

## The potential of enhanced weathering in the UK

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### Abstract

Enhanced weathering is the process by which carbon dioxide is sequestered from the atmosphere through the dissolution of silicate minerals on the land surface. The carbon capture potential of enhanced weathering is large, yet there are few data on the effectiveness or engineering feasibility of such a scheme. Here, an energy/carbon balance is presented together with the associated operational costs for the United Kingdom as a case study. The silicate resources are large and could theoretically capture 430 billion tonnes (Gt) of CO<sub>2</sub>. The majority of this resource is contained in basic rocks (with a carbon capture potential of ~0.3 tCO<sub>2</sub> t<sup>-1</sup> rock). There are a limited number of ultrabasic formations (0.8 tCO<sub>2</sub> t<sup>-1</sup> rock) with a total carbon capture potential of 25.4 GtCO<sub>2</sub>. It is shown that the energy costs of enhanced weathering may be 656 - 3501 kWh tCO<sub>2</sub><sup>-1</sup> (net CO<sub>2</sub> draw-down, which accounts for emissions during production) for basic rocks and 224 - 748 kWh tCO<sub>2</sub><sup>-1</sup> for ultrabasic rocks. Comminution and material transport are the most energy intensive processes accounting for 77-94% energy requirements collectively. The operational costs of enhanced weathering are £44-361 tCO<sub>2</sub><sup>-1</sup> (\$70 - 578 tCO<sub>2</sub><sup>-1</sup>) and £15-77 tCO<sub>2</sub><sup>-1</sup> (\$24 - 123 tCO<sub>2</sub><sup>-1</sup>) for basic and ultrabasic rocks respectively. Full exploitation of this resource is not possible given the environmental and amenity value of some of the rock formations. Furthermore, the weathering rate and environmental impact of silicate mineral application to the land surface is not fully understood, and further investigation in this area is required to reduce the uncertainty in the estimated costs presented here.

### 1. Introduction

The discontinuity between anthropogenic carbon emissions and atmospheric levels which are necessary to prevent dangerous climate change has led to the concept of geoengineering (Keith, 2000). Geoengineering is the 'deliberate large-scale intervention in the Earth's climate system, in order to moderate global warming' (The Royal Society, 2009). This umbrella term is used to cover a range of technologies with varying capacity and risk.

This paper presents the potential capacity of 'enhanced weathering' and the engineering challenges associated with such a scheme, at a national (UK) scale to inform policy development. This is particularly relevant in the context of recent discussions in the UK Parliament (House of Commons: Science and Technology Committee, 2010) which tentatively calls for additional research on geoengineering. By presenting the potential capacity for enhanced weathering in the

UK, this paper justifies additional research in this area.

Enhanced weathering involves the dissolution of silicate minerals so that calcium and magnesium are leached out, and react with dissolved carbon dioxide (HCO<sub>3</sub><sup>-</sup>, CO<sub>3</sub><sup>2-</sup>). Precipitation of carbonate minerals (e.g. calcite; CaCO<sub>3</sub>, magnesite; MgCO<sub>3</sub>, dolomite; CaMg(CO<sub>3</sub>)<sub>2</sub>) out of solution is possible. However, if the solution was transported to the ocean prior to precipitation then the dissolved components would contribute to ocean alkalinity without precipitation, and draw down additional CO<sub>2</sub> (Harvey, 2008, Kheshgi, 1995, Köhler et al., 2010).

Silicate minerals occur naturally at the surface in igneous rock formations with varying chemical compositions (Figure 1). Furthermore, silicates are produced as a by-product of some human activities (mine waste, cements, ashes, slags; Renforth et al., 2011). Over geological time the weathering of silicate minerals, their flux into the

oceans and the precipitation of carbonate minerals is a natural negative feedback mechanism in the Earth's climate (Berner and Kothavala, 2001). As silicate dissolution rate is enhanced with increased temperature, elevated atmospheric CO<sub>2</sub> results in additional CO<sub>2</sub> draw-down. Enhanced weathering schemes propose to accelerate this for appreciable CO<sub>2</sub> draw down on human relevant time scales.

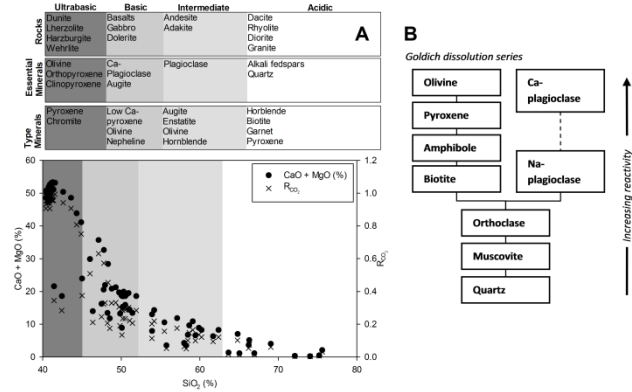
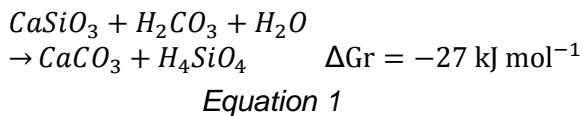


Figure 1: A) Total Alkaline Earth metals (CaO + MgO %) and maximum CO<sub>2</sub> capture potential (R<sub>CO<sub>2</sub></sub>) plotted against SiO<sub>2</sub> %. Some of the common rocks and their associated essential and type minerals are presented above. B) The Goldich dissolution series (Goldich, 1938).

Equation 1 presents a simplified reaction between the calcium silicate mineral wollastonite (CaSiO<sub>3</sub>) and dissolved CO<sub>2</sub> (carbonic acid; H<sub>2</sub>CO<sub>3</sub>). The reaction varies depending on what silicate mineral is used which is expanded upon in Lackner et al. (1995) and Renforth et al. (2011).



where  $\Delta G_r$  is the free energy of reaction (a negative value suggest that the reaction is thermodynamically likely to proceed).

The extensively mapped geology, well developed infrastructure and geoengineering regulation discussions in parliament (House of Commons: Science and Technology Committee, 2010) makes the UK a particularly pertinent case study.

### 1.1 Silicate minerals for enhanced weathering and mineral carbonation

Reactive and cation rich igneous rocks are the most suitable for carbon dioxide sequestration, which are chemically classified as acidic, intermediate, basic and ultrabasic depending on their silica content (Figure 1A). The relationship between the silica and alkaline Earth cation content is represented in Figure 1A.

The maximum carbon dioxide capture potential of a rock ('R<sub>CO<sub>2</sub></sub>' in tonnes CO<sub>2</sub> per tonne of material) is defined in Equation 2:

$$R_{\text{CO}_2} = \frac{M_{\text{CO}_2}}{100} \left( \frac{\% \text{CaO}}{M_{\text{CaO}}} + \frac{\% \text{MgO}}{M_{\text{MgO}}} \right) \cdot \omega$$

Equation 2

where M<sub>CaO</sub>, M<sub>MgO</sub> and M<sub>CO<sub>2</sub></sub> are the molecular masses of CaO, MgO and CO<sub>2</sub> respectively (adapted from O'Conner et al 2005)  $\omega$  represents the additional draw down from cation flux into the ocean (approximately 1.7; see below). As expected R<sub>CO<sub>2</sub></sub> correlates closely to the total alkaline cation content of the material (Figure 1A), the small discrepancy arising from the differences in Mg and Ca molecular masses. During enhanced weathering, it is possible for the cations to be transported to the ocean where they remain dissolved. This flux of cations into the ocean results in approximately 1.7 moles of CO<sub>2</sub> removed from the atmosphere for every mole of Mg or Ca (Harvey, 2008, Kheshgi, 1995, Rau, 2011). This equates to an approximate maximum R<sub>CO<sub>2</sub></sub> of 0.4 and 1.2 for basic and ultrabasic rocks respectively (Figure 1A).

An equally important parameter to consider is the rate at which the material dissolves (or 'weathers'). There has been substantial research investigating the weathering rate of silicate minerals (White and Brantley, 1995). Generally, minerals formed at higher temperatures undergo more rapid dissolution as outlined in the Goldich dissolution series (Figure 1B). In reality, their weathering rate is a function of a number of variables including the proportion of 'active' surface area, the temperature at which they weather, and solution chemistry.

A simplified expression for weathering rate (W<sub>r</sub> in mol m<sup>-2</sup> s<sup>-1</sup>) is presented in Palandri and Kharaka (2004) Equation 3

$$W_r = \left[ \begin{aligned} &k_{acid} e^{\frac{-E_{acid}}{R} \left( \frac{1}{T} - \frac{1}{298.15} \right)} a_{H^+}^{n^1} (1 - \Omega) \\ &+ k_{neutral} e^{\frac{-E_{neutral}}{R} \left( \frac{1}{T} - \frac{1}{298.15} \right)} (1 - \Omega) \\ &+ k_{base} e^{\frac{-E_{base}}{R} \left( \frac{1}{T} - \frac{1}{298.15} \right)} a_{H^+}^{n^2} (1 - \Omega) \end{aligned} \right]$$

Equation 3

where  $k$  is the rate constant for acid ( $\text{mol m}^{-2} \text{s}^{-1}$ ), neutral and base mechanisms of dissolution at  $25^\circ\text{C}$   $\text{pH} = 0$ ,  $E$  is the activation energy ( $\text{kJ mol}^{-1}$ ) for the different dissolution mechanisms,  $T$  is the absolute temperature ( $^\circ\text{K}$ ),  $a_{H^+}$  is the activity of hydrogen ions ( $\text{mol l}^{-1}$ ),  $\Omega$  is the saturation state, and  $n$  the reaction order.

