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# Carbon dioxide efficiency of terrestrial enhanced weathering

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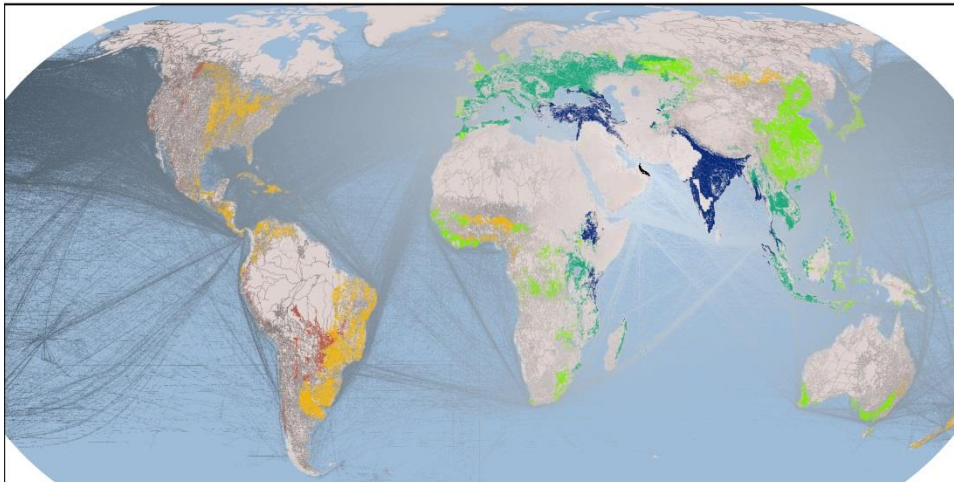
## Abstract

Terrestrial enhanced weathering of ultramafic silicate rocks, the spreading of rock flour to enhance natural weathering rates, has been suggested as part of a strategy to reduce global atmospheric CO<sub>2</sub> levels. Here, we assess the net CO<sub>2</sub> removal of terrestrial enhanced weathering, by budgeting potential CO<sub>2</sub> sequestration against the associated CO<sub>2</sub> emissions. We combine global spatial datasets of potential source rocks, transport networks and application areas with

CO<sub>2</sub> emissions associated with source rock processing in an optimistic and a pessimistic scenario.

Terrestrial enhanced weathering consumes more CO<sub>2</sub> than it emits for mining, comminution, transport, and application in most locations. The CO<sub>2</sub>-efficiency is dominated by the choice of source rocks and material comminution. CO<sub>2</sub> emissions from transport have a small effect on the overall budget (on average 0.5-3% of potentially sequestered CO<sub>2</sub>) and the emissions of material mining and application are negligible. After all emissions, 0.5-1.0 t CO<sub>2</sub> can still be sequestered on average per tonne of rock. However, very large amounts of rock would be needed to control or reduce the atmospheric CO<sub>2</sub> concentrations substantially with enhanced weathering. Before enhanced weathering could be applied at large scales, more research is needed to assess weathering rates, potential side effects, social acceptability, and mechanisms of governance.

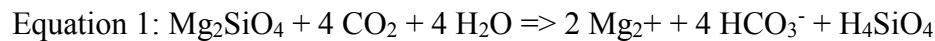
## TOC Art



## Introduction

Rising levels of atmospheric CO<sub>2</sub> may cause substantial challenges for human society. Stagnation in efforts to cut anthropogenic CO<sub>2</sub> emissions has led to the proposal of technological solutions for capturing and storing atmospheric CO<sub>2</sub><sup>1-4</sup>. Terrestrial enhanced weathering was suggested as one of these solutions<sup>5-7</sup>. The term “terrestrial enhanced weathering” is commonly used for the application of ultramafic silicate rock powder to suitable application areas to increase natural chemical weathering rates<sup>6-10</sup>.

Natural chemical silicate rock weathering is a major geological sink of atmospheric CO<sub>2</sub><sup>11-13</sup>. The release of cations during mineral weathering binds dissolved CO<sub>2</sub> to form bicarbonate and carbonate ions, which are then transported to the ocean. Annually, natural chemical silicate weathering consumes about 0.86 to 1.06 Gt of atmospheric CO<sub>2</sub><sup>14-16</sup>. Chemical weathering of forsterite is exemplary of the process (Eq. 1):



Theoretically, based on stoichiometry and mass, the weathering of a 4 mm thick layer of forsterite (Mg-olivine) spread over the entire terrestrial land mass could consume all atmospheric CO<sub>2</sub><sup>6</sup>. In practice, there are limits to sequestration potential, e.g. due to saturation effects<sup>17</sup>. Silica saturation, for example, could limit the CO<sub>2</sub> sequestration in humid tropical regions to 3.7 Gt CO<sub>2</sub> a<sup>-1</sup><sup>17</sup>, although an abiotic limitation by forsterite saturation may substantially reduce that value<sup>7</sup>. Furthermore, carbonate minerals may precipitate which would liberate up to half of the sequestered CO<sub>2</sub> to the atmosphere[manning ref, manning and renforth], but is unclear if the weathering rates are sufficient to supersaturate solutions with respect to carbonate phases. If carbonates precipitate, it is likely the containing CO<sub>2</sub> will be sequestered for millions of years<sup>2, 11</sup>. All of the key technologies required for terrestrial enhanced weathering are mature and already used on regional scale for fertilization or pH-management of agricultural and forest soils. However, the industry and associated environmental impact of up-scaling this technology require

consideration. Possible side effects of terrestrial enhanced weathering on e.g. river pH and alkalinity<sup>17</sup>, or release of metals<sup>10, 18</sup> are related to the source rock composition as well as the deployment extent and method. In addition, the availability of suitable land may be a major limiting factor<sup>4</sup>, and the infrastructure requirements of transporting large volumes of rock could inherently limit the available application area.

