Developing Lifecycle Inventory Indices for Estimating the Carbon Sequestration of Artificially Engineered Soils and Plants

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Abstract
Recent research into soil engineering indicates that increased carbon sequestration through the conversion of atmospheric CO_2 to a pedogenic carbonate mineral (calcite, $CaCO_3$) may help mitigate global warming through artificially engineering a variety of soils with selected materials and vegetation so that they have a photosynthesis-driven carbon capture function. Non-biological processes of carbonation also occur, at high pH. In both cases, CO_2 partitions into soil porewaters as dissolved carbonate, and precipitates by combining with Ca derived from portlandite $(Ca(OH)_2)$ and weathered cement-derived calcium silicates, derived from materials generated by the demolition process. This paper aims to illustrate a method for capturing lifecycle data by quantifying the stocks and flows of the process through a series of possible experiments which if proved successful will allow a deeper understanding of the ability of soil calcite to act as a stable CO_2 sink. The challenge is to identify non-calcined (i.e. calcium silicate) rocks, that weather sufficiently rapidly to provide a net sink for CO_2, taking into account all emissions during production. The results will lead to normalised impact assessment data which can be applied globally within the fields of soil science and civil engineering. Furthermore, if the results indicate a strong uptake in terms of CO_2 sequestration there is the potential to assist the United Kingdom's 80% CO_2 reduction target by 2050 through widespread adoption of the technology, provided that there is a positive cost-benefit ratio.

Keywords: Climate change, Soil science, Carbon sequestration, Environmental policy

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1 Introduction

The potential of urban plants and soils to store C has long been recognised (IPCC, 1996; Lal et al., 2007) however, the magnitude and significance of this store with regard to climate change have only recently been quantitatively estimated (Smith, 2004; Smith et al., 1997). In a global context, soils are now recognised as an important potential tool for mitigation of rising atmospheric CO_2 concentrations. The 1997 Kyoto Protocol highlighted soils as a 'major carbon store' and recognised that processes underlying soil function should be considered in CO_2 emissions accounting.

In the UK, the Department for Environment, Food and Rural Affairs (DEFRA, 2009) highlighted a pressing need to 'develop a better understanding of steps that can be taken to protect or enhance levels of soil carbon'. Given that there is an 80% emissions reduction target by 2050 in the UK, it is imperative that further research into the development of engineered soils with a carbon function is maintained. The scale of the problem of compensating for anthropogenic emissions of CO_2 is clear: by 2050, global emissions will exceed 2000 levels by 7 Gt CO_2. There is no single process or activity that could compensate for this, and it has been suggested that 7 different approaches or carbon wedges, each accounting for 1 Gt C (approx. 3.7 Gt CO_2), might be a reasonable target (Pacala & Socolow, 2004). Current UK emissions are 456 mt CO_2 (590 mt CO_2 in 1990; www.decc.gov.uk), and so the UK's target for CO_2 emissions reduction, whilst ambitious, is small compared with potential global targets.

The aim of this paper is to report the design and development of an indexed inventory-based lifecycle assessment (LCA) framework that results in the creation of normalised impact assessment data. This also involves expanding initial embedded lifecycle CO_2 anthropogenic estimates into Global Warming Potentials (GWP) through CO_2 equivalency (CO_2e). It is the first lifecycle assessment that addresses organic and inorganic soil carbon sequestration; no primary lifecycle inventory data on carbon sequestration rates for minerals exist, possibly due to the wide variability in site location, the quantity of non-calcined rock types as well as logistical and production based dimensions. Following a short overview of current engineered and mineral based carbon sequestration methods, the paper is then illustrated focusing on the scope of the field study, data collection and comparison of rock types from brownfield sites and an illustration of the study location.

2 Carbon Sequestration of Engineered Soils

Recent evidence of soil based chemical processes indicate a rapid carbon sequestration function, removing CO_2 from the atmosphere and storing it as organic (soil organic matter) and inorganic (pedogenic) forms of carbon (C). During silicate weathering, calcium (Ca) and magnesium (Mg) silicate minerals naturally react with dissolved carbon dioxide (CO_2) to form carbonates (Berner et al., 1983) effectively capturing and fixing atmospheric C. Weathering involves leaching and transport of Ca^{2+} and Mg^{2+} in solution, some of which reacts with carbonate anions formed by CO_2 dissolution in soil pore waters to precipitate pedogenic (soil-formed) carbonates. This process contributes to the stabilisation of atmospheric CO_2 concentrations over various geological timescales (Berner & Lasaga, 1989; Berner et al., 1983) and forms pedogenic carbonates in both natural (Nettleton, 1991) and artificial soils (Renforth &

Manning, 2011). A simplified version of the carbonation reaction for artificial calcium silicates is given in Reaction 1.

$$CaSiO_3 + CO_2 + 2H_2O \rightarrow CaCO_3 + H_4SiO_4 \tag{1}$$

2.1 Mineral Carbonation for Carbon Capture and Storage

Reaction (1) involves passive capture and storage of C. The addition of Ca and Mg-rich silicates to soils may promote the accelerated draw-down and storage of CO_2 from the atmosphere as relatively stable carbonate minerals (Manning, 2008; Schuiling & Krijgsman, 2006). Recently investigations into the role of urban soils as a sink for atmospheric CO_2 have shown that pedogenic carbonate minerals, normally regarded as a phenomenon of natural arid soils (Landi et al., 2003) form in *artificial* urban soils in the UK and North America (Manning & Renforth, 2012). For example, work at the 10 ha Newcastle Science Central site (Figure 1) formerly occupied by the Newcastle Brewery, has shown that the **soils onsite remove 85 t of CO_2/ha annually** (Washbourne et al., 2015). Overall, the $CaCO_3$ content to 100 mm depth over 18 months (2010−2012) increases from 22.0 to 39.0 wt %. Assuming that the UK has 1.7 million ha of urban land, only 12,000 ha of land in a similar state to that of Science Central will remove 1 Mt CO_2 annually.

