



# Carbonate precipitation in artificial soils produced from basaltic quarry fines and composts: An opportunity for passive carbon sequestration<sup>☆</sup>



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## ABSTRACT

The proportions of different carbon pools within artificial soils prepared by blending composts with dolerite and basalt quarry fines has changed over a period of 7 years, accumulating inorganic carbon as carbonate minerals newly formed within the soils. With no artificial energy inputs following construction, this is regarded as a passive mineral carbonation process. Carbon isotope data show that up to 40% of the carbon within the soil carbonate is derived from photosynthesis, mixed with carbon from geological sources (limestone present in the quarry fines). Organic matter within the soils shows very variable composition, with an apparent increase with time in the relative proportion of labile carbon relative to more stable forms, reflecting a change in the soil organic matter composition associated with the establishment of new plant communities. The rate of accumulation of inorganic carbon as carbonate minerals is estimated to be equivalent to  $4.8 \text{ t C ha}^{-1}$  annually to a depth of 0.3 m, consistent with rates observed for accumulations of carbonate carbon in urban soils containing demolition wastes (annually  $3.0 \text{ t C ha}^{-1}$  to 0.3 m). There appears to be substantial potential for artificial soils to be designed expressly for the purpose of carbon capture. The process is analogous to the use of reed beds for the removal of pollutants from contaminated waters.

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## 1. Introduction

There is little doubt that atmospheric concentrations of  $\text{CO}_2$  and other greenhouse gases have increased as a consequence of human activity, possibly dating back to the dawn of agriculture (Kutzbach et al., 2010) as well as through the comparatively recent combustion of fossil fuels since the industrial revolution (Denman et al., 2007). It is widely accepted that the increase in atmospheric greenhouse gas concentrations is responsible for a change in climate; the geological record suggests that our present interglacial period has extended for longer than previous interglacials (Kutzbach et al., 2010), and global temperatures overall are rising with implications for sea level and the frequency of extreme weather events (Denman et al., 2007).

In the context of climate change mitigation, there is a need to improve our understanding of natural processes that remove atmospheric  $\text{CO}_2$  in ways that begin to compensate for artificial emissions. Work of this type provides a basis for decisions to be made that enable sustainable carbon dioxide removal systems to be trialled, and possibly then implemented. In this context, the 'excess'  $\text{CO}_2$  that needs to be removed in order to compensate for anthropogenic emissions and so maintain atmospheric levels at a target of 500 ppmv is of the order of  $8 \text{ Gt C y}^{-1}$  by 2050 (Pacala and Socolow, 2004). These authors envisage that no single technique or approach will achieve this, and suggest that individually some approaches may potentially remove a 'wedge' of  $1 \text{ Gt C y}^{-1}$ .

Near complete elimination of anthropogenic greenhouse gas emissions is the only long-term sustainable solution. However, the intrinsic relationship between fossil fuel energy and developed economies, and the slow progress that has been made to decouple this dependency, suggests that atmospheric  $\text{CO}_2$  concentration will continue to rise, beyond a current value of approximately 400 ppmv. Even if emissions were drastically reduced to zero today, it would take hundreds of years before atmospheric  $\text{CO}_2$  would stabilize back to pre-industrial concentrations (Lowe et al., 2009). In

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these circumstances it is prudent to develop mitigation technologies that remove CO<sub>2</sub> from the atmosphere.

It is well known that the coupled plant–soil system plays a fundamental role in the global carbon cycle (e.g. Lal, 2004). Although its size as a reservoir is small compared with the ocean, the coupled plant–soil system contains 2261 Gt C (1990s figures; Denman et al., 2007), of which approximately 600 Gt C is in plants and 1600 Gt C in soil. This is well in excess of the atmospheric reservoir (762 Gt C; Denman et al., 2007). Most importantly, the fluxes of carbon to and from the coupled plant–soil system are greater than those between the atmosphere and the ocean (approximately 150 Gt C y<sup>-1</sup>). An amount equivalent to all atmospheric CO<sub>2</sub> will pass through the coupled plant–soil system in 7 years. In these circumstances, and given the recent geological record of agricultural influence on atmospheric CO<sub>2</sub> concentrations (Kutzbach et al., 2010), it seems self evident that any influence that humans may wish to have should be based on ways in which we can intercept and preserve in soils carbon that has been removed from the atmosphere by photosynthesis.

Many studies have investigated the application of organic matter to soils, as composts, manures etc. (Diacono and Montemurro, 2010). It takes many years to build soil carbon stocks using this approach. For example, Triberti et al. (2008) report annual increases in soil organic carbon of 0.16–0.26 t C ha<sup>-1</sup> over a 30 year period in a cropped soil system. It is equally well known that soil organic matter turns over at rates that vary for different SOM pools (Jones et al., 2005; Smith, 2004, 2008 and references therein). Large-scale studies have shown that, overall, as much as 90% of carbon input as compost is lost from a soil system within 10 years (Lin et al., 1997). Overall, where data are available, loss of organic matter from soil over time is potentially a serious problem (Smith et al., 2005). Restoration of organic carbon concentrations in modified soils (agriculture and urban environments) has the potential to be an important tool for mitigating anthropogenic climate change (Lal, 2003), as long as organic inputs are maintained to compensate for turnover. Organic carbon in soils is controlled largely by land management practices, and these may change depending on a number of social and economic drivers (e.g. feeding the increasing global population).