Ultramafic igneous rocks have the largest carbon sequestration potential by mass and fastest dissolution rates of silicate rocks, and are thus likely to be the most applicable for terrestrial enhanced weathering<sup>19-22</sup>. Several studies have already investigated their carbonation potential at elevated temperatures and under elevated pCO<sub>2</sub> in reactors<sup>23-25</sup>, which can be referred to as “accelerated weathering”, although “mineral carbonation” is used here to delineate. For mineral carbonation, formalized life cycle studies exist<sup>26</sup>. The capital investment required for mineral carbonation (e.g. for creating a large reactor), may limit deployment [REF]. The terrestrial enhanced weathering assessed here conceptually uses soil as a ‘reactor’, potentially negating some of the capital expenditure for mineral carbonation. However, these technologies are not necessarily mutually exclusive, and it may be that mineral carbonation is applicable alongside, or preponderate to, terrestrial enhanced weathering.

As a first step to investigate the feasibility of the method at the global scale, a basic carbon budget of terrestrial enhanced weathering has been performed to identify key areas of uncertainty for future research. Expanding on the study of Renforth<sup>9</sup>, which focused on the United Kingdom, this study globally constrains the net-CO<sub>2</sub>-efficiency of terrestrial enhanced weathering by applying optimistic and pessimistic scenarios of a spatially explicit carbon budget. Rock properties, mining, comminution, transport, and application are included in the analysis presented here.

## **Data and methods**

Spatial datasets representing source rocks, transport pathways and potential application areas were combined with associated CO<sub>2</sub> emissions (into the atmosphere) and sequestration (from the atmosphere) to develop a spatially explicit CO<sub>2</sub> budget of terrestrial enhanced weathering, and identify its net efficiency on areas suitable for its application. This does not account for a potential increase in biomass or crop production due to the release of geogenic nutrients during the dissolution process<sup>7</sup>. The main CO<sub>2</sub> emissions associated with terrestrial enhanced weathering are generated by mining, crushing/milling, transport and spreading of the rock material, for which a pessimistic and an optimistic scenario were defined (Table 1). The technically simple process of spreading rock flour on agricultural areas simplifies the CO<sub>2</sub> budget compared to the technically more complex accelerated weathering in reactors, where CO<sub>2</sub> budgets in addition to the here assessed aspects have to account for chemical conversion, beneficial reuse, transport of used minerals and disposal<sup>26</sup>. The CO<sub>2</sub> budget of enhanced weathering is calculated after Equation 2:

Equation 2:  $\Delta\text{CO}_2 = \text{Potential CO}_2 \text{ sequestration (based on source rock properties)} - \text{CO}_2 \text{ emissions (Mining + Comminution + Transport + Application)}$

$\Delta\text{CO}_2$  will later be referred to “CO<sub>2</sub> available for sequestration”, which implies that this is the amount of CO<sub>2</sub> which could effectively be sequestered (without giving a statement of the time that takes) after subtracting the emissions from the maximum potential sequestration. The datasets representing the individual parts of the budget are provided in Table 1 and illustrated in Figure 1.

All datasets were combined in a global GIS and resampled to a grid resolution of 1 x 1 km (all GIS functionality implemented in the software ArcGIS 10 by ESRI®).

**Table 1:** Factors affecting the CO<sub>2</sub> budget of terrestrial enhanced weathering and their assumed CO<sub>2</sub> emissions and sequestration. In the budget, negative values indicate CO<sub>2</sub> emissions (into the atmosphere); positive values indicate CO<sub>2</sub> sequestration (from the atmosphere).

Theme	Spatial Data Reference	CO <sub>2</sub> budget reference	Condition optimistic	Condition pessimistic	Value optimistic	Value pessimistic	Unit <sup>a)</sup>	Comments
Source material	<sup>27</sup>		Ultramafic rocks	Ultramafic rocks	736	736	10 <sup>3</sup> km <sup>2</sup>	
Potential maximum CO <sub>2</sub> sequestration		<sup>9</sup>	Upper limit for ultramafic rocks in Fig. 1 of the reference	Lower limit for ultramafic rocks in Fig. 1 of the reference	1.10	0.80	t CO <sub>2</sub> t <sup>-1</sup>	
Mining		<sup>8</sup>	Estimated energy need (18.8 MJ t <sup>-1</sup> rock) times CO <sub>2</sub> emission per MJ provided by <sup>9</sup>	Estimated energy need (18.8 MJ t <sup>-1</sup> rock) times CO <sub>2</sub> emission per MJ provided by <sup>9</sup>	-0.007	-0.007	t CO <sub>2</sub> t <sup>-1</sup>	
Crushing / milling		<sup>9</sup>	0.6 GJ / t energy demand (see supplemental	2 GJ / t energy demand (see supplemental	-0.07	-0.22	t CO <sub>2</sub> t <sup>-1</sup>	

