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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 CO_2 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 CO_2 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 (CO_2) 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 CO_2 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 CO_2 will have to be removed from the atmosphere by 2100 (IPCC, 2018, 2021; Psarras et al., 2017; Rockström et al., 2017). Decarbonization roadmaps show that NETs must be deployed quickly and at large scale: CO_2 removal would need to reach about 5 Gt CO_2 y^{-1} by 2050, and increase further to about 10 Gt CO_2 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; Goll et al., 2021; Köhler et al., 2010; Strefler et al., 2018). The mechanism of CO_2 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 CO_2 and water to form bicarbonates which can either leach out of the soil into the groundwater, rivers and eventually the ocean, or precipitate in the soil, forming pedogenic carbonates (Fig. 1). The latter reduces short-term C storage approximately by half, but in both cases, C is stored for hundreds of years and longer (Hartmann et al., 2013; Köhler et al., 2010).

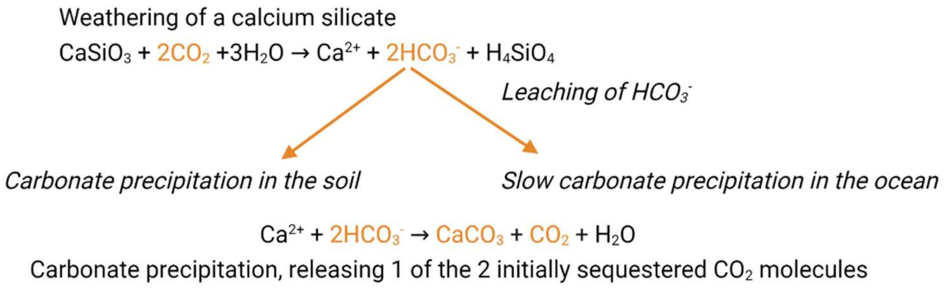


Figure 1: *Simplified silicate weathering reaction indicating the two pathways: bicarbonate leaching out of the system and carbonate precipitation in the soil.*

The proof of principle that silicate weathering draws down atmospheric CO_2 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 CO_2 concentrations raise temperatures and increase rainfall, thereby accelerating silicate weathering rates and atmospheric CO_2 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 (Goll et al., 2021; 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 (Likens, 2017; Peters et al., 2004; Taylor et al., 2021).