Figure 1: Science Central; St James Park in background (Newcastle upon Tyne, UK)

Isotopic analysis of carbonate C and oxygen (O) shows that pedogenic carbonates in urban soils have similar isotopic compositions to carbonates from natural soils in which photosynthesis contributed to their formation (Cerling, 1984), and confirms that a significant proportion of the C has been captured from the atmosphere (Renforth et al., 2009; Wilson et al., 2009). This may be a characteristic phenomenon of urban brownfield sites and other anthropogenic soils, as construction activities in urban soils usually involve the addition of Ca/Mg-rich substrates. If added to such soils, construction and demolition (C&D) waste, fly ashes, iron and steel slag etc. may enhance C capture and storage in the urban environment (Morales-Flórez et al., 2011; Renforth et al., 2011a; Renforth et al., 2009; Renforth et al., 2011b) thus the value of

materials which may otherwise be regarded as 'wastes' is increased. In principle, carbonation of these materials could be used to offset the C emissions associated with their production. There is potential for C capture and storage to become a routine design consideration in the engineering of anthropogenic soils, with minimal additional energy input, little change in current management practise and minimal translocation of materials. Figure 2 illustrates recently completed trial plots at Cockle Park Farm in Northumberland, UK, which use a mixture of different materials to test carbon capture.

Figure 2: Carbon Capture Trial Plots at Cockle Park Farm (Northumberland, UK)

2.2 Organic and Plant-Based Reactions
A number of organic chemical reactions exist which have an effect on soil's ability to act as a carbon sink (Schmidt et al., 2011). According to Davidson and Janssens (2006) the decomposition of microbial matter in soil along with root respiration almost entirely leads to expulsion of CO_2 to the atmosphere leading to a positive feedback loop on climate change. This so called C fluxing must be managed in order to reduce its exposure in the atmosphere as much as possible. Carbonation of minerals in soil breaks this feedback loop. Soil stores up to four times as much carbon as plant biomass (Stocker et al., 2013) and soil microbial respiration releases about 60 Gt of carbon per year as carbon dioxide (Shao et al., 2013). While initial experiments have shown that soil microbial respiration increases exponentially with temperature (Davidson & Janssens, 2006) more recent work has attempted to predict the magnitude of this reactivity. Karhu et al (2014) studied a range of different ecosystems from the Arctic to the Amazon to determine how microbial community responses affect the typical 90 day mid to long-term sensitivity to temperature change. They discovered that at colder climates the sensitivity is enhanced by a factor of 1.4 compared to instantaneous responses. While the assessment of carbon flux is out of scope for this initial study, subsequent experiments will attempt to take such reactions into account in order to determine its impact on carbon sequestration.

3 Methodology

The methodology consists of an approach which enables the design of an LCA inventory index to determine the 'default' carbon sequestration rates of calcium carbonate in brownfield sites. In addition, a lifecycle assessment of one of the key plots within the SUCCESS carbon capture site is simulated using dolerite (diabase) and basalt as calcium silicate additives to soil.

3.1 Scope of the Study

The scope of the study is based around maximizing the effectiveness of carbon sequestration and $CaCO_3$ formation within brownfield sites or engineered soils associated with construction activity, by adding artificial or natural calcium silicate materials. The reason for using this type of material as described earlier is the abundance of calcium and magnesium substrates, which in part is a direct consequence of the construction and demolition process. In order for this to happen a variety of different experiments consisting of soil types and vegetation is being explored. Figure 3 illustrates a comparison of a typical carbon cyclic process of C between greenfield lands and the artificially engineered experimental soils at Cockle Park Farm using crushed concrete and dolerite, collected locally, to maximize sequestration effectiveness. It should be noted that a combination of feedback processes in both greenfield and carbon capture sites will be occurring such as photosynthesis in the plot trials etc.
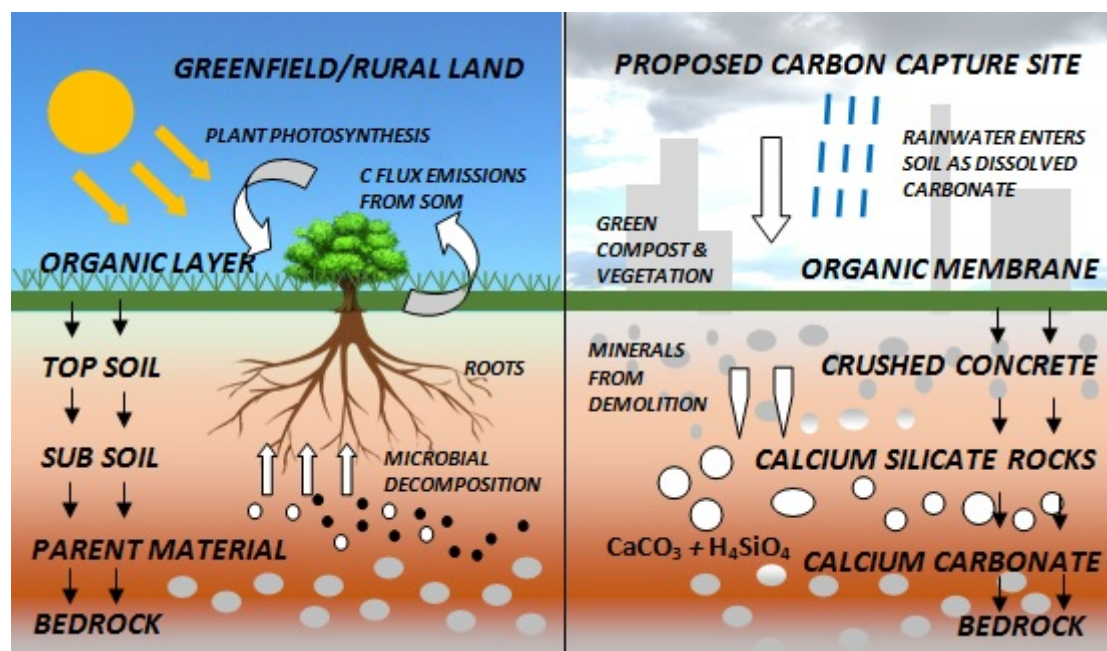