			information)	information)				
Road	<sup>28</sup>	<sup>29</sup>	Lower estimate	Upper estimate	-59	-109	g CO <sub>2</sub> km <sup>-1</sup> t <sup>-1</sup>	
Railroad	<sup>28</sup>	<sup>29</sup>	Lower estimate	Upper estimate	-7	-26	g CO <sub>2</sub> km <sup>-1</sup> t <sup>-1</sup>	
Trail	<sup>28</sup>	<sup>29</sup>	Lower estimate	Upper estimate	-59	-109	g CO <sub>2</sub> km <sup>-1</sup> t <sup>-1</sup>	Values taken from class “road”
Rivers	<sup>30</sup>	<sup>29</sup>	Lower estimate	Upper estimate	-28	-35	g CO <sub>2</sub> km <sup>-1</sup> t <sup>-1</sup>	Used major world rivers
5 km buffer around roads, railroads, trails and rivers	Self- defined	<sup>29</sup>	Lower estimate	Upper estimate	-59	-109	g CO <sub>2</sub> km <sup>-1</sup> t <sup>-1</sup>	Values taken from class “road”
Shipping lines	<sup>31</sup>	<sup>29</sup>	Lower estimate	Upper estimate	-5	-20	g CO <sub>2</sub> km <sup>-1</sup> t <sup>-1</sup>	
Application emissions		<sup>32</sup>	1 t per ha, 80 ha per field	3 t per ha, 1 ha per field	-0.0011	-0.004	t CO <sub>2</sub> t <sup>-1</sup>	Using a factor of 2.6 to calculate CO <sub>2</sub> emissions from the



								reported diesel volume.
Arable land	<sup>33, 34</sup>		Upper estimate of proportions on cells, outside polar or arid climates.	Lower estimate of proportions on cells, outside polar or arid climates.	14.7	11.8	10 <sup>6</sup> km <sup>2</sup>	

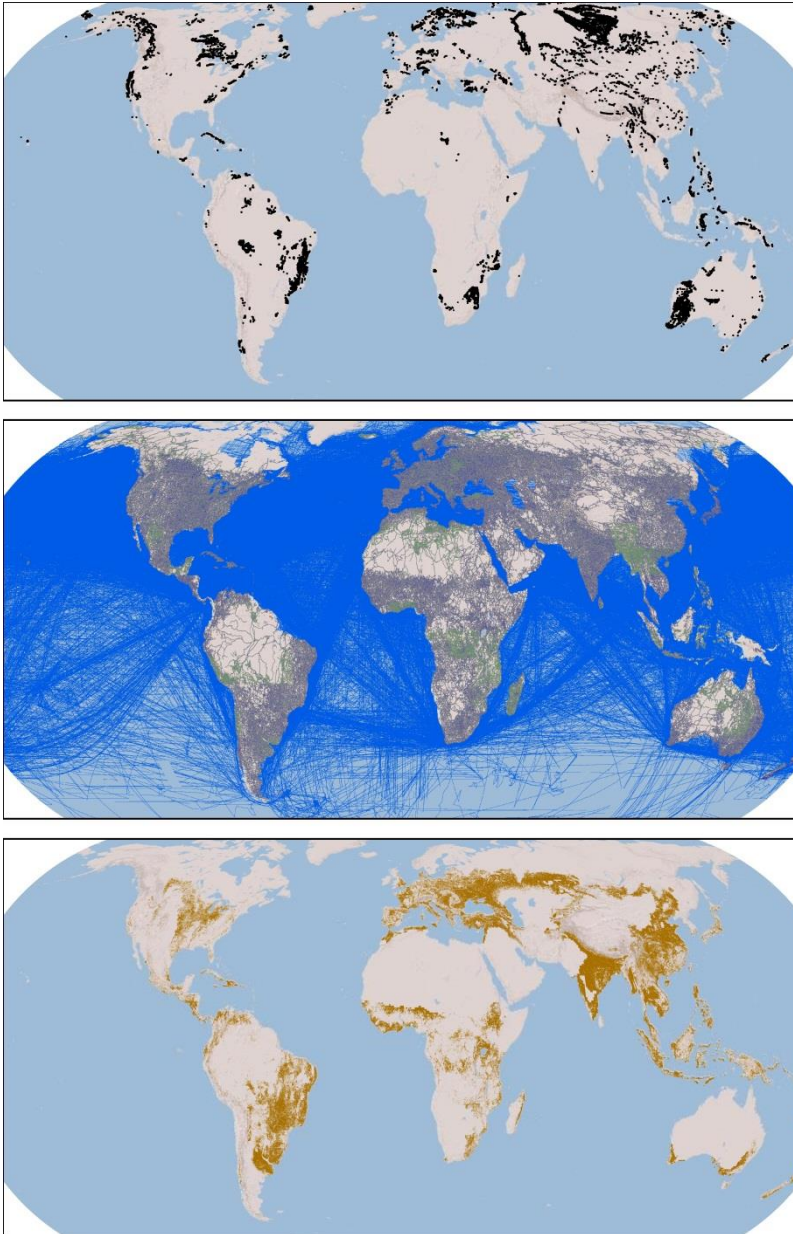
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103    <sup>a)</sup> t<sup>-1</sup> refers to tons of rock.

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107

108 **Figure 1:** Spatial input data of the study. a) Source rock locations according to the GLiM (black,  
109 area exaggerated for visibility, coverage of potential source rocks is not exhaustive), b) Transport  
110 routes (colors indicate different modes of transport, but the grid is too fine to be resolved in the  
111 image), c) Application area.

