Combating Climate Change Through Enhanced Weathering of Agricultural Soils

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Rising levels of atmospheric carbon dioxide (CO_2) are driving increases in global temperatures. Enhanced weathering of silicate rocks is a CO_2 removal technology that could help mitigate anthropogenic climate change. Enhanced weathering adds powdered silicate rock to agricultural lands, accelerating natural chemical weathering, and is expected to rapidly draw down atmospheric CO_2. However, differences between enhanced and natural weathering result in significant uncertainties about its potential efficacy. This article summarizes the research into enhanced weathering and the uncertainties of enhanced weathering due to the key differences with natural weathering, as well as future research directions.

KEYWORDS: enhanced weathering, chemical weathering, climate change, carbon dioxide removal technologies, negative emission technologies, agriculture

ENHANCED WEATHERING AS A CARBON DIOXIDE REMOVAL TECHNOLOGY

Anthropogenic emissions of carbon dioxide (CO_2) are increasing global temperatures via the greenhouse effect. The Intergovernmental Panel on Climate Change (IPCC) "business as usual" emission scenarios [the representative concentration pathway (RCP) model that considers radiative forcing values of 8.5 W/m^2 by the year 2100] project that global temperatures could rise by between 2.6 °C and 4.8 °C, compared to the reference period 1986–2005, by the end of the 21st century (IPCC 2013). The 2015 Paris Agreement, under the United Nations Framework Convention on Climate Change, established the target of limiting global warming to below 2 °C. To meet this goal, carbon dioxide removal technologies are required to actively withdraw CO_2 from the atmosphere. Enhanced weathering (EW) of silicate rocks is a carbon dioxide removal technology that could help mitigate anthropogenic climate change by reducing atmospheric CO_2 levels.

The principle behind EW is similar to that of natural, geologic-timescale chemical weathering: when silicate minerals are dissolved in a reaction with atmospheric CO_2 and water, the products of this reaction, which include calcium (Ca^{2+}), magnesium (Mg^{2+}), and bicarbonate (HCO_3^-) ions, are transported to the ocean by rivers (FIG. 1) and are stored as carbonate minerals on geological timescales, thereby locking up atmospheric CO_2 in ocean sediments (Kasting 2019 this issue). Natural chemical weathering

occurs slowly and consumes ~0.26 PgC y^{-1} of atmospheric CO_2 as HCO_3^- (Hartmann et al. 2009), as compared to the ~10 PgC of atmospheric CO_2 released per year by fossil fuel combustion. Enhanced weathering speeds up chemical weathering rates such that CO_2 is scrubbed from the atmosphere in meaningful quantities on human timescales (FIG. 2). Estimates suggest that if deployed on a sufficiently large scale, EW could draw down from <1.0 PgC y^{-1} up to ~30 PgC y^{-1} as HCO_3^- (Köhler et al. 2010; Taylor et al. 2016; Strefler et al. 2018) and reduce the rise in atmospheric temperatures predicted for 2100 (assuming "business as usual" emission scenarios) by between 0.2 °C (Köhler et al. 2010; Taylor et al. 2016) and 1.6 °C (Taylor et al. 2016).

Enhanced weathering operates through the application of powdered silicate rock to soils (FIG. 1), although application directly into the ocean is also under investigation (Renforth and Henderson 2017). The theory behind EW has developed from the extensive literature on the controls on natural chemical weathering rates. Laboratory trials have shown that finely ground, unweathered rock has a high reactive surface area, leading to increased weathering rates (White and Brantley 2003). Powdered mafic rock (e.g., basalt) and ultramafic rock are those most commonly proposed for EW due to their relatively high proportion of olivine, a rapidly weathering, Mg-bearing silicate mineral (Hartmann et al. 2013). Agricultural lands are now the target for EW because interactions with plant roots and fungal hyphae, as well as organic acids, increase weathering rates, and because these sites are equipped with the infrastructure to apply rock powder on large spatial scales. Strefler et al. (2018) suggest that 5.1 Mkm^2 of tropical and 2.8 Mkm^2 of temperate croplands are suitable for treatment. Enhanced weathering is predicted to be most effective in tropical climates due to the relationship between higher temperatures and/or rainfall and higher chemical weathering rates (Edwards et al. 2017).