Figure 3: Comparison of dominant carbon cyclic processes in greenfield/rural land and the carbon capture site

Figure 4 outlines the scope (system boundary) of the LCA model. The model considers the initial supply of raw material including natural rock and crushed concrete fines in which they both have their individual collection process. The concrete fines for this study are classified as a waste product from crushing associated with demolition. These are sourced from the same locations as the natural rock. Logistical transport, excavating and mixing is measured in CO_2 equivalency using the

Global Warming Potential (GWP) characterisation factor. The so-called global warming potential (GWP_i) of GHG emission i measures its relative global warming strength in terms of its absorbed radiation in comparison to a CO_2 emission (Solomon, 2007). To obtain a global warming impact score GW for all GHG emissions m_i along a life cycle, the amounts m_i of all GHG emissions i are multiplied by their characterization factors GWP_i and summed:

$$GW = \sum_i m_i GWP_i \qquad (1)$$

The distinction between GWP_i and GW is essential: GW describes the actual global warming impact of a product's lifecycle (also known as Carbon Footprint), whereas GWP_i characterizes the radiation absorption of a single GHG emission independently from any product life cycle. Within the life cycle, material processing includes the mechanical handling (excavation) and mixing of the material at site which leads to the eventual application to the soil plots. The CO_2 release of the soil organic matter due to microbial and vegetation respiration while out of scope for this analysis should be estimated in future, based upon previous studies.
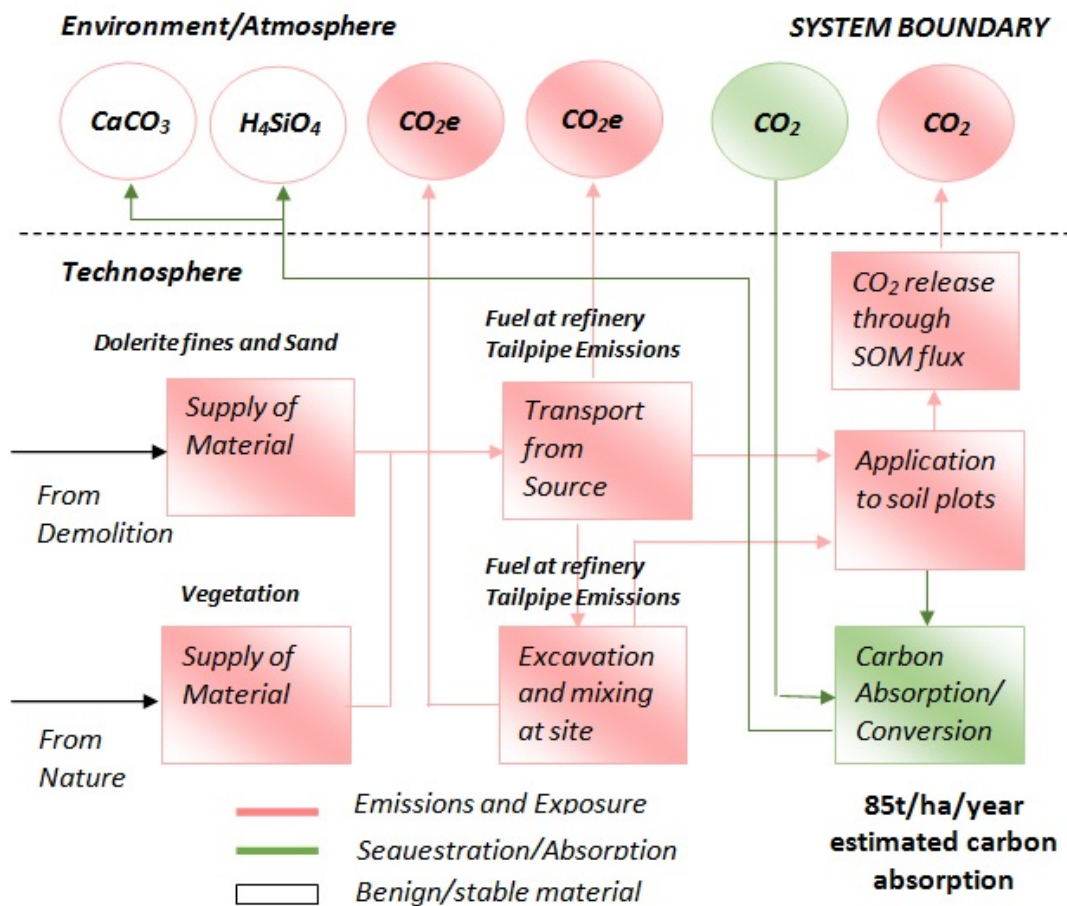

Figure 4: LCA Boundary Analysis

3.2 Data collection

The trial at Cockle Park Farm consists of 14 distinct plots each with a mixture of different types of material, mainly dolerite, sand and crushed concrete. In order to select the appropriate plot to simulate it was first necessary to perform a comparison of materials between several source quarries in the region. Figure 5 illustrates the sample region and collection.