112

### 113 **Material source rocks**

114 Ultramafic rocks suitable for terrestrial enhanced weathering are generally rich in the mineral  
115 forsterite (the olivine Mg-end member) which was previously recommended for this technique <sup>6</sup>,  
116 <sup>35</sup>. The Global Lithological Map (GLiM)<sup>27</sup> contains 736,000 km<sup>2</sup> of rock units in which  
117 ultramafic rocks occur (Figure 1) but does not identify all ultramafic rocks globally. The source  
118 maps of the GLiM emphasize ultramafic rocks differently, and e.g. in Japan, Iceland or eastern  
119 Africa, additional ultramafic rock occurrences are likely. The maximum potential CO<sub>2</sub>  
120 sequestration per tonne of rock material is represented by upper (optimistic scenario) and lower  
121 (pessimistic scenario) literature values of the CO<sub>2</sub> sequestration of ultramafic rocks <sup>9</sup>. In the case  
122 that the source rock consists of pure forsterite, the potential maximum CO<sub>2</sub> sequestration (at  
123 neutral pH) would be even higher, namely 1.25 t CO<sub>2</sub> t<sup>-1</sup>, based on the stoichiometry of equation  
124 1.

### 125 **Material application areas**

126 Potential application areas need to 1) provide a suitable environment for chemical weathering  
127 and 2) be accessible for terrestrial material spreading. Suitable environments are moist and  
128 warm, based on the assumption that moisture is needed for a dissolution reaction and  
129 temperature increases chemical weathering rates<sup>36</sup>. These conditions are here represented by  
130 omitting areas from dry and very cold climate zones (Main classes “Arid climates” and “Polar  
131 climates”<sup>34</sup>) for application. Areas suitable for terrestrial spreading of rock powder are defined  
132 by arable land cover<sup>33</sup> (Figure 1). Arable land seems most suitable for application because it is  
133 already intensively managed and spreading crushed rock would, notionally, require only limited  
134 new infrastructure. The land cover data provide a proportion range to which each cell is covered  
135 by arable land<sup>33</sup>. The upper (optimistic) and lower (pessimistic) ends of that range are used here

(Table 1). Land cover data were converted from raster into polygon data using the “Raster to Polygon” tool implemented in ArcGIS. This combines neighboring cells with the same attributes into one polygon with a unique identifier. These were reconverted into raster cells with that identifier to define individual agricultural areas and link them to the transport datasets.

#### **Material extraction**

Extraction and application require minimal amounts of energy, which are included in the budget. The energy demand for surface extraction is  $18.8 \text{ MJ t}^{-1}$ <sup>8</sup>, which was translated into CO<sub>2</sub> emissions (Table 1).

#### **Material comminution**

Size reduction of rock is achieved in a number of steps including at least one instance of crushing, followed by milling or grinding<sup>9</sup>. Crushing, required for particle size reduction to 1 mm diameter, can be achieved with minimal energy input ( $5\text{-}10 \text{ MJ t}^{-1}$ <sup>9</sup>). It is likely that additional size reduction to  $<100 \text{ }\mu\text{m}$  will be required, which necessitates grinding. The energy required in this process is directly related to the surface area created<sup>8, 9, 23, 37</sup>. We use a shrinking core model (see supporting information) to calculate the initial particle diameter required to achieve complete weathering within 1 year given a specific weathering rate. The grinding energy to produce this particle size is then calculated. An optimistic (and pessimistic) log weathering rate of  $-12$  (and  $-18$ )  $\text{mol m}^{-2} \text{ s}^{-1}$ <sup>9</sup>, was used to calculate a grinding energy of

$0.6$  ( $2.0$ )  $\text{GJ t}^{-1}$ , which emits  $0.07$  ( $0.22$ )  $\text{t CO}_2 \text{ t}^{-1}$  due to electrical energy use. As we have not included a temporal dimension into our analysis, the weathering rates remain effectively constant in the shrinking core model. The 6 orders of magnitude range in weathering rates between the optimistic and pessimistic scenarios is indicative of the range of values between laboratory determined kinetics of ‘fresh’ material and heavily weathered material in catchment scale

studies. Few experimental data exist that investigate silicate minerals added to the environment. As such, the treatment of kinetics for terrestrial enhanced weathering is highly uncertain.

### **Material transport**

Rock material for terrestrial enhanced weathering could be transported on shipping routes (oceans, certain rivers and channels), train lines and roads. Airfreight is disregarded here because of its high associated CO<sub>2</sub> emissions. This study combines various global datasets to generate a routing raster from the source rock areas to the application areas. Shipping lanes are represented by a dataset of known ship positions<sup>31</sup>. Each grid cell with at least one documented ship position in the original data is considered a potential shipping lane. For river transport, 98 major rivers of the world<sup>30</sup>, with an average length of 2660 km were included and assumed to be navigable. This assumption was verified against available maps of navigable waterways. Only the upper reaches of the rivers may be too small for ships to pass, but the resulting underestimation of transport CO<sub>2</sub> emissions should be very small, as 1000 km transportation on rivers emits only 0.03 (0.07) t CO<sub>2</sub> t<sup>-1</sup> less than on roads, in the optimistic (pessimistic) scenario. Land transport routes were derived from the VMAP0 dataset<sup>28</sup>, which includes global vector maps of roads, pathways, railroads, structures, and trails. The vector maps were converted to raster datasets with a 1 x 1 km grid resolution. Some data provided different subtypes of roads (e.g. the information that a route is under construction or its status is unsure). This information was generalized to the classes in Table 1. All routes represented in the datasets allow material transport. The class “Structure” was omitted, because it contained only few datasets. The routes represented in the VMAP0 “pathway” dataset are interpreted as small roads and thus classed as roads. To ensure connectivity within the transport network, a 5 km buffer around all mapped transport lines is assumed to be usable for transport. Areas without transport routes are considered impassable. CO<sub>2</sub> emissions per kilometer vary widely between different modes of transport<sup>29</sup>, even for given