Using empirically derived values from Palandri and Kharaka (2004), Figure 2 presents the variation in weathering rate as a function of reaction temperature (assuming a reaction far from equilibrium). Intuitively, the weathering rate of all silicate minerals increases with temperature. However, this varies substantially between minerals. Simplification of weathering rates in Equation 3 masks the substantial variation that is observed in the laboratory under apparently similar experimental conditions (White and Brantley, 2003).

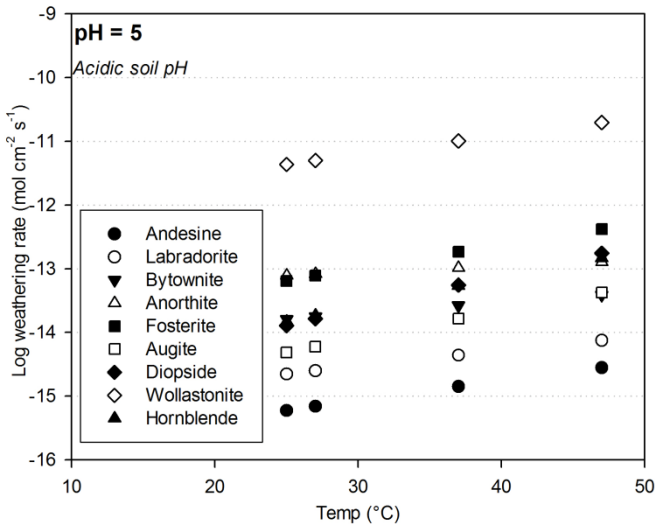


Figure 2: The weathering rate against temperature for a number of calcium and/or magnesium bearing silicate minerals. Andesine, labradorite, bytownite and anorthite are plagioclase feldspars  $((\text{Ca}, \text{Na})\text{Al}_x\text{Si}_y\text{O}_8)$  where the value of  $x$  and  $y$  is between 0-2 depending on the concentration of Ca or Na in the mineral. Fosterite ( $\text{Mg}_2\text{SiO}_4$ ) is the magnesium silicate end member of olivine. Augite (a mixture of diopside ( $\text{MgCaSi}_2\text{O}_6$ ) and hedenbergite  $\text{FeCaSi}_2\text{O}_6$ ), and wollastonite ( $\text{CaSiO}_3$ ) are members of the pyroxene group of

minerals. Calculated from Palandri and Kharaka (2004).

A number of methods have been suggested to promote the dissolution of silicates on human relevant time scales including elevated temperature 'reactor' methods (Lackner et al., 1997, Lackner et al., 1995, O'Connor et al., 2005), underground injection into silicate rock formations (Gislason et al., 2010, Kelemen and Matter, 2008, Kelemen et al., 2011, Matter et al., 2011, McGrail et al., 2006, Goldberg et al., 2008) and enhanced weathering on the land surface (Köhler et al., 2010, Manning, 2008, Schilling and Krijgsman, 2006). As of yet, there has been little work investigating enhanced weathering. This paper focuses on the engineering requirements of an enhanced weathering scheme, using the United Kingdom as a case study. The aim of this paper is to quantify the raw material that may potentially be used for enhanced weathering and to understand the cost of the technology.

### 1.3 Enhanced Weathering

Putting ground silicates onto the land surface has been proposed as a potential ex-situ alternative to high temperature/pressure reactors (Schilling and Krijgsman, 2006), referred herein as 'enhanced weathering'. Biogeochemical activity in soil naturally accelerates weathering which could be exploited globally (Manning, 2008; Manning and Renforth 2012).

In a feasibility analysis of enhanced weathering in coastal environments, Hangx and Spiers (2009) use a shrinking core model (Equation 4) of particle dissolution to estimate reaction times ( $t$ ), which for 100% dissolution of  $10\mu\text{m}$  olivine particles spread onto beaches is approximately 23 years,

$$X(t) = \frac{d_0^3 - (d_0 - 2W_r V_m t)^3}{d_0^3} \times 100 \quad \text{Equation 4}$$

where  $d_0$  is the initial diameter of the particle (m),  $W_r$  is the dissolution rate ( $\text{mol m}^{-2} \text{s}^{-1}$ ),  $X$  is the % of mineral dissolved, and  $V_m$  is the molar volume of the mineral ( $\text{m}^3 \text{mol}^{-1}$ ). Fine grinding of particles never produces a single particle size, but creates a size distribution. Recalculating a shrinking core model based on Hangx and Spiers (2009) methodology for a 'typical' particle size distribution suggests the reaction time to be around 30% less (Table 1).

**Table 1: Adaptation of the shrinking core model to account for a typical particle size distribution with a  $P_{80}$  of approximately 10 $\mu$ m.**

Particle Size (mm)	% Retained	% Passing	Time for 95% dissolution (d(t); in years)	Weighted Average (%Retained/100 *d(t); in years)
0.09770	0.8	100	227.89	1.82
0.06910	0.9	99.2	161.18	1.45
0.04880	0.8	98.3	113.83	0.91
0.03450	1.1	97.5	80.47	0.89
0.02440	4.2	96.4	56.91	2.39
0.01730	4.1	92.2	40.35	1.65
0.01220	10.4	88.1	28.46	2.96
0.00863	6.2	77.7	20.13	1.25
0.00400	12.8	71.5	9.33	1.19
0.00200	11.1	58.7	4.67	0.52
0.00100	10.2	47.6	2.33	0.24
0.00050	10.8	37.4	1.17	0.13
0.00010	13.9	26.6	0.23	0.03
0.00009	12.7	12.7	0.21	0.03
<b>Sum</b>				<b>15.46</b>
<i>(<math>W_r</math> 1.58 x 10<sup>-10</sup> mol m<sup>-2</sup> s<sup>-1</sup> and <math>M_m</math> 43.02 x10<sup>-6</sup> m<sup>3</sup> mol<sup>-1</sup>)</i>				

Modifying  $W_r$  to reflect steady state rates at a soil pH of 5 (10<sup>-9</sup> mol m<sup>-2</sup> s<sup>-1</sup>) results in a decrease in weathering time from 15.5 to 2.4 years.

The value of considering weathering reactions of single minerals in steady state is limited as it is substantially removed from realistic environmental conditions, in which silicate dissolution may vary by several orders of magnitude (White and Brantley, 2003) depending on solution flux, pH, temperature, saturation, and surface area for a range of minerals in a rock mass. Köhler et al. (2010, 2011) highlight the importance of silica

(H<sub>4</sub>SiO<sub>4</sub>) saturation on limiting the dissolution rate. They suggest that the global potential of olivine enhanced weathering is around 3.7 GtCO<sub>2</sub> per year based on a silica saturation of 2 mol m<sup>-3</sup> in solution. Applying this method to UK agricultural soils, a maximum carbon capture potential 11.8 MtCO<sub>2</sub> per year is posited (equivalent to 2% of UK CO<sub>2</sub> emissions). Others have contended this by suggesting biological uptake of Si in soils and drainage waters limits silica saturation (Schuiling et al., 2011). These preliminary assessments of enhanced weathering vary substantially in their conclusions, and additional work is required to understand the maximum weathering potential of the land surface. That withstanding, enough is known or can be assumed to make a preliminary feasibility assessment of the engineering requirements of enhanced weathering.

## 2. Theoretical maximum carbon capture potential of natural silicates in the UK

Natural silicate rock formations are located close to the surface in Scotland (Isle Mull, Isle of Skye, Western Isles, Central Lowlands, Grampian Mountains), England (Northumberland, County Durham, The Lake District, Cornwall), Wales (Pembrokeshire, Northern Wales), and Northern Ireland (Figure 3). This section presents a summary of these rock formations and their carbon capture potential. For a comprehensive review of igneous rocks in the UK see Sutherland (1982). While the focus of this paper is enhanced weathering, the quantification of resources has been extended to include those rocks that may be used for CO<sub>2</sub> sequestration by mineral carbonation or underground injection of CO<sub>2</sub>. The terms enhanced weathering (EW) or mineral carbonation (MC) 'ex-situ' refer to resources that can be exploited at the surface either through enhanced weathering or mineral carbonation. 'In-situ' refers to resources that require CO<sub>2</sub> injection to be exploited.