Figure 5: Left – A map illustrating quarries in Northumberland (Source: Simkin, 2012), UK; Top-Right –'Disturbed' samples (Site A); Middle-Right – Site B (possible calcium carbonate visible on surface); Bottom-Right – 'Disturbed' samples (Site B) of 100mm applied substrate depth; Bottom-Left - SUCCESS team on-site collecting samples from 'Site B'

The initial data collection consisted of collecting a variety of 'disturbed' test samples from site A (UK Grid Reference NY 92643 75682). 6 random 'disturbed' samples were also taken at site B (Grid Reference NU 13163 34162). Figure 5 illustrates the site locations and sample collection process. The estimated rate of C sequestration (85t/ha/year) for demolition/waste material is derived from the study undertaken at Science Central (concrete-based) by Washbourne et al (2015). Site B plot trials have been operating for around 4 years while Site A is estimated to have remained untouched for 15 years. Figure 6 illustrates the layout of the plots as well as the different treatments that were given (Simkin, 2012). A disturbed sample was taken from each plot in order to estimate the current level of carbonization of CO_2 into calcium carbonate. The collected samples would assist in providing an understanding of which types of soil material perform best at absorbing carbon and therefore estimating the carbon sequestration rate without the addition of other materials.
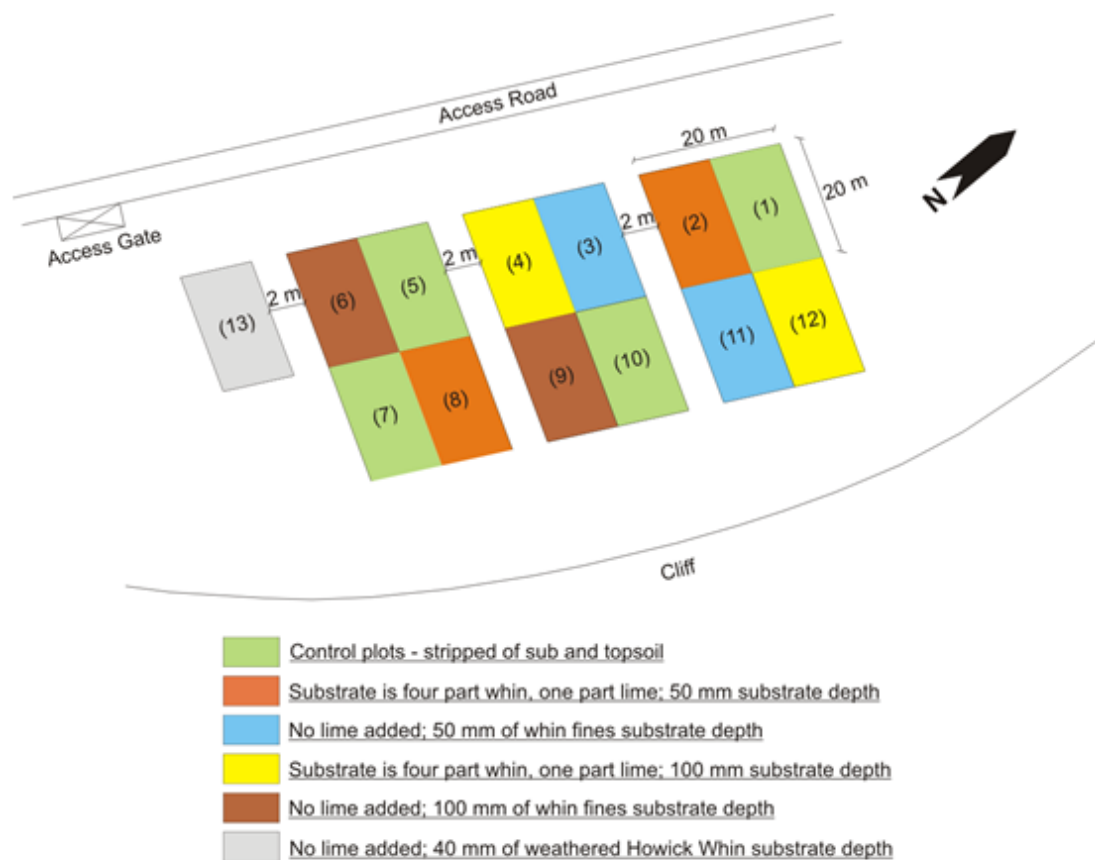

Figure 6: 'Site B' plot layout and treatments (Source: Simkin, 2012)

The first set of results consist of an organic and inorganic sequestered carbon comparison of local quarries i.e. site A and site B. As the two locations feature carbonation at different time scales, a time series analysis was applied to the results. An LCA of the SUCCESS carbon capture plot that theoretically has the most potential to absorb carbon was then performed using a mixture of materials that is considered to be optimal for sequestration. This is based on past studies of basalt showing a rapid carbon absorption rate. Table 1 and Figure 7 illustrates a carbon analysis of the Site A quarry sampling. Two samples were collected for each specific location within the

quarry. From field observation, the quarry floor indicates a rough mix of clay, crushed concrete and dolerite/basalt fines.

