

Using carbonates for carbon removal

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The application of limestone to croplands has the potential to remove atmospheric CO₂ while improving crop yields and restoring ecosystems from the acidification associated with industrialization.

There is increasingly broad acceptance that large-scale carbon dioxide removal (CDR) may be required as a part of an overall effort to limit the damaging effects of anthropogenic climate change. The extent of carbon capture required to meet climate goals is, of course, dependent on how thoroughly and quickly we can reduce greenhouse gas emissions. However, estimates of required CDR even in optimistic emissions reduction pathways are -5–10 gigatons (Gt; 10⁹ ton) of CO₂ per year¹. Current rates of CDR are -2 GtCO₂y⁻¹, and this is overwhelmingly dominated by conventional land management². Given increasing debate about the effectiveness and verity of additional CDR, there has been increasing recent focus on ‘durable’ carbon removal – typically geochemical modes of CDR that last hundreds or thousands of years. However, the feasibility of these removal pathways at scale are still debated and currently amount to at most -0.001 GtCO₂y⁻¹ globally². Given the massive gap between current and targeted rates of carbon dioxide removal from the atmosphere and accelerating timelines due to persistently high emissions, we should be evaluating all possible options for near-term deployment of durable CDR.

Enhanced weathering is one durable CDR strategy that has garnered extensive recent attention. The carbon removal potential of enhanced weathering, although still poorly defined, may rival or surpass methods based on sequestration in organic carbon (for example, afforestation or soil organic carbon storage) and is potentially as high as >5 Gt CO₂ per year³. However, essentially all recent dialogue around enhanced weathering has focused on silicate feedstocks. Enhanced weathering of carbonate minerals has largely been dismissed so far, based on assumptions about the way in which carbonate minerals dissolve in soils and the potential reversibility of carbonate dissolution over longer timescales. Here, we make a case that enhanced weathering using carbonate feedstocks should also be considered and evaluated as an important component in a portfolio of climate solutions.

Enhanced carbonate weathering as a means of carbon capture

There are three basic premises that underlie arguments against enhanced carbonate weathering as a means of durable carbon removal. First, the mining, crushing, distribution and spreading of calcium carbonate requires energy, which at present is mostly or entirely derived from the burning of fossil fuels. This argument also holds for silicate feedstocks, although moving to non-fossil energy sources for transport and grinding costs will improve the efficiency of all enhanced chemical weathering approaches. Second, in many agricultural regions where liming is most often practised, a variable but occasionally significant proportion of the inorganic carbon in carbonate minerals can be

degassed to the atmosphere as CO₂ through reaction with nitric acid, a byproduct of nitrogen fertilization (Box 1). Indeed, carbonate mineral weathering in agricultural soils is conventionally considered a carbon source rather than a sink⁴, with existing IPCC guidelines for greenhouse gas inventories stipulating that all of the carbon in finely ground carbonate minerals used for liming ultimately becomes CO₂ that escapes to the atmosphere. Finally, CaCO₃ can re-precipitate downstream of initial dissolution, releasing CO₂ previously sequestered as dissolved bicarbonate and carbonate ions. Below we develop a rationale for considering enhanced carbonate mineral weathering a carbon-negative process despite these concerns.

On-site CO₂ capture from liming materials

The widespread liming of agricultural soils is performed primarily to modify initially low-pH soils or to buffer soil acidification caused by the oxidation of nitrogen-based fertilizers to create conditions more favourable for crop nutrient uptake and production. However, if initial soil pH is low, or if high rates of nitric acid production from fertilizer drive soil pH low, nitric acid can convert the inorganic carbon originally bound in carbonate to CO₂, which can re-enter the atmosphere (Box 1). This release of CO₂ to the atmosphere is directly related to the pH-dependent distribution of aqueous carbon species. At low pH a significant proportion of dissolved inorganic carbon is present as aqueous CO₂, which can exchange with the atmospheric carbon dioxide reservoir. With additional lime application soil pH can be raised to a pH in which the majority of the carbon is instead present as bicarbonate preventing CO₂ outgassing. Critically, there is clear support for this framework from field observations demonstrating that extensive on-site degassing in limed agricultural soils is not the predominant fate of added carbonate minerals, and that heavier liming results in a greater proportion of carbon sequestered as bicarbonate ions⁵. Even traditional liming practices not optimized for CDR have resulted in significant increases in alkalinity export in agricultural regions⁵.

