

Eion Corp Whitepaper

# Terrestrial Enhanced Rock Weathering: Fundamental Principles

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# 1 Summary

Enhanced rock weathering (ERW) is the process by which mineral additions via rock weathering store carbon on geologic time scales. Adding mineral-derived alkalinity to soil waters drives an increase in the storage of dissolved inorganic carbon (DIC). This DIC eventually flows via freshwater into the ocean, where it has a lifetime of approximately 50,000 years. Here, we present the fundamental carbonate budget, demonstrating the theoretical foundations of ERW. We follow the flow of DIC backwards, starting in the ocean, to freshwaters, to soil solution to calculate the storage potential and leakage of  $\text{CO}_2$ . We also describe a trace-element approach to measure the progress of mineral weathering and associated carbon dioxide removal.

## 2 Introduction

When mafic rocks are applied to the soil system, Mg is added, which influences the carbonate chemistry of the soil solution. Water that leaches out of the soil (and corresponding DIC) enters freshwater systems. Some of this DIC then reaches the saltwater of the ocean. In this section, we will follow the DIC from the soil to the sea to analyze the stability of the stored C and the impacts of the DIC on the pH balance of the fresh and sea water.

Here, we describe the influence of Mg addition on the pH, alkalinity, and carbonate chemistry of the soils, freshwater systems, and the ocean. We start in the ocean and work backwards, from simplest to most complex systems.

## 3 Geochemical Fundamentals

In this section, we walk through the carbonate system parameters, explorations of carbonate chemistry, carbonate chemistry in the ocean, freshwater carbonate chemistry, and carbonate chemistry in the soil system.

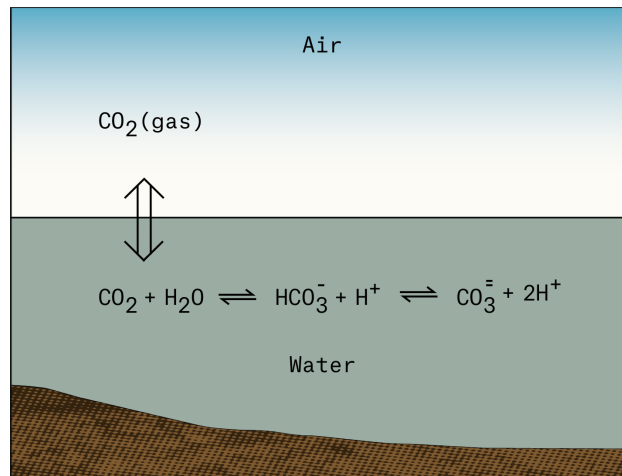


Figure 1: Figure showing that  $\text{CO}_2$  in the atmosphere is in equilibrium with the  $\text{CO}_2$  in the ocean. The ocean  $\text{CO}_2$  equilibrates with the carbonate and bicarbonate in the ocean [1].

### 3.1 Carbonate System Parameters

$\text{CO}_2$  in the atmosphere is dissolved into the ocean, where it can speciate into other forms including carbonate ( $\text{CO}_3^{2-}$ ) and bicarbonate ( $\text{HCO}_3^-$ ). The dissolution of gaseous, atmospheric  $\text{CO}_2$  into dissolved  $\text{CO}_2$  is based on a solubilization coefficient that is inversely related to the temperature of the surface ocean and is proportional to the partial pressure of  $\text{CO}_2$  in the surface ocean. The

equation follows Henry's law, which is that the dissolved gas in solution is proportional to its partial pressure. Thus,  $K_h$ , the solubility of  $\text{CO}_2$  in water is defined as:

$$[\text{CO}_2] = K_0(T, S) \cdot p\text{CO}_2 \quad (1)$$

Computed as:

$$\ln K_h = 9345.17/T - 60.2409 + 23.3585 \ln(T/100) + S[0.023517 - 0.00023656T + 0.00047036(T/100)^2] \quad (2)$$

where  $T$  is temperature in  $^\circ\text{K}$ ,  $S$  is salinity in UNITS, and units are mol/kg. The partial pressure,  $p\text{CO}_2$  can be closely approximated by the fugacity of  $[\text{CO}_2]$ ,  $f[\text{CO}_2]$  [2].