Table 1: Site A Quarry Carbon Analysis (Organic/Inorganic)

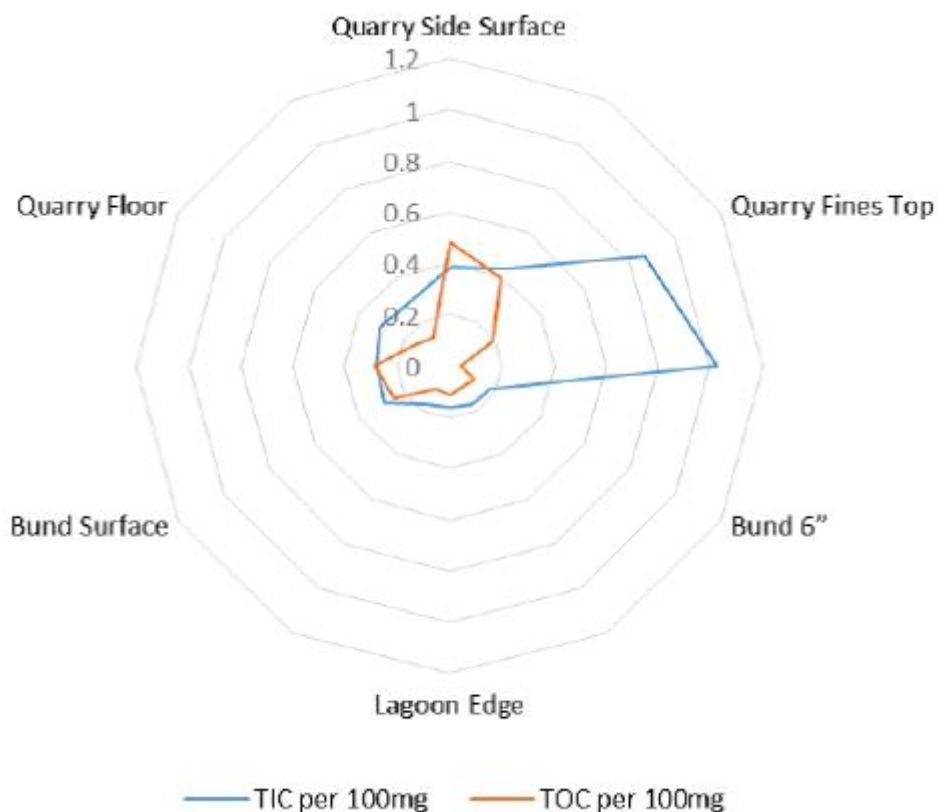| Sample ID	Sample Description	TOC per 100mg (%)	TIC per 100mg (%)	TC per 100mg (%)
Qu Si Su	Quarry Side Surface	0.38	0.48	0.87
		0.43	0.39	0.83
Qu Fi To	Quarry Fines Top	0.86	0.19	1.05
		1.02	0.04	1.07
Bu 6"	Bund 6"	0.18	0.10	0.28
		0.17	0.09	0.26
La Ed	Lagoon Edge	0.16	0.11	0.27
		0.17	0.10	0.27
Bu Su	Bund Surface	0.28	0.24	0.52
		0.27	0.28	0.56
Qu Fl	Quarry Floor	0.30	0.16	0.47
		0.29	0.12	0.42

Figure 7: Comparison of Site A carbon analysis

Table 2 and Figure 8 illustrate the analysis of the site B carbon content. Three samples in total from each plot were analysed with the mean TOC/TIC illustrated in the table.

From the results it appears that the plots which feature significant levels of carbon also feature a large quantity of dolerite material (Plots 2, 4, 8 and 12) in addition to 20% of these plots also containing lime. All of the materials originate from the site.

Table 2: Site B Carbon Analysis (Organic/Inorganic)