The formation of pedogenic carbonates offers a sink for plant-derived carbon that is effectively permanent from a human perspective (Manning, 2008; Pustovoytov, 2003). Globally, the amount of inorganic carbon held within soil carbonates is estimated to be between 720 Gt (Batjes, 1996) and 950 Gt (Schlesinger, 1985). Pedogenic carbonates readily form in specific environments with appropriate conditions that relate (for example) to depth and rainfall as well as the availability of calcium (Jenny, 1941). In general, the occurrence of pedogenic carbonates may have been underestimated, partly because of the scale of the task of mapping soils to greater depths than is conventional (typically 1 m), and partly because of the need for rigorous chemical or mineralogical determination of carbonate and its carbon and oxygen isotopic composition (Cerling, 1984).

We have recently shown (Renforth et al., 2009; Washbourne et al., 2012; Manning and Renforth, 2013) that pedogenic carbonate formation commonly occurs within urban soils that carry a burden of fine-grained mineral matter derived ultimately from concrete and Portland cement (in the UK, this type of soil is sometimes known as 'made ground'). Total inorganic carbon (TIC) contents of such soils may achieve 300 t C ha<sup>-1</sup> to a depth of 3 m, compared with 175 t C ha<sup>-1</sup> to a depth of 1 m for TOC within agricultural soils (e.g. Smith et al., 2005), the majority of which is in the top 0.3 m. What is more, urban soils accumulate TIC very rapidly (i.e. within 5–10 years) following demolition or landscaping, after which restoration may involve initial seeding of an installed top-soil followed by opportunistic revegetation. Washbourne et al.

(2012) have shown that the 10 ha Science Central site in Newcastle removed 36,000 t CO<sub>2</sub> from the atmosphere in 3 years following demolition. This is equivalent to 100 t CO<sub>2</sub> ha<sup>-1</sup> monthly, for a site with no restoration following demolition.

A key control on the formation of pedogenic carbonates in urban soils is the availability of calcium. Arisings of artificial calcium silicates with the potential to undergo carbonation are estimated by Renforth et al. (2011) to have an annual global sequestration potential equivalent to 190–332 million t C (a sixth to a third of a wedge; Pacala and Socolow, 2004) which would have the potential to mitigate 4–7% of emissions over the next 100 years (Allen et al., 2009).

In the context of considering ways in which soils can be managed to enhance the accumulation of inorganic carbon derived ultimately from the atmosphere, this paper describes artificial plots that have developed significant pedogenic carbonate within a blend of compost and finely ground basic igneous rock. It is important to note that the plots were originally constructed with other purposes in mind, as part of a semi-commercial study designed to assess the potential for blending quarry fines (a by-product of primary aggregate production) with composts in the formulation of artificial soils. Unexpectedly, the experimental plots have provided evidence of rates and mechanisms of carbon mineralization through reaction between compost and the calcium silicate minerals present within the quarry fines, and because of the importance of these observations they are reported here. The work presented in this paper augments reports of carbonation of synthetic calcium silicate minerals (Renforth and Manning, 2011), and is particularly significant because the carbonation of natural calcium silicate minerals through 'enhanced weathering' within soils could, by virtue of their widespread natural occurrence, be much more significant (Schuiling and Krijgsman, 2006; Hartmann and Kempe, 2008; Hartmann et al., 2013; Köhler et al., 2010; Renforth, 2012).

## 2. Rock dust-compost experimental plots

In 2003, Mineral Solutions Ltd ([www.mineralsolutions.co.uk](http://www.mineralsolutions.co.uk)), with support from the Minerals Industry Research Organisation (MIRO; [www.miro.co.uk](http://www.miro.co.uk)), investigated the development of artificial soils for land restoration purposes. The investigation focused on the use of quarry fines (generally <0.5 mm), which arise as a normal by-product of crushing of hard rocks mined for construction aggregates. Mineral Solutions Ltd. carried out pot trials, lysimeter trails and field trials, reporting in the public domain through MIRO and the company's website (Mineral Solutions Ltd., 2004).

At Barrasford Quarry (Lafarge Tarmac Ltd., Tyne Valley, England; Ordnance Survey grid reference NY914745), five trial plots were prepared with two replicates of each treatment and a single control (Fig. 1). These involved use of two quarry fines (Kelso sub-suite basalt from Craighouse quarry, Lafarge Tarmac Ltd., Melrose, Scotland; NT 600 362; dolerite from Barrasford quarry), and two composts (commercially produced composted food industry [FI] waste and material from an anaerobic digester that had subsequently been composted [AD]). Compositional and textural information for the quarry fines and compost is summarized in Tables 1–3.

The purpose of the field trial at Barrasford was to investigate, on a pilot basis, the suitability of blends of composts and quarry fines as an artificial growing medium suitable for land restoration and other purposes. Carbon capture was not part of the design. However, work carried out on the plots since their establishment in the winter of 2003 has shown, unexpectedly, that they have gained and accumulated soil carbonate minerals. This paper takes the opportunity to describe and evaluate the carbon-capture benefits of the Barrasford plots, so that the experience thus gained can be used in future work expressly intended to investigate carbonation of basic rocks as a carbon dioxide removal mechanism.

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