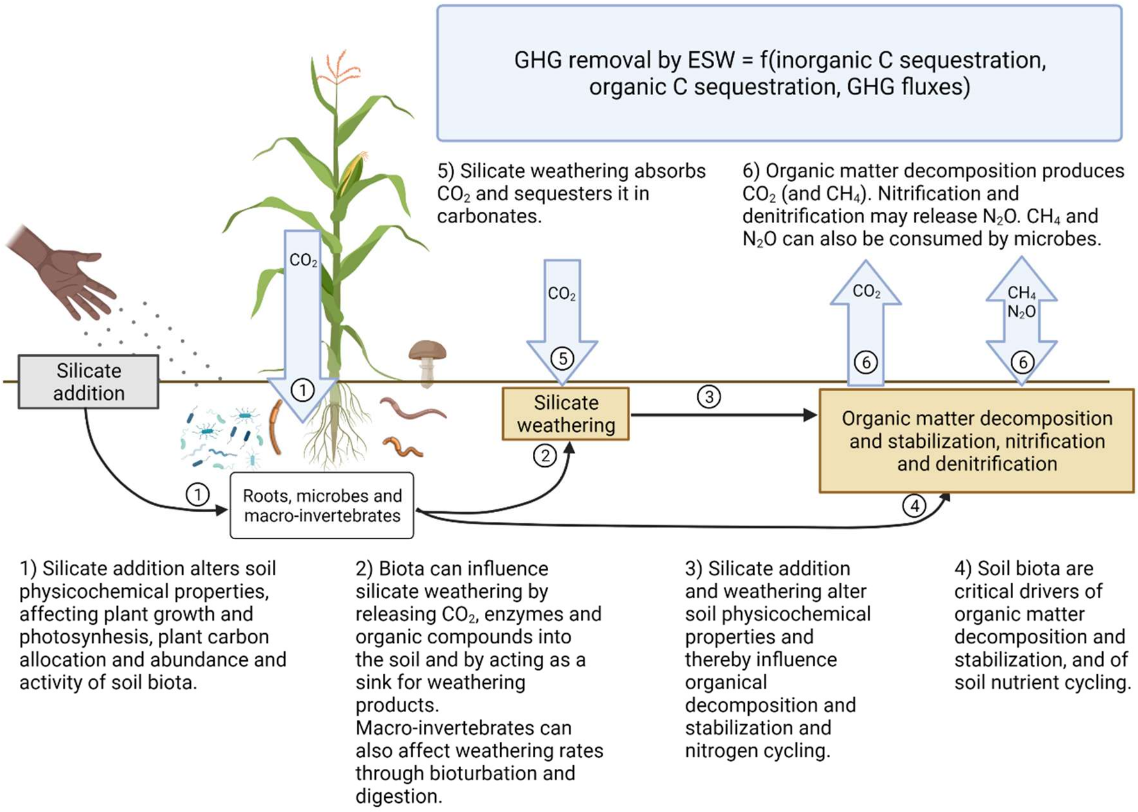
Early lab experiments and modelling indicate the highest potential for ESW on cation depleted soils in humid and warm environments (Amann & Hartmann, 2019). Estimates of the global inorganic C sequestration potential of ESW range widely between 0.5 and 5 Gt CO_2 y^{-1} (depending on cost assumptions, among others; Beerling et al., 2020; Fuss et al., 2018; Goll et al., 2021). 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 uncertainties concern the *in natura* weathering rate, the co-benefit of increased plant growth and associated C sequestration (Fuss et al., 2018; Goll et al., 2021). 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 the 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 might 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.



5) Silicate weathering absorbs CO_2 and sequesters it in carbonates.

6) Organic matter decomposition produces CO_2 (and CH_4). Nitrification and denitrification may release N_2O. CH_4 and N_2O can also be consumed by microbes.

1) Silicate addition alters soil physicochemical properties, affecting plant growth and photosynthesis, plant carbon allocation and abundance and activity of soil biota.

2) Biota can influence silicate weathering by releasing CO_2, enzymes and organic compounds into the soil and by acting as a sink for weathering products. Macro-invertebrates can also affect weathering rates through bioturbation and digestion.

3) Silicate addition and weathering alter soil physicochemical properties and thereby influence organical decomposition and stabilization and nitrogen cycling.

4) Soil biota are critical drivers of organic matter decomposition and stabilization, and of soil nutrient cycling.

***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 (Burghelea 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). Note that this plant uptake can also affect estimation of weathering rates based on soil concentrations of these elements (and not on inorganic C pools and fluxes). By releasing protons and CO_2, roots reduce soil pH and increase the CO_2 concentration in the rhizosphere (Lenzewski et al., 2018), both of which stimulate mineral weathering (Harley & Gilkes, 2000). Plant roots also exude organic compounds such as malate or citrate that can for example protect the plant from Al intoxication (Ryan et al., 2001), while also stimulating mineral weathering by chelating reaction products and dissolving silicate minerals (Dontsova et al., 2020; Drever, 1994; Zhang & Bloom, 1999). 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.

Plant effects on ESW are expected to differ among species, and this is likely (in part) related to nutrient acquisition strategy. Haque et al. (2019b), for example, reported that in soils treated with wollastonite, weathering rates were higher with leguminous beans than with non-leguminous corn or for bare soil without plants. Most leguminous plants such as beans and soybean live in symbiosis with nitrogen fixing bacteria and the H^+ excreted during N_2 fixation by legumes acidifies the soil. This acidification is more pronounced for temperate than for tropical legumes (Bolan et al., 1991), which may lead to differences in their effect on ESW between climatic regions. Moreover, exudation of proteins, phenols, sugars and free amino acids may even differ among genotypes, as has been reported for soybean (Krishnapriya & Pandey, 2016) and maize (Gaume et al., 2001). This may open possibilities for engineering of plant-soil combinations optimized for climate change mitigation through ESW.

