in turn depends on soil pH (Sinsabaugh et al., 2008). Many C- and N-acquiring enzymes increase in potential activity after application of lime to acid soils (Acosta-Martínez & Tabatabai, 2000) and increased pH upon silicate addition may stimulate SOM decomposition and stabilization (Leifeld et al., 2013). Turnover of new C depends strongly on the recalcitrance of litter and rhizodeposits. Although decomposition of recalcitrant litter is slower than that of labile litter, cumulative C losses from recalcitrant litter are generally higher than C losses from more labile inputs because high-quality, labile C is stabilized more through microbial processing (Cotrufo et al., 2013). As with liming, silicate addition may increase litter quality and plant C inputs (Forey et al., 2015; Melvin et al., 2013; Paradelo et al., 2015) and hence increase SOM stabilization. The literature review by Paradelo et al (2015) showed that this generally results in increased SOC stocks following liming, although increased mineralization rates upon liming may also reduce SOC stocks, especially in organic soils and (acid) organic soil horizons (Lundström et al., 2003; Paradelo et al., 2015).

Aggregate formation is also key SOM stabilization mechanism that can be increased by mineral weathering (Doetterl et al., 2018) and is influenced also by soil organisms (Lehmann et al., 2017;

Thomas et al., 2020). Given that aggregates are hotspots of biological activity and biogeochemical processes (Or et al., 2021), weathering rates may be higher inside aggregates than in the surrounding soil. On the other hand, reduced water flow may lead to saturation of the water inside the aggregates, reducing weathering rates. The release of Ca from basalt can stimulate aggregation through enhanced flocculation of clay minerals, an effect possibly enhanced by earthworm activity (Shipitalo & Protz, 1989), and the formation of complexes between Ca and high-molecular weight organic compounds (Baldock & Skjemstad, 2000; Rowley et al., 2018). Furthermore, carbonate minerals are known to improve soil structure and can act as cementing agents in the occlusion of SOM, although uncertainty exists on the importance of this mechanism for field SOC stocks (Fernández-Ugalde et al., 2014; Rowley et al., 2021).

Besides litter recalcitrance and aggregate formation, interactions between silicate minerals and SOM can impact SOC sequestration. Ca released during weathering impacts organo-mineral association via mediation of complexation processes (Rowley et al., 2021) and during the weathering of some silicates such as basalt, substantial amounts of Fe- and Al-oxi-hydroxides are formed. The latter have a strong SOM stabilization potential and the presence of such reactive minerals can increase SOC sequestration (Abramoff et al., 2021; Cotrufo et al., 2013; Or et al., 2021).

Finally, changes in SOM decomposition, e.g. due to altered litter quality or aggregate formation, may also impact ESW, creating a feedback loop. For example, faster turnover of higher-quality litter will increase the amplitudes of soil CO₂ concentration, impacting mineral dissolution. At the same time, increased litter turnover will lead to a higher conversion ratio of litter C to dissolved organic matter (Cotrufo et al., 2013), and thus increases the potential of organic compounds to either form stable organo-mineral complexes or aid in the weathering.

In determining the net effect of ESW on soil C budgets, it is important to consider both inorganic and organic C sequestration and the interactions among the different processes involved. In doing so, the various timescales at which sequestration mechanisms are active need to be considered. Mean residence times of soil organic and inorganic C differ by orders of magnitude, and the persistence of SOC varies widely depending on the location and form of SOC (Schmidt et al., 2011; Zamanian et al., 2016). Moreover, biological responses to silicate weathering might reach saturation on shorter timescales, depending on silicate applications and environmental conditions (Goll et al., 2021). This calls for a better understanding of the extent to which amplifying and dampening biotic responses saturate, as well as the respective timescales. A combination of targeted field experiments and theoretical modelling is required to span the large range of timescales from responses of microbes to SOM stabilization. Soil development chronosequences could provide information on long-term impact of ESW (Doetterl et al., 2018) as ESW-focused studies are still scarce and (yet) of short duration.

ESW effects on other GHG emissions

Silicate addition has been suggested to affect soil emissions of GHGs other than CO₂, especially nitrous oxide (N₂O; Fig. 1; Beerling et al., 2018). N₂O emissions from soils, in both natural and agricultural systems, represent 56–70% of all global N₂O sources (Tian et al., 2020). Agricultural soils are a major source of N₂O to the atmosphere due to the high amount of mineral fertilizers that increase microbial

N availability (Guenet et al., 2021). Also soil acidification can contribute to the N₂O emissions. Low soil pH decreases the activity of N₂O reductase, stimulating the release of N₂O as an intermediate product of the denitrification process (Cayuela et al., 2013). As with liming, silicate addition to acid soils is expected to buffer pH and could thus reduce N₂O emissions (Blanc-Betes et al., 2021; Hénault et al., 2019), although this is not always the case (Nadeem et al., 2020).