vehicle types with varying load or driving style<sup>38</sup>. The CO<sub>2</sub> budget includes upper (pessimistic) and lower (optimistic) ends of the provided CO<sub>2</sub> emission ranges per km and tonne<sup>29</sup> (Table 1). These values are similar to CO<sub>2</sub> efficiency in road freight transportation from other sources<sup>39</sup>.

Transport CO<sub>2</sub> emissions between source rocks and application areas were calculated as minimum (mean) of a cost distance raster (ESRI ArcGIS functionality) per continuous area of arable land in the optimistic (pessimistic) scenario. The calculations were performed using the “Zonal Statistics” tool implemented in ArcGIS. The optimistic scenario considers the minimum transport emissions per area, following the argument that as soon as the material arrives at the application area, the application emissions cover the transport on the field (possibly an underestimation for large agricultural areas consisting of many fields). The pessimistic scenario considers the mean transport emissions per application area, which certainly overestimates the transport emissions in many cases.

## Results

Globally 736,000 km<sup>2</sup> of suitable source rock areas are mapped. Potential application areas for terrestrial enhanced weathering amount to 14,700,000 (11,800,000) km<sup>2</sup> in the optimistic (and pessimistic) scenario (Figure 1). Throughout the text, results for the pessimistic scenario are represented in parentheses.

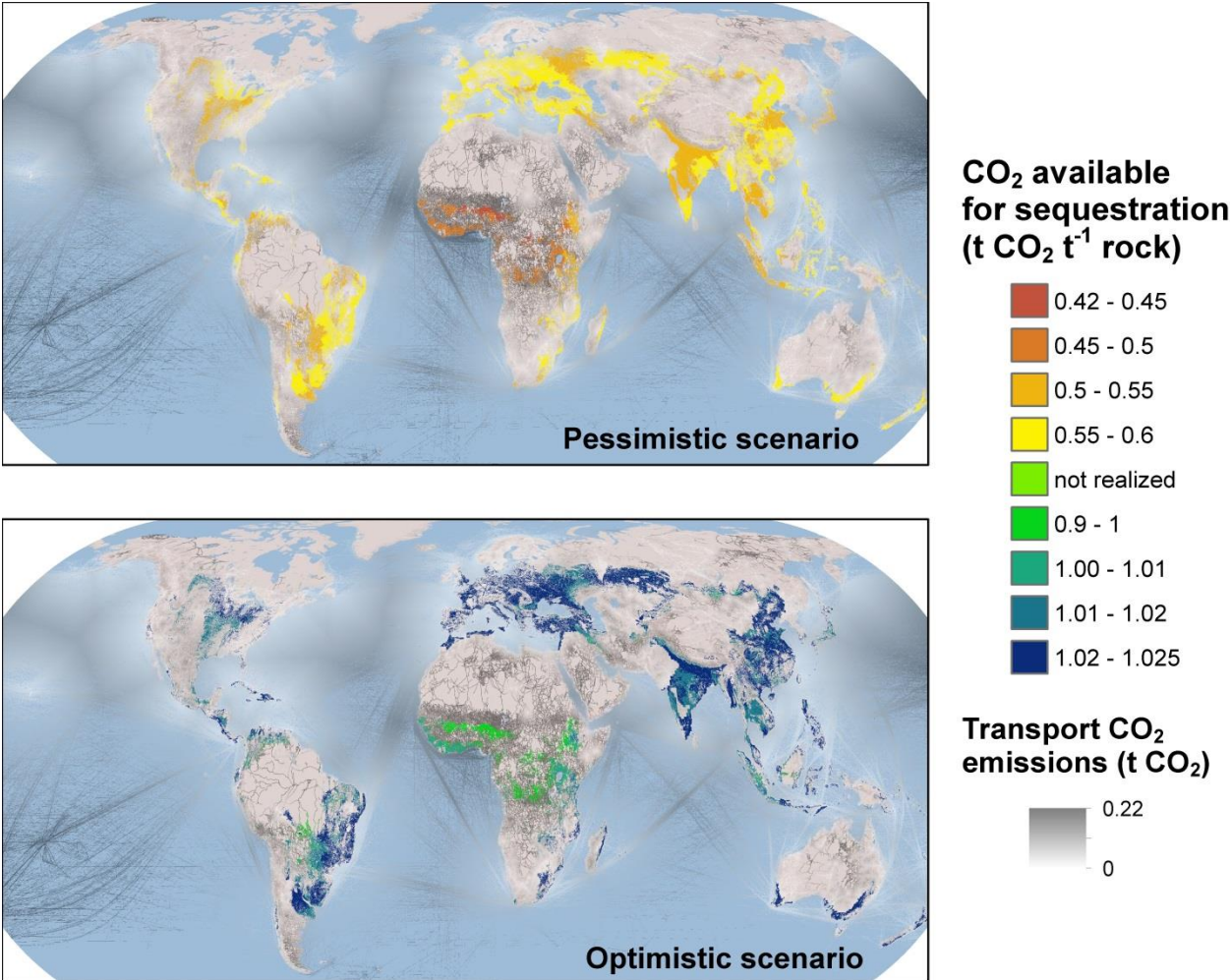
The grains spread on the application areas can potentially sequester up to 1.1 (0.8) t CO<sub>2</sub> t<sup>-1</sup> (t<sup>-1</sup> means “per tonne of rock”) in the optimistic (and pessimistic) scenario. Before transport to the application areas, the source rocks need to be mined (extraction) and their grain size sufficiently reduced (comminution), which emits 0.074 (0.229) t CO<sub>2</sub> t<sup>-1</sup>. Spreading the material on the application areas (excluding transport) emits 0.001 (0.004) t CO<sub>2</sub> t<sup>-1</sup>. The emissions associated