| Sample ID	Sample Description	TOC per 100mg (%)	TIC per 100mg (%)	TC per 100mg (%)
1	Control Plot – Stripped of sub and topsoil	1.06	0.72	1.78
2	Four part Dolerite, one part lime, 50mm substrate depth	0.17	8.62	8.80
3	No lime added; 50 mm of Dolerite fines substrate depth	0.70	1.16	1.86
4	Substrate is four part Dolerite; one part lime; 100 mm substrate depth	0.20	8.22	8.43
5	Control Plot – Stripped of sub and topsoil	1.20	0.49	1.70
6	No lime added; 100 mm of Dolerite fines substrate depth	0.12	0.48	0.60
7	Control Plot – Stripped of sub and topsoil	0.74	0.91	1.65
8	Four part Dolerite; one part lime; 50 mm substrate depth	0.25	8.09	8.35
9	No lime added; 100mm of Dolerite fines substrate depth	0.03	0.47	0.50
10	Control plot – Stripped of sub and topsoil	0.71	0.68	1.39
11	No lime added; 50mm of Dolerite fines substrate depth	0.46	0.57	1.04
12	Four part Dolerite, one part lime	0.36	8.08	8.44
13	No lime added; 40mm of weathered Howick Dolerite substrate depth	1.06	0.09	1.16
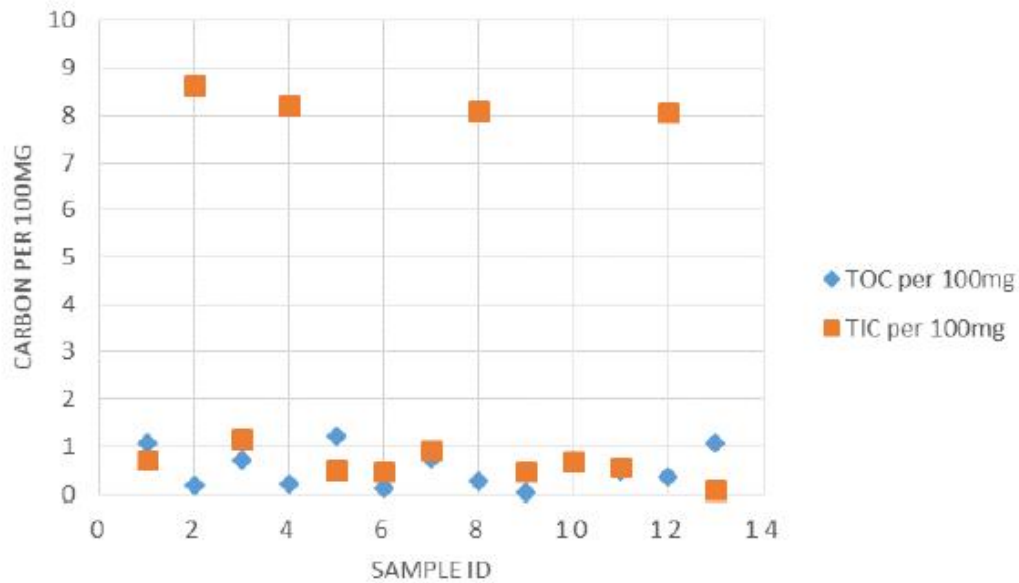
Figure 8: Comparison of Site B plot samples illustrating both total organic and inorganic carbon

Figure 8 illustrates the difference between total organic and inorganic carbon. For Site B, the inorganic carbon results for plots with added lime (limestone) include the carbon that is part of the lime used to build the plots, but the maximum TIC contribution from this source is 2.4%. The excess above this is due to the absorption of CO_2 from air into the soil.

3.3 Inventory Analysis and Key Assumptions
SimaPro v8.1 was used in the analysis of the estimated embedded emissions of the plot trials using a combination of previous studies and general assumption. SimaPro is a dedicated LCA software tool for undertaking LCA studies according to EN ISO 14040 (International Standards Organisation, 2006) and EN ISO 14044 (International Standards Organisation, 2006). The Ecoinvent inventory database (v3) was used to derive emission factors for each stage of the lifecycle. Table 3 shows the inventory analysis as input to the LCA, extrapolated to 1 ha ground area. The supply of raw material consists of dolerite, sand and compost from green waste. Sand was used as an unreactive mineral base, analogous to the sand fraction found in soils. Compost from green waste sources were added and sowed with a wildflower seed mix in order to encourage vegetation to growth and therefore allow increased absorption of CO_2 through photosynthesis to occur.

Table 3: Plot trial sub processes and inputs extrapolated to 1 ha.

Categories	Factors considered	Input
Supply of Raw Material	Dolerite (30% composition)	5,291.3 tonnes (weight)
	Sand (70% composition)	9,078.5 tonnes (weight)
	Compost from green waste (Top soil – 4% composition)	169.2 tonnes (weight)
Transport to site	Dolerite from Site B using Truck >20 tonnes	tkm = 338,066.2 (max load of 20 * total distance of 16903.31 km)
	Sand from Thrislington using Truck >20 tonnes	tkm = 664,774.5 (max load of 20 * total distance of 33238.7271)
	Compost from Newcastle upon Tyne using Truck >20 tonnes	tkm = 5,718.2 (max load of 20 * total distance of 285.91)
Excavation	Excavator	16,000 tonnes (weight)
Mixing at site	Loader	416.6 hours loader is active (time)

The input for each material is based upon the total material composition for a proposed trial plot of volume $10,000*1m^3$ (1ha). The weighting of the material was calculated by multiplying the density of each material with the volume of the plots and their composition. The estimated transport of the materials to the site was calculated in tkm (tonnes * total kilometres driven) with a max load of 20 tonnes for each trip (return journeys with an empty load were automatically calculated using the appropriate Ecoinvent dataset). The excavation of the area requires 16,000 tonnes of Holocene sand to be removed. This area was once again calculated using the density of the land to be excavated. Emissions of the excavator were calculated with an engine rating of 100kw. Diesel consumption is .000556 kg per kg of excavated material. Finally, the mixing at the site was carried out by a standard loader with front facing bucket. As the vehicle remains stationary during the mixing process the engine is in an idle state, therefore this dataset is based upon duration of mixing where baseline emissions are calculated from the engine remaining active but on standby.

4 Lifecycle Impact Assessment Results

4.1 Analysis
The use of impact categories gives the ability to compare the environmental impacts of the different options. Characterization factors, or equivalency factors, describe the relative impact of the different environmental flows (ISO 2006). A larger characterization factor means a larger impact for that flow. Characterization factors are multiplied by each of the environmental flows to convert all them into an equivalent amount of the category indicator. The category indicator is the flow that is usually associated with that particular impact category (CO_2 equivalency for global warming). The environmental impact categories which are required for LCI inventory involve SimaPro V. 8.1; this study used the category to assess the environmental impacts from the creation of passive carbon capture trial plots in the north-east of England.