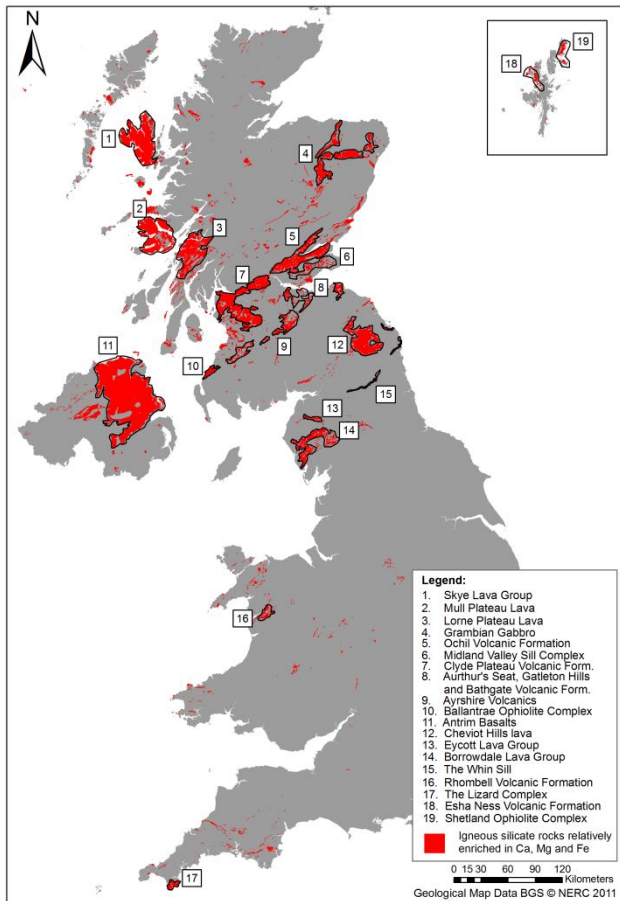


Figure 3: Basic and ultrabasic rock distribution in the UK

The depth to which a quarry or open cast mine is operational is based on an economic cost-benefit analysis between the value of the extracted material and the cost of extraction at depth. Some mines that extract particularly valuable ore ( $\sim \text{£}100 \text{ t}^{-1}$ ) have depths of up to 4000m. Assuming that an appropriate value of carbon may be  $\text{£}10\text{-}10^2 \text{ tCO}_2^{-1}$  ( $\sim \text{£}20 \text{ t}^{-1}$  of rock based on  $R_{\text{CO}_2} = 0.2$ ), it is unlikely that substantially deep mines would be viable. The values presented below have been calculated assuming that an extraction depth similar to those used in aggregate quarries ( $\sim 50\text{m}$ ) is viable (Mann, 1996). However, a more detailed evaluation is required for each potential outcrop against a range of technical, economic, and socio-environmental drivers.

Additional work is required to assess the most efficient injection depth and the proportion of the rock that could be used for carbon capture. However, based on McGrail et al. (2006) maximum injection depth of 1000 m has been assumed. McGrail et al. (2006) and Goldberg et al. (2008) calculated in-situ carbon capture potential

by assuming the porosity in a volume of rock (10-15%) will be completely filled with  $\text{CO}_2$  at injection pressures ( $\sim 100$  at with a density of  $1 \text{ g cm}^{-2}$ ) rather than the chemical limitation of  $R_{\text{CO}_2}$  for ex-situ dissolution. Gislason et al. (2010) go further and assume that only 10% of this volume will be filled with carbonate minerals. In this study, a conservative estimate of 1% of the rock volume (Gislason et al., 2010) can be used for carbon sequestration (where precipitated carbonate minerals have the density of  $2.7 \text{ t m}^{-3}$ ). The equations used for calculating carbon capture potential for the range of aforementioned techniques have been collated in the Appendix.

## 2.1 The Whin Sill and Midland Valley Sill Complex

The Whin Sill is an intrusive dolerite rock that outcrops at the surface in several places across County Durham and Northumberland. Approximately  $67 \text{ km}^2$  of Whin Sill is exposed at the surface (measured from geological bedrock maps, excluding drift deposits), assuming a typical depth of material (30m; Liss et al., 2004), and density ( $3 \text{ t m}^{-3}$ ; Sloane, 1991), it is estimated that  $6.2 \text{ Gt}$  are near the surface. The majority of the Whin Sill is located 10 - 1000 m below the surface running parallel to existing sedimentary strata and is thicker and deeper towards the North Sea (Thorpe and MacDonald, 1985). It is estimated that  $5000 \text{ km}^2$  (Thorpe and MacDonald, 1985) of North East England is underlain by the sill (which equates to  $600 \text{ Gt}$  using the same thickness and rock density as above). The Midland Valley Sill (which is geologically similar to the Whin Sill) outcrops in a number of places to the north and west of the Firth of Forth, and underlies much of the area to a depth of 1300 m. The thickness of the sill is 140m in the north and decreases to 0-30 m south of Edinburgh. The dolerite Whin Sill and Midland Valley Sills are primarily composed of Ca plagioclase (70-80% An in some places), and pyroxene (orthopyroxene, pigeonite, and Ca-rich pyroxene). Amphiboles, biotites and chlorites are also present in minor amounts (Randall, 1989). This material has a total carbon capture potential of  $4.9 \text{ GtCO}_2$  (EW ex-situ),  $2.9 \text{ GtCO}_2$  (MC ex-situ) and  $4.3 \text{ GtCO}_2$  (in-situ), using the chemical composition presented in Table A1.

The extraction industry currently operates at a number of places along the Whin Sill. The British Geological Survey Resource Map (Mankelov et al., 2008a), and the calculations made in this

paper suggest that approximately 65% of the outcrop is under land-use with an environmental designation. The Midland Valley Sill is also well exploited for high quality road stone due to its homogeneity (Bide et al., 2008), but only 13% lies under land with an environmental designation.

## 2.2 *British Tertiary Volcanic Province*

The Isles of Mull, Rum and Skye are the eroded remnant of the British Tertiary Volcanic Province (which also includes basalts in Northern Ireland, described below, and other smaller formations) extruded during the early Paleogene 60 million years BP (Emeleus, 1982). The central complexes located in south east Mull, west Ardnamurchan, Rum and south east Skye are predominantly granitic, felsic and gabbroic. Large lava fields, with varying olivine, plagioclase feldspars, augite and pyroxene content extend North West of the central complexes covering the majority of present day Mull and Skye.

The Skye lava field covers approximately 1060 km<sup>2</sup> and is 1.2 km thick (England, 1994), equating to 1270 km<sup>3</sup> of rock. The Mull lava field covers approximately 900 km<sup>2</sup> (Kerr, 1995) with variable lava thickness (150 - 250 m is typical), extending to 900 m on Central Mull. Assuming a typical lava thickness of 200 m it is estimated that 180 km<sup>3</sup> of lava is present. Using a rock density of 3 t m<sup>-3</sup>, an extraction depth of 50m, and a chemical composition presented in Table A1, it is calculated that Mull and Skye lava groups have a total EW ex-situ carbon capture potential of 44.7 and 37.5 GtCO<sub>2</sub> respectively (MC ex-situ of 26.3 and 22.1 GtCO<sub>2</sub>). Assuming an injection depth between 50-1000 m, the in-situ carbon capture potential is approximately 2.1 and 11.9 GtCO<sub>2</sub>.

Similar to other rocks of the British Tertiary Volcanic Province, the Antrim lava group is primarily composed of basaltic material (olivine, plagioclase and clinopyroxene; Barrat and Nesbitt, 1996). Current exposed igneous rock covers approximately 3500 km<sup>2</sup> to a depth of 300 m (Barrat and Nesbitt, 1996). Assuming a density of 3 t m<sup>-3</sup>, and a chemical composition presented in Table A1, it is estimated that 3150 Gt of material are present with an EW ex-situ carbon capture potential of approximately 159.5 GtCO<sub>2</sub> (93.8 GtCO<sub>2</sub> MC ex-situ).

## 2.3 *Red Sandstone Volcanics*

There are a number of volcanic formations in Scotland and Northern England that are composed of 'Old Red Sandstone Lavas' including the Ochil Volcanic Formation, Cheviot Hills, Lorne Plateau, Ayshire Volcanics, and Esha Ness volcanic Formation, which were extruded during the Devonian (359-416 MaBP). These lavas are porphyritic and contain combinations of olivine, plagioclase, clinopyroxene, orthopyroxene and K-feldspar, in which olivine and plagioclase are Fo<sub>65-75</sub> and An<sub>53-65</sub> respectively (Gandy, 1975). Some of the volcanic formations associated with the Old Red Sandstone are reported to be over 1000 m thick. For instance, the Lorne Plateau, and Ochil Volcanic Formation are estimated to be approximately 2400 and 3300 m respectively (Gandy, 1975). The cumulative areal coverage of these formations is approximately 2470 km<sup>2</sup>. The cumulative carbon capture potential of the Red Sandstone Lavas is approximately 67.6 GtCO<sub>2</sub> (EW ex-situ), 51.5 GtCO<sub>2</sub> (MC ex-situ), and 24.1 GtCO<sub>2</sub> (in-situ).

Exploitation of these resources for aggregate is wide spread, with a number of quarries existing on each formation. Exploitation of the Ochil Volcanic and Ayshire Volcanic formations is economically limited by their distance from local markets (Smith et al., 2008c). Between 5 and 44% of these formations are located on land with an environmental designation.

## 2.4 *Carboniferous volcanics of the Midland Valley*

Carboniferous (and Permian) volcanic rocks of the Midland Valley include those present in the Aurther's Seat, Garleton Hills, Bathgate Hills Group, and Clyde Plateau Volcanic Formations. Thickness of the formations is variable but is reported to be between 300-420 m for Aurther's Seat, Garleton Hills, and Bathgate Hills, and Clyde Plateau, although, some report the Clyde Plateau to extend up to 800 m in depth (Browne et al., 1999). The mineralogical composition is particularly dominated by Ankaramite basalt (-olivine+clinopyroxene + plagioclase type minerals; Macdonald et al., 1977). Hawaiite-mugearite (-olivine+apatite) is also reported.

Collectively, the areal extent of carboniferous volcanic rocks of the Midland Valley is approximately 1310 km<sup>2</sup> (equating to a carbon capture potential of 54.3 GtCO<sub>2</sub> EW ex-situ, 31.9 GtCO<sub>2</sub> MC ex-situ and 6.4 GtCO<sub>2</sub> in-situ).

The Clyde Plateau and the Bathgate Hills are not worked for road stone or aggregate due to the 'higher quality' material that is worked from adjacent dolerite sills (Smith et al., 2008b). However, the Bathgate Hills have been mined for silver, nickel, cobalt and lead (Smith et al., 2008a). Authur's Seat Formation is not extracted due to its high scenic/amenity value.