An important additional point is that acidity produced by the oxidation of nitrogen-based fertilizer can lead to CO₂ outgassing regardless of whether liming occurs. For instance, in the absence of liming, acidity produced by the nitrification of fertilizers can react with bicarbonate in surface waters (Box 1), leading to CO₂ outgassing. As a result, it is fertilization and subsequent production of nitric acid by nitrification that should be thought of as a potential CO₂ source rather than liming itself. Similarly, liming should only be considered a net carbon source if the acid from fertilizer application would have interacted with silicate minerals in the background/counterfactual case. This scenario should be relatively rare due to much faster reaction rates with bicarbonate ions in most soils and inland waters.

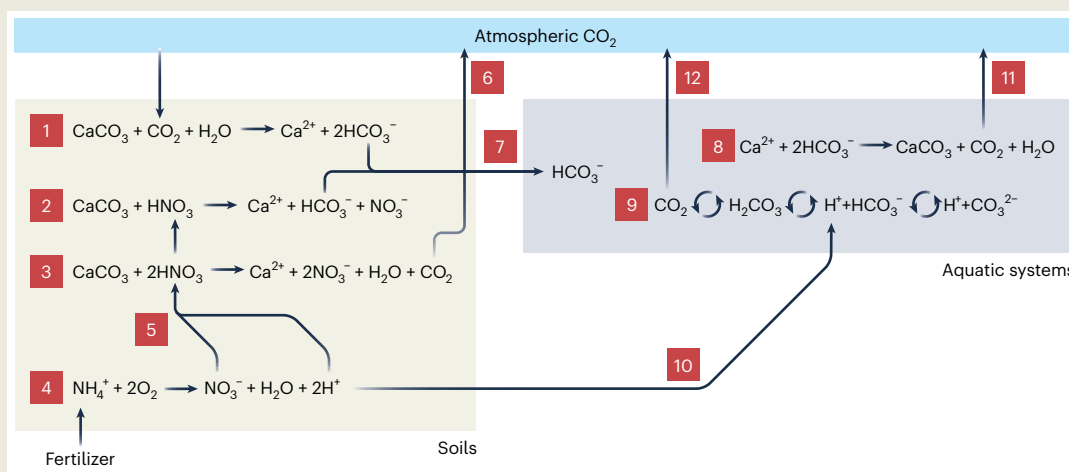
Finally, liming can be considered carbon-positive, -neutral or -negative depending on the timeframe being considered and the status of the soil exchange complex. Soils with added limestone may initially be carbon-positive if Ca²⁺ ions from weathering move onto a poorly buffered soil exchange pool and liberate acidity. However, once Ca²⁺ ions are released they will be charge-balanced by the production and export of two bicarbonate ions. The large cation exchange capacity of

BOX 1

Biogeochemistry of liming

The uptake of CO₂ occurs during reaction of carbonic acid (CO₂ + H₂O) with a carbonate mineral (1). However, in agricultural settings acid produced by nitrification (4) can also react with carbonate creating bicarbonate when soils are buffered (2) and CO₂ when they are not (3). When CO₂ is created by the dissolution of a carbonate by nitric acid it can be a source of CO₂ to the atmosphere (6). Net bicarbonate produced by these soil processes can be

transferred to inland and coastal waters (7). The acid produced by nitrification can also be delivered to inland waters (10) where it can lower the pH and create CO₂ from HCO₃⁻ from carbonate system equilibria (9), leading to CO₂ degassing (12). Finally, if concentrations of Ca²⁺ and HCO₃⁻ are high, CaCO₃ can precipitate in inland or coastal waters (8), leading to the production of CO₂, which can enter the atmosphere (11).



soils complicates tracking CO₂ removal fluxes over time, and further research is needed on the timescale and variability of the response of the soil exchange complex to large fluxes of anthropogenic alkalinity. Nonetheless it is expected that on decadal (or shorter) timescales liming in many regions is likely to be carbonate-negative. Critically this framing builds from tenets of aqueous geochemistry—foremost charge balance in all surface waters.