Other interactions with biota arise here as well. For example, mycorrhizal fungi have been shown to reduce N₂O emissions (Storer et al., 2018), potentially enhancing this anticipated co-benefit of ESW, whereas earthworms have been reported to increase N₂O emissions (Augustenborg et al., 2012; Lubbers et al., 2013). In some soils, earthworm activity may account for more than 50% of the total soil N₂O emissions (Augustenborg et al., 2012) due to the increase of substrate availability resulting from their activity, the anaerobic environment in their casts as well as their effect on macropore formation (Lubbers et al., 2013; Nebert et al., 2011). The interactive effect of soil biota and silicate-weathering on N₂O emissions is yet unexplored but could provide ways to increase the climate change mitigation effect of ESW. For example, growing N fixing plants, especially temperate legumes, typically acidifies the soil (Bolan et al., 1991), possibly leading to high N₂O emissions. This effect could be countered by an increase in pH upon silicate addition. Furthermore, potential improvements of soil structure through the combination of silicate addition and biotic activity may increase soil aeration and thus reduce denitrification.

Whereas N₂O can be of huge importance in agricultural soils, methane (CH₄) typically is not. CH₄ production is a strictly anaerobic process. In aerobic soils CH₄ oxidation typically exceeds CH₄ production, making these soils modest CH₄ sinks (Dutaur & Verchot, 2007). Rice fields, however, are an important source of CH₄ emissions due to their waterlogged anaerobic soils (Saunio et al., 2020). Some studies have reported a decrease in CH₄ emissions when adding silicates (Ali et al., 2008; Wang et al., 2018b), while others reported an increase (Ku et al., 2020). Silicate addition can reduce CH₄ emissions by reducing methanogenesis and/or increasing CH₄ oxidation (Das et al., 2019b). Silicates containing Fe can stimulate Fe-reducing bacteria at the expense of methanogens, as Fe is a more favorable electron acceptor than CO₂ (Das et al., 2019b; Gwon et al., 2018). Increased Si availability can also increase plant O₂ transport to roots by enlarging aerenchyma gas channels (Liang et al., 2017), increasing CH₄ oxidation. On the other hand, enlarged aerenchyma can also increase CH₄ funneling to the atmosphere and increased plant productivity in response to silicate addition may increase CH₄ emissions by increasing plant belowground C input quantity and quality (Ku et al., 2020). Hence, the net effect of silicate addition on CH₄ emissions will depend on the balance between these counteracting processes.

Future outlook and research needs

We illustrated that the weathering rates and the GHG removal potential of ESW depend not only on abiotic conditions, but is potentially strongly influenced by biota, which have been largely overlooked in ESW research. The multiple soil biota/silicate-weathering interactions imply that the ultimate GHG removal effect of ESW will depend on the balance between positive and negative influences of silicates on biota, and their subsequent joint effects on inorganic and organic C and N fluxes. Further unraveling and quantifying the impact of biota on ESW will be critical for planning widespread use of ESW as a

climate change mitigation strategy. If biological processes are indeed critical in determining GHG removal by ESW, this may imply that the biota-silicate interaction determines the location of ESW hotspots, possibly overriding current assumptions regarding (climate-driven) ESW hotspots in the tropics.

Taking into account biological processes will also be critical to anticipate synergistic effects between ESW and environmental or climatic changes. For example, elevated CO₂ concentrations often increase plant growth and belowground C inputs (Terrer et al., 2021), which could in turn stimulate ESW and SOC sequestration. Warming can be expected to increase weathering rates, but may also decrease SOC sequestration as a result of increased microbial activity and decomposition (Davidson & Janssens, 2006). Moreover, as droughts increase in frequency and intensity, silicate application may reduce some of its impacts through improving soil aggregation, if confirmed, through Si accumulation in plants reducing plant water losses (Guntzer et al., 2012) and/or K release improving water use efficiency (Battie-Laclau et al., 2016). In-depth research is needed to quantify the effects of ESW on plants, soil and GHG removal with an emphasis on environmental moderators subjected to global change.

Further interest in exploring the biota/silicate weathering interaction lies in the potential benefits for agriculture and nature restoration. The potential of ESW as a NET and feasibility of widespread application is not only determined by its GHG removal potential, but also by its potential for increasing crop yield and biomass production, while at the same time avoiding environmental and health risks. Silicate rock powders and other silicate materials (e.g. concrete fines and steel slags) are already being used to “rejuvenate” soils and to provide slow-release bioavailable nutrients. Currently, however, the positive properties of the slow-leaching rock powder nutrients are also the limitations of the material, because their low solubility may render the material cost-inefficient as a fertilizing agent. By increasing the weathering rate with the help from biota, drawdown of CO₂ and the soil fertilizing effects could improve, increasing the potential profit to be made with ESW application. Concerns about the release of toxic trace elements also put a constraint on application of ESW. Here, the possibility for phytoremediation and immobilization of heavy metals contained in some fast-weathering silicate minerals such as olivine could be explored to moderate these risks. We conclude that in order to determine the true potential of ESW as a NET as well as to maximize its climate change mitigation effect, the biotic context must be comprehensively evaluated in lab and in field settings.

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