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; Burghelea et al., 2015; Burghelea et al., 2018; Zaharescu et al., 2020). Given that mycorrhizal fungi depend on their host for C, their influence on ESW is likely to be strongly related to plant activity and plant 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). As for plants, fungi and other microbes can also stimulate weathering by acting as a sink for weathering products (Oelkers et al., 2015). Fungal hyphae are very thin and can therefore interact with surfaces 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 anhydrases (CA) which are found within all domains of life and play a fundamental role for respiration, CO_2 transport and photosynthesis, have a combined effect of both increasing silicate weathering and carbonate precipitation. A few studies have been able to show increased weathering of silicates and carbonates with added CA (Xiao et al., 2015; Zaihua, 2001). CA catalyzes the equilibrium reaction between CO_2 and bicarbonate ions, which in contact with the free metal ions from weathering of silicates, combine to form solid carbonate precipitates such as calcite ($CaCO_3$), magnesite ($MgCO_3$), dolomite ($CaMg(CO_3)_2$) or siderite ($FeCO_3$). The abiotic process of carbonate precipitation is slow and requires pH values higher than 8, whereas the addition of CA accelerates this reaction considerably (Bose & Satyanarayana, 2017). In fact, CA is one of the fastest enzymes, performing up to 10^6 CO_2 conversion reactions per second. CA is most efficient at high pH and may thus be especially important for ESW in alkaline soils.

Recently, there has been an increased interest in using CA in the industrial and agricultural sector for C sequestration and enhanced crop growth. Industrial slag waste from the steel industry is regularly used as a soil fertilizer due to its composition of bio-essential nutrients such as phosphates, silicates and trace elements, which can increase crop productivity (Reddy et al., 2019; Wang et al., 2018b). Das et al. (2019b) suggested that the use of CA-containing bacteria in slag-fertilized soils could accelerate the weathering of the silicate-containing slags and hence C sequestration.

An enhanced microbial expression of CA genes will promote the generation of H_2CO_3 and a concomitant increased silicate weathering and a release of the bio-necessary nutrients. Some organisms adapt to increasing CO_2 levels by downregulating the gene expression of CA (Xiao et al., 2015). Experiments with fungi have shown that one way to keep an upregulated CA expression despite high CO_2 concentrations is to limit K availability and adding K-feldspar as the only available source of K (Sun et al., 2013; Xiao et al., 2012). This can be seen also in environments with Ca deficiency, where silicates are the only available source for Ca (Xiao et al., 2015). High concentrations of Zn and Fe, on the other hand, stimulate CA activity, while complex binding of Zn is a strong inhibitor of zinc metalloenzymes such as CA (Borja et al., 1998).

Urease is another enzyme used by prokaryotes and eukaryotes for efficient biomineralization. Urease is a nickel metalloenzyme that catalyzes the conversion of urea to ammonia with the side-effect of raising pH, which in turn stimulates carbonate precipitation. As with CA, urease increases pH locally and is inhibited by low pH. 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.

On the other hand, high urease activity can be undesirable in agriculture. Urea ammonium nitrate (UAN) is a commonly used fertilizer. When added to soil, UAN is quickly converted to ammonia and volatilized to the atmosphere (Wang et al., 2020b), leading to fertilizer losses and increasing emissions of the potent greenhouse gas N_2O. There is thus a great need for a more efficient use of N in fertilizers and since urease is the main enzyme responsible for conversion of urea to NH_3, urease inhibitors have effectively been used for lowering the volatilization of urea and increasing crop yield (Drury et al., 2017; Mira et al., 2017; Wang et al., 2020b). Humic acids are among the more efficient inhibitors of urease. Humic acids irreversibly inhibit the hydrolytic decomposition of urea (Liu et al., 2019) and concomitantly reduce urease-induced carbonate precipitation. On the other hand, the natural concentration of humic acids in soils is likely too low to have a profound impact on the precipitation capacity of urease (Al-Taweel & Abo-Tabikh, 2019; Moghal et al., 2020).