Furthermore, EW is an attractive potential carbon dioxide removal technology because, in addition to removing CO_2 from the atmosphere, there may be substantial co-benefits. An extensive literature exists on the application of powdered basalt to croplands. This work has largely investigated rock powder as a sustainable and cheap alternative to chemical fertilizers and demonstrated that it improves soil quality and increases crop yields (Van Straaten 2007; Beerling et al. 2018). Through increased uptake of silicon from soil following rock powder treatment, crops also have increased

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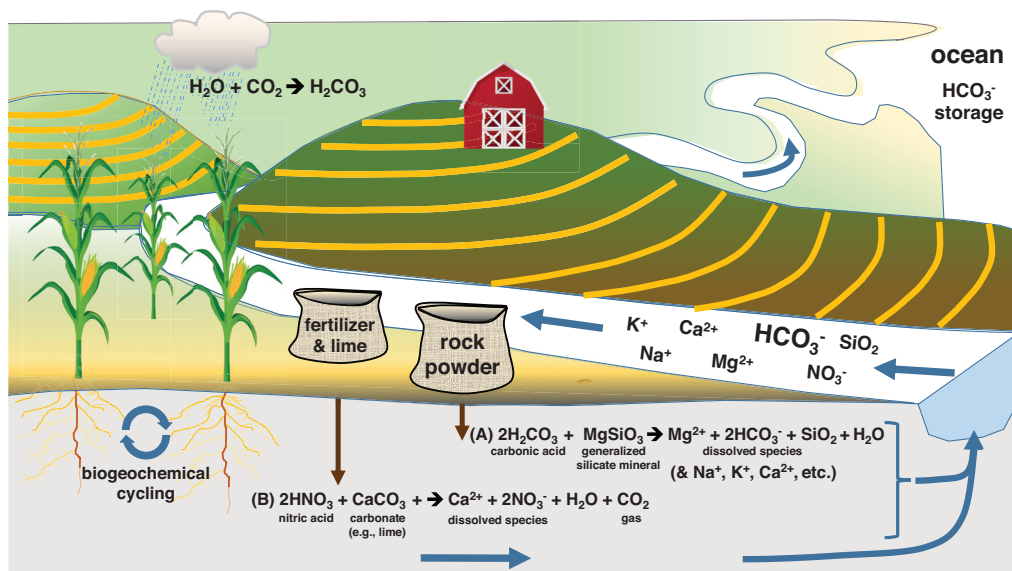 E-mail: L.L.Taylor@sheffield.ac.uk

FIGURE 1 Diagram of enhanced weathering. Equation A is a generalized version of the fundamental weathering reaction behind enhanced weathering, that of silicate rock powder weathered by carbonic acid. Equation B is an example of weathering between nitric acid (derived from fertilizer) and carbonate, such as that which comprises lime. For a more complete roster of possible weathering reactions associated with enhanced weathering, see Hartmann et al. (2013).

resistance to pests and disease (Beerling et al. 2018). Due to these interactions, EW may help alleviate the threat to global food security posed by climate change, as well as reduce the need for harmful fertilizers.

Ocean acidification is another negative side effect of climate change. It causes the dissolution of carbonate structures in marine organisms such as corals, as well as plankton and some shellfish (e.g. oysters, scallops) (IPCC 2013). Some fisheries are threatened by ocean acidification, as are economies based on organisms further up the food chain through disruption to the marine food web. Through the addition of HCO_3^- to the ocean, EW could help reverse ocean acidification in coastal waters (Köhler et al. 2010; Taylor et al. 2016; Strefler et al. 2018).

Here, we summarize the different approaches scientists are taking to assess EW as an efficient and safe carbon dioxide removal technology. We also examine the key differences between EW and natural chemical weathering and how these differences introduce challenges and uncertainties to quantifying atmospheric CO_2 sequestration via EW. Finally, we discuss essential areas for further research.

