Sequestration of CO_2 by concrete and natural minerals - current status, future potential, and additional benefits

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Abstract

Concrete structures are some of the largest constructions in human civilization. Their manufacture releases CO_2 into atmosphere, which is partially readsorbed by standing structures, and further release occurs when they are demolished. Concrete is chemically similar to basaltic minerals, both adsorb CO_2 where they are exposed on the earth's surface. Sequestration of CO_2 is beneficial to reduce atmospheric concentrations, and thus limit future temperature increases. Therefore, multiple options are being examined for CO_2 sequestration. For the first time, we compare the CO_2 sequestration capacity of these two materials. We review previous work quantifying CO_2 sequestration capacity of both materials and for the first time, compare their potential quantitative roles. Costs of that are compiled, to the extent they have been examined. Costly grinding of these materials to small particle sizes accelerates CO_2 sequestration, and mycorrhizae in agricultural soils might reduce the associated costs. Both these materials can improve nutrient status in agricultural soils, and limit acidification from external nitrogen fertilization. Limitations are discussed in terms of land-use and material availability, and soil pH conditions. We call for further experiments with these materials that compare CO_2 sequestration and other biogeochemical processes in agricultural systems across climates, carried out especially where such materials are conveniently available.

Introduction

Basaltic minerals contain calcium and magnesium. They are formed by deep-earth processes at temperatures far exceeding 1,300 °C, where chemical bonds with carbonate are thermodynamically unstable. When those minerals appear at the earth's surface, they encounter much lower temperatures, and those minerals can react with and sequester carbon dioxide (CO_2). Rates of that process, called carbonation, are limited by mineral chemistry, surface areas, and access to carbon dioxide. Over millennia, temperature sensitivity and water limitations modulate rates of sequestration, and appear to act as a negative feedback thermostat on the Earth's climate[1].

An interesting parallel exists for concrete, the most widely used construction material on Earth. Ingredients of this anthropogenic material also include calcium, and its formation at temperatures exceeding 1,300 °C expels carbonate in the same way. Half of the CO_2 resulting from cement manufacture results from those chemical reactions, with the other half due to energy required for heating[2].

Sequestration capacity of CO_2 by all such materials is ultimately controlled by their disposition at the earth's surface, their chemistry, and by their surface areas. Accelerated C-sequestration by all such materials is being explored. Their current rates of CO_2 sequestration, and opportunities to increase those rates are reviewed here.

Chemical reactions by which these materials can sequester CO_2 have been described in earlier publications[3,4] and are not repeated here. Maximal C-sequestration occurs when added minerals' effects reach marine environments, and if limited to terrestrial effects, only half of that C sequestration occurs[5]. Sequestration is ineffective in areas with pH < 5[6]. Terrestrial applications can increase soil alkalinity, cation exchange capacity, and availability of limiting soil nutrients[7,8]. While some terrestrial experiments have been done, no marine or land-margin mineral application experiments have been published. We also address future work on that topic.

