

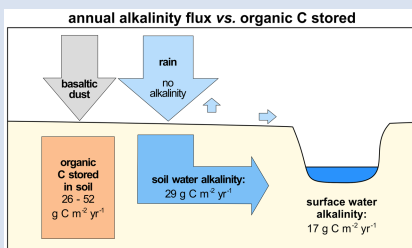
# Direct evidence of CO<sub>2</sub> drawdown through enhanced weathering in soils

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## Abstract



The ability of engineered enhanced weathering to impact atmospheric CO<sub>2</sub> has been challenging to demonstrate due to the many processes occurring in soils and the short time span of current projects. Here we report the carbon balance in an Icelandic Histic/Gleyic Andosol that has received large quantities of basaltic dust over 3300 years, providing opportunity to quantify the rates and long term consequences of enhanced weathering. The added basaltic dust has dissolved continuously since its deposition. The alkalinity of the soil waters is more than 10 times higher than in equivalent basalt dust-free soils. After accounting for oxidation and degassing when the soil waters are exposed to the atmosphere, the annual CO<sub>2</sub> drawdown due to

alkalinity generation is 0.17 t C ha<sup>-1</sup> yr<sup>-1</sup>. This study validates the ability of fine grained mafic mineral addition to soils to attenuate increasing atmospheric CO<sub>2</sub> by alkalinity export. Induced changes in soil organic carbon storage, however, likely dominate the net CO<sub>2</sub> drawdown of enhanced weathering efforts.

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## Introduction

The natural weathering of basaltic and ultramafic rocks has been demonstrated to have a relatively large role in the drawdown of CO<sub>2</sub> from the atmosphere (Dessert *et al.*, 2003; Gislason *et al.*, 2009; Taylor *et al.*, 2021). Such observations have motivated several proposals to use these rocks to remove CO<sub>2</sub> directly from the atmosphere through a process called Enhanced Weathering (EW) (Moosdorf *et al.*, 2014; IPCC, 2018; Beerling *et al.*, 2020). Enhanced weathering involves amending soils with crushed fine grained, fast reacting Ca-Mg silicate rocks and minerals such as basalts and peridotites (Strefler *et al.*, 2018). To date, enhanced weathering field experiments have demonstrated improved crop vigour, organic and inorganic carbon storage and decreased N<sub>2</sub>O degassing (Haque *et al.*, 2019a, 2020; Beerling *et al.*, 2020). One of the goals of EW is to increase the alkalinity export of waters that drain from soils and enter rivers and streams. The quantification of carbon drawdown by enhanced weathering has been challenging to identify or quantify due to the large number of processes that occur in soils and the short duration of existing field studies.

One approach to investigate the long term behaviour and consequences of EW is *via* natural analogues. Enhanced weathering experiments to date have tested the addition of up to 400 t ha<sup>-1</sup> yr<sup>-1</sup> of crushed Ca-Mg silicate rocks to agricultural soils (Gillman *et al.*, 2002; Amann *et al.*, 2018; Haque *et al.*, 2019b). This crushed rock flux is orders of magnitude higher than the average global desert dust deposition on Earth, which is estimated to be 0.5 t ha<sup>-1</sup> yr<sup>-1</sup> (Mahowald *et al.*, 2005). In the vicinity