While urease can stimulate silicate weathering through carbonate biomineralization, agricultural practices aimed at reducing urease activity can limit this effect. An alternative pathway that would reconcile the interest in C sequestration and reduction of N losses is to inhibit the total conversion of urea to gas by increasing the efficiency by which plants and/or microorganisms make use of the added urea fertilizer. Interestingly, the addition of Ni - the urease co-factor and present in several silicate materials - may aid in this regard. Laboratory studies have shown that supplementation of Ni to the soil increased health and growth rate of lettuce plants (Khoshgoftarmanesh et al., 2011; Oliveira et al., 2013). Adding silicate materials containing Ni may thus stimulate biomineralization of $CaCO_3$ by urease, and hence C sequestration (Bachmeier et al., 2002), while at the same time stimulating plant growth and reducing urea volatilization. It is, however, not yet fully understood how the net fertilizer efficiency and gas exchange rate will develop on a larger timescale (Tosi et al., 2020) and more research is needed to investigate the effects of combined urease and silicate addition on greenhouse gas emissions and plant growth.

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 of 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 estimated dissolution rates that were 60 to 330 times higher than in the control plots. On the one hand, ants may thus be 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 unravel the mechanisms involved, including interactions with other biota, and to quantify their potential effect on ESW.

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 environmental, 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; Kelland et al., 2020; Swoboda et al., 2021; Taylor et al., 2021; Van Straaten, 2006), although this is not always the case (Haque et al., 2020c; Swoboda et al., 2021; 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).

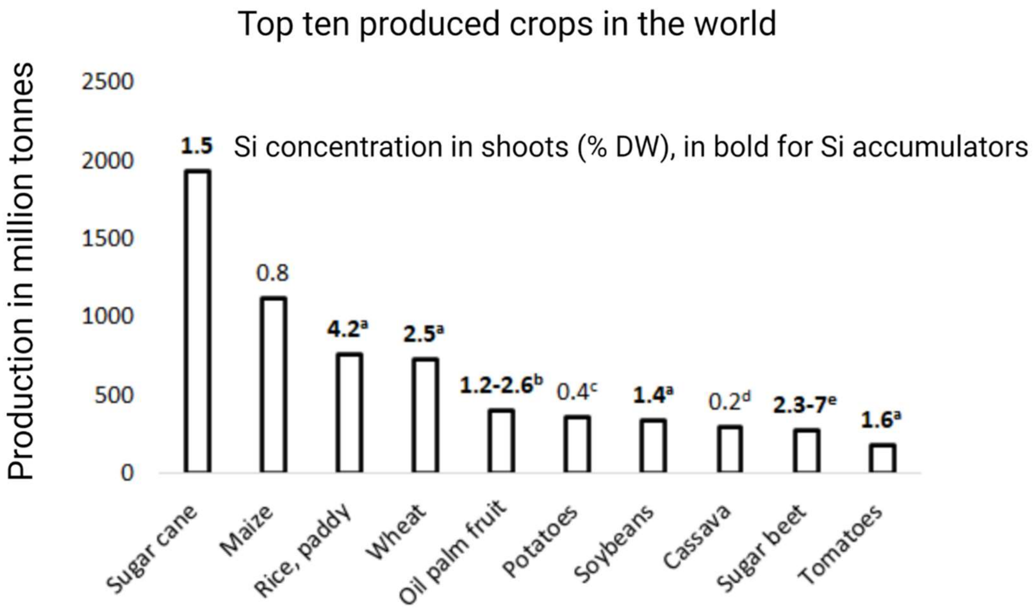


***Figure 3**: Top ten produced crops in the world in 2018 (FAOSTAT, 2018). Seven of these crops are classified as Si accumulators (>1.0% Si of dry weight (DW)). The values above the bar are average shoot Si concentrations. [a] compiled from Hodson et. al (2005); [b] averages compiled from Munevar & Romero (2015); [c] estimated averages of the data (Solanaceae) compiled by Hodson et. al (2005); [d] estimated averages of the data (Euphorbiaceae) compiled by Hodson et. al (2005); [e] averages computed from the data of 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).