DOES ENHANCED WEATHERING WORK?

At present, very few laboratory or field studies have been conducted with an explicit focus on EW. Studies that examined rock powder as a fertilizer typically report crop growth and yield under variable rates of basalt powder application, but only occasionally provide ion concentrations of soil water or exchangeable soil fractions from which chemical weathering rates can be gleaned (e.g., Anda et al. 2013, 2015; ten Berge et al. 2012). Despite significant inter-study differences in rock powder composition, grain size, and application rates, as well as starting soil material, vegetation, and climate conditions, such studies converge on three overarching conclusions. First, the application of powdered basalt to soil increases total chemical weathering rates (FIG. 3). Second, higher basalt application rates yield higher total chemical weathering rates (FIG. 3). Third, weathering efficiency declines with increased basalt load (FIG. 4). It is worth noting, however, that the experimental design of these studies is distinct from that commonly proposed for EW in that rock powder was applied only at the onset of each experiment, whereas

EW models often assume repeated, annual application of rock powder. The impact of repeated rock powder application on soil saturation and chemical weathering rates over longer timescales, therefore, remains unclear. The studies above also monitored ion concentrations at some depth in the soil column. Chemical weathering and CO_2 consumption rates from EW derive from ion concentrations in discharge (FIG. 1), as this captures the net carbon sequestration following all chemical processes which occur throughout the soil column and during hillslope transport (e.g., CO_2 degassing).
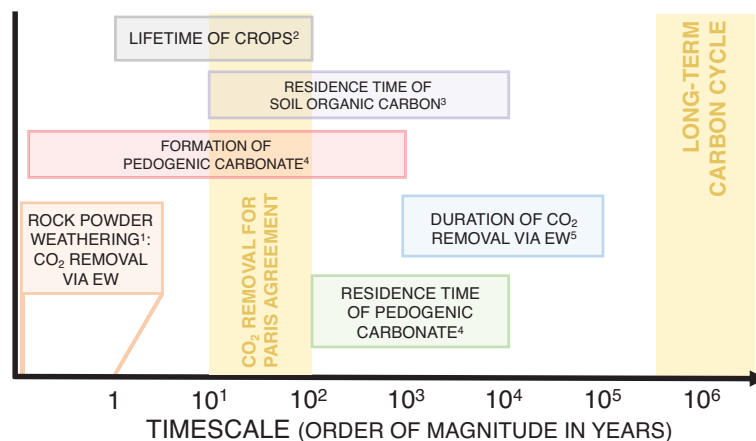