## 2.5 Gabbros of North East Scotland

A number of gabbroic complexes outcrop in North Eastern Scotland including the Morven-Cabrach, Knock, Inch, Haddo House, Huntly, Coyles of Muick, Boganclogh, Belhelvie, and Maude which collectively cover approximately 595 km<sup>2</sup>. The 'Newer Gabbroic' formations are remnants of basic plutons that were intruded approximately 480 – 500 million years ago (Wasdsworth, 1982). The mineralogy of the formations is complex, and includes combinations of olivine, clino- and orthopyroxene, and plagioclase.

The thickness of these formations is likely to extend well beyond 1000 m, and the carbon capture potential is 19.9 GtCO<sub>2</sub> (EW ex-situ), 11.7 GtCO<sub>2</sub> (MC ex-situ) and 7.1 GtCO<sub>2</sub> (in-situ).

## 2.6 Ophiolite Complexes – The lizard complex, Bellatrea and Shetland

Ophiolites are assemblages of mafic and ultramafic rocks, particularly consisting of a basal serpentinitised ultramafic complex, a gabbroic complex, and a mafic dyke/volcanic complex (Coleman, 1977). The Lizard Complex Cornwall, S.W. England is the remnant of the Devonian Lizard Ophiolite Complex which is dominated by serpentinitised peridotites, gabbros, hornblende schists, banded gneisses, and dolerite dykes (Kirby, 1984). Approximately 77 km<sup>2</sup> of the Lizard Complex is exposed at the surface, and is 500 m thick (Rollin, 1986). This material has a carbon capture potential of 8.1 GtCO<sub>2</sub> (EW ex-situ), 4.7 GtCO<sub>2</sub> (MC ex-situ), and 0.5 GtCO<sub>2</sub> (in-situ). Although this may be an over estimation given the large quantity of gabbroic rock at the surface covering a substantial proportion of the formation.

The Ballantrae complex is located at the Ayshire Coast, Scotland, adjacent and north of Ballantrae. The Shetland Ophiolite complex outcrops on the Shetland islands of Unst and Fetlar. All of these formed during Caledonian orogeny 500 MyBP (Flinn and Oglethorpe, 2005). Similar to the Lizard Complex, they are composed of a range of

ultrabasic (dunite, harzburgite, wehrlite) and basic (gabbro, dolerite) rocks (Church and Gayer, 1973). Approximately 70 km<sup>2</sup> of the formation is exposed at Ballantrae, and 71 km<sup>2</sup> on Shetland, and have a collective EW ex-situ carbonation potential of 17.3 GtCO<sub>2</sub> (assuming similar rock chemistry to that of Shetland Ophiolite for both formations) and 1.7 GtCO<sub>2</sub> (in-situ).

There is limited extraction of the Lizard, Ballantrae and Shetland Ophiolite complexes due to their remoteness from markets, although Mankelow et al. (2008b) shows two active quarries on the Lizard Complex.

## 2.7 Borrowdale and Eycott Volcanic Formations

The Borrowdale and Eycott Volcanic formations in the Lake District, England, are remnants of a volcanic arc that was extruded during the Ordovician period (444-488 MaBP; Moseley, 1982). The Borrowdale Volcanic Group is composed of a suite of lavas, tuffs and ignimbrites (Fitton, 1972). The extrusive volcanic rocks are dominated by plagioclase feldspar (particularly Ca-rich minerals) and orthorhombic pyroxenes (hypersthene). Approximately 750 km<sup>2</sup> of rock outcrops in the Lake District, and has a thickness of 8km (Millward et al., 2000). Approximately 15 km north of the Borrowdale Volcanic Group is the outcrop of the Eycott Volcanic Group, which covers approximately 45km<sup>2</sup> and it around 3.2 km thick (Millward et al., 2000). Most of the formation comprises of tabular sheets of basalt, basaltic andesite, andesite and dacite. Plagioclase minerals dominate the rock, with 5-10% orthopyroxene and 3% orthopyroxene (Millward et al., 2000). Collectively, the Borrowdale and Eycott Volcanic formations have a theoretical maximum carbon capture potential of 14.0 GtCO<sub>2</sub> (EW ex-situ), 8.2 GtCO<sub>2</sub> (MC ex-situ) and 9.4 GtCO<sub>2</sub> in-situ.

All of the Borrowdale and 80-90% of the Eycott Volcanic formations out crop on land with an environmental designation (predominantly the Lake District National Park), which prevents current and probably future exploitation of this resource.

## 2.8 Summary

The carbon sequestration potential of UK basic rocks is large (Table 2). Extracting material at the surface for enhanced weathering could capture 430.1 GtCO<sub>2</sub>. It is calculated that these rocks could capture 253 GtCO<sub>2</sub> and 81 GtCO<sub>2</sub> for mineral carbonation and underground injection respectively, but no further discussion of these technologies is made in this paper, and the reader is referred to O'Connor et al., (2005), Kelemen and Matter (2008) and McGrail et al., (2006) for more information. While no ranking of resource has been made here, it is clear that some resources may be favourable for exploitation. For instance, the greater R<sub>CO2</sub> values in the ophiolite complexes would require less material to be extracted and processed for the same carbon draw down using basic rocks. However, the quantity of this resource is smaller (25.4 GtCO<sub>2</sub>) and there would be environmental and amenity restrictions to exploitation. Some basic rock formations that contain lower R<sub>CO2</sub> values are more plentiful and exploitation may be less restricted (basic rocks in Northern Ireland, Ochil Volcanic Formation, Clyde Plateau) but may be prohibitively expensive to process. However, the quantity of extractable material does not limit this technology. The cost of activities required to fulfil this potential, and the environmental implications for extraction and application will be limiting, the former is covered in the following sections of the paper. Additional work is required to assess the environmental implications of extraction and application.

### 3. Artificial silicates

The term artificial silicate is used here to refer to silicate bearing materials produced from human processes. Generally, these materials are a by-product of an energy/carbon intensive process. Therefore, the subsequent carbonation is effectively compensating some of the emissions realised during production.

This section is expanded from Renforth et al. (2011; and references therein), associated production statistics are available from ACS Publications

(<http://pubs.acs.org/doi/suppl/10.1021/es103241w>). The UK produces approximately 86 Mt of silicate material annually as a by-product of cement manufacturing (cement kiln dust, CKD; 2.0 Mt a<sup>-1</sup>), construction and demolition (C and D waste; 77 Mt a<sup>-1</sup>), iron and steel processing (slag; 4.5 Mt a<sup>-1</sup>), and coal combustion (fuel ash; 2.6 Mt a<sup>-1</sup>).

#### 3.1 Cement waste

Cement is manufactured by heating calcium carbonate (CaCO<sub>3</sub>; quarried as limestone) in a kiln at 1400°C with a source of silica (SiO<sub>2</sub>; from clay or fly ash) to produce a mixture of calcium silicate minerals. During this, CKD is produced which is too fine to be sold. There are 10 cement production sites across the UK (Figure 4B). A discussion of cement carbonation is found in (Huntzinger et al., 2009). Approximately 655 Mt of cement has been produced since the early 1960s (Renforth et al., 2011) across 169 sites (historic kiln locations supplied by Dylan Moore; <http://cementkilns.co.uk/>), creating an estimated 131 Mt of cement kiln dust.

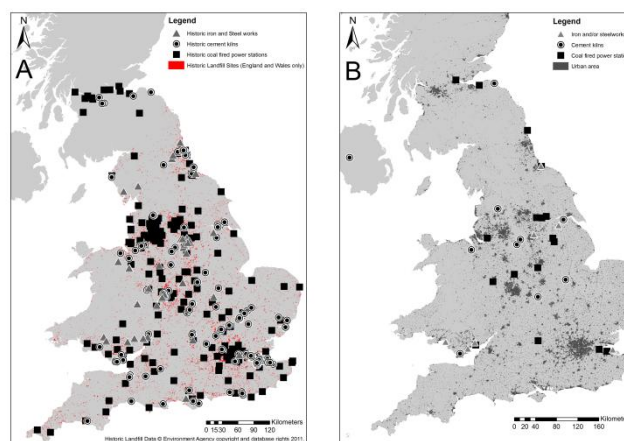


Figure 4: A) Historic and B) current production sites of artificial silicate material

#### 3.2 Construction and demolition waste

Cement is hydrated during construction producing hydrated calcium silicate gels. Cement together with that of aggregate (limestone or igneous rock) and sand are the components of concrete. C and D waste is produced as a product of the construction and demolition industries. The constituents depend on material sorting practices, but usually contains a substantial quantity of concrete (25%; Lawson et al., 2001), which is typically recycled and sold as secondary aggregate. Approximately 0.559 t of C and D waste is produced each year per capita. Figure 4B presents the major (>400,000 popn) urban centres in the UK. Approximately 4.1 Gt of C and D waste may have been produced since the early 1950s (Renforth et al., 2011), the majority of which since