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). An increase of xylanase, phosphatase, and protease activity - essential enzymes for the breakdown of organic matter - could increase soil CO_2 emissions. However, even though rock powder additions increased the protease content of both the Stagno-Mollic Gleysol and the Calcaric Regosol and Cambisol, CO_2 emissions and microbial biomass only increased for the Regosol and Cambisol. The rock powders also increased the nitrification and nitrate contents of the Regosol and Cambisol, which could increase N_2O emissions. Simultaneous N_2O reductions might however be achieved through a reduction of soil acidity, as discussed in detail below. These findings illustrate that the effect of ESW on microbial communities depends on 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 CH_4 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., 2020a), 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 and higher SOC stocks. Moreover, 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 (Paradelo et al., 2015).

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: turnover of new C inputs and modification of organic matter present in the soil. 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 during decomposition of recalcitrant litter are generally higher than C losses from more labile inputs (Cotrufo et al., 2013). This is because a larger fraction of the labile C can be converted into microbial biomass and microbial products. The close association between microbes and soil mineral surfaces then explains the greater stabilization of labile C inputs than of recalcitrant C inputs (Cotrufo et al., 2013). As with liming, silicate addition may increase plant C inputs and/or its nutrient concentrations (Forey et al., 2015; Melvin et al., 2013; Paradelo et al., 2015) and hence increase SOM stabilization.

Liming and silicate addition can affect SOM formation and decay via altered activity of extracellular enzymes, driven by the modified 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). Increased pH upon silicate addition can thus accelerate the decomposition of plant litter and SOM (Leifeld et al., 2013), resulting in reduced litter and SOC stocks, but the improved living conditions are likely to result in enhanced microbial growth and thus formation of stabilized SOM.

Aggregate formation is also a key SOM stabilization mechanism that can be increased by the presence of secondary minerals formed during 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, enzyme activities 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 weathering rates, creating a feedback loop. For example, faster turnover of higher-quality litter will increase the soil CO_2 concentration, impacting mineral dissolution. At the same time, increased litter turnover enhances dissolution of organic matter (Cotrufo et al., 2013), and thus increase the potential of organic compounds to either form stable organo-mineral complexes or aid in the weathering. Overall, the balance between the effects on plant C inputs, litter decomposition and SOM stabilization will determine the net effect of silicate addition on SOC sequestration. In the case of liming, a literature review by Paradelo et al (2015) showed that SOC stocks generally increased with liming in mineral soils. In organic soils and (acid) organic soil horizons, increased mineralization rates upon liming appear more likely to reduce SOC stocks (Lundström et al., 2003; Paradelo et al., 2015).

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 CO_2, especially N_2O (Fig. 1; Beerling et al., 2018). Total annual N_2O emissions from soils in natural and agricultural systems together represent about 55% of all global N_2O sources (Tian et al., 2020). Agricultural soils are a major source of N_2O to the atmosphere due to the high amount of mineral fertilizers that increase microbial N availability (Guenet et al., 2021). Soil moisture is a key determinant of soil N_2O emissions (Firestone & Davidson, 1989) and changes in soil hydrology following silicate addition can thus influence N_2O emissions (among others depending on soil texture and size of the silicate grains). Also soil pH influences N_2O emissions; low pH decreases the activity of N_2O reductase, stimulating the release of N_2O as an intermediate product of the denitrification process (Hu et al., 2015; Liu et al., 2010). Silicate addition to acid soils is expected to buffer pH and thus reduce N_2O emissions by increasing the N_2:N_2O ratio (i.e., enhancing complete denitrification; Blanc-Betes et al., 2021), similar to what has been reported for liming (Hénault et al., 2019). In aerobic soils, however, reduced N_2O release from denitrification may be counterbalanced by increased N_2O release during nitrification, as pH increases stimulate nitrification and favor ammonia oxidizing bacteria over ammonia oxidizing archaea, with the former producing more N_2O (Nadeem et al., 2020).