Once dissolved,  $\text{CO}_2$  speciates into the other forms of DIC (including carbonate  $\text{CO}_3^{2-}$  and bicarbonate  $\text{HCO}_3^-$ ) based on equilibrium kinetics.  $K_1^*$  is the stoichiometric equilibrium constant of the first dissociation of carbonic acid, between carbon dioxide and bicarbonate, defined as:

$$K_1 = \frac{[\text{H}^+][\text{HCO}_3^-]}{[\text{CO}_2]} \quad (3)$$

and  $K_2^*$  is the stoichiometric equilibrium constant of the first dissociation of carbonic acid, between bicarbonate and carbonate, defined as:

$$K_2 = \frac{[\text{H}^+][\text{CO}_3^{2-}]}{[\text{HCO}_3^-]} \quad (4)$$

These constants can be computed as:

$$\ln K_1 = 2.83655 - 2307.1266/T - 1.5529413 \ln T - (0.207608410 + 4.0484/T)\sqrt{S} + 0.0846834S - 0.00654208S^{3/2} + \ln(1 - 0.001005S) \quad (5)$$

and

$$\ln K_2 = -9.226508 - 3351.6106/T - 0.2005743 \ln T - (0.106901773 + 23.9722/T)\sqrt{S} + 0.1130822S - 0.00846934S^{3/2} + \ln(1 - 0.001005S) \quad (6)$$

where  $T$  is in  $^\circ\text{K}$  and units are mol/kg. [3]

Dissolved organic carbon (DIC) is the sum of all dissolved forms, including  $\text{CO}_2$ ,  $\text{HCO}_3^-$ , and  $\text{CO}_3^{2-}$ , and is notated as DIC or  $\sum \text{CO}_2$ , as defined in the following equation:

$$\text{DIC} = [\text{CO}_2] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}] \quad (7)$$

The relative proportion of these three species of DIC is driven by temperature and salinity. The speciation of DIC in turns impacts the concentration of  $[\text{H}^+]$  in the water, and therefore impacts the pH. These relationships are shown in Figure 2 below.

Carbonate alkalinity (CA) is the sum of the total charges on the carbon forms, which is:

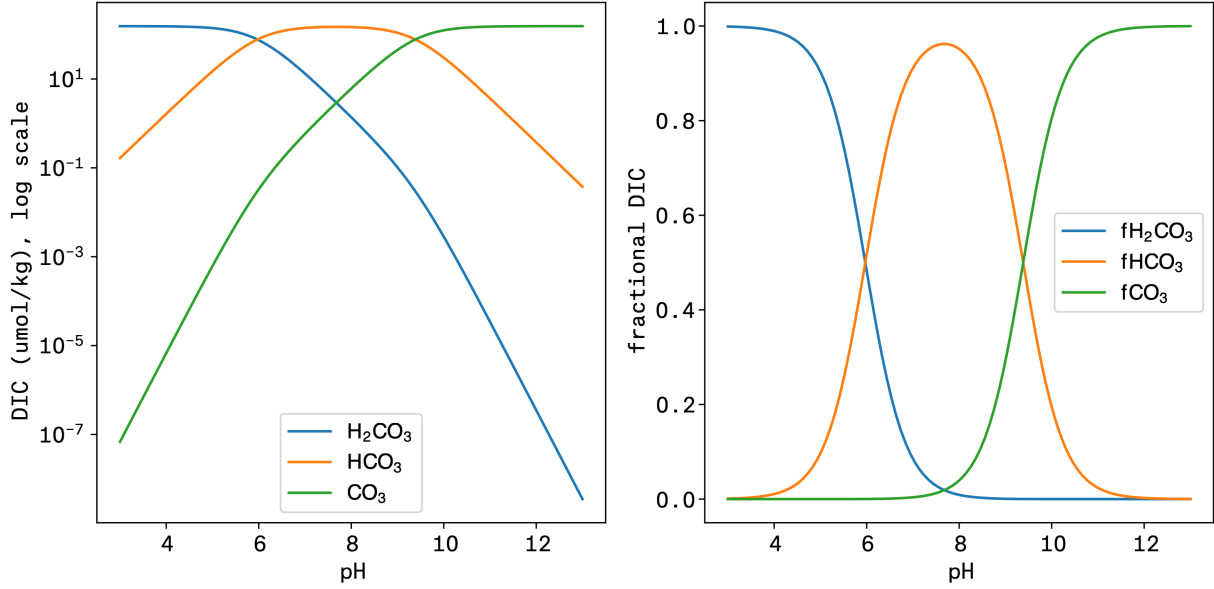


Figure 2: DIC speciation vs. pH, showing  $\text{CO}_2$ ,  $\text{HCO}_3^-$ , and  $\text{CO}_3^{2-}$ , on a log scale (left) and as a fraction of total DIC (right)

$$\text{CA} = [\text{HCO}_3^-] + 2 \cdot [\text{CO}_3^{2-}] \quad (8)$$

$\text{CO}_3^{2-}$  has twice the influence on CA as  $\text{HCO}_3^-$  because it has two negative charges, while  $\text{HCO}_3^-$  only has one.