of the dust “hot spots”, such as South Iceland, however, the mass of deposited fine grained basaltic dust can be as high as 8 t ha<sup>-1</sup> yr<sup>-1</sup> (Arnalds, 2010; Arnalds *et al.*, 2014, 2016). Although this natural mass flux of basalt is less than that of current EW efforts, 1) this basaltic dust flux has been added continuously to the soil of this region over at least the past 3300 years, such that in total over 16,500 t ha<sup>-1</sup> of basaltic dust has been added over this time, and 2) the specific surface area of natural basaltic dust is likely higher than that added to soils in current EW experiments due to its finer grain size. The grain size of the crushed rocks used in EW applications, if reported, is commonly less than 150 μm (Haque *et al.*, 2019a; Gillman *et al.*, 2002). In contrast, the average size of basaltic Icelandic dust ranges from 10 to 62 μm (Arnalds *et al.*, 2014; Liu *et al.*, 2014). For these reasons, the mineral rich Histic/Gleyic Andosols (Arnalds, 2015) considered in this study located in South Iceland provide an insightful natural analogue to illuminate the long term effect of EW applications performed under similar climate, vegetation, and soil conditions. The studied soil receives large amounts of air borne volcanic material during 1) explosive volcanic eruptions in the form of glassy volcanic ash fallout, and 2) dust storms (Shoji *et al.*, 1995; Arnalds *et al.*, 2016). Explosive eruptions lead to evident tephra horizons that can be used to date these soils. The more frequently deposited windblown dust is finer grained than the tephra and intermingled with the soil organic carbon (see “Soil classification and soil evolution in Iceland” in Supplementary Information). Based on palaeoecologic research (Gísladóttir *et al.*, 2008; Arnalds, 2015; Möckel *et al.*, 2017), in the absence of volcanic dust input, the Histic/Gleyic Andosols of

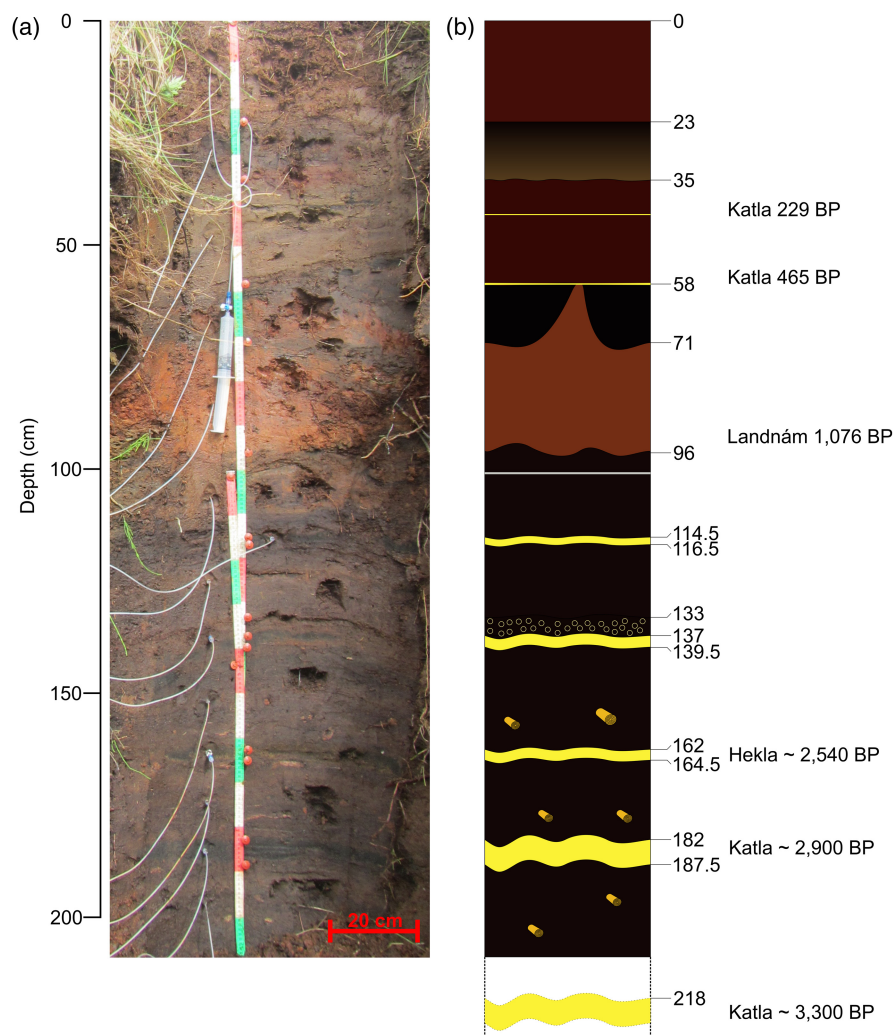
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**Figure 1** (a) Photograph of the studied soil system. The dark layers correspond to tephra originating from historic volcanic eruptions. (b) Schematic illustration of the soil profile with depths and ages (in years before present) of identified tephra layers shown in yellow for better visibility, the Landnám Layer that occurred around the time of the Icelandic settlement could not be clearly identified because of alteration. Plant remnants are visible in the lower part of the profile, indicating high organic content.