Current carbonation by basaltic minerals

Carbon sequestration by basaltic minerals was estimated to be 100 million tonnes of carbon per year[9]; equivalent to 0.1 Petagrams of carbon per year (10^9 tonnes; Pg C yr^{-1}). That estimate was later revised[10] to show that volcanic and crystalline minerals together sequester 0.08 Pg C yr^{-1} and sedimentary and carbonate minerals together sequester 0.16 Pg C yr^{-1}, for a total of 0.24 Pg C yr^{-1}. Based on geochemical exports from global rivers[11], these minerals sequester 0.25 Pg C yr^{-1}. Newer work[12] asserts that mineral weathering sequesters 0.3 Pg C yr^{-1}. Mineral weathering sequesters 0.4 Pg C yr^{-1}, according to the IPCC[13]. These estimates are presented together in Fig. 1. These carbon sequestrations remain outside global carbon-cycle accounting[14] as they are not anthropogenic. Carbona-
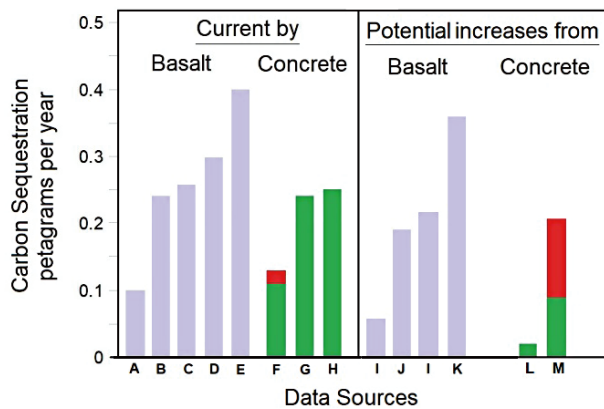
Fig. 1 Current global C sequestration rates by basaltic minerals and concrete, and potential increases from applying those materials to croplands. Data sources are Seifritz[9], Hartmann et al.[10], Gaillardet et al.[11], Strefler et al.[12], Ciais et al.[13], Cao et al.[20] with the red part of the bars showing C sequestration by ancillary concrete materials, Guo et al.[21], Xi et al.[19], Goll et al. using 20 million km^2 of global hinterland[22], Beerling et al.[5] (using 11% of global cropland), Goll et al. using 100 million km^2 of global hinterland[22], Beerling et al.[5] (using 22% of global cropland); potential from more extensive cropland utilization is not presented; higher estimates of potential C sequestration rates by basaltic minerals of 1.34 Pg C yr^{-1} by Strefler et al.[12] and 1.09 Pg C yr^{-1} by Fuhrman et al.[23] are excluded as those authors did not consider cropland area that would be required, Zhang et al. [24], Renforth et al.[25] with the red part of the bar showing C sequestration by ancillary concrete materials.

tion of basaltic minerals is increasing, along with surface temperatures. Rate of increase was estimated to represent 40% of the increase in global land C uptake since 1750[15]. That estimate was later revised downward to 0.008 Pg C yr^{-1}[16].

Basaltic minerals are present at the surface over broad continental areas, but many of those are far from agricultural areas[17]. Specifically, those authors showed that surface basalt exposures are rare in China but more proximal to agriculture in other regions, particularly in India[17]. Potential carbon sequestration rates by basaltic minerals are limited by particle surface areas and land available to apply them as crushed material, not by their global distributions.

Injection of CO_2 into deep basalt was (in 2019) sequestering less than 0.01 Pg C yr^{-1}[18]. This amount is too small to be shown in Fig. 1. It follows the same C sequestration chemistry as others reviewed here, but it is not further examined because CO_2 injection can only be done in few geological settings. Expansion of this C-sequestration much beyond 0.01 Pg C yr^{-1} would require developing additional sites[18] at costs that have not yet been estimated.

Current carbonation by concrete materials

A comprehensive examination of C sequestration by concrete in its various forms, during and after built service life has been made[19]. Those authors reported global production of cement was 4 billion tonnes in 2013. They reported carbonation of cement materials over their life cycle was 0.25 Pg C yr^{-1}, with China contributing 0.14 Pg C yr^{-1} in 2013. Global concrete in 2014 was sequestering 0.49 gigatons of CO_2 in total, of which 0.06 was by cement kiln dust, 0.02 was by construction waste, and 0.04 was by buried concrete demolition waste (CDW), with

the rest sequestered in standing concrete structures[20]. Global concrete in 2019 was sequestering 0.89 Pg CO_2 yr^{-1} in total. The latter two estimates were averaged for a total carbonation sink of 0.2 Pg C yr^{-1} in the most recent annual global carbon budget[14]. All these global estimates are presented in Fig. 1.

Removal of concrete structures produces concrete demolition waste (CDW). Production of CDW in 2017 was 3.1×10^9 tonnes globally, with 2.3×10^9 tonnes in China[24]. This represents a C sequestration capacity of 0.02 petagrams of C yr^{-1}[24], or 0.09 petagrams of C yr^{-1}[25]. This uncertainty could be decreased with additional field studies. Cement production, concrete building construction, and CDW generation rates will all increase through year 2100[3], illustrating an increasing potential for C sequestration by these pathways in the future.