The equivalence point is defined as the point at which carbonate alkalinity is 0, so

$$[\text{H}^+] = [\text{HCO}_3^-] + 2 \cdot [\text{CO}_3^{2-}] + [\text{OH}^-]. \quad (9)$$

The equivalence point is also known as the proton condition, which can be experimentally determined via titration.

Total alkalinity is similar but includes the influence of other ions, including Boron. Define total alkalinity (TA) as the sum of proton acceptors minus proton donors (for now ignoring minor species), and is interpreted as a charge imbalance:

$$\text{TA} = [\text{HCO}_3^-] + 2 \cdot [\text{CO}_3^{2-}] + [\text{OH}^-] - [\text{H}^+] \quad (10)$$

This definition of TA has units of meq/kg, where meq is the charge weighted molar concentration of the species under consideration:

$$\text{TA} = \sum_j z_j [c_j] \quad (11)$$

where  $c_j$  is an ion of interest,  $c$  is the concentration of that ion (units mol/kg)  $z$  is the valence of that ion (1, 2, or 3, positive or negative), hence 2 for  $[\text{CO}_3^{2-}]$  and -1 for  $[\text{H}^+]$ .

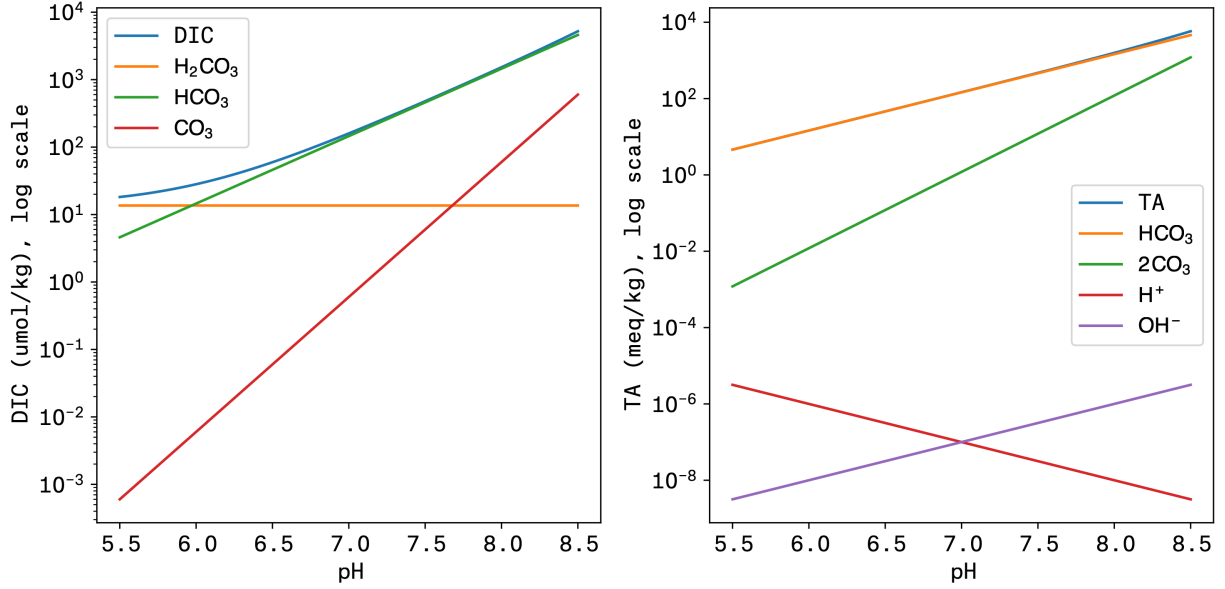


Figure 3: Carbonate species as they comprise DIC (left) and total alkalinity (right) across a range of pH.