**Table 2: The theoretical maximum carbon capture potential of a number of igneous rock formations in the UK.**

Name	Areal extent (km <sup>2</sup> )	Depth or thickness of exploitable material (m)		Total volume of exploitable material (km <sup>3</sup> )		Total Mass of material (Gt)	Total volume for carbon capture (km <sup>3</sup> )		Total carbon capture potential (GtCO <sub>2</sub> )		Environmental designation (%)	R <sub>CO2</sub>	Potential		
		Ex-situ	In-situ	Ex-situ	In-situ	Ex-situ	In-situ		Ex-situ	In-situ			Chemistry	Quantity	Environmental
Lizard Ophiolite	77	50	500	3.85	38.50	11.55	0.39		8.06	0.46	100	0.70	++	○	-
Ballantrae Ophiolite	70	50	1000	3.50	70.00	10.50	0.70		8.58	0.83	26	0.82	++	○	+
Shetland Ophiolite	71	50	1000	3.55	71.00	10.65	0.71		8.71	0.84	46	0.82	++	○	-
Whin Sill	69	30	-	2.07	200.00	6.21	2.00		1.47	2.38	65	0.24	○	-	-
Midland Valley Sill	97	50	100	4.85	160.00	14.55	1.60		3.45	1.90	13	0.24	○	-	++
Is of Mull Lava	900	50	200	45.00	180.00	135.00	1.80		44.74	2.14	28	0.33	+	++	+
Is of Skye Lava	1060	50	1000	53.00	1000.00	159.00	10.00		37.53	11.88	22	0.24	○	++	+
Antrim, N Ireland	3500	50	300	175.00	1050.00	525.00	10.50		159.54	12.47		0.30	+	++	
Arthur's Seat Volcanic Formation	49	50	300	2.45	14.70	7.35	0.15		2.03	0.17	60	0.28	+	-	-
Garleton Hills Volcanic Formation	78	50	380	3.90	29.64	11.70	0.30		3.23	0.35	15	0.28	+	-	++
Clyde Plateau Volcanic Formation	1123	50	420	56.15	471.66	168.45	4.72		46.43	5.60	11	0.28	+	++	++
Bathgate Hills Volcanic Formation	62	50	320	3.10	19.84	9.30	0.20		2.56	0.24	21	0.28	+	-	+
Ochil Volcanic Formation	699	50	1000	34.95	699.00	104.85	6.99		21.41	8.30	5	0.20	○	++	++
Cheviot Lava	700	50	500	35.00	350.00	105.00	3.50		11.29	4.16	44	0.11	-	+	-
Shetland Esha Ness	60	50	60	3.00	60.00	9.00	0.60		1.96	0.71	12	0.22	○	-	++
Lorne Plateau	787	50	800	39.35	629.60	118.05	6.30		24.11	7.48	19	0.20	○	++	++
Ayshire Volcanics	287	50	1000	14.35	287.00	43.05	2.87		8.79	3.41	9	0.20	○	○	++
Grampian Hills basic plutons	595	50	1000	29.75	595.00	89.25	5.95		19.94	7.07	31	0.22	○	+	○
Rhombell Volcanic Formation	73	50	960	3.65	70.08	10.95	0.70		2.33	0.83	100	0.21	○	-	-
Borrowdale Volcanic Formation	750	50	1000	37.50	750.00	112.50	7.50		13.09	8.91	100	0.12	-	+	-
Eycott Volcanic Formation	45	50	1000	2.25	45.00	6.75	0.45		0.86	0.53	84	0.13	-	-	-
<b>Total</b>	<b>11152</b>			<b>556.22</b>	<b>6791.02</b>	<b>1668.66</b>	<b>67.91</b>		<b>430.13</b>	<b>80.68</b>			++ (high potential) - (low potential)		

the Environment Act, has been disposed of in landfill. Chemical analysis of landfill leachate suggests that this material has, at least, partially carbonated and sequestered CO<sub>2</sub> as carbonate minerals (Fleming et al., 1999, Manning, 2001). Similarly, analysis of soils in brownfield sites that have been mixed with demolition waste also show substantial carbonate formation (Renforth et al., 2009).

### 3.3 *Iron and steel slag*

Similar to cement manufacturing, calcium or magnesium carbonate is heated with iron ore and calcium and magnesium rich silicate glass is produced (slag). Slag from iron production is readily reused as a secondary aggregate in construction. However, due to presence of hydroxide minerals (Ca(OH)<sub>2</sub>), steel slag is stockpiled and 'weathered'. There are 7 sites in the UK that produce iron and/or steel (Figure 4B). A discussion of iron and steel slag carbonation is found in (Huijgen et al., 2005). Approximately 566 Mt of slag have been produced since the end of the 19<sup>th</sup> Century, some of which is stockpiled adjacent to former and current steelworks.

### 3.4 *Coal ash*

Ash is produced as a by product of coal combustion. The UK consumes approximately 60 Mt of (primarily bituminous) coal a<sup>-1</sup>, producing 2.6 Mt of ash (Renforth et al., 2011) across 18 sites. The material is rich in silica and a number of metals and is often reused as a pozzolan in cement and concrete production. Approximately 630 Mt of fuel ash has been produced since the early 20<sup>th</sup> century, most of which has probably been reused, but a substantial quantity may still be available. The UK Quality Ash Association suggests that this quantity is around 50Mt (<http://www.ukqaa.org.uk/> accessed 27 November 2011).

### 3.5 *Total carbon capture potential of wastes*

The total carbon capture potential of artificial silicates in the UK is approximately 5.6 MtCO<sub>2</sub> a<sup>-1</sup>. Figure 4A and 4B shows the historic and current production sites respectively. Urban areas are included in Figure 4B as 'production sites' for construction and demolition waste. Historic landfills are presented in Figure 4A as they are a potential repository of un/partially-carbonated waste material.

## 4. **Extraction, Processing, Transport and Application**

Developing the carbon capture potential outlined in the previous two sections into a fully operational industry is a substantial engineering challenge. This section presents the key processes of enhanced weathering, and their associated energy, carbon and financial costs. Financial costs are expressed in GB£, but have also been converted to US\$ assuming £1 = \$1.6. The carbon intensity of electricity was assumed to be the recent average UK value ~400 gCO<sub>2</sub> kWh<sup>-1</sup> (The Energy and Climate Change Committee, 2010). The carbon intensity of road fuel/diesel was calculated using the cumulative emissions from road transport in the UK (DfT, 2011b) against associated fuel consumption (DfT, 2011a), and is approximately 237 gCO<sub>2</sub> kWh<sup>-1</sup>. Both values are conservative for a future technology considering that the prospective conditions that promote the creation of an industry to remove CO<sub>2</sub> from the atmosphere will also likely catalyse carbon intensity reductions in the energy industry. However, a sensitivity analysis has been included in Section 4.7 to assess the variability of cost with reasonable changes in these assumptions.

### 4.1 *Current extraction of igneous rock in the UK*

Extraction of igneous rock in the UK employs 3667 people and processes 47 Mt of material a<sup>-1</sup>, the total value of which is approximately £420 million (\$672 million; approximately \$14.3 t<sup>-1</sup>). A small quantity of granite (~1 Mt) is imported each year (BGS, 2009). The primary use of igneous rock is sub base or roadstone (46.7 %), other unspecified construction purposes (31.4 %), aggregate for concrete (14.0 %), and railway ballast (~6 %). Due to incomplete reporting between 7-27 % of igneous rock production is unaccounted, although this has decreased substantially in recent years (Mankelov et al., 2011; data for England and Wales). Given that the majority of the rocks in the UK have an R<sub>CO2</sub> of around 0.2-0.4, a substantially expanded extraction industry would be required to make a significant contribution to CO<sub>2</sub> drawdown. For instance, to capture the equivalent of 10% of UK CO<sub>2</sub> emissions (~50 MtCO<sub>2</sub>), at least 125 Mt of material will need to be extracted and processed every year. While this comparison with mitigation of emissions is problematic (see discussion in Keith, 2009), it is used here to represent a 'significant' quantity of CO<sub>2</sub> sequestration.

## 4.2 Comminution

The size reduction of rock may be the primary energy consuming operation in enhanced weathering. Comminution occurs in a number of discreet steps (Figure 5), namely blasting/drilling, primary, secondary and tertiary crushing, and grinding. Blasting, drilling and short range haulage within the extraction site are relatively low energy operations and using approximately  $5 \text{ kWh t}^{-1}$  (Hangx and Spiers, 2009), which is an  $R_{\text{CO}_2}$  reduction of 0.0012.

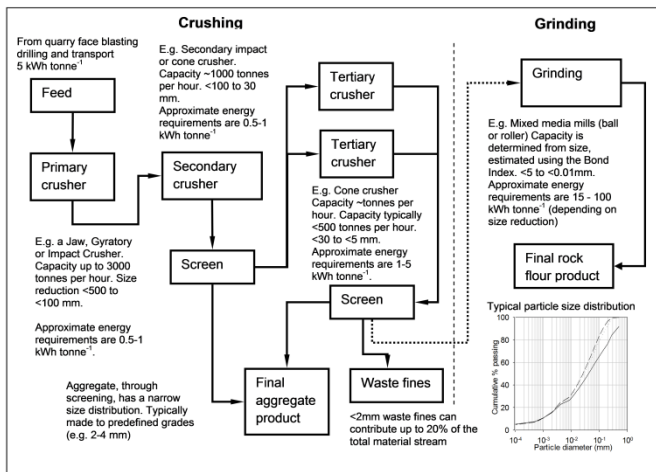


Figure 5: Typical flow path of material during comminution. Size reduction data from Metso (2010) and typical energy requirements taken from Metso (2010) and Wang and Fossberg (2003).