In China, > 90% of CDW goes to landfills[26,27]. In contrast, about 60% of concrete is recycled in the US, with the remaining 40% sent to landfills[28,29]. In Europe, 61% of CDW is recycled and 39% sent to landfills[30,31]. Landfilled CDW and that recycled into road construction cannot sequester carbon[19]. Some amount of recycled CDW is added into new concrete manufacture[32,33] where it can continue C sequestration during the subsequent 'built phase' and its later 'CDW phase'. We have found no information on how much CDW is now being recycled into new concrete manufacture in any region.

To some extent, CDW has already sequestered carbon during the concrete-structure built phase. If CDW is to be widely added to soils for carbon sequestration, it is useful to know what fraction of CDW can continue to do this. Standing concrete structures are carbonated from exposed surfaces inward to greater depths. Concrete carbonation depth in small samples averaged < 3 mm yr^{-1}[34] and 1 mm yr^{-1} in the Itaipu dam[2]. Therefore, larger and younger concrete structures will present more carbon sequestration potential after being converted to CDW. This potential can readily be determined by simple chemical testing. The pH indicator phenolphthalein shows distinctive colors where CO_2 has not yet been sequestered[2]; useful for sorting CDW by potential for use in experimental-scale studies. However, depth of carbonation was on average twice that determined with the phenolphthalein indicator[35]. Forty two-year old concrete had carbonated to a depth of 1.2 mm yr^{-1}[36] at 9 °C annual T, compared to 1 mm yr^{-1} at 23 °C at Itapu. It seems possible that carbonation in built structures may be underestimated, and conversely, that potential future carbonation in CDW may be overestimated. It has further been estimated that carbonation depth increased with the square root of time[37], so that linear comparisons may be misleading. Reducing CDW to small particle sizes accelerates carbon sequestration[31], and additional assistance from soil mycorrhizae is considered below. Here it is important to also note that plant-nutritional benefits of CDW in soils persist even if minerals undergo prior carbonation.

Large-scale application of CDW to soils may still reveal remaining carbonation potential, if such tests are also used in experimental designs. In contrast, basaltic minerals are dark in color and cannot be sorted in this way.

Future potential carbonation by basaltic minerals - insights from experiments and models

The paucity of experimental work here has been noted, and guidance for interdisciplinary field trials presented[38]. Artificial

soils were made by them from CDW, dolerite (diabase) and quartz sand mixtures. AM fungi were not inoculated to soils, and were sampled from all treatments. Red fescue (*Festuca rubra*) was the target plant here. Both soil inorganic and soil organic carbon increased across treatments. Particularly in treatments with less sand (thus, more Ca + Mg). CDW showed higher increases that dolerite, consistent with its higher (Ca + Mg, 16.2% in CDW and not reported there for dolerite, generally 21% vs 12%; see Supplemental File 1). This was not a natural soil experiment, but it showed plants and fungi could grow in purely inorganic 'non biological' starting materials[39]. Yield increases of oats and maize were reported after addition of basaltic minerals sized from 4.8 to 0.3 mm[40]. Basaltic mineral powder additions to cropland soils were examined in mesocosms in the United Kingdom[41]. They reported 0.65 tonnes C ha^{-1} yr^{-1} of net C sequestration and 21% seed-yield increase in the basalt-addition treatments. The crop was *Sorghum bicolor* (a C4 plant) and a commercial mycorrhizal inoculum was also added.

Following application of 100 tonnes basalt ha^{-1} (125–250 μm particle sizes) to soil cores, herbaceous plants naturally grew in the soil cores and were neither removed nor measured, nor were mycorrhizae. This led to a net treatment removal of 10.2 ± 0.8 Kg CO$_2$ ha^{-1} yr^{-1}; markedly less than in similar experimental studies[42]. This was attributed to relatively low soil moisture availability, however as lime (calcium)-rich soils were used, there may have also been a background level of ongoing carbonation that limited detectable effects of new mineral additions. Both of those factors could relate to site-selection strategies for future large-scale mineral carbonation.

In a paired-catchment study in an oil palm plantation in Sabah, Malaysia, 50 tonnes ha^{-1} (andesitic basalt, particle size ~2 μm) was added to treatment catchments[43]. One of three catchment pairs showed net C sequestration of ~1 tonne CO$_2$ ha^{-1}, while no net effect was detected in the other two pairs. Effects were detected by analyses of exiting streamwater chemistry and soil geochemical analyses. Streamwater sampling was not done for 4 months during the second post-treatment year. All catchments also received Mg-containing fertilizers[43] which may have dampened net C sequestration responses.