Southern Iceland would have developed into Histosols (Arnalds, 2008). Consequently, the comparison of the behaviour of South Icelandic Histic/Gleyic Andosols with that of volcanic dust-free Histosols, located in similar climatic zones provides insight into the consequences of adding fine grained basaltic material to soils as part of enhanced weathering efforts.

One of our motivations to focus on the addition of volcanic material to Histosols/peat soils to drawdown  $\text{CO}_2$  from the atmosphere stems from the role of these soils in the global carbon cycle. Although peatlands cover only about 3 % of the continents (Xu *et al.*, 2018), they store  $\sim 10$  % of all non-glacial freshwater and roughly 30 % of the land-based organic carbon (Mitra *et al.*, 2005; Bragazza *et al.*, 2013). Man made drainage and burning of peat areas worldwide releases  $0.5\text{--}0.8 \text{ Gt C yr}^{-1}$ , which is equivalent to 5–8 % of global anthropogenic carbon emissions (Hooijer *et al.*, 2006; Parish *et al.*, 2008). Carbon dioxide emission from the drainage of peat areas is estimated to be the largest anthropogenic source of  $\text{CO}_2$  emissions in Iceland (Keller *et al.*, 2020). The addition of reactive silicate rock dust to peat soils might help increase carbon storage being otherwise lost due to peatland draining.

This manuscript is one of two exploring the long term efficiency and consequences of enhanced rock weathering efforts

through the study of a South Iceland Gleyic/Histic Andosol. The first manuscript (Linke *et al.*, 2024) reports the composition of fluids and solids collected over two field seasons to 1) quantify the saturation state of the primary and secondary mineral phases with respect to the soil solutions, 2) determine the processes controlling the mobility of heavy metals, and 3) assess the rate at which basalt dissolved in the soils. In this manuscript we present a comparison of the alkalinity export from this soil with corresponding results from volcanic dust-free Histosols to quantify the ability of enhanced weathering efforts to drawdown  $\text{CO}_2$  from the atmosphere. Results are then used to estimate the efficiency of enhanced weathering at a larger scale. The purpose of this paper is to present the results of this study and use the results to gain insight into the consequences of current and future enhanced weathering efforts.

## Field Site Description

The field site is located above the source of the Rauðalækur (“Red creek”) river at  $63^\circ 53' 42.5'' \text{ N}$ ,  $20^\circ 21' 15.9'' \text{ W}$ , which is approximately 7 km north of the town of Hella in South Iceland. This site consists of an upper Gleyic Andosol and a lower Histic Andosol (see Supplementary Information for further

details). The study site receives an annual aeolian dust flux of 5–8 t ha<sup>-1</sup> yr<sup>-1</sup>, consisting of mostly basaltic glass (Arnalds *et al.*, 2016). Additional basaltic material is added during irregular volcanic events. During the past several decades, drainage trenches have been cut into the nearby soils; the closest drainage ditch is located more than 150 metres from the study site. The studied soil contains several prominent horizontal tephra layers, with thicknesses ranging from a few mm to few cm and deposited during the past 3300 years. An image and schematic illustration of the system is provided in Figure 1 and a detailed description of the field site is provided in the Supplementary Information and Table S-1 therein.