FIGURE 2 Carbon-cycle timescales relevant to enhanced weathering (EW). Superscript notes (1–5) are as follows: [1] Under field conditions, rock powder is likely to dissolve on timescales ranging from nearly instantaneous (e.g., calcite) to multiple years for some silicate minerals. [2] Crops such as maize are harvested in their entirety annually, whereas tree crops typically have their commercial products (e.g., fruits) harvested (sub-)annually but the tree itself could live for over a hundred years (e.g., apple trees). [3] Although the mean residence time of topsoil organic carbon is on decadal to centennial timescales, the residence time of deep soil organic carbon can extend to >10^4 years (Schmidt et al. 2011). [4] Pedogenic carbonate forms on timescales ranging from days to 10^3 years, and persists in the soil. The mean residence time of pedogenic carbonate is ~80,000 years (Zamanian et al. 2016). [5] Some C removed from the atmosphere by EW will become part of the long-term C cycle and remain sequestered for >10^5 years.
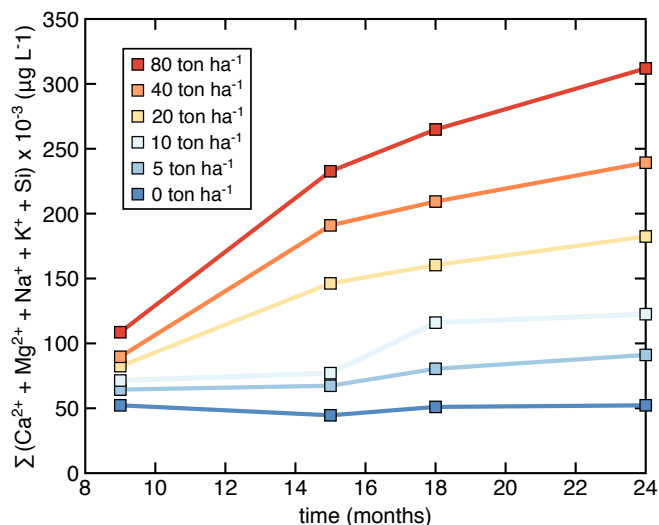
FIGURE 3 Total dissolved cation load through time for a series of abiotic soils with variable rates (figure key) of basalt powder (<50 μm) application (Anda et al. 2015). Total dissolved cation load is proportional to total chemical weathering rate (W_{total}), when W_{total} (g km^2 y^{-1}) = $\sum$(Ca^{2+} + Mg^{2+} + Na$^+$ + K$^+$ + Si) (μg L^{-1}) × runoff (mm y^{-1}), when runoff is assumed to be equal across all manipulations because of the identical pot size and uniform addition of water to treatments. The data indicate that total dissolved cation load, and therefore total chemical weathering rates, increase with time and with greater basalt powder application rates.
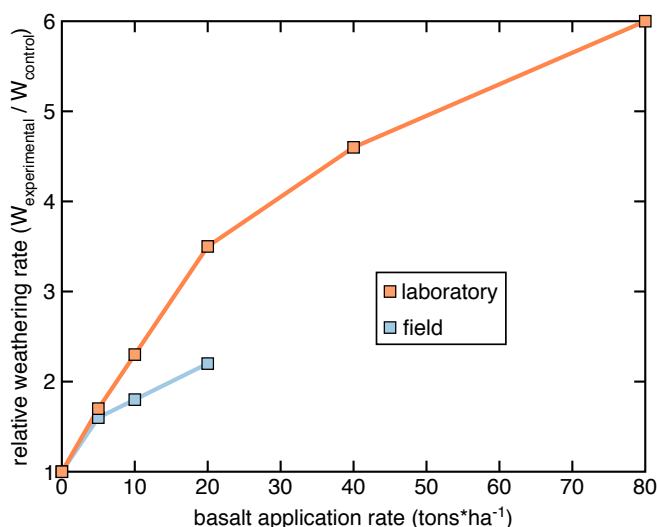

FIGURE 4 Decline in weathering efficiency (i.e., weathering per ton of basalt powder) with increasing basalt powder application rates in both an abiotic laboratory trial ("laboratory"), having particle sizes <50 μm (Anda et al. 2015), and a small-scale field trial with cocoa plants ("field") having particle sizes ≤250 μm (Anda et al. 2013). Weathering rates (see FIG. 3 caption) of experimental manipulations ($W_{experimental}$) have been normalized to their respective control data ($W_{control}$) to generate a relative weathering rate and to facilitate inter-study comparison. In calculating a relative weathering rate, runoff is canceled out based on the assumption that runoff is identical in all manipulations. This assumption is based on either the identical pot size and uniform addition of water to treatments under laboratory conditions (Anda et al. 2015) or the equal area, similar slopes, and close geographic proximity of field plots (Anda et al. 2013).

A few ongoing field trials of EW are currently underway (e.g., Andrews et al. 2018; McDermott et al. 2019) (FIG. 5). Following two years of rock powder treatment to maize crops in the American Midwest, preliminary data from drainage waters do not demonstrate an increase in total weathering rate following basalt application (Andrews et al. 2018). Conversely, basalt powder treatment to soils in Northern Ireland demonstrated a doubling of pore-water cation concentrations (e.g., Ca, Mg) in less than a year (McDermott et al. 2019). These apparently contradictory results may be, in part, reconciled by considering the different water reservoirs which were analyzed, as well as the impact of crops in the former study. A useful analog is an experiment conducted at the Hubbard Brook Experimental Forest (New Hampshire, USA), where, following application of the silicate mineral wollastonite ($CaSiO_3$) to a temperate, forested catchment, treatment-derived Ca from soils was not observed in stream water for the first three years due to Ca uptake by vegetation (Shao et al. 2016). Ultimately, the efficacy of EW at any locality will depend on many site-specific factors related to the existing biology, geology, and hydrology.