A mesocosm study used high-pH (~7.7) soils with 50 tonnes ha^{-1} amendment of mixed basalt minerals containing 23.7% Ca + Mg[44] (see also Supplemental File 1). Particle sizes were dominated by 500 μm or below. Mesocosms were planted with potatoes (*Solanum tuberosum*) and soil mycorrhizae were not considered. Carbon sequestration was modeled as 1.83 and 4.48 tonnes CO$_2$ ha^{-1} after 1 and 5 years, respectively[44].

Application of crushed wollastonite (a calcium silicate mineral) to a temperate forest at Hubbard Brook, New Hampshire, USA at 3.44 tonnes ha^{-1} to caused direct C sequestration over 15 years of 0.0014 tonne C ha^{-1} yr^{-1}[45]. While that is a notably small amount, researchers also accounted for increased tree growth and (carbon) penalties associated with mineral supply and application, and found that total net C sequestration was 0.18 tonne C ha^{-1} yr^{-1}. Obviously, a single study cannot show how global forests might sequester C after mineral applications, especially as this one in northeastern US has been heavily impacted by atmospheric acidic deposition for decades. But it may represent an alternative where mineral supplies exceed cropland areas available for application.

Pot cultures of beans and corn (*Phaseolus vulgaris* L. and *Zea mays* L.) in Ontario, Canada soils were amended with crushed wollastonite[46]. They found mineral addition to have increased early crop growth and total soil carbon contents. They asserted such mineral additions sequester C, but did not present data that can be compared to other studies cited here.

Beyond implications for carbon, a modeling study predicted that applications of crushed basalt could reduce N$_2$O emissions by 16% in maize and 9% in *Miscanthus*, with several mechanisms being invoked[47]. This secondary effect on radiative forcing by greenhouse gases could come into play for other crops, and for CDW applications. It has not yet been examined by field experiments.

A modeling study of Brazil's Sao Paulo state suggested that applying crushed basalt (~5 mm) at 1 tonne ha^{-1} could capture 1.3 to 2.4 tonnes CO$_2$ eq y^{-1}[48]. They cited no site-based experimental studies, but they included energy and carbon costs of mining, transport distance, and particle-size reductions.

Basaltic minerals are also dilute sources of phosphorous. It has been modeled that 150 Pg of basaltic minerals powder application would be needed to close global P supply gaps in agriculture, and sequester 190 Pg C during 2006–2099[49].

Application of at least 1.1 Pg yr^{-1} of powdered basalt is required to sequester 0.35 Pg C yr^{-1} from the atmosphere[22].

After applying 50 tonnes ha^{-1} of powdered basaltic minerals, between 1.3 and 8.5 tonnes of CO$_2$ ha^{-1} was sequestered[50]. Authors suggested this should be considered illustrative for agricultural soils[50].

Enhanced basalt mineral weathering could provide sequestration of 6 to 30 million tonnes of CO$_2$ yr^{-1} for the United Kingdom by 2050[51]. They did not specify UK cropland area, but it is elsewhere reported as 4.3 million ha[52]. Based on that total area, their C sequestration estimate would be 0.4 to 1.9 tonnes C ha^{-1} yr^{-1}.

Croplands in China, India, the USA and Brazil combined have potential to sequester 0.14 to 0.55 Pg C y^{-1} with mineral application, depending on how much agricultural land is used[5]. Smaller contributions could come from other countries. Here we simplify their Table 1 within Fig. 1 to show global (specifically for their selected countries of Brazil, Canada, China, France, Germany, India, Indonesia, Italy, Mexico, Poland, Spain and USA) scenarios of carbon sequestration for different levels of effort by 2050. Their information is converted here from petagrams of CO$_2$ to petagrams of carbon, for consistency with other global studies. They reported carbon sequestration to scale linearly for cropland area dedicated, and for mineral supplies required. It may be surprising that costs are only weakly affected by project scales, but that supports authors' view that carbon sequestration by mineral application to croplands is a viable tool for global C sequestration. Their study contributes to a quantitative basis to compare C sequestration by natural minerals and concrete, even though it does not consider the latter. Global agricultural land area is approximately five billion hectares[53], implying that the use of 55 million hectares of agricultural land can sequester 0.2 Pg C yr^{-1} by application of crushed basaltic minerals, or 3.6 tonnes C ha^{-1} yr^{-1}. That is at least twofold higher than other site-based studies considered above, and that discrepancy cannot be resolved without additional field studies. Finally, this approach may be limited by cropland availability, as basaltic minerals are available in excess. Their highest scenario uses about 54% of global cropland to sequester 0.72 Pg C yr^{-1}.