Hangx and Spiers (2009) estimate energy requirements up to  $180 \text{ kWh t}^{-1}$  to produce  $10 \mu\text{m}$  particles. This estimation is based on the Bond Work Index (Equation 5), and is approximately supported by direct measurements (O'Connor et al., 2005).

$$W = 10 W_i \left( \frac{1}{\sqrt{P_{80}}} - \frac{1}{\sqrt{F_{80}}} \right)$$

Equation 5

where  $W_i$  is the work index (approximately  $20 \text{ kWh t}^{-1}$  for a number of silicate minerals).  $P_{80}$  and  $F_{80}$  represent the size of particle in which 80% of the product or feed respectively passes. Part of the  $P_{80}$  is often referred to as 'fines' (material that is too small for use as aggregate usually  $<4 \text{ mm}$ ), which is a substantial problem in quarries. The size, and subsequent 'quality', of the aggregate is carefully controlled by crusher operational parameters and screens that separate out the required size fraction. Effort is made to reduce the production of the waste fines, which are stockpiled

and sometimes used in redevelopment of the site (Woods et al., 2004).

An alternative approach, one that is probably more practical given the difficulties of particle size distribution measurements on ultrafine material, is to relate crushing energy to surface area creation (Figure 6). The advantage of this approach is that surface area can be related directly to dissolution rates. There are a limited number of studies that investigate energy requirements for surface area generation in calcium or magnesium silicates (Baláz et al., 2008, Haug et al., 2010), which primarily focus on high energy grinding. There is a need to expand the dataset for low energy grinding (shaded region in Figure 6) for a range of silicate minerals.

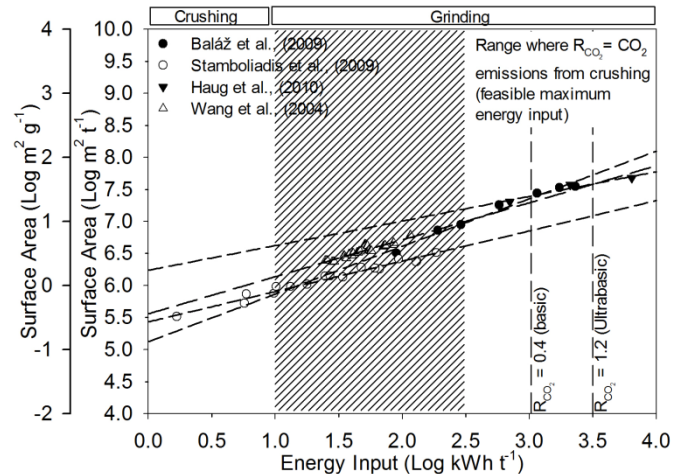


Figure 6: Relationship between energy input into comminution and the surface area generated.  $R_{\text{CO}_2}$  were estimated assuming  $\text{CO}_2$  emissions from energy generation were  $400 \text{ gCO}_2 \text{ kWh}^{-1}$ .

Energy requirements for grinding are variable (manifested in substantial gradient differences in the Log-Log relationship in Figure 6), which are influenced by mineralogy and crushing practices. Crushing and grinding equipment is largely inefficient at generating new surface area (typically less than 10%) and remains a continuing area of research for the comminution community (Wang and Forssberg, 2003).

To remove  $50 \text{ MtCO}_2$  per year using a silicate rock with a weathering rate between  $10^{-14} \text{ mol(cation) cm}^{-2} \text{ sec}^{-1}$ , would require  $2.12 \times 10^{14} \text{ m}^2$  of active surface area. This would require approximately  $200 \text{ Mt}$  of material crushed to a surface area of  $1 \text{ m}^2 \text{ g}^{-1}$ . While this oversimplifies the weathering

processes to a single rate, it is useful for identifying the potential requirements for material surface area, which may be of the order of  $1\text{-}10\text{ m}^2\text{ g}^{-1}$ . If weathering rate is limiting, then additional material may need to be placed which would require additional energy. It is unknown what rate silicates mixed into the soil, or placed on the land surface, will weather. Figure 2 presents rates derived from laboratory studies which show weathering rates varying between minerals ( $10^{-15}$  to  $10^{-10}\text{ mol cm}^{-2}\text{ sec}^{-1}$ ) and temperature (an order of magnitude between  $20$  and  $50^\circ\text{C}$ ). The comminution energy requirements for enhanced weathering may lie within the shaded box in Figure 6 (between  $10$  and  $316\text{ kWh t}^{-1}$ ). This represents a reduction in  $R_{\text{CO}_2}$  by  $0.0040$  to  $0.1264$  if the energy was derived from conventional fossil fuels ( $400\text{ gCO}_2\text{ kWh}^{-1}$ ).

Table 3 collates ‘typical’ operational and capital cost information for comminution practices. To capture equivalent to 10% of UK  $\text{CO}_2$  emissions, approximately  $125\text{ Mt}$  of material would need to be processed every year. This represents a substantial expansion of the current extraction industry. Extrapolating the information in Table 3, it is estimated that the total capital expenditure could be approximately  $\text{£}1.7 - 6.7\text{ B}$  ( $\text{\$}2.7\text{-}10.7\text{ B}$ ). Operational costs vary substantially and could be between  $\text{£}0.4\text{ B}$  to  $\text{£}5.1\text{ B}$  ( $\text{\$}0.6$  to  $\text{\$}8.2\text{ B}$ ) per year, depending primarily on the energy requirements of grinding. Assuming a discount rate of 4% over a 40 year time period, an even distribution of capital over that time period would add an additional  $\text{£}1.6\text{-}6.5\text{ t}^{-1}$  material. Total cost for comminution is therefore  $\text{£}3.4 - 40.68\text{ t}^{-1}$  material.

**Table 3: Capital and operational costs for mineral comminution (Source: Huijgen et al., 2007, Brown et al., 2010)**

Operation	For a 0.5 to 5 million tonne per annum operation (CAPEX)	Operation	£ tonne <sup>-1</sup> material (OPEX)
Crushing	£12.97M – 35.27M	Fixed plant, buildings and equipment (crushing)	0.16-0.63

Screening	£5.66M – £8.09M	Diesel (crushing)	0.03 - 0.08
Buildings/development	£1.94M - £4.93M	Electricity (crushing)	0.15-0.72
Mobile Plant	£3.65M – £12.62M	Personnel	0.27-0.85
Grinding and accessories	£2.69M - £5.68M	Fixed plant, buildings and equipment (grinding)	0.19-0.34
		Energy (grinding)	1.00-31.60*
Total CAPEX	£26.91M – £66.59	Total OPEX	1.80-34.22
Total CAPEX per tonne	£53.82 - £13.32	OPEX + Discounted CAPEX+	3.40-40.68 <sup>+</sup>

\*Assuming an energy cost of  $\text{£}0.1\text{ kWh}^{-1}$ , for energy requirements of  $10\text{-}316\text{ kWh tonne}^{-1}$ .

+Assuming a discount rate of 4% over a 40 year time period

#### 4.5 Transport costs

Movement of millions of tonnes of rock per year poses substantial logistical challenges. Transport technologies have a range of energy efficiencies,  $\text{CO}_2$  emissions, and capital and operational costs (Table 4).

**Table 4:  $\text{CO}_2$  emissions, energy consumption, and operational and capital expenditure for a range of material transport options**

Transport Method	$\text{CO}_2$ emissions $\text{gCO}_2\text{ tonne-km}^{-1}$	Energy $\text{kWh tonne-km}^{-1}$ *	OPEX (£ tonne-km <sup>-1</sup> )	Current freight transportation inside the UK ( $\text{Mt a}^{-1}$ )
Road heavy goods vehicle	62	0.365	0.0496	1489
Rail freight	22	0.058	0.0284 - 0.0342	44
Inland waterways	31			43

Large Ship distribution	7	0.060	0.0010	
<p>Emissions data from CEFIC (2011). *Energy data derived from Davis et al., 2011. Heavy goods vehicle OPEX calculated using <a href="http://www.freightmetrics.com.au/Calculators/TruckOperatingCostCalculator">http://www.freightmetrics.com.au/Calculators/TruckOperatingCostCalculator</a> – Accessed March 2012, and assuming a fuel cost of £1.41 l<sup>-1</sup>, and a haulage distance of 1288 km day<sup>-1</sup>, a 3 axis rigid truck with 30 tonnes payload. This does not include infrastructure modification/maintenance. Maritime shipping based on values from UNCTAD (2010).</p> <p>Current UK rail freight statistics from <a href="http://www.rfg.org.uk">http://www.rfg.org.uk</a> – Accessed March 2012.</p> <p>Statistics for road and inland water freight from the Department for Transport (DfT, 2012, DfT, 2011c).</p>				

For enhanced weathering, the material would be transported from the outcrop to the application site. In the UK the application site would be agricultural land; although other studies have focus on application in the tropics (e.g. Köhler et al., 2010). Figure 7 shows the distribution of major ports and arable farmland in the UK. Most arable land and silicate rock outcrops are located within 100 km of a major port. Assuming a conservative ocean transport distance of approximately 2000 km (approximately 2/3 of the circum-navigable distance around the British Isles), a land haulage distance of 75 km using rail freight and 25 km using heavy good vehicles requires approximately 133 kWh t<sup>-1</sup>. The operational cost of transporting material is presented in Table 4.