## Results

**Fluid compositions.** Soil fluid samples were collected using suction cup samplers from May to November 2018. The compositions of all fluid samples are provided in Table S-2 (Supplementary Information) and selected dissolved constituents are shown as a function of depth in Figure 2. The pH of the samples was recalculated using PHREEQC (Parkhurst and Appelo, 1999) to the *in situ* soil temperature of 7 °C. This is the average soil temperature at 76–260 cm depth during the summer months (Petersen and Gerber, 2018). The concentrations of major elements increase continuously with depth suggesting the continuous dissolution of the basaltic dust in the soil. The soil waters become increasingly anoxic with depth as indicated by the Eh values shown in Figure 2b.

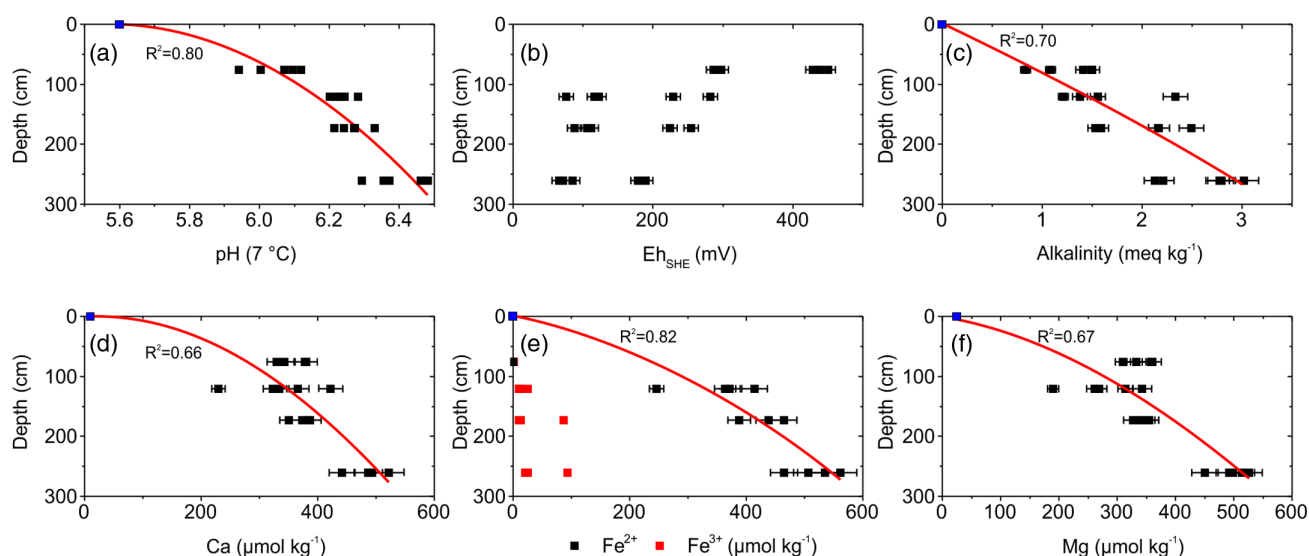
The alkalinity of the soil waters increases from 0 to 3 meq kg<sup>-1</sup> with depth. Once these waters exit the soils, they will equilibrate with the O<sub>2</sub> and CO<sub>2</sub> in the atmosphere. PHREEQC calculations indicate that the alkalinity of the soil waters will decrease on average to 1.53 ± 0.2 meq kg<sup>-1</sup> due to iron oxidation/precipitation reactions when they come in contact with the atmosphere as they flow into local rivers (further details of this calculation are provided in the Supplementary Information).