with these three aspects are spatially static – they do not change with distance between source rock and application areas.

Subtraction of the named spatially static emissions leaves CO<sub>2</sub> for 17,000 (5,000) km transport on road or 140,000 (21,000) km on railroad after which the emissions would exceed the potential maximum CO<sub>2</sub> sequestration. 89% of the application areas are connected to the transport network. The transportation CO<sub>2</sub> emissions from source to application areas average 0.007 (0.022) t CO<sub>2</sub> t<sup>-1</sup>, which amounts to 0.7% (4.0%) of the potential CO<sub>2</sub> sequestration after the emissions of the spatially static parameters. The available CO<sub>2</sub> for sequestration (maximum potential CO<sub>2</sub> minus emissions, equation 1) differs strongly for both scenarios. Even the maximum transport emissions in the optimistic scenario do not compensate for the difference between the static emissions of the optimistic and pessimistic scenarios. This implies that the smallest available CO<sub>2</sub> for sequestration at any application area in the optimistic scenario is higher than the highest available CO<sub>2</sub> in the pessimistic scenario (Figure 2). This highlights that transport costs are not a major constraint to the effectiveness of the enhanced weathering technique. In addition, source rock occurrences, which may not be represented in the GLiM units containing ultramafic rocks could shorten transport routes and reduce the average transport emissions at global scale even further. After subtracting all emissions, on average 1.02 (0.54) t CO<sub>2</sub> t<sup>-1</sup> are available for sequestration at the application areas in the optimistic (pessimistic) scenario.

If terrestrial enhanced weathering were used to sequester 10% of the 9.1 Gt CO<sub>2</sub>-C emitted by fossil fuel combustion and cement production in 2010 <sup>40</sup>, 0.8 (1.7) Gt of ultramafic rock material would needed to be weathered according to the optimistic (pessimistic) scenario. All this material would need to be mined, crushed and transported to the application regions. For

comparison, the estimated present total mass movement by humans is 40 to 45 Gt a<sup>-1</sup> <sup>41</sup>. Certainly, moving these large additional masses would have strong socioeconomic and environmental consequences.



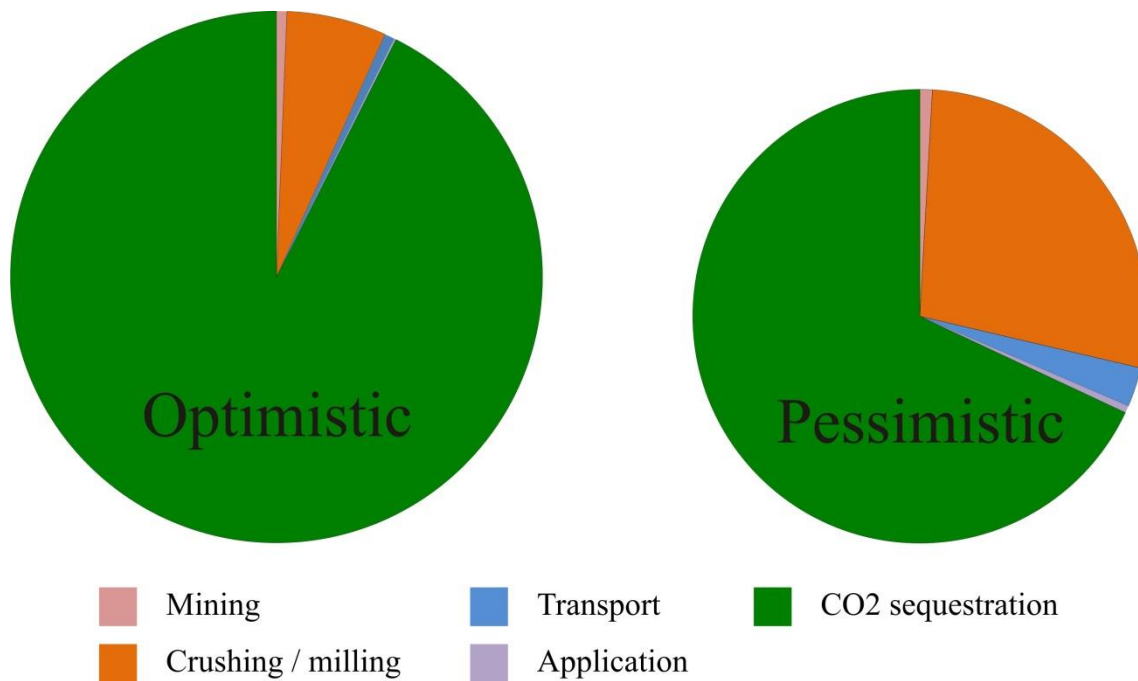
**Figure 2:** Available CO<sub>2</sub> for sequestration in the application areas. The emissions by transport are shown for comparison. The class “not realized” represents the values between 0.6 and 0.9 which do not occur in any of the maps. Everywhere in the optimistic scenario, more CO<sub>2</sub> is available for sequestration than anywhere in the pessimistic scenario.