We interpret alkalinity as the charge imbalance of conservative cations over conservative anions, and therefore that alkalinity is a conserved quantity. The charge imbalance between the cations and the anions in the ocean is responsible for the total alkalinity of the ocean. The chemical reactions in the ocean include photosynthesis and respiration, which remove or add  $\text{CO}_2$ , respectively. The charges remain constant even though the forms of carbon change.

Alkalinity defined by the conservative ion budget equals the alkalinity defined by the proton acceptor/donor budget. The proton acceptor/donor budget definition of the total alkalinity is the number of moles of  $\text{H}^+$  ions equivalent to the excess of proton acceptors over proton donors in one kg of sample. Bases formed from acids with  $\text{pK} \geq 4.5$  are proton acceptors, while acids with  $\text{pK} < 4.5$  are considered proton donors.

Electroneutrality is the property where the sum of the positive charges of the cations equals the sum of the negative charges of the anions. The mass budget of conservative cations and conservative anions can be computed as a charge. To maintain electroneutrality, the sum of the positive charges must equal the sum of the negative charges.

$$\begin{aligned}
 & [\text{Na}^+] + 2 * [\text{Mg}^{2+}] + 2 * [\text{Ca}^{2+}] + [\text{K}^+] + [\text{H}^+] \\
 & - [\text{Cl}^-] - 2 * [\text{SO}_4^{2-}] - [\text{NO}_3^-] - [\text{HCO}_3^-] - 2 * [\text{CO}_3^{2-}] - [\text{B}(\text{OH})_4^-] - [\text{OH}^-] = 0
 \end{aligned} \tag{12}$$

This equation can also be expressed as  $\sum z_j [\text{c}_j] = 0$  where  $[\text{c}_j]$  is concentration of a compound,  $z_j$  is the charge of the compound, and  $j$  is the compound.

### 3.2 Explorations of Carbonate Chemistry

An important principle arises here, namely that the charge balance of water itself is zero, whereas TA is positive. What accounts for the charge imbalance? It turns out that TA is also the charge

imbalance of conserved cations (positively charges) over conserved anions (negative charges). Conserved in this context means that they don't vary with temperature or pressure, nor are they proton donors or acceptors at the pH threshold of 4.5 used in the definition of TA above. Thus, an alternative expression for total alkalinity is by way of these conserved species:

$$TA = [\text{Na}^+] + 2 [\text{Mg}^{2+}] + 2 [\text{Ca}^{2+}] + [\text{K}^+] + \dots + \\ - [\text{Cl}^-] - 2 [\text{SO}_4^{2-}] - [\text{NO}_3^-] \quad (13)$$

The importance of this expression is that it relates a change in TA by way of a conserved cation, such as  $[\text{Mg}^{2+}]$  or  $[\text{Ca}^{2+}]$  originating from mineral dissolution, to a change in TA that includes carbonate terms, i.e.  $[\text{HCO}_3^-]$  and  $[\text{CO}_3^{2-}]$ . The sum of total charges remain constant even while the forms of DIC change. Below, we will develop an expression for this change,  $dDIC/dTA$ , which quantifies a "stoichiometry" between cations and carbonate species, based on the fundamental equations provided above.

The complexity of the stoichiometry is due to the presence of both monovalent  $[\text{HCO}_3^-]$  and divalent  $[\text{CO}_3^{2-}]$ , which causes the stoichiometric ratio to value between 1:2 (two  $[\text{HCO}_3^-]$  per  $[\text{Mg}^{2+}]$  or  $[\text{Ca}^{2+}]$ ) at lower pH down to 1:1 (one  $[\text{CO}_3^{2-}]$  per divalent cation) at higher pH. Moreover, the original dissolution event will likely take place in an environment (i.e. soil) that is more acidic, not saline, and at a different temperature, than the ultimate sink for cations and carbonates (i.e. the ocean), which is less acidic, very saline, and differs by temperature. Because the dissolution constants of carbonic acid vary with temperature and salinity, and the distribution of carbonates between  $[\text{HCO}_3^-]$  and  $[\text{CO}_3^{2-}]$  varies with acidity, the answer is not straightforward. Nonetheless, we will arrive at an analytical expression for  $dDIC/dTA$  that can be used in terrestrial, riverine, and marine environments. We will show that the stoichiometry at the source is what can be measured and verified as the site of sequestration; that the stoichiometry at the ocean sink is what will be stored on millennial timescales; and the difference between these two can be interpreted as leakage of  $[\text{CO}_2]$  back into the atmosphere.