Rock carbonation can sequester 0.68 Pg C yr^{-1} in non-agricultural areas[22], or 0.82 Pg C yr^{-1} without describing total areas that would be needed[54].

Weathering rates of basalts reduced to a grain size of 20 microns yield a total global C sequestration potential of 1.34 Pg C yr^{-1}[12]. It is clear that additional C sequestration by basaltic minerals can be increased beyond their current natural rate of 0.4 Pg C yr^{-1}, but additional field studies are needed to increase confidence in estimating benefits from upscaling.

A much more expansive potential assessment was presented[7]. Application of 50 tonnes ha^{-1} of basaltic minerals to 2×10^9 ha of tropical land (far exceeding tropical agricultural land areas) could sequester 550 to 1,300 Pg C during the 21st century, or about 5.5 to 13 Pg C yr^{-1}. Those estimates are excluded from Fig. 1, because of their large geographical extent and with direct and indirect costs far exceeding any efforts that could now be realistically considered.

Some basaltic minerals are 'ultramafic' meaning that they have high concentrations of magnesium (ma) and iron (fe), with olivine being often examined. They have particularly high sequestration potentials[55,56] and their global distribution can be seen at https://smartstones.nl/about-olivine/. However, their high concentrations of possibly toxic nickel and chromium argue against agricultural use[57].

Future potential carbonation by concrete minerals

Soil C sequestration by concrete construction and demolition waste (CDW) was shown to be 25 tonnes C ha^{-1} yr^{-1} in a UK urban 'brownfield' site with little vegetation cover, where CDW had previously been been incorporated into soils at depths up to 3 m[58]. In a subsequent similar UK study[59], CDW led to sequestration of 23 tonnes C ha^{-1} yr^{-1}. These similar values are substantially larger than any C sequestration reported for basaltic mineral applications to soils (see above), but amounts of CDW in soils was not measured in these studies. Based on chemistry, there is some reason to suppose that CDW has a higher C sequestration potential than basaltic minerals by weight. Carbon sequestration is limited by calcium and magnesium contents of minerals and mixtures involved. Those are summarized for basaltic minerals, ultramafic minerals, and concrete in Supplemental File 1. Briefly, concrete can have similar or higher Ca + Mg than basalt, but it is not normally measured during concrete production. Instead, these disparate results point to a need to perform cropland experiments using both materials, with chemical compositions measured, across a range of climates.

The lowest current CDW carbonation estimate was 0.014 Pg C yr^{-1}[60], accompanied by the highest estimate of future estimate of indirect carbonation of 0.58 Pg C yr^{-1}. Their indirect future estimate included reusing global CDW in new concrete manufacture and includes avoided CO_2 release by taking such future actions. As such, it is difficult to assess as to what plausibly can be done.

In laboratory incubations, freshly made concrete crushed to 1 to 8 mm size was exposed to 3,500 ppm CO_2 or higher. It absorbed 60% to 80% of the CO_2 released during calcination (cement manufacture at high temperature) within 20–35 d of exposure. Such high CO_2 concentrations can occur in soils, but no soils, plants or microbes were present in those incubations[61]. Those experiments confirmed that prompt and complete carbonation of cement minerals can occur under optimal conditions.

Artificial soils were constructed from different mixtures of CDW, dolerite (basalt) and sand (quartz) mixtures[39]. Meadow-seed mixtures were planted and AM fungi were sampled but not inoculated to soils. AMF colonized all treatments. Both soil inorganic and soil organic carbon increased across treatments; particularly so in treatments with less sand (more Ca + Mg). CDW showed higher increases than dolerite, consistent with its higher (Ca + Mg was 16.2% in CDW and not reported for dolerite, generally about 11% for basalt, see Supplemental File 1). It showed plants and fungi could grow in these purely inorganic 'non biological' starting materials[39].