FIGURE 5 Contrasting methods of basalt powder application in two field trials of enhanced weathering for the Leverhulme Centre for Climate Change Mitigation at the University of Sheffield (UK). (LEFT) Basalt powder is dispersed in a corn field by a machine traditionally used for lime spreading. Location: Energy Farm at the University of Illinois at Urbana-Champaign (USA). PHOTO CREDIT: ILSA KANTOLA. (RIGHT) At an oil palm plantation, basalt powder is broadcast by hand in line with standard fertilizer application practice. Location: Sabahmas Oil Palm Plantation (Wilmar International Ltd.), Sabah (Malaysia). PHOTO CREDIT: CHRISTOPHER PEARCE.

Future laboratory and field trials will provide valuable opportunities to characterize the controls on EW and its capacity to sequester atmospheric CO_2. Laboratory studies allow for comparative tests that examine the impact of isolated variables on EW before scaling up to more involved and costly field trials. Variables that are likely best investigated in the laboratory include the impact of rock powder grain size and mineral composition, depth of rock powder transport under various tillage practices, as well as the effect of different crop species on weathering. To understand the potential efficacy of EW in the coming decades, future climate change scenarios can also be simulated in the laboratory, including higher atmospheric pCO$_2$ levels. On the other hand, it is commonly observed that weathering rates derived in the laboratory substantially exceed those documented in the field (White and Brantley 2003) and riverine chemistry can change during downstream transport towards the ocean. Field trials are, therefore, essential for determining the true effectiveness of EW as a carbon dioxide removal technology. Quantification of chemical weathering rates and CO_2 consumption rates from EW field

trials will require targeted approaches that monitor, at a minimum, discharge amounts and chemical properties such as major cation and anion concentrations, dissolved organic- and inorganic carbon concentrations, and likely cation source tracers such as radiogenic Sr isotope ratios ([87]Sr/[86]Sr). Given that silicate and carbonate weathering reactions balance HCO_3^- production with different cation stoichiometry, then mass balance equations that disentangle the proportion of dissolved species from each of these weathering reactions (in particular those pertaining to nitric acid weathering) must be developed to generate an accurate estimate of CO_2 sequestration from EW field data. Field trials also provide an opportunity to trouble-shoot the logistics of implementing EW. These trials also allow for the monitoring of environmental and public health concerns, such as biodiversity and water quality (e.g., heavy metal release, water turbidity). We note that ultramafic rocks may contain substantial quantities of heavy metals (Taylor et al. 2016; Beerling et al. 2018).

DIVERGING FROM NATURE

Timescales of Weathering Reactions

Although EW is conceptually developed from our understanding of geologic-timescale chemical weathering, the two processes differ in ways that introduce challenges to quantifying the ability of EW to sequester atmospheric CO_2. A key difference is the timescale of weathering. Whereas natural chemical weathering works on hundred thousand- to million-year timescales (i.e., geologic), EW is largely concerned with CO_2 sequestration on decadal to centennial timescales (FIG. 2). Short-term processes that can be dismissed in the study of natural chemical weathering rates are, therefore, significant to EW. For example, on geologic timescales, atmospheric CO_2 is not considered truly sequestered until it is locked into marine carbonate, limiting the relevant silicate mineral weathering reactions to minerals containing Ca and Mg. Although more research is needed on the longevity of C storage in the ocean (Renforth and Henderson 2017), both the millennial timescales of $CaCO_3$ compensation (Archer 2003) and the HCO_3^- residence time of ~10^5 years (Renforth and Henderson 2017), suggest sufficient longevity to be relevant for climate change mitigation, equal to or greater than the terrestrial sinks of biomass and soil (FIG. 2). On these timescales, any weathering reaction that transfers atmospheric CO_2 to riverine HCO_3^-, such that it is ultimately stored in the ocean (FIG. 1), is a pertinent atmospheric CO_2 sequestration pathway for EW. Thus, HCO_3^- generation via weathering of non–Ca- and Mg-bearing silicate minerals, as well as weathering of carbonates, will sequester atmospheric CO_2. One important consequence of this is that carbonate weathering, which results in no net change to atmospheric CO_2 levels over geological timescales (Kasting 2019 this issue), can lead to drawdowns of CO_2 that are significant over the timescales of anthropogenic climate change. The above accounting of weathering pathways assumes that carbonic acid (H_2CO_3) is the weathering agent, as is typically the case with natural chemical weathering (FIG. 1) but that may not be exclusively true with EW (see below). Additionally, pedogenic (soil) carbonate is a secondary mineral which forms from chemical weathering. It has a mean residence time in soil of ~80,000 years (Zamanian et al. 2016), which makes its formation inconsequential to geologic-timescale weathering but a relevant additional CO_2 sink to EW, as long as the carbon comprising the pedogenic carbonate originated from silicate mineral weathering (FIG. 2). If, on the other hand, pedogenic carbonate carbon was derived from carbonate mineral weathering, then its formation has no impact on atmospheric CO_2 reserves.