First, simplify the expressions for DIC and TA to be functions of  $[\text{CO}_2]$  and  $[\text{H}^+]$  alone. For convenience (and consistency with Zeebe [1]) we'll use notation where  $s = [\text{CO}_2]$  and  $h = [\text{H}^+]$ .

From (3.1) express  $[\text{OH}^-]$  as:

$$[\text{OH}^-] = \frac{K_w}{h} \quad (14)$$

From (3.5) express  $[\text{HCO}_3^-]$  as:

$$[\text{HCO}_3^-] = s \cdot \frac{K_1}{h} \quad (15)$$

From (3.5) and (3.6) express  $[\text{CO}_3^{2-}]$  as:

$$[\text{CO}_3^{2-}] = s \cdot \frac{K_1 K_2}{h^2} \quad (16)$$

These allow us to express DIC and TA as:

$$DIC = s \cdot \left[ 1 + \frac{K_1}{h} + \frac{K_1 K_2}{h^2} \right] \quad (17)$$

$$TA = s \cdot \frac{K_1}{h} + s \cdot 2 \cdot \frac{K_1 K_2}{h^2} + \frac{K_w}{h} - h \quad (18)$$

With these definitions in place, we can develop an estimate of  $dDIC/dTA$ . First, compute the derivative  $dTA/dh$ :

$$\frac{dTA}{dh} = -s \cdot \left( \frac{K_1}{h^2} + 4 \cdot \frac{K_1 K_2}{h^3} \right) - \frac{K_w}{h^2} - 1 \quad (19)$$

Next compute the derivative  $dDIC/dh$ :

$$\frac{dDIC}{dh} = -s \cdot \left( \frac{K_1}{h^2} + 2 \cdot \frac{K_1 K_2}{h^3} \right) \quad (20)$$

Finally multiply  $dDIC/dh$  by the inverse of  $dTA/dh$  to calculate  $dDIC/dTA$ :

$$\frac{dDIC}{dTA} = \frac{dDIC}{dh} \cdot \frac{dh}{dTA} \quad (21)$$

What these calculations show is that (at constant temperature and salinity) at lower pH values, the C storage per cation introduced is higher than the C storage per cation at higher pH. At pH 5.5, the change in DIC per change in TA is nearly 1:1, though at higher pH such as 8.3 the balance is lower because some of the charge is balanced by  $\text{CO}_3^{2-}$ , not just  $\text{HCO}_3^-$  (Figure 4a). At lower pH, the change in pH is also more significant per unit TA than at higher pH (Figure 4b).