## Discussion

In the optimistic scenario, associated emissions reduce the actual CO<sub>2</sub> sequestration only slightly below the potential maximum CO<sub>2</sub> sequestration (Figure 3). The pessimistic scenario shows a substantial reduction of the actual CO<sub>2</sub> sequestration, mainly because of the assumed less favorable rock composition (and the resulting smaller maximum CO<sub>2</sub> consumption) and increased CO<sub>2</sub> emissions of comminution (Figure 3). The effect on the difference between the optimistic and pessimistic scenarios is largest for the potential maximum CO<sub>2</sub> sequestration. It is responsible for 80.6% of the variability of the available CO<sub>2</sub> for sequestration in a Monte Carlo Simulation (100,000 draws, Oracle Crystal Ball software, assuming a uniform distribution between optimistic and pessimistic values of all parameters). The second most sensitive parameter is comminution, contributing 19.2% to the variability. Efficiency improvements and renewable energy usage could reduce the associated CO<sub>2</sub> emissions below the optimistic scenario assumed here. Uncertainty in comminution requirements is largely down to uncertainty in weathering rates. The slower the weathering rate, the more processing is required to produce the same dissolution per mass. Experimental evidence is needed that examines the dissolution kinetics in various potential application environments. Chemical weathering rate constants (per surface area of rock) have also been shown to increase with finer material as a result of ‘mechano-chemical activation’, but only a small number of studies explore this in silicate minerals and the impact on enhanced weathering remains unclear<sup>8, 23, 37</sup>. The emissions of mining, application, and also transport (on average) are negligible for the variability of CO<sub>2</sub> emissions of terrestrial enhanced weathering (0, 0, and 0.2%, respectively). The sensitivity analysis suggests that investigations of suitable source rocks, energy requirements of comminution and mineral reactivity in the environment are critical in assessing the potential of terrestrial enhanced weathering. The timeframe of current CO<sub>2</sub> policy might even qualify

carbonates as enhanced weathering source rock<sup>42</sup>, which weather faster than ultramafic silicates<sup>22</sup>.



**Figure 3:** CO<sub>2</sub> budgets per tonne rock material according to the optimistic scenario and pessimistic scenario. The area of the optimistic pie is 1.4 times that of the pessimistic, representing the different potential maximum CO<sub>2</sub> sequestration.

One specific geological unit, the Semail Ophiolite in Oman, was suggested as location for carbon management by in-situ-carbonation, and its carbon sequestration potential is therefore well researched<sup>43</sup>. To explore its potential for enhanced weathering, and as an example for the effect of transportation, we ran the transport coast routing model with the Semail Ophiolite, represented in the Geological map of the Middle East<sup>44</sup>, as single source rock in the GIS (Figure 4). Abundantly available Harzburgites from the Semail Ophiolite contain 25 % Mg by weight<sup>45</sup>, which translate to a potential maximum CO<sub>2</sub> sequestration of 0.89 t CO<sub>2</sub> t<sup>-1</sup>. After deducting the

pessimistic scenario emissions, treating the large agricultural areas in Europe and most parts South-East Asia with material of the Samail Ophiolite would still sequester half a tonne CO<sub>2</sub> per tonne of rock (Figure 4). Even transport to the Corn Belt in North America would still allow some CO<sub>2</sub> sequestration and only in the remotest areas of the Americas the CO<sub>2</sub> sequestration is reduced below 0.3 t CO<sub>2</sub> t<sup>-1</sup> by transport emissions (Figure 4).