Carbon-sequestration potential of several industrial byproducts, dominated by those from cement manufacture was examined[25]. Those authors asserted 7 to 17 billion tonnes of global production could have an annual sequestration potential of 0.19 to 0.33 Pg C. Direct and indirect potential C sequestration by waste materials from cement manufacture have been estimated as 0.26 Pg C yr^{-1} in China and 0.56 Pg C yr^{-1} globally[60]. Global potential C sequestration by ancillary concrete minerals are shown in Fig. 1 by the red parts of the bars relating to concrete.

Another way to accelerate C sequestration by concrete is to inject CO_2 during its manufacture. This increases concrete strength, but can promote degradation of steel reinforcing materials also used[62]. It also requires pure CO_2 to be provided during manufacture, which adds cost. Another way to improve C sequestration by concrete is to include CDW in its manufacture[62]. A similar approach is to add sodium bicarbonate to concrete during manufacture[63]. These could reduce CDW disposal in landfills, reduce amounts to transfer to agricultural or other soils, and allow C sequestration to continue in a subsequent built phase.

Future global CDW production is affected by extraordinary events

Xiao et al. estimated that 3×10^7 tonnes of CWD was generated by the Chi-chi earthquake in Taiwan (1999 September, magnitude 7.6) and that 2×10^8 tonnes of CWD was generated by the Wenchuan earthquake in Sichuan, China (2008 May, magnitude 7.9)[64]. The Tohoku earthquake in Japan (2011 March, magnitude 9.0) caused more than US$200 billion in damages, but < 9% of affected buildings were of concrete or masonry construction[65] as those are uncommon there. No assessment appears to have been made of CDW generation by that event. There have recently been 57 magnitude 7.5 or higher earthquakes globally per decade[66], some of which occur near populated areas and may generate large amounts of CDW. A global assessment of CDW from such events is lacking. The 2008 Wenchuan earthquake is the largest known addition of CDW by a seismic event, and may have added 7% to global annual CDW production in a small geographic area in 2008. It is expected that seismic events generally make smaller annual CDW additions (for example 1% addition to global annual CDW production by the Chi-chi earthquake in 1999), but they are notable for producing CDW over relatively small geographic areas. The Philippines suffered a magnitude 7 earthquake in 2022 July, but its structural damages including CDW

production cannot yet be assessed. A very recent swarm of earthquakes in Turkey and Syria destroyed > 10,000 large concrete buildings. Use of CDW for C-sequestration pales in comparison to needs for humanitarian assistance, and this large CDW source event may provide limited C-sequestration benefits because of regional climates and agricultural patterns in the region[67,68].

Such events can be viewed as opportunities for C sequestration by CDW application to soils, completely apart from their negative consequences.

The current military conflict in Ukraine is generating CDW at rates that cannot be accurately assessed, and it will continue for an unpredictable time. Yet it might be assumed that > 50,000 concrete buildings of 1,000 m³ have been demolished so far, which could represent > 3% of annual global CDW generation rate. This estimate is notable for occurring over hundreds of km near to highly productive agricultural areas in Ukraine, where CDW might later be beneficially deployed. High accuracy of our current estimate is not being asserted, but we suggest that large military conflicts can generate CDW over larger areas than seismic events, and that both (beyond all their other consequences) could be seen as local or regional C-sequestration opportunities.

Estimated costs of C sequestration by these minerals

Increased mineral sequestration of atmospheric CO_2 carries costs estimated by earlier research, and those estimates are summarized here. It must be stressed that C sequestration can be expressed in terms of C or CO_2, with the latter being 3.667 higher, or for costs, equally lower. Here we describe all based on C.

Costs of sequestering C in Scotland ranged from US$70 to 6,000 per tonne of C sequestered, increasing with larger efforts[69]. It was proposed that finely ground basaltic minerals could be placed within structures in Japan to absorb CO_2 from 415 ppm ambient air. Fine grinding (to size < 3 microns) constituted more than half of estimated energy costs. Overall costs were estimated as about US$350 per tonne C trapped[70]. The process proposed was abiotic; basaltic minerals would interact directly with CO_2 without soil, plant root or mycorrhizal processes.