Re-precipitation of CaCO₃ and CO₂ degassing

Carbon dioxide is released from surface waters if calcium carbonate re-precipitates from the products of enhanced carbonate weathering and is then buried as carbonates in the sediments of inland waters (Box 1). It is important to note that precipitation may only lead to a temporary CO₂ source, since this reaction is reversible. Inland waters would need to permanently bury large amounts of calcium carbonate for this to become a net CO₂ source. However, extensive calcium carbonate burial does not seem to be widespread in areas of intensive liming⁶, probably due to the large inputs of respiration-derived CO₂ in the sediments and bottom waters of inland water systems. Release of CO₂ during formation of CaCO₃ from dissolved HCO₃⁻ can also occur in shallow/coastal marine environments. However, ongoing ocean uptake of anthropogenic CO₂ from human emissions will tend to reduce CaCO₃ precipitation in shallow marine systems especially where rivers discharge, and these sediments may also favour dissolution of carbonate phases precipitated in surface waters. On longer timescales, carbonate burial in the ocean will balance cation and alkalinity input from carbonate dissolution (Box 1), but this feedback operates on

timescales of ~10⁵ years⁷. Taken together, these considerations suggest that although enhanced carbonate weathering may not represent a permanent carbon sink on geologic timescales (millions of years) the potential storage timescales are exceptionally long from a societal point of view (thousands of years).

Other arguments for enhanced carbonate mineral weathering

There are also several straightforward biogeochemical arguments for exploring enhanced carbonate mineral weathering as a carbon dioxide removal strategy. Calcium carbonate dissolution kinetics are orders of magnitude faster than those of silicates⁷, and the dissolution of carbonate minerals is more likely to be congruent across a wide range of ambient environmental conditions – in contrast, silicate weathering is commonly slowed by production of clays and oxides. In addition, silicate mineral feedstocks have much higher concentrations of potentially toxic trace metals than carbonates. Olivine, for instance, is a target silicate mineral for enhanced weathering due to its fast dissolution rates and high availability, yet there is concern that accumulation of trace heavy metals from olivine in soils, inland waters and coastal marine habitats may have unintended consequences. In contrast, feedstocks for conventional agricultural liming are relatively pure, containing little to no potentially harmful trace elements. Because they contain much less magnesium than silicate feedstocks carbonates are also much less likely to promote the formation of secondary clay minerals⁸, a potential efficiency concern for enhanced weathering deployments using natural silicate rocks as feedstock. Lastly, carbonate mining is already

ubiquitous – over six billion tons of carbonates are mined annually, largely for the cement industry⁹.

Co-benefits of enhanced carbonate mineral weathering

There are multiple potential co-benefits to large-scale enhanced carbonate weathering as a carbon dioxide removal strategy. For instance, liming has consistently been shown to have positive effects on the growth rates of a wide range of crops¹⁰. In fact, current underutilization of liming has been proposed to result in significant losses in agricultural yields and profits. For example, sub-optimal use of liming has been estimated to lead to a drop in agricultural profits in Australia of ~25%¹¹. Thus, liming could lead to large crop production increases in areas like Africa, where the pH of soils are sub-optimal yet liming is not broadly implemented as a land-use practice.

The increase in pH associated with liming can also provide benefits to the nitrogen cycle. There has been increasing attention on reducing methane and nitrous oxide (N₂O) emissions from agricultural systems to help meet climate mitigation targets. It is well-known that agricultural liming, by inducing an increase in the pH of soil porewaters, reduces N₂O emissions¹², and there is also recent work that suggests liming can reduce rice field methane emissions¹⁰. Thus, increased liming has the potential to reduce emissions of other potent greenhouse gases – in addition to serving as a means of durable carbon removal.

Lastly, the solutes produced from enhanced carbonate weathering will increase carbonate saturation states in surface waters and shallow marine systems. In this sense, enhanced carbonate weathering can also partially offset the ‘other CO₂ problem’ – acidification. If enhanced carbonate weathering increases in scale, inland waters will receive large quantities of weathering products. In poorly buffered regions that are impacted by acid deposition, the use of limestone to counteract acidification has historically been seen as a form of restoration. Thus, in acidified regions a major co-benefit of liming is rapid pH restoration in inland waters. The widespread CO₂-induced acidification of the ocean has far-reaching implications for marine ecosystems and the services they provide to humanity. Thus, even with potential downstream CO₂ leakage, any enhanced weathering activity in watersheds draining to the ocean will contribute to the buffering of ocean acidification and could, arguably, still be considered as a natural climate solution. Indeed, ocean uptake of anthropogenic CO₂ since the preindustrial period amounts to more than 150 GtC, initially as carbonic acid, and this ‘acidification debt’ needs to be rebalanced before one should consider carbonate mineral weathering on land to be a zero-sum gain in terms of atmospheric CO₂ balance.