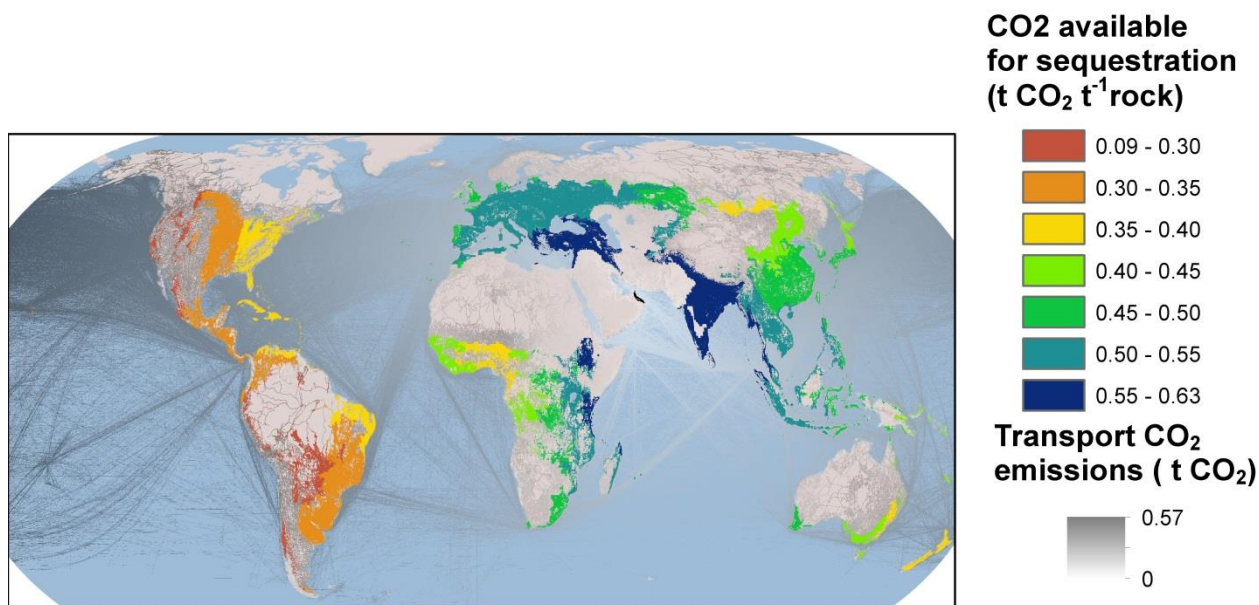


Figure 4: Conservative scenario of terrestrial enhanced weathering CO<sub>2</sub> sequestration efficiency using the Samail Ophiolite as source rock (marked in black in the map, located in Oman).

The logical next step, after identifying the total potential of enhanced weathering is to identify the rates at which the rock flour weathers in application areas and add these rates to the evaluation. Mineral reactivity, and thus weathering rates, depend on a number of environmental parameters<sup>7</sup>. While, chemical weathering rates determined in controlled laboratory experiments, (e.g. far from equilibrium and strongly influenced by temperature<sup>46</sup> and pH<sup>47</sup>) can be used as an upper estimate, the large difference of rates compared with catchment studies prevents precise

assessment of terrestrial enhanced weathering.. Only dedicated experiments conducted to assess dissolution kinetics in specific conditions could quantify extrinsic environmental impacts (e.g. temperature) on chemical weathering rates in the field <sup>19, 48</sup>. Pertinent to this is the influence of plants and microorganisms on chemical weathering (see Manning and Renforth for discussion). A comparison of five catchments in Iceland showed that in the vegetated catchments, chemical weathering was increased between 2 and 10 times depending on vegetation type and mineral<sup>49</sup>. Similarly, a correlation between vegetation type and weathering induced bicarbonate fluxes was shown for 338 stream catchments in North America <sup>20</sup>. Field studies also showed the potential of microorganisms<sup>50</sup> and fungi<sup>51, 52</sup> to increase weathering rates, which are nearly ubiquitous in natural environments<sup>53</sup>. The biological increase of chemical weathering rates could allow comminution to larger grain sizes, which would save energy and improve the CO<sub>2</sub> budget. However, a lot more research on the controls of potential rates of enhanced weathering is needed, before a qualified quantification of the temporal dimension of this technique will be feasible.

In addition, enhanced weathering was reported to improve soil and plant productivity in agriculture. One widely applied method of enhanced weathering of carbonate rocks is agricultural liming to raise pH values of acidic soils, which already significantly impacts e.g. alkalinity flux into the Gulf of Mexico<sup>54</sup>. Enhanced weathering of silicate rocks could benefit the primary agricultural use of the areas where it may be applied. The released silicon is a beneficial nutrient for many plants<sup>55</sup>, it enhances resistance of rice to certain diseases<sup>56</sup> and helps some grasses to defend against herbivores<sup>57</sup>. In addition, trace contents of phosphorus and other elements in the weathering rock powder could increase the productivity of some agricultural areas<sup>58</sup>. Alternatively, metals released by the weathering rock powder could inhibit plant growth

and use<sup>10</sup>. These effects particularly impact the choice of source rocks to optimize the mineral content according to the needs in the application areas. The small proportion of transport emissions to the CO<sub>2</sub> budget suggests a choice of source rocks based on their optimal mineral content rather than based on their proximity to the application areas. The effect of enhanced weathering on agricultural output will be one of the main factors determining the success of the method. Thus, not only regarding enhanced weathering as a CO<sub>2</sub> removal method but also regarding alternative fertilization methods in the face of dwindling phosphate rock resources<sup>59</sup>, the potential effects of enhanced rock weathering on agricultural productivity<sup>60</sup> need more research.

Regarding the huge logistics necessary for global application, terrestrial enhanced weathering can only be one piece of the puzzle to control or reduce atmospheric CO<sub>2</sub> levels. This study highlights that associated emissions not exceed the carbon sequestration potential for most application areas even under pessimistic assumptions. However, as costs are usually expressed in terms of net carbon sequestration potential (e.g. US\$ tCO<sub>2</sub><sup>-1</sup>, or GJ tCO<sub>2</sub><sup>-1</sup>) the closer the budget approaches zero, the higher the unit cost. Combining the assumptions of energy use for extraction, comminution and spreading (Table 1), converting the transport emissions into energy requirements (assuming an emissions intensity of 77 gCO<sub>2</sub> MJ<sup>-1</sup> REF), and normalizing against net carbon sequestration, the total energy requirements of terrestrial enhanced weathering is 1.6 (9.9) GJ per tonne of CO<sub>2</sub> sequestered. This range is similar to other technologies that propose to remove carbon dioxide from the atmosphere (e.g. Renforth et al 2013).

Terrestrial enhanced weathering could be targeted in regions with high potential weathering rates or on soils depleted in cations and subject to biological carbon management e.g. afforestation, where suitable rocks could provide nutrients for biological carbon storage. Large

uncertainties in the budget, the weathering rates, and the possible side effects on soil productivity highlight the need for more targeted research before practical application might commence.

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