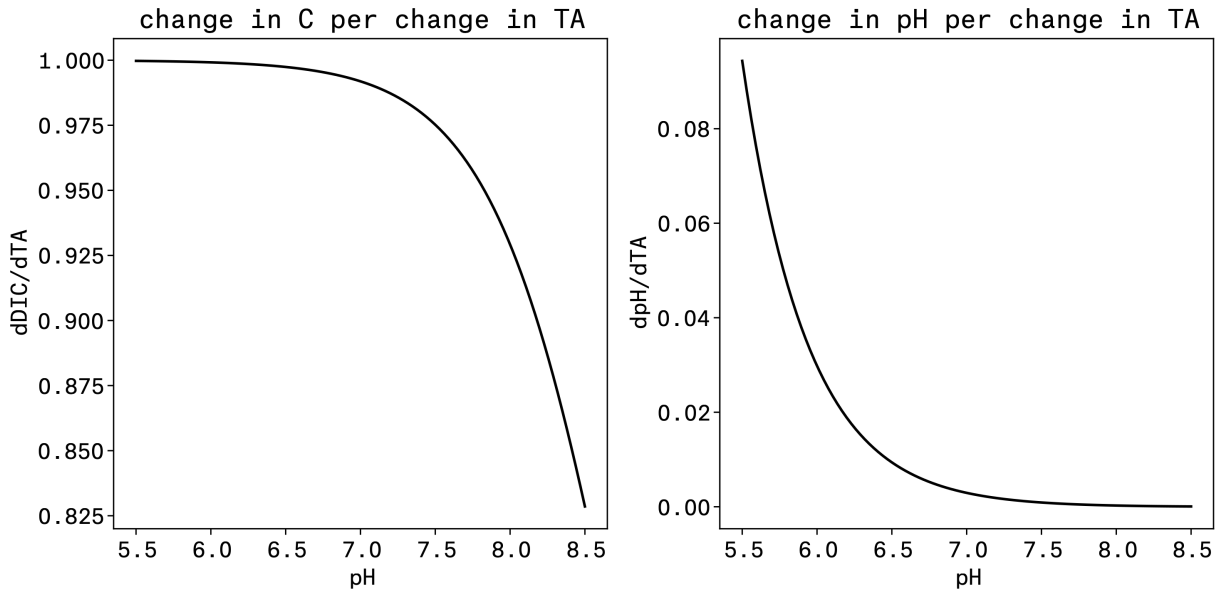


Figure 4: Change in derivatives of carbonate system with respect to changes induced by a change in alkalinity. Derivatives of DIC (left) and pH (right).



Additionally, these calculations can illustrate the impact of alkalinity on pH, which surfaces the counter-intuitive phenomenon that alkalinity is not simply the inverse of acidity.

$$\frac{dpH}{dTA} = -\log_{10}(e) \cdot \frac{1}{h} \cdot \frac{dh}{dTA} \quad (22)$$

Figure 4 shows that across any value of pH, additions in alkalinity always result in positive increases in pH and thus reductions in acidity.

### 3.3 Carbonate Chemistry in the Ocean

In the ocean, dissolved boric acid contributes to the alkalinity budget. Total boron, that is the sum of  $B(OH)_3$  and  $B(OH)_4^-$ , is proportional to salinity, which itself varies with the fresh-water budget, both influx of freshwater from rivers, and evaporation of pure water from the surface.

The constant for boric acid is defined as:

$$K_B = \frac{[H^+][B(OH)_4^-]}{[B(OH)_3]} \quad (23)$$

It is computed as:

$$\begin{aligned} \ln K_B = & \left( -8966.9 - 2890.53S^{1/2} - 77.942S + 1.728S^{3/2} - 0.0996S^2 \right) / T \\ & + 148.0248 + 137.1942S^{1/2} + 1.62142S \\ & - \left( 24.4344 + 25.085S^{1/2} + 0.2474S \right) \ln T + 0.053105S^{1/2}T \end{aligned} \quad (24)$$

In marine settings (where  $S$  is large and  $pH > 8$ ), the boric acid term ( $\frac{K_B B_T}{K_B + h}$ ) is added to the equation for TA, and the derivative of TA with respect to  $h$  becomes:

$$\frac{dTA}{dh} = -s \cdot \left( \frac{K_1}{h^2} + 4 \cdot \frac{K_1 K_2}{h^3} \right) - \frac{K_B B_T}{K_B + h^2} - \frac{K_w}{h^2} - 1 \quad (25)$$

In the ocean, the alkalinity is more or less defined by the consistent cation/anion budget, proportional to salinity. Alkalinity measures the charge concentration of anions and cations in the solutions, and is equal to the number of moles of acid (e.g. HCl) to add to neutralize the anions of the weak acid equals the carbonate alkalinity. The carbonate system is one contributor to total alkalinity, though there are other contributors including boric acid ( $B(OH)_4^-$ ), phosphoric acid, and water. Practical alkalinity (pA) is a simplified equation for alkalinity, which includes carbonate alkalinity, borate alkalinity, and water alkalinity, following the equation:

$$pA = [HCO_3^-] + 2 * [CO_3^{2-}] + [B(OH)_4^-] + [OH^-] - [H^+] \quad (26)$$

The equilibrium constants in the carbonate equations depend on pressure and temperature, which vary throughout the water column. Thus, as carbon sinks from the surface to deeper waters, these values change. However, when only the temperature and pressure change (and not salinity), DIC and TA are constant.

Ocean alkalinity is roughly -2.5. As pH increases, with no other changes to the system, carbonate alkalinity will decrease (Figure 5).