The alkalinity of soil waters in our studied dust-rich soil are compared to the corresponding alkalinities of basalt

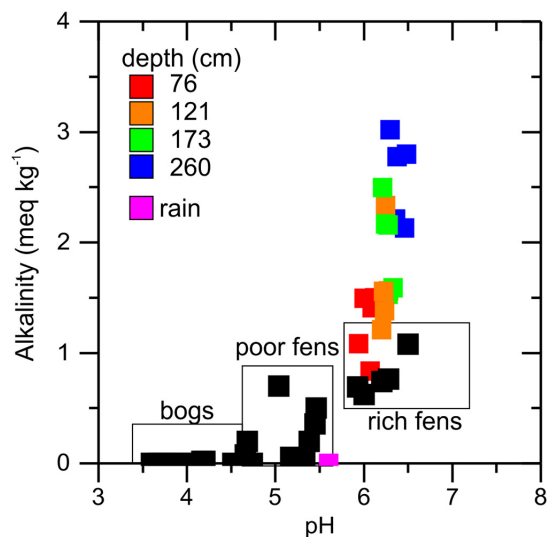
dust-free Histosols located in non-carbonate terrains in Figure 3. Our field site, mostly fed by rainwater, shows considerably higher alkalinity and pH values than observed in corresponding basalt dust-free Histosols. Histosols located in carbonate terrains are not included in this comparison as the presence of carbonate minerals leads to a pH and alkalinity increase due to carbonate dissolution, a process which has no long term net effect on atmospheric carbon drawdown. The comparison in Figure 3 shows that the addition of volcanic dust to our soil increased substantially the alkalinity in its soil waters, most notably deep in the soil column. This observation confirms the ability of enhanced weathering by the addition of basaltic dust to soils to drawdown CO<sub>2</sub> from the atmosphere.

A noteworthy observation is that the basaltic dust in the studied soil column persists and is reactive throughout the soil column, despite the fact that some of this dust has been present in the soil for 3300 years. This observation is consistent with mass balance estimates of the import and export of metals to the soil column. The study site receives an average annual dust flux of 500–800 g m<sup>-2</sup> yr<sup>-1</sup>. This basalt flux adds 0.96–1.54 mol Ca and 0.72–1.16 mol Mg per m<sup>2</sup> yr<sup>-1</sup> to the soil. In contrast, the average Ca and Mg concentration of the deep soil water is 5 × 10<sup>-4</sup> and 4 × 10<sup>-4</sup> mol kg<sup>-1</sup> for Ca and Mg, respectively. Taking account of the estimated 925 ± 150 kg m<sup>-2</sup> yr<sup>-1</sup> of water that flows through, and is exported annually by our studied soil (see Supplementary Information for details of this water flow estimate), we estimate that 0.47 ± 0.07 and 0.37 ± 0.06 mol yr<sup>-1</sup> of Ca and Mg, respectively, are removed from the soil per square metre of soil surface area at present. The input of Ca and Mg by volcanic dust addition is, therefore, approximately 2–3 times more than that removed by soil water export. The results of this comparison are consistent with the persistence of the reactive dust throughout the soil column and suggest the long term viability of enhanced weathering efforts.

**Carbon Storage via alkalinity export by the addition of basaltic material to soils.** The rate of carbon drawdown due to alkalinity export by enhanced weathering in our studied field site can be estimated by combining the annual water flux through the soil and the measured alkalinity, as demonstrated in



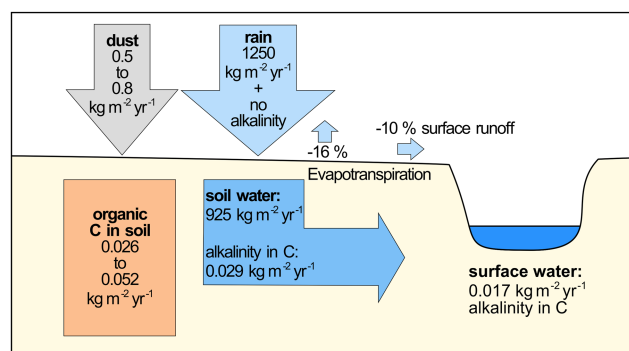
**Figure 2** Measured soil water concentrations determined in the present study in all samples collected from May to November 2018, as a function of depth. The pH values are normalised to a 7 °C reference temperature and Eh values to a Standard Hydrogen Electrode. The black squares represent measured water concentrations, whereas the blue squares show the composition of rainwater. The error bars correspond to a ±5 % uncertainty on the measured concentrations; error bars do not appear if the uncertainty is smaller than the symbol size. The red curves in the figure show 2<sup>nd</sup> order polynomial fits of all the measured concentrations with the corresponding R<sup>2</sup> values next to each curve.