In order to interpret the results, a characterisation method was selected. As only global warming potential was appropriate for this study there was no particular requirement to display the results of all the characterisation factors, therefore the

GWP factor from the CML baseline method was chosen. This method is an update of the CML 2 baseline 2000 and was released by CML in April 2013 (version 4.2).

4.2 Main results
The results of the LCA are illustrated in Figure 9. The diagram shows the embedded GHG emissions from the selected plot construction. As anticipated, carbon dioxide has a total share of 85% global warming potential, primarily from the logistics and transport requirements of the materials to the plot site. The other highest emission is the combustion of fuel required to run the trucks and loaders (marked as fossil). The majority of CO_2, methane, and dinitrogen monoxide emissions derive from the exhaust of the trucks used to transport the materials to the site. The middle row consists of the estimated take back of carbon against the embedded emissions after 1 year of carbon sequestration. The left pie chart illustrates the comparison between embedded and absorbed CO_2 while the right hand pie chart illustrates absorption against total GWP. The bottom row illustrates carbon sequestration after 2 years where the plot is estimated to become CO_2 negative.
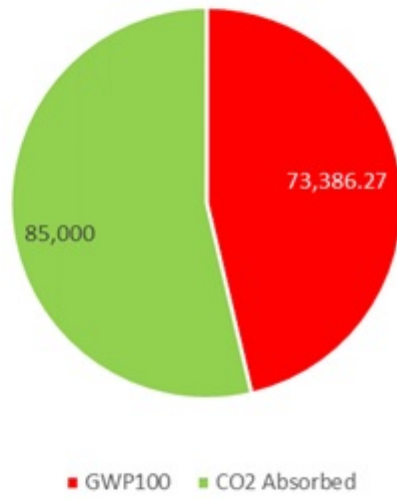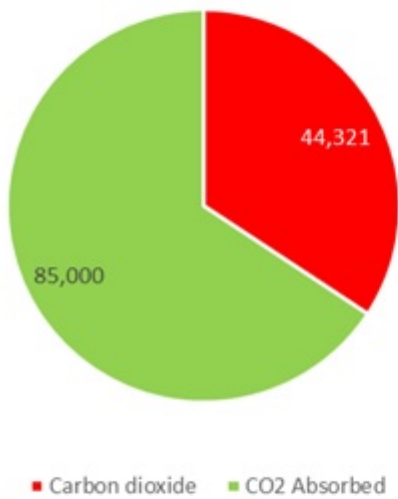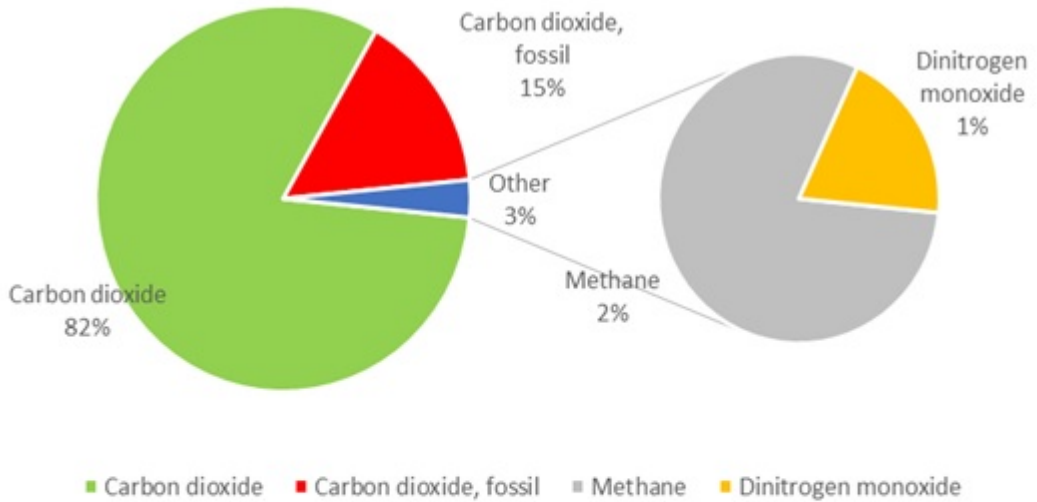
Figure 9: Top row: Embedded GHG emissions from selected plot construction (extrapolated to 1 ha); Middle row Estimated carbon sequestration level after 1 year compared to total embedded CO₂ (left) and total GWP100 (right); Bottom row:

projected carbon absorption after 2 years - experiment becomes carbon negative.
The mixing of materials, excavation of the area and the transport materials are illustrated below. All four areas of analysis illustrate the levels of GHG emissions based upon the functional unit of 1 hectare/10,000m^2 of carbon capture land per 1 year's operation. A cut off of 0.036% has been applied in order to illustrate only the most prominent emissions within the lifecycle. Note that the carbon sequestration function is shown here to compare with the total negative lifecycle emissions (i.e. 85t/ha/annum). From these results, it would take roughly around 18 months to offset the embedded emissions of 1 hectare of carbon capture plot using the materials provided.
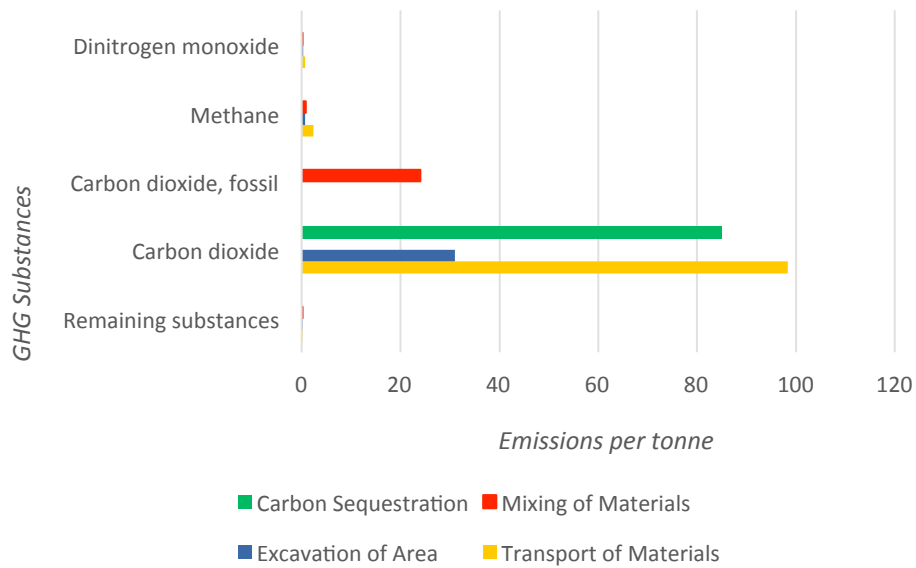