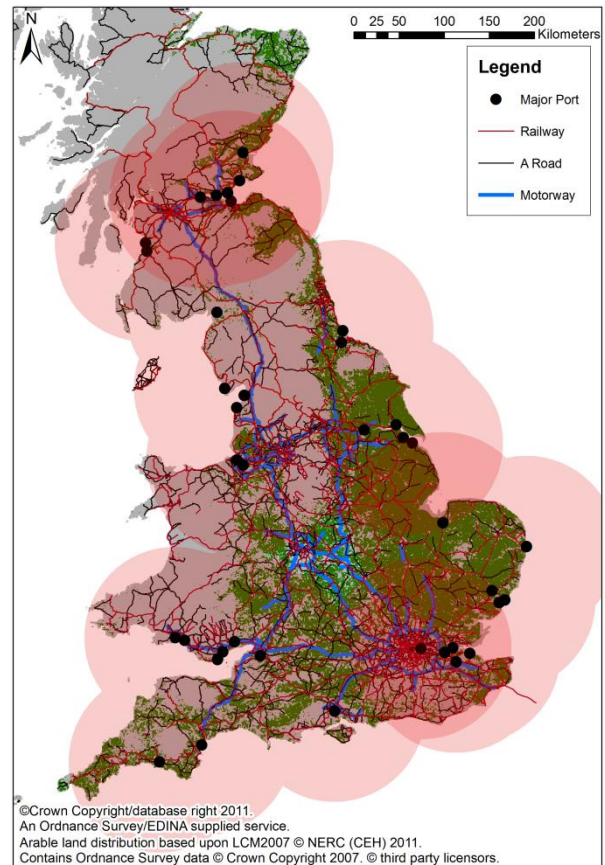


Figure 7: Major port, road, and arable land distribution in the United Kingdom. Shaded circles represent 100km radius around a major port. Shaded regions on land represent arable farming.



Figure 8: Sites of ultrabasic industrial mineral deposits in Norway. (Source: National Geological Survey of Norway <http://www.ngu.no> – Accessed March 2012)

Given that road, rail and inland water ways transport approximately 1500 Mt of material  $\text{a}^{-1}$ , additional capital investment may be required, particularly in rail infrastructure. Although, spare capacity is available in some sections of infrastructure (DfT, 2004). A full assessment of current utilisation and the required additional infrastructure is beyond the scope of this paper.

#### 4.6 Application of silicates to land

Spreading crushed rock onto the land surface is widely practiced in agriculture (addition of limestone). Typical application rates are around  $0.4 \text{ tonnes ha}^{-1} \text{ a}^{-1}$ , which is distributed using farming machinery capable of material spreading (Davies et al., 2001). Essentially, this requires a mobile material storage bin with rotating discs at an aperture, kinetic energy imparted by the discs randomly sprays the particulate material onto the field. Approximately  $21 \text{ kWh t}^{-1}$  is required for a spreading density of  $2.5 \text{ t ha}^{-1}$  (Chudleigh and Greet, 1983), although this is substantially more than the  $0.2 \text{ kWh t}^{-1}$  quoted in ASAE (1999) or 0.5-

$1.5 \text{ kWh t}^{-1}$  calculated from emission data in Lal (2004). Of course, this value will change with respect to spreading density. Spreading 125 Mt of material over 50% of the 5 million ha of arable land in the UK (Figure 7), would require a spreading density of  $5 \text{ kg m}^{-2}$  ( $50 \text{ tonnes ha}^{-1} \text{ a}^{-1}$ ; similar to manure application rates).  $21 \text{ kWh t}^{-1}$  has been used in the calculations in this paper (equivalent to a reduction in  $\text{RCO}_2$  by 0.005).

#### 4.7 Cost Summary

There is limited information on the costs associated with enhanced weathering so a preliminary estimate has been made. Using the range of possible values for material processing, transport and distribution an estimate of operational cost for enhanced weathering is provided in Table 5. It is clear that there is substantial variation in the cost, which is primarily due to the uncertainties associated with energy use in mineral comminution ( $39\text{--}2327 \text{ kWh tCO}_2^{-1}$  for basic rocks and  $13\text{--}497 \text{ kWh tCO}_2^{-1}$  for ultrabasic rocks). While this cost model takes into account  $\text{CO}_2$  production from this technology, it also assumes that the maximum potential of carbon capture is achieved (applying an efficiency factor here would be arbitrary). The estimated energy requirements for enhanced weathering are  $656\text{--}3501 \text{ kWh tCO}_2^{-1}$  for basic rocks and  $224\text{--}748 \text{ kWh tCO}_2^{-1}$  for ultrabasic rocks. The operational costs of enhanced weathering, assuming national transport, are  $\text{£}44\text{--}361 \text{ tCO}_2^{-1}$  ( $\text{\$}70\text{--}578 \text{ tCO}_2^{-1}$ ) for basic rocks and  $\text{£}15\text{--}77 \text{ tCO}_2^{-1}$  ( $\text{\$}24\text{--}123 \text{ tCO}_2^{-1}$ ) for ultrabasic rocks. The use of basic rocks for enhanced weathering is likely to be limited unless they undergo rapid weathering when placed onto the land surface (subsequently requiring minimum comminution energy input). Substantial comminution would release nearly as much carbon dioxide than is sequestered through weathering, which is responsible for the high unit cost above. Exploitation of ultrabasic rocks consumes less energy per unit of  $\text{CO}_2$  sequestered. It is tempting to compare these cost estimates with conventional mitigation or underground carbon capture and storage. However, it should be noted that some emissions may be difficult to mitigate (e.g. air travel and land use change), which would provide a niche opportunity for carbon dioxide removal from the atmosphere (Keith, 2009). Furthermore, it is unknown what level of  $\text{CO}_2$  in the atmosphere is to be considered 'dangerous', and technologies should be developed (concurrent with, but

independent of conventional mitigation) for potential deployment should this limit be exceeded. In this sense, a comparable technology to enhanced weathering would be direct capture from the air. Estimates for air capture are variable ranging from \$100- \$1000 tCO<sub>2</sub><sup>-1</sup> (£63-625 tCO<sub>2</sub><sup>-1</sup>; House et al., 2011, Keith, 2009), which are at least comparable to the values proposed in this paper.

[Table 5 after references]

Additional work is required to assess the effectiveness of enhanced weathering by investigating the geochemical limitations to achieving the maximum carbon capture potential, together with the energy requirements of mineral comminution, and strategies for material distribution.

This study has focused on exploiting mineral resources in the UK. It may be possible to import ultrabasic rocks from regionally local sources. For instance, the west coast of Norway has substantial ultrabasic resources which could be extracted, transported by sea freight, and used by the agricultural industry in the UK (3000 km), the Baltic States (3500 km), northern Germany (2500 km), the Netherlands and Belgium (2000 km), northern/western France (3500 km), and northern Spain (3500 km).

Figure 9 presents a sensitivity analysis for reasonable changes in key assumptions. The top three panels demonstrate cost variation due to changes in transport distance, in which the cost range increases dramatically for material with lower  $R_{CO_2}$  values. The lower three panels show changes in cost associated with grinding energy cost and carbon intensity of electricity and fuel. These also demonstrate greater sensitivity in materials with lower  $R_{CO_2}$ , particularly for the maximum estimates (the minimum estimates remain largely unresponsive).

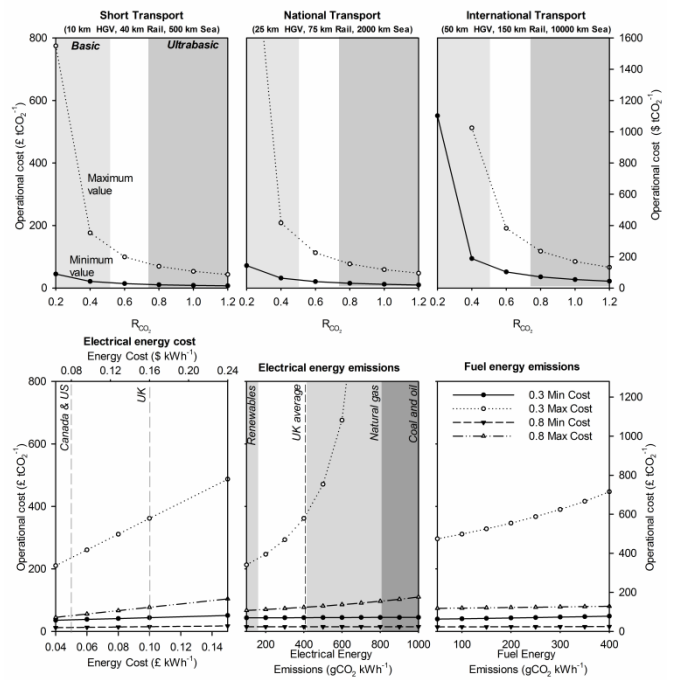


Figure 9: Sensitivity analysis of the key assumptions (energy cost, energy carbon intensity) and  $R_{CO_2}$  variation in enhanced weathering

## 5. Social and Environmental impacts

It is probable that the economic, environmental, and social impact of enhanced weathering will increase as a function of utilisation (e.g. discussion in Keith, 2000). These factors are likely to impose capacity limits that are lower than the theoretical maximum of a technology (Figure 10). For instance, over exploitation of enhanced weathering may impact on food and biomass production (though modification of soil chemistry) or generate an unacceptable amount of dust during comminution or application. Therefore, there are social, economic, and environmental limits of this technology, which are independent of its theoretical maximum storage potential. As new technologies, social priorities, or environmental conditions develop, these limits will change. Understanding the relationship between each of these limits and how they could change over time should be a key area for enhanced weathering (and geoengineering) research. This paper focuses on a number of the key technical limitations and does not explore environmental or social limitations of enhanced weathering.