Fuss et al. estimated annual costs for a global potential of 0.5 to 1 Pg C yr⁻¹ by 2050 to be from US$180 to 730 per tonne of C sequestered[54]. Carbon sequestration by new use of basaltic minerals could cost US$200 to 1,580 per tonne C[25]. It could cost US$550 per tonne C, for 1.8 Pg C yr⁻¹ trapping[12]. It could cost US$200 to 710 per tonne C, for 0.14 Pg C yr⁻¹ trapping, to US$300 to 810 per tonne C, for 0.56 Pg C yr⁻¹ trapping[5]. Global costs range from US$370 to 1,800 per tonne of C sequestered[22]. It could cost US$880 to 1,100 per tonne C in 2020 (with lower costs anticipated in later years)[51]. Corresponding costs of CDW sequestration have not yet been estimated.

These wide-ranging cost estimates provide little guidance, and could be improved by experiments using basaltic minerals and CDW crushed to various sizes, with addition to agricultural soils across climates, while also exploring effects of soil mycorrhizae. Even though CDW has less global potential than basaltic minerals (Fig. 1), it may be available closer to croplands.

Strefler et al. reported costs exceeding US$150 per tonne for grinding rocks to 2 microns[12]. As mineral crushing to sub-cm size adds substantial costs[31] we suggest that much can be learned from adding basalts and CDW of different sizes to agricultural soils across climates, and with consideration of different crops and their mycorrhizal counterparts. Proximity of CDW to agricultural soil target areas (as occurs in China) would be a factor in minimizing transport cost and its associated carbon footprint[71].

What further experiments need to be done?

No previous studies consider C sequestration by coarse aggregate minerals included in manufacture of concrete or CDW, that comprise more than half by weight, which are sometimes C-sequestering basalt (see Supplemental File 1), when especially high strength is required[72,73]. Inclusion of basaltic minerals in concrete (and thus later CDW) represents an additional C sequestration pathway, if they are reduced to small particle sizes, or if mycorrhizae can improve access for carbonation.

No field research efforts have yet manipulated soil mycorrhizae in relation to C sequestration by mineral applications to croplands. That is surprising, as soil mycorrhizae gave the earliest land plants capability to alter soil minerals, and they have continued in this role throughout biological evolution[74].

Ectomycorrhizal fungi have been shown to create channels into basaltic rocks[75−77]. Fungi, bacteria and archea all degrade organic minerals and mobilize products with excreted acids and chelating agents[78−80]. Microbes are < 1/10 the size of plant root hairs and so can access smaller existing defects in mineral assemblages[81]. By hydrostatic expansion they increase mineral surface areas. Arbuscular and ectomycorrhizal fungi both participate[80]. Ectomycorrhizal fungi in conjunction with gymnosperms and angiosperms release calcium from basalt at twice the rate of arbuscular fungi in conjunction with gymnosperms[82]. It is not currently known if some are superior in both accelerating mineral degradation and improving crop or forest productivity. Further research should be done on arbuscular fungi in conjunction with crops and basaltic mineral and CDW applications[83]. Soil fungi, bacteria, and nitrogen-fixing bacteria have all been shown to accelerate carbonation of Ca- and Mg-containing minerals[4].

Most but not all crops rely upon mycorrhiza[84]. Soil fungal interactions with minerals added to agricultural soils may remove the need for minerals to be reduced to small sizes (and their associated costs). Future work should explore this possibility. Some plant families include crops that do not rely upon mycorrhiza, and both types of crops should be compared together in field experiments measuring crop production as well as C sequestration.

Experiments with basaltic minerals have typically crushed those to 10 to 100 microns (0.01 to 0.1 millimeter size). Costs of that have been estimated to be a large part of total processing[51]. While rock-crushing equipment is widely used in CDW disposal, its main products are of cm-or-larger sizes. It may not be feasible to deploy smaller-crushing equipment in global efforts considered here.

A case study in Austria found that enhanced mineral weathering will only significantly contribute to net CO_2 drawdown if particle sizes < 10 microns are used[85]. Previous work thus emphasizes the importance of comparing a range of particle

sizes for both basaltic minerals and CDW, with and without mycorrhizal assistance.