Agricultural Lands

Whereas natural chemical weathering research has almost exclusively focused on pristine environments, EW is being largely considered for agricultural lands (Hartmann et al. 2013). These areas are often subject to heavy applications of nitrogen (N) fertilizers. The nitrogen cycle is of little importance to natural chemical weathering, but it has a strong influence on the chemistry of agricultural soils. Nitrogen fertilizers can generate nitric acid (HNO_3) and participate in chemical weathering (Fortner et al. 2012; Hartmann et al. 2013) (FIG. 1). Nitric acid weathering of silicate minerals will have no impact on atmospheric CO_2 levels, but under high concentrations of nitric acid, weathering of carbonate minerals will add CO_2 to the atmosphere (Hartmann et al. 2013) (FIG. 1). Carbonate minerals are widespread in agricultural lands due to the use of lime, and calcite is a common trace mineral in silicate rocks.

Nitrogen fertilizers also indirectly affect weathering in the soil column because root uptake of ammonium (NH_4^+) acidifies the surrounding soil, but nitrate (NO_3^-) uptake acts to raise the local pH (Taylor et al. 2017). Furthermore, although inorganic N is released from decomposing organic matter as NH_4^+, which may be retained on soil exchange sites, nitrification processes convert NH_4^+ to non-exchangeable, highly mobile NO_3^-, potentially leading to nitric acid weathering. Easily leached NO_3^- may also compromise the greenhouse-gas balance for a watershed by reducing dissolved inorganic carbon export and by possible in situ or downstream denitrification and release of nitrous oxide (N_2O), a potent greenhouse gas, to the atmosphere. The impact of N fertilizers on weathering will vary with crop type. Legumes, for example, host N-fixing bacteria on their roots such that N is adsorbed directly by the plant, eliminating the need for N fertilizers. Conversely, crops such as maize inefficiently recycle N and, thus, require substantial applications of N fertilizer. Waters draining maize fields exhibit high NO_3^- concentrations as a result of both the extensive application of N fertilizers and annual tillage (Smith et al. 2013).

The natural weathering literature has established that plants can dramatically increase chemical weathering rates (Porder 2019 this issue), but agricultural ecosystems contain different plant species, and significantly reduced biodiversity, than natural settings. Under monoculture conditions, it will be particularly important to identify which crops best induce chemical weathering. Biological enhancement of grain-scale weathering rates will be highest at depths where high microbial activity and root respiration of atmospheric CO_2 lead to high soil CO_2 concentrations, yet nutrient and water cycling beneath crops may cause significant delays between weathering in soils and increased fluxes of weathering products to the ocean. Additionally, biomass can sequester some weathering products which are then removed from the landscape at harvest, thereby disrupting natural recycling of carbon and nutrients. Productivity of crops may increase in response to EW before a significant carbon dioxide removal effect can be demonstrated from stream water chemistry.