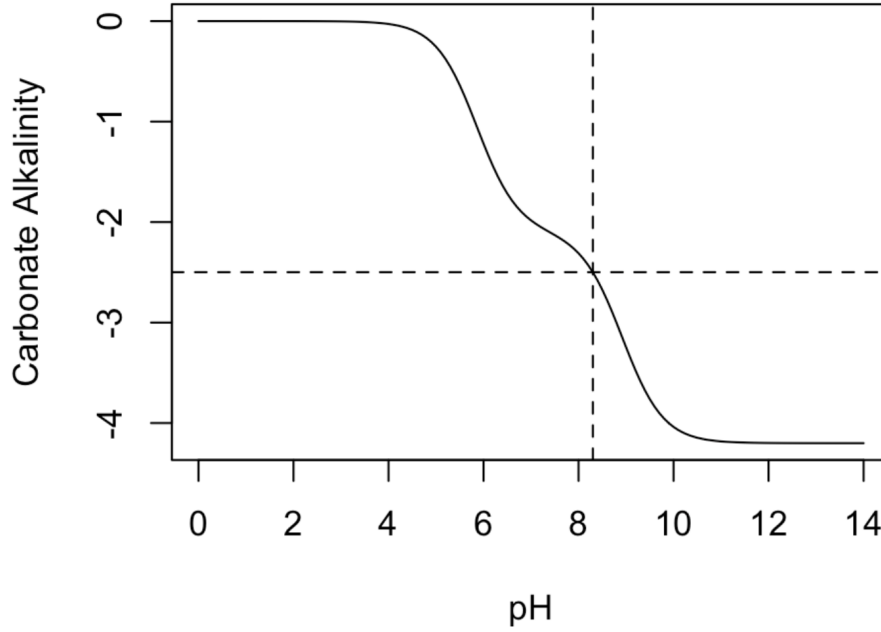
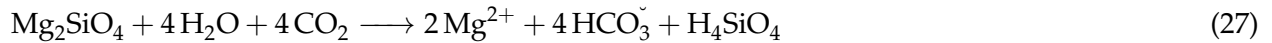


Figure 5: This figure shows carbonate alkalinity vs. pH. The dashed lines show where pH=8.3, and carbonate alkalinity = -2.5.

## 4 Quantifying Potential Geochemical CO<sub>2</sub> Removal

### 4.1 Mineral Potential

Once crushed silicate or carbonate rocks are spread on agricultural fields the weathering reaction takes place. This is a natural process that does not require any energy inputs and is thermodynamically irreversible. However, the weathering chemistry varies based on the mineral feedstock and each feedstock has a Mineral Potential (MP), which is the amount amount of CO<sub>2</sub> it can stoichiometrically sequester. For magnesium silicate materials, the archetypal weathering reaction takes the following form:



In this reaction, one mole of magnesium silicate reacts with four moles of CO<sub>2</sub>, so two negatively charged bicarbonate HCO<sub>3</sub><sup>-</sup> are created for every one divalent Mg<sup>2+</sup> weathered. Given the molecular weight of Mg<sub>2</sub>SiO<sub>4</sub> (140g/mol) and the molecular weight of CO<sub>2</sub> (44g/mol), weathering one metric tonne of Mg<sub>2</sub>SiO<sub>4</sub> removes 1.25 metric tons of CO<sub>2</sub> from the atmosphere. Beerling (2020) [4], citing Renforth (2012) [5], citing O'Connor (2004) [6], citing Goff and Lackner (1997) [7] provides mineral potential (MP) for CO<sub>2</sub> removal based on feedstock chemical composition in terms of MgO% and CaO%, shown in Equation 28.

$$\text{MP} \equiv \frac{t\text{CO}_2\text{e}}{t\text{Ore}} = \frac{\text{MW}_{\text{CO}_2}}{100\%} \cdot \left( \frac{\text{MgO}\%}{\text{MW}_{\text{MgO}}} + \frac{\text{CaO}\%}{\text{MW}_{\text{CaO}}} \right) * V \quad (28)$$