**Figure 3** Comparison of pH and alkalinity of soil waters collected from our studied soil with similarly composed, but volcanic dust-free soils reported in other studies. The black symbols correspond to alkalinity values reported in the literature for Histosols from bogs, poor or rich fens located in non-volcanic regions and in the absence of carbonate bedrock. The black boxes around the black symbols represent the commonly reported pH-alkalinity ranges of bogs and fens respectively. The red, orange, green and blue symbols represent soil water samples measured in the present study at the depths indicated in the figure. The purple symbol shows the composition of rainwater at our field site. The sources and location of the literature data are provided in Table S-3 of the Supplementary Information.

**Figure 4.** By taking account of the rainfall, evaporation, and surface runoff it is estimated that  $925 \pm 150 \text{ kg m}^{-2} \text{ yr}^{-1}$  of water pass through and are exported from the studied soil annually. Multiplying this number by the  $1.53 \pm 0.2 \text{ meq kg}^{-1}$  average alkalinity of the deepest water samples of our study area, after its equilibration with the atmosphere, yields an estimated alkalinity export from our soils of  $1.43 \pm 0.3 \text{ meq m}^{-2} \text{ yr}^{-1}$ . Multiplying this number by the atomic weight of carbon yields an annual carbon addition to our river water of  $17 \pm 3.6 \text{ g m}^{-2} \text{ yr}^{-1}$ , which equals  $0.17 \pm 0.036 \text{ t ha}^{-1} \text{ yr}^{-1}$  of C. The degree to which this carbon drawdown rate is influenced by the rate of basalt input to the soil and its surface area has yet to be quantified. Although, dissolution rates are commonly thought to be proportional to the fluid-mineral surface area, these rates are also influenced by fluid compositions, including approach to equilibrium and fluid flow paths in the soil column (Schott *et al.* 2009; Linke *et al.*, 2024).

It is insightful to extrapolate this annual rate of carbon drawdown to a larger scale. If the results of our studied field site are representative, the removal of  $1 \text{ Gt yr}^{-1} \text{ CO}_2$  from the atmosphere through alkalinity production would require a total of 16 million  $\text{km}^2$  of surface. This is larger than the total surface area of the United States. Moreover, the mass of basaltic dust required to provoke this rate of carbon removal may be unrealistically large. The average annual flux of basaltic dust into the studied South Iceland soils is  $5\text{--}8 \text{ t ha}^{-1} \text{ yr}^{-1}$ . Adding this mass of basalt over 16 million  $\text{km}^2$  of surface would require 8 to 13 Gt of finely ground basalt annually. This mass of ground basalt is larger than the world's annual cement production of 4.3 Gt in 2020 (<https://iea.org/reports/cement>). This conclusion, based on the alkalinity export from our studied soil, which contains more organic matter (12 % to >20 % C) than most soils globally (<5 % C; *e.g.*, Stockmann *et al.*, 2015), is nevertheless supported



**Figure 4** Schematic illustration of the processes drawing down  $\text{CO}_2$  at our field site. The site receives  $\sim 1250 \pm 200 \text{ kg m}^{-2} \text{ yr}^{-1}$  of rainfall. Of this rainfall 16 % is estimated to evaporate and 10 % is estimated to be lost to surface runoff. As the remaining  $925 \pm 150 \text{ kg m}^{-2} \text{ yr}^{-1}$  of water passes through the soil, its alkalinity increases on average from 0 to  $2.59 \pm 0.34 \text{ meq kg}^{-1}$  at depth. Once these waters equilibrate with the atmosphere, this fluid oxidises and some  $\text{CO}_2$  is released such that the alkalinity decreases to  $1.53 \pm 0.2 \text{ meq kg}^{-1}$  resulting in an annual export of  $17 \pm 3.6 \text{ g C per m}^2$  soil surface area. At the same time  $26\text{--}52 \text{ g m}^{-2} \text{ yr}^{-1}$  of C is drawn down from the atmosphere by organic carbon production and stored in the soil.