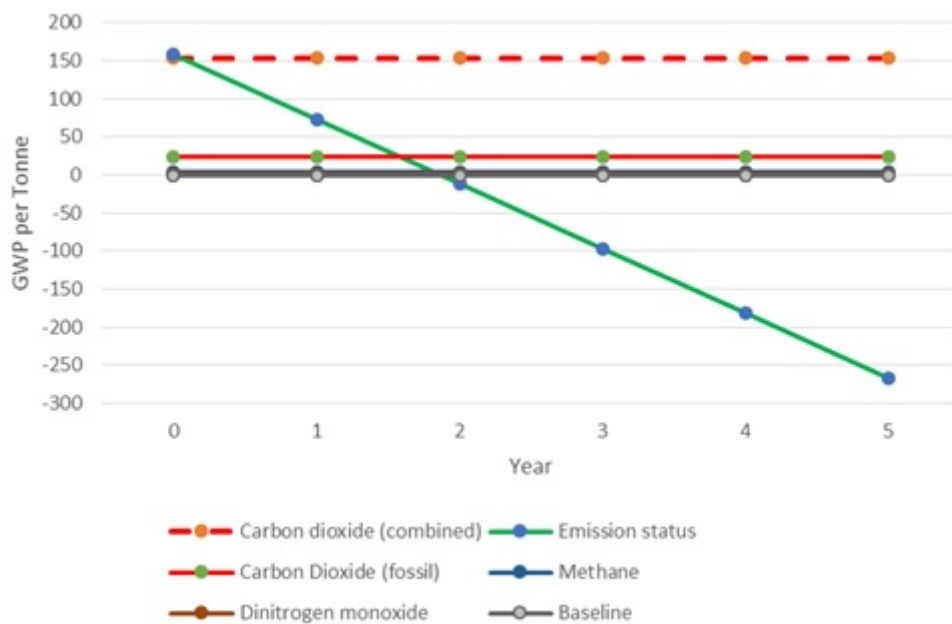Figure 10: GHG contribution to the extrapolation of a SUCCESS trial plot to 1 hectare

Figure 11: Embedded GWP and captured carbon over time

4.3 Discussion and Interpretation

From the results, it appears that the lifecycle scenario is highly sensitive towards the placement of the suppliers and the actual location of the carbon capture site. This is due to the high levels of emissions arising from transport of the material to the site. In order to achieve 85t carbon sequestration per year, it is necessary for $CaCO_3$ production in the soil to be approx. 39% present within the soil based on previous studies in Washbourne et al (2015) . However, using a dolerite material as opposed to crushed concrete effectively allows CO_2 absorption based upon past studies of using this material (Manning et al., 2013). It appears the material dolerite has a very high potential to capture carbon, particularly when the material is crushed and processed into fines.

In order to keep embedded CO_2 emissions to a minimum the placement of the site with regard to the suppliers should be at its shortest distance (no longer than 200 km each way), otherwise the duration of the plots to become CO_2 negative in a reasonable time will increase drastically

The logistical chain should use environmentally friendly vehicles in order to reduce the payback rate of the embedded emissions within the lifecycle.

An additional carbon capture component should be installed in order to provide extra sequestration support in the form of vegetation.

Any additional mixing should be carried out on-site and thereby avoiding carbon intensive processes. This is due to industrial mixing processes being more carbon intensive due to the additional energy requirements as opposed to mobile loaders which require less power, and produce less emissions due to improvement in recent euro 5 emission standards.

5 Conclusions and Future Work

The paper has applied a lifecycle assessment to a trial plot as part of the SUCCESS projects primary carbon capture experiment. It was important to illustrate the embedded emissions burden during the initial construction to determine the level of offset required for the plot lifecycle to become carbon negative.

Future work includes the continued monitoring of the SUCCESS carbon plot trials over an 18 month period in order to determine the actual rate of sequestration and the capacity of the material. However, what is unknown is the point where the soil becomes saturated with calcium carbonate such that CO_2 absorption ceases. This is important, as when this occurs intervention is necessary though either ploughing the trial or replacing the calcite-saturated material with minimal $CaCO_3$ so that the process can begin again. Work is ongoing in also adapting the carbon capture trial plots to a possible highway based setting where plot trials are implemented on road-side verges. Finally, a more in-depth study on carbon fluxes, in particular the role of microbe decomposition and respiration will be explored in order to fully capture the carbon cycle.

Acknowledgments

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