## 4.2 Leakage

In equation 28,  $V$  is the valence of the cation (2 for Mg and Ca) and  $MW$  is the molecular weight of the mineral. To estimate the net removal of  $\text{CO}_2$  from silicate weathering, we need to consider the fate of dissolved inorganic carbon (DIC) as it moves through the environment to the ocean, and account for any  $\text{CO}_2$  leakage. Following the derivation in 3 the amount of  $\text{CO}_2$  initially absorbed from the atmosphere when the mineral is dissolved in an acidic soil, ( $\text{CDR}_{\text{gross}}$ ) is determined by the mineral potential  $MP$  of the rock, multiplied by the fraction of dissolved inorganic carbon (DIC) uptake per unit of total alkalinity  $TA$  increase (Eq 21), at the temperature and pH conditions of the aqueous environment on land, as shown in equation 29. This equation represents the equilibrium conditions of perturbations to the carbonate system due to the mineral dissolution. The DIC then moves through terrestrial waterways to the ocean where the pH changes from the acidic conditions found on land (pH 5-6) to the colder and more alkaline conditions of the ocean (pH  $\sim 8$ ). Most saliently here, The DIC of the ocean has about 10%  $\text{CO}_3^{2-}$  compared to only 0.001% in soils (Figure 2). This means 10% of the  $\text{Mg}^{2+}$  or  $\text{Ca}^{2+}$  in solution is matched 1:1 with a  $\text{CO}_3^{2-}$  ion and a portion of  $\text{CO}_2$  is lost to the atmosphere due to this pH forcing. We interpret this as a form of leakage of sequestered carbon, and it is a standard feature of the literature on this topic [4,5,8]. The leakage is calculated as shown in Equations 30 and 31. With these expressions in place, we can compute  $\text{CDR}_{\text{net}}$  as  $\text{CDR}_{\text{gross}}$  minus the leakage, as shown in equation 32.

$$\text{CDR}_{\text{gross}} = MP \cdot \left. \frac{d\text{DIC}}{dTA} \right|_{\text{land}} \quad (29)$$

$$f_{\text{Leakage}} = \left. \frac{d\text{DIC}}{dTA} \right|_{\text{land}} - \left. \frac{d\text{DIC}}{dTA} \right|_{\text{sea}} \quad (30)$$

$$\text{Leakage} = -MP \cdot f_{\text{Leakage}} \quad (31)$$

$$\text{CDR}_{\text{net}} = \text{CDR}_{\text{gross}} - \text{Leakage} \quad (32)$$

## 4.3 Worked Example

Consider a standard reference material, a magnesium silicate with 47.45% MgO and 0.22% CaO. The Mineral Potential of this silicate is 1.047  $\text{tCO}_2/\text{tOre}$  per Eq 28. Let us apply to a soil with pH 5.5 at a standard temperature of 25°C, with an ultimate destination in the ocean at pH 8.08 and 16.1°C (global mean oceanic values), and a boundary condition for atmospheric  $\text{pCO}_2$  of 400ppm (Fig 6). Applying the equations in Section 3, we compute the constants and  $d\text{DIC}/dTA$  for land (Fig 7) and ocean (Fig 8). These calculations result in a leakage of 14.3% (Fig 9). In reality, these reactions will not be under conditions representing the mean ocean, nor will they be under conditions at an exact transition boundary between a river and the ocean. Instead, the parameters will depend on a dynamic turbulent mixing process that is intermediate between the two extremes. Research is underway within the scientific community using numerical simulation models combining biogeochemistry and ocean fluid dynamics to refine the understanding of this parameter.

<b>Land Parameters</b>	
Land pH	5.5
Land Temperature (°C)	25
Land Temperature (°K)	298.2
<b>Ocean Parameters</b>	
Ocean pH:	8.08
Ocean Salinity:	35
Ocean Temperature (°C)	16.1
Ocean Temperature (°K)	289.3
pCO <sub>2</sub>	400

Figure 6

<b>Land Calculations</b>	
Kh	0.03402
pKw	12.543
pK1	5.971
pK2	9.385
Kw	2.86E-13
K1	1.07E-06
K2	4.12E-10
<i>h</i>	3.16228E-06
<i>s</i>	1.36E+01
dTA/dh	-1.46E+06
dDIC/dh	-1.46E+06
dDIC/dTA	0.99974

Figure 7

<b>Ocean Calculations</b>	
Kh	0.04395
pKw	14.643
pK1	5.940
pK2	9.078
pKb	8.702
Kw	2.28E-15
K1	1.15E-06
K2	8.36E-10
Kb	1.98E-09
<i>h</i>	8.31764E-09
<i>s</i>	1.76E+01
TB	4.16E-04
dTA/dh	-4.09E+11
dDIC/dh	-3.51E+11
dDIC/dTA	0.85666

Figure 8

<b>Net Calculations</b>	
Gross CDR, tCO <sub>2</sub> e / tOre:	1.047
Leakage, %:	0.143
Net CDR, tCO <sub>2</sub> e / tOre:	0.897

Figure 9

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