Rock Powder Surface Area and Mixing Depth

The dependence of chemical weathering rates on temperature, runoff, and biology has been extensively documented in laboratory experiments, as well as through empirical relationships based on field data at the regional or global scale (Li et al. 2016; Porder 2019 this issue). Crucially, weathering rates are also constrained by the reactive surface area of mineral grains, which increases as particles are ground to smaller sizes. Although freshly milled, unweathered rock powders should initially have reactive

surface areas comparable to those used in laboratory experiments, the mineralogy, particle size and shape distributions will evolve with time and will change with soil depth. Processes such as secondary mineral precipitation may block reactive surfaces over time. The depth distribution of particles is also an important constraint on weathering because carbonic acid weathering depends on biologically respired CO_2, as well as CO_2 dissolved in rainwater (FIG. 1). Modelling results (Taylor et al. 2016) suggest that mixing depths shallower than the depth of greatest biological CO_2 production could lead to lower carbon sequestration rates. The depth distribution of powder in agricultural soils is likely to be tillage- and site specific because the retention of particles depends on their size relative to the soil porosity. It is possible that repeated large applications of rock powder may change the soil permeability due to accumulation of slower-weathering phases where tillage pans restrict particle transport. Particle loss to stream water is also possible in soils that experience significant surface runoff, macro-porosity and shallow flow paths.

Given the complex set of processes that impact rock grains within the soil column, initial total surface area measurements using gas adsorption or estimating geometric surface area from the particle size distribution would need to be repeated with samples from several depths to elucidate grain-scale weathering processes. Changes in total mass, secondary phases or overall chemistry might be quantified by burying small bags of powder or by collecting soil cores for examination by X-ray fluorescence or X-ray diffraction. For detailed examination of individual surfaces, a variety of techniques such as atomic force microscopy, vertical scanning interferometry, or scanning electron microscopy could be employed, but it may be necessary to prepare pre-scanned mineral surfaces with fiducial marks. The depth distributions of particles, reactive surface area, pore-water chemistry and soil CO_2 would provide the detailed picture of weathering that is needed for process modelling and to estimate optimal carbon dioxide removal.

GOING GLOBAL

For EW to be implemented globally, field and laboratory trials must demonstrate that EW effectively and safely sequesters atmospheric CO_2. Enhanced weathering–specific models must also indicate that EW can remove sizeable quantities of CO_2 from the atmosphere across large spatial and temporal scales. However, the literature on natural chemical weathering has revealed the complexity associated with extrapolating brief, single catchment studies to larger scales (Li et al. 2016). For EW, the climate, soils, hydrology, crops, agricultural practices, and rock powder mineralogical composition will vary considerably not only from watershed to watershed but, in some cases, from farm to farm. The use of short-term time-series data from individual field sites may provide unexpected results in regional or global models because of spatial and temporal discrepancies between grain-scale weathering and dissolved ion fluxes. As crop harvesting repeatedly removes cations from agricultural soils, considerable discrepancies are expected between grain-scale weathering rates and stream water ion fluxes following EW treatments, even if biomass stubble is left on the fields. Global and regional models may not account for such discrepancies if they ignore productivity and nutrient cycling by the crops.

It is worth noting that annually, croplands emit an estimated 0.5–3.1 Mg CO_2-C m^{-2} to the atmosphere through soil respiration (Muñoz et al. 2010), far exceeding C fluxes associated with weathering. Thus, changes in soil respiration resulting from EW treatments could potentially form a major part of the greenhouse gas balance but such savings have not been previously considered in carbon budgets of EW (FIG. 6). In any case, cropland greenhouse gas emissions depend on factors such as management practices of fertilization and tillage, which are outside the scope of this review.