This review compares C sequestration by soil burial of basaltic minerals and CDW in experiments that have been done separately, and gave dissimilar results. Side-by-side comparisons of both mineral types would greatly help in planning improvement of C sequestration and agricultural productivity with these under-utilized resources.

Finally, crops are grown globally under average temperatures spanning at least 5 to 28 °C, but very little of that climatic range has yet been examined for C sequestration by materials considered here. We envision agricultural experiments distributed across global climates using both basaltic minerals and CDW, exploring a range of particle sizes, and with both mycorrhizal and non-mycorrhizal crops. Obviously this represents substantial research efforts. Those appear to be justified by global-scale C sequestration potentials summarized here.

Other applications – C sequestration in marine areas

No experiments have apparently been carried out on applying minerals for C sequestration to marine or land-margin areas, even though C sequestration there is potentially higher and accompanied by beneficial pH increases. A model of enhanced rock weathering in marine systems showed potential sequestration of 2.7 Pg C yr^{-1}[86]. A scoping study considered dispersal of crushed olivine to coastal environments[87]. It presented a framework to consider carbon sequestration and ocean de-acidification, and effects on marine biota. It did not consider potential negative effects of heavy metal releases to the marine food chain. Effects of basaltic minerals added to marine pelagic communities have also been examined[88].

Ocean acidification is seen as an important future risk[89], but experimental mineral applications have not been performed there. We suggest that while basaltic minerals and CDW are being developed for terrestrial applications, some of those resources could be directly applied near continental margins.

Overview - how does this carbonation compare to other global efforts?

Smith et al. examined impacts of land-based greenhouse gas removal options, including mineral carbonation, on ecosystem services, and addressing United Nations Sustainable Development Goals[90]. Haque et al. describe the potential of basaltic-mineral applications to croplands to serve as a climate change stabilization wedge[91]. Zomer et al. asserted that increasing tree cover on agricultural land by 10% globally, that is, by 1% per year for the next 10 years, could sequester more than 18 Pg C, or 1.8 Pg C yr^{-1}[92]. In comparison, C sequestration by basaltic minerals might require application to half of global croplands to achieve 0.7 Pg C yr^{-1}, and full utilization of the concrete supply chain could sequester an additional 0.2 Pg C yr^{-1}.

All estimates of what can be achieved thereby need confirmation from local research done widely. Even as direct C-sequestration benefits accrue, we are poorly informed on how improved soil fertility can also benefit plant growth. Surficial deposits of basaltic minerals can be seen as essentially infinite for C sequestration, but not all are conveniently located with respect to croplands. CDW, while less, comes from urban areas and may be closer to target areas.

Basaltic minerals are currently sequestering about 0.4 Pg C yr^{-1} and concrete minerals are currently sequestering about 0.25 Pg C yr^{-1}. Both of those can be increased by dispersed applications to soils, particularly croplands, but their global potentials will remain uncertain until field experiments with crops and mycorrhizae are widely performed. Mineral sequestration of C may have benefits similar in magnitude to strategies that increase plant growth by burying biochar in soils[5]. In addition, it increases soil fertility and can de-acidify both terrestrial and marine systems[5]. Large amounts of basaltic minerals and CDW are available and represent untapped resources, and their value can only be assessed by future experiments. Basaltic minerals and CDW can sequester additional CO_2 in soils. This can be done most efficiently where source materials are close to target soils, and where mycorrhizae can act in place of particle grinding to facilitate access to CO_2.

No single approach can offset all fossil-CO_2 emissions. Many approaches need to be considered in terms of costs, practicality and potential benefits. Mineral applications to croplands examined here are constrained in terms of C-sequestration potential, but are accompanied by soil nutrition and de-acidification benefits.

Author contributions

The authors confirm contribution to the paper as follows: study conception and design: Schaefer DA; draft manuscript preparation: Schaefer DA, Gui H; supervision and editing: Xu J. All authors reviewed the results and approved the final version of the manuscript.

Data availability

Data sharing not applicable to this article as no datasets were generated during the current study.

Conflict of interest

The authors declare that they have no conflict of interest.

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