by other recent enhanced rock weathering studies. Our result of  $0.17 \pm 0.036 \text{ t ha}^{-1} \text{ yr}^{-1}$  of carbon drawdown from alkalinity export by enhanced rock weathering is within the range of the handful of large scale cropland EW studies of  $0.0005\text{--}0.5 \text{ t C ha}^{-1} \text{ yr}^{-1}$  (Haque *et al.*, 2020; Taylor *et al.*, 2021; Larkin *et al.*, 2022). It should be noted, however, that the alkalinity generated in our studied Histic/Gleyic Andosol was the consequence of the dissolution of the basalt added to this soil annually over the past 3300 years. This annual addition has led to a buildup of basaltic material over time. The results shown in Figure 2 indicate that the presence of older basaltic dust, located deep in the soil profile is an important contributor to alkalinity production. As such, it seems likely that substantially more than  $5\text{--}8 \text{ t ha}^{-1} \text{ yr}^{-1}$  would need to be added to soils near-term as part of enhanced weathering efforts to provoke a similar rate of alkalinity production as observed in our study area.

**Carbon drawdown by alkalinity production versus soil organic carbon.** The total mass of organic carbon in our studied soil is estimated to be  $86\text{--}172 \text{ kg C m}^{-2}$  with average net annual rate of carbon drawdown estimated to be  $26\text{--}52 \text{ g C m}^{-2} \text{ yr}^{-1}$  (see Supplementary Information). This rate of  $\text{CO}_2$  drawdown is substantially larger than the corresponding  $17 \pm 3.6 \text{ g C m}^{-2} \text{ yr}^{-1}$  drawdown due to alkalinity export in our studied soils. These estimates are in agreement with previous studies (Taylor *et al.*, 2021). These estimates also suggest that the amount of  $\text{CO}_2$  removed by the addition of basaltic dust to the soil in one year by alkalinity export is more than 3 orders of magnitude less than the total  $\text{CO}_2$  stored as organic carbon in the soil. This latter observation should serve as a warning to those attempting atmospheric  $\text{CO}_2$  drawdown by enhanced weathering in soils. If the addition of basaltic dust to soil leads to the accelerated decomposition of organic material in soils, the latter process could readily dominate leading to a net increase of  $\text{CO}_2$  released to the atmosphere due to enhanced weathering efforts.

## Conclusions

The results of this study confirm the ability of fine grained basaltic rock added to soils to enhance  $\text{CO}_2$  drawdown directly from the atmosphere due to alkalinity production. It is estimated

that  $17 \pm 3.6 \text{ g C m}^{-2} \text{ yr}^{-1}$  is currently drawn down and added to rivers by alkalinity production from our South Iceland field site. The enhanced alkalinity production of our soils was produced by the addition of approximately  $1.7\text{--}2.6 \text{ t m}^{-2}$  of basaltic dust to this soil over 3300 years. Upscaling of this process to address even a small fraction of the mass of anthropogenic  $\text{CO}_2$  emissions to the atmosphere, however, may be challenging because 1) this enhanced weathering process is slow and would require more land than is available for a sizeable drawdown of  $\text{CO}_2$  through alkalinity production, and 2) the currently unquantified effect of adding basalt powder to soils on soil organic matter. So, although this study demonstrates the potential of enhanced weathering efforts to contribute to attenuating atmospheric  $\text{CO}_2$  concentrations, the degree to which this approach will prove successful at a larger scale remains unclear.

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## Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2415>.



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