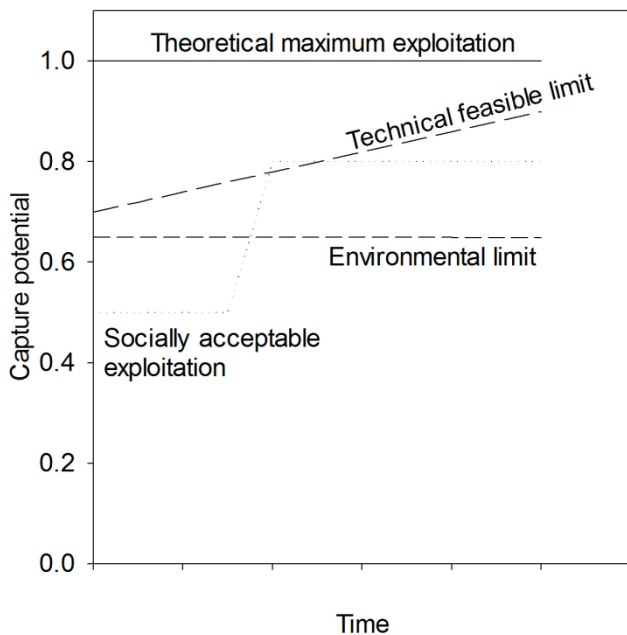


Figure 10: Conceptual diagram of exploitation limits, and potential changes over time.

## 5. Conclusions

The UK has substantial silicate resources that may be exploited for enhanced weathering (430 GtCO<sub>2</sub>). The majority of this is in the form of basic rocks with an  $R_{CO_2} \sim 0.3$  (tCO<sub>2</sub> per t of rock). The uncertainty in energy requirements, specifically for mineral comminution, reflects the uncertainty in how this material will weather when placed on the land. However, it is estimated here that enhanced weathering using basic rocks will require between 656 and 3501 kWh tCO<sub>2</sub><sup>-1</sup> and cost approximately £44-361 tCO<sub>2</sub><sup>-1</sup> (\$70-578 tCO<sub>2</sub><sup>-1</sup>). The UK has less ultrabasic resources (25.4 GtCO<sub>2</sub>), which generally exist in environmentally sensitive locations. However, given the higher concentrations of cations in the material, the unit cost of exploitation is considerably cheaper (£15-77.2 tCO<sub>2</sub><sup>-1</sup>; \$24-124 tCO<sub>2</sub><sup>-1</sup>) and requires considerably less energy (224-748 kWh tCO<sub>2</sub><sup>-1</sup>).

Some rock formations underlie environmentally protected land (e.g. The Borrowdale Formation in the Lake District National Park, or the Whin Sill adjacent to protected areas along the Northumberland coast), and in some cases are likely to be unavailable for extraction. However, substantial potential may still be available (Ochil Volcanic Formation, Clyde Plateau, Isles of Mull and Skye, Northern Ireland basalts). The environmental impact of extraction or silicate

application to land has not been comprehensively assessed.

The potential of this technology warrants further investigation, particularly understanding the rates and limitations to weathering of silicates placed onto the land surface. However, this should be done concurrently with an assessment of the engineering requirements of enhanced weathering, particularly the energy demand of comminution. If enhanced weathering was deployed in the future, it is likely that it will form part of a portfolio of carbon dioxide removal techniques.

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**Table 5: Energy and operational cost of enhanced weathering**

Operation	Energy cost (kWh t <sup>-1</sup> )	Reduction in R <sub>CO2</sub>	Energy cost kWh tCO <sub>2</sub> <sup>-1</sup>		OPEX (£ t <sup>-1</sup> )	OPEX (£ tCO <sub>2</sub> <sup>-1</sup> )	
			R <sub>CO2</sub> = 0.3 (basic)	R <sub>CO2</sub> = 0.8 (ultrabasic)		R <sub>CO2</sub> = 0.3	R <sub>CO2</sub> = 0.8
Excavation, drilling, blasting and short range haulage	5	0.0012	19.4-36.8	6.6-7.9	0.5 <sup>†</sup>	1.9-3.7	0.7-0.8
Comminution	10-316 (as electricity)	0.0040 to 0.1264	38.7-2326.9	13.2-497.0	3.4-40.7	13.2- 299.5	4.5-64.0
Transport (national)*	133	0.0316	516.9-4- 982.8	176.0-209.9	5.4-5.8	20.8- 42.7	7.1-9.1
Spreading	21	0.0050	81.3-154.6	27.7-33.0	2.1-2.1 <sup>†</sup>	8.1-15.5	2.8-3.3
Total	169-475	0.0418 to 0.1642	656.4- 3501.2	223.5-747.8	11.4-49.1	44.0- 361.4	15.0-77.2

\*Sea freight 2000 km, land using rail 75 km, land using HGV 25 km

<sup>†</sup>Calculated using a value of energy ~£0.1 kWh<sup>-1</sup>

## Appendix

### Equations for estimating carbon capture potential

The maximum carbon dioxide capture potential (R<sub>CO2</sub> in tCO<sub>2</sub> per t of material) of enhanced weathering is defined by the chemistry of the silicate material. It can be estimated through Equation A1:

$$R_{CO_2} = \frac{M_{CO_2}}{100} \left( \frac{\% CaO}{M_{CaO}} + \frac{\% MgO}{M_{MgO}} \right) \cdot \omega \quad \text{Equation A1}$$

where M<sub>CaO</sub>, M<sub>MgO</sub> and M<sub>CO2</sub> are the molecular masses of CaO, MgO and CO<sub>2</sub> respectively.  $\omega$  represents the additional draw down from cation flux into the ocean (approximately 1.7). The carbon capture potential of mineral carbonation is calculated using Equation A1 assuming that  $\omega = 1$ . Equation A1 is used in Section 2 to estimate the carbon capture potential of rock formations through Equation A2

$$\text{Total Capture Potential of Rock formations} = R_{CO_2} \cdot S \cdot z \cdot \rho_{\text{silicate}} \quad \text{Equation A2}$$

where S, z and  $\rho_{\text{silicate}}$  are the spatial extent, thickness and typical density of the silicate rock formation respectively.

Estimation of the in-situ carbon dioxide capture potential of a rock formation was calculated by volumetric analysis of the rock rather than the chemistry of the material Equation A3.

$$\text{Total Capture Potential of Rock formations} = S \cdot z \cdot \phi \cdot \rho_{\text{carbonate}} \cdot \frac{44}{100} \quad \text{Equation A3}$$

Where  $\phi$  is the fraction of the volume available for carbon storage (~1%), and  $\rho_{\text{carbonate}}$  is the density of the precipitated carbonate.

**Table A1: Chemical compositions of igneous rocks in the UK**

	Mull lava field (Kerr 1995)	Whin Sill dolerite Randall 1989	Antrim Basalts (Barrat and Nesbitt 1996)	Central lowland Carboniferous (Macdonald et al., 1977)	The Lizard Complex (Green 1964)	Skye Lava field (Bell et al., 1996)	Cheviot lava (Thirlwall 1981)	Old Red Sandstone Lava, Cheviot (Thirlwall	Grampian Gabbro (Viète et al., 2010)	Borrowdale Lava (basaltic andesite; Fitton	Rhombell lava (Kokelaar 1986)	Shetland Ness (Thirlwall 1981)	Shetland ophiolite (BGS 1985)	Eycott (Millward et al., 2000)
SiO <sub>2</sub>	46.35	49.85	48.90	45.55	39.90	46.06	63.20	54.62	50.77	55.74	47.98	52.57	40.46	58.95
TiO <sub>2</sub>	1.36	2.28	1.33	2.39	0.14	2.40	0.91	1.34	1.76	1.09	1.24	1.03	0.01	1.15
Al <sub>2</sub> O <sub>3</sub>	16.14	14.17	14.96	14.70	3.74	16.38	16.01	17.33	15.71	17.81	17.86	17.08	0.44	16.58
Fe <sub>2</sub> O <sub>3</sub>	12.49	12.56	12.09	11.72	8.01	11.28	4.92	7.49	10.39	8.84	10.35	9.40	10.63	8.18
MnO	0.18	0.18	0.19	0.18	0.08	0.18	0.07	0.11	0.14	0.20	0.18	0.27	0.15	0.12
MgO	10.01	6.01	9.18	7.92	36.21	4.63	3.00	5.59	6.01	2.83	7.86	6.22	43.51	3.17
CaO	10.80	9.33	9.90	9.55	1.54	11.19	3.85	7.47	8.32	4.75	4.96	7.61	0.28	5.13
Na <sub>2</sub> O	2.30	2.42	2.46	2.79	0.10	3.88	3.57	3.89	3.72	2.67	4.19	3.88	0.01	3.35
K <sub>2</sub> O	0.19	0.93	0.53	1.14	0.02	0.24	3.94	1.51	0.80	2.97	0.68	1.22	0.01	1.90
P <sub>2</sub> O <sub>5</sub>	0.13	0.28	0.16	0.48	0.04	1.92	0.31	0.27	0.30	0.19	0.11	0.15		0.14
H <sub>2</sub> O <sup>+</sup>	-	1.31	-	2.83	10.11	-	-	-						
H <sub>2</sub> O <sup>-</sup>	-	-	-	-	0.33	-	-	-						
CO <sub>2</sub>	-	-	-	0.40		-	-	1.10			4.58			
Total	99.95	99.29	99.70	99.65	100.22	98.17	99.78	100.72	97.91	97.09	99.99	99.43	95.49	98.67
R <sub>CO2</sub>	0.33	0.24	0.30	0.28	0.70	0.24	0.11	0.20	0.22	0.12	0.21	0.22	0.82	0.13