Other interactions with biota arise here as well. For example, mycorrhizal fungi have been shown to reduce N_2O emissions (Storer et al., 2018), potentially enhancing this anticipated co-benefit of ESW, whereas earthworms have been reported to increase N_2O emissions (Augustenborg et al., 2012; Lubbers et al., 2013). In some soils, earthworm activity may account for more than 50% of the total soil N_2O 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 N_2O 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 N_2O 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 N_2O can be of huge importance in agricultural soils, methane (CH_4) typically is not. CH_4 production is a strictly anaerobic process. In aerobic soils CH_4 oxidation typically exceeds CH_4 production, making these soils modest CH_4 sinks (Dutaur & Verchot, 2007). Rice fields, however, are an important source of CH_4 emissions due to their waterlogged anaerobic soils (Saunois et al., 2020). Some studies have reported a decrease in CH_4 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 CH_4 emissions by reducing methanogenesis and/or increasing CH_4 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 CO_2 (Das et al., 2019b; Gwon et al., 2018). On the other hand, increased plant productivity in response to silicate addition may increase CH_4 emissions by increasing plant belowground C input quantity and quality (Ku et al., 2020) and enlarged aerenchyma due to higher root biomass might further increase CH_4 funneling to the atmosphere (Kim et al., 2018; Ku et al., 2020). Hence, the net effect of silicate addition on CH_4 emissions will depend on the balance between these counteracting processes.

As illustrated above, silicate addition can have diverging effects on the release of CO_2, CH_4 and N_2O, from soils and ecosystems. Reductions in the emission of one of these GHGs might be counteracted by increases in another. Ku et al. (2020), for example, reported a reduction in N_2O emissions from a rice field amended with a calcium silicate, but CO_2 and CH_4 emissions increased more, leading to an increase in global warming potential of the cumulative GHG emissions. This illustrates the importance of considering the emissions of all three of these GHGs when assessing the climate change mitigation potential of ESW, and its interaction with the biota.

Advances in modelling ESW

Models are based on the theoretical understanding of the involved processes and observational data to parameterize mathematical formulations. As a consequence, few modelling studies address interactions between ESW and biota. Most studies are limited to the dissolution reactions, removal of weathering products, abiotic CO_2 drawdown (e.g., Rinder & von Hagke, 2021; Strefler et al., 2018), and impact on soil hydrology (de Oliveira Garcia et al., 2020). Nonetheless, first models are emerging which include interactions between biota and weathering rates. Goll et al. (2021) used a comprehensive land surface model coupled to a model of mineral dissolution to simulate the effect of nutrient release from basalt on plant growth and ecosystem carbon storage. Cipolla et al. (2021) coupled an ESW component to a ecohydrological-biogeochemical soil model to investigate the combined contributions of hydrology and plants to weathering rates. Beerling et al. (2020) used a one-dimensional vertical reactive transport model with steady-state flow, and a source term representing rock grain dissolution which includes an empirical formulation for the combined effect of biotic processes that accelerate the physical breakdown and chemical dissolution of minerals.

Land surface models which resolve the water, energy and biogeochemical cycles in plant and soils coupled to weathering models can provide the means to study the full effect of ESW on biota and vice versa. The increasing realism of belowground processes in such models provide the basis to integrate the emerging data from experiments in biologically active soils, mesocosm and field experiments (e.g., Kelland et al., 2020).

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 CO_2 concentrations often increase plant growth and belowground C inputs (Terrer et al., 2021), which could in turn stimulate ESW and SOC sequestration. In addition, the nutrient limitation on the CO_2 fertilization effect may be (partly) alleviated by ESW treatments (Goll et al., 2021; Terrer et al., 2019). 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. Si accumulation in plants can reduce plant water losses (Guntzer et al., 2012) and K release through weathering may improve plant 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 and this should consider interactions with nutrient cycling (Vicca et al., 2018) and other important 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 or GHG emission reduction 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 or alkaline 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 (Amann & Hartmann, 2019). By increasing the weathering rate with the help from biota, drawdown of CO_2 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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