The nitrogen cycle is also a key component of EW soil chemistry that is largely missing from current weathering models (Taylor et al. 2017). In addition to the impact of nitric acid on cropland weathering, EW treatments may also enhance stream water NO_3^- fluxes (Rosi-Marshall et al. 2016). Nitrate-reducing bacteria are largely responsible for the production of N_2O, a greenhouse gas. Thus, increased stream water NO_3^- fluxes may drive increased N_2O emissions in near-shore environments, offsetting some of the climate change mitigation impacts of CO_2 drawdown by EW. Conversely, pH buffering resulting from addition of HCO_3^- to soils may decrease cropland N_2O emissions (Kantola et al. 2017). Both CO_2 and N_2O fluxes to and from the atmosphere should be considered in models of EW; N_2O emissions are now being built into the land surface components of general circulation models. The net effect of EW on climate will depend on the fluxes of all greenhouse gases.

Finally, CO_2 emissions associated with implementing EW globally must also be considered when assessing its net
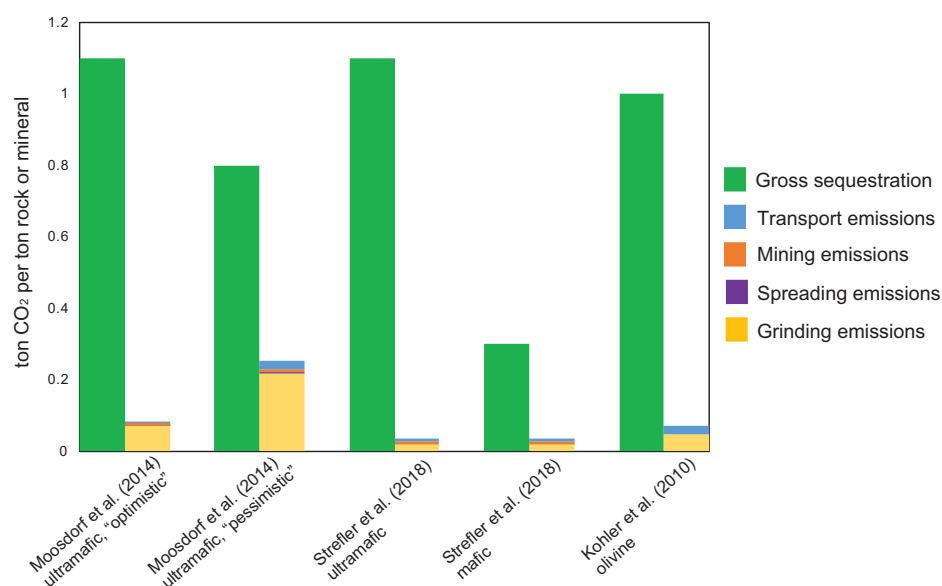
FIGURE 6 Model estimates of the enhanced weathering CO_2 budget for different rocks and minerals (see axis captions). Note that Strefler et al. (2018) reference Moosdorf et al. (2014) for mining, transport, and spreading emission estimates. Köhler et al. (2010) combine mining and grinding emissions and so is here represented as grinding.

impact on atmospheric CO_2 levels. The major processes that emit CO_2 are grinding of rock into powder, and transport of powder from source point to application point (FIG. 6). Preliminary calculations suggest that transport could emit up to 4% of the CO_2 sequestered by EW, while rock grinding may emit as much as 25% of the CO_2 sequestered by EW (Moosdorf et al. 2014). On balance, however, model calculations suggest that EW still removes significant quantities of CO_2 from the atmosphere (FIG. 6). Nevertheless, meaningful reductions in CO_2 emissions could be achieved

through the use of basalt powder currently stockpiled as quarrying waste product or through mining operations that use green energies.

SUMMARY

Enhanced weathering of silicate rocks is a carbon dioxide removal technology that may be capable of removing sizeable quantities of CO_2 from the atmosphere relatively rapidly and, therefore, help mitigate anthropogenic climate change. However, current understanding of EW is largely based on previous studies of long-timescale natural chemical weathering. The true ability of enhanced weathering to sequester atmospheric CO_2 will depend on a multitude of weathering pathways and biogeochemical interactions that are unique to enhanced weathering due to the short timescale on which it operates, its agricultural landscape setting, and the central role of rock-powder weathering. Laboratory and field trials will play a critical role in better understanding these unique controls, determining the real-world efficiency of enhanced weathering, and driving global models. But these trials have only just begun.

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