Next steps

Enhanced carbonate weathering has long been widely implemented in many agricultural settings and has been economically feasible even without subsidies because of beneficial impacts on crop yield. As such, there is a significant amount of existing physical and economic infrastructure to support widespread enhanced carbonate weathering. Importantly, there are also many operational limestone quarries associated with concrete and aggregate production, making it easier to predict costs associated with scaling up deployment compared with other negative emission technologies. Since the early 1900s there has been nearly unanimous buy-in to the concept of liming from the agricultural community – making it a rare case where the key stakeholders most required for implementation of a negative emissions technology are already demonstrably supportive. Globally, agricultural lime application rates are ~30 megatons (Mt; 10⁶ ton) of carbon per year⁴. These rates are far from optimized in agricultural regions that use liming as

a regular practice, and there are large agricultural areas of the globe that could benefit from greatly enhanced liming in the future. Initial studies indicate that the cost of enhanced carbonate weathering may rival or be lower than enhanced silicate weathering¹³. Beyond driving durable CDR, ramping up liming should improve yields and reduce other on-farm greenhouse gas emissions.

The long history of liming, however, makes financing a challenge. Liming can be most rapidly scaled up in areas that have historically practised liming. With many paths of paying for CDR (for example, voluntary carbon markets) it is essential to define how a change in practice would lead to additional carbon removal. Thus, defining counterfactual scenarios in some regions is challenging. Some potential counterfactual scenarios, for instance, would reward farmers with past practices of under-liming. However, limited soil pH management – that is, limited liming – leads to more nutrient runoff and thus pollution. Therefore, payment based on individual farm counterfactuals would put farmers that have been responsibly managing their soil health and limiting pollution at an economic disadvantage. As importantly, there is also a long history in carbon markets of problems tracking individual or project level practices instead of regional trends. The more pragmatic alternative approach is to define counterfactual scenarios based on regional pH trends. For instance, farmers could be paid through a market or incentive programme for any liming beyond what is needed to match the regional mean. This provides a simple incentive for additional liming – providing the co-benefits of addressing soil acidification while driving CDR. Subsidies and a pay-for-practice approach provides another potential path forward. Given that agricultural subsidies are ubiquitous, they could be used to drive agricultural soil pH values that optimize nutrient uptake and crop yields, while regulating agricultural greenhouse gas emissions. In this case, the benefits from modifying liming could then be tied to regional, state or national greenhouse gas accounting schemes – regardless of the additionality of the practice.

Croplands are the obvious target for rapidly scaling up liming. However, enhanced carbonate weathering can also be deployed in a range of systems beyond croplands. For example, extensive enhanced carbonate weathering could be deployed as part of the management of urban and suburban turf grass cover. Managed forests and forests impacted by acid rain are also limed and represent another underutilized landscape with the co-benefit of increased tree growth, although there are some associated risks¹⁴. Given the rapid and well-understood dissolution kinetics of ground limestone, it may be possible to have robust model-based estimates of CDR from ground limestone addition along roadsides and in managed non-agricultural settings. Further, carbonate could be the feedstock for controlled reactor approaches, an idea that was extensively explored before large-scale durable carbon markets existed¹⁵ and that has not been deployed at any meaningful scale. Although the overall potential scope, cost and CO₂ balance associated with a range of approaches need to be assessed in detail, multiple deployment approaches for enhanced carbonate weathering could potentially result in gigaton-scale CDR.


We suggest that it is time to put enhanced carbonate mineral weathering on the table as a potential durable carbon removal strategy. Liming new areas and paying farmers to lime their fields more heavily is a potentially effective and inexpensive means of CDR, and one that is likely to increase agricultural yields and make farmers more accustomed to enhanced weathering approaches as the potential for silicate mineral approaches expands in the coming decades. There is an obvious need to rethink enhanced carbonate mineral weathering, provide new constraints on the carbon capture potential of this

strategy. Scaling up carbonate weathering to help mitigate the impacts of anthropogenic climate change while improving agricultural yields and decreasing the impacts of anthropogenic activities on surface waters. Carbonate weathering can provide durable carbon removal, decrease nutrient runoff from farms, and mitigate surface water acidification – the questions we should be focusing on are scale, costs and efficiency of these impacts.

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Competing interests

The authors declare that N.P. and C.T.R. were co-founders of Lithos Carbon but have no financial ties to the company. P.R. and C.T.R. are scientific advisors to CREW Carbon, which N.P. founded and advises but has no financial ties to.

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