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Recycling Paper to Recarbonise Soil

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Abstract

Soil organic carbon can be increased through sympathetic land management and/or directly by incorporating carbon rich amendments. Herein, a field experiment amended paper crumble (PC) to soil at a normal deployment rate of 50 t ha^{-1}, and at higher rates up to 200 t ha^{-1}. The nominal 50 t ha^{-1} PC amendment resulted a mean increase in soil carbon of 12.5 g kg^{-1}. Using a modified ROTH-C carbon fate model, the long-term (50 years) carbon storage potential of a 50 t ha^{-1} PC amendment was determined to be 0.36 t_C ha^{-1}. Modelling a rotational (4 yearly) 50 t ha^{-1} PC amendment indicated 6.65 t_C ha^{-1} uplift would accrue after 50 years. Contextualised for the average farm in the East of England (~120 ha, with 79% as arable), PC derived increases in SOC would be equivalent to 2310 t CO_2e. These results support the use of PC to deliver significant levels of soil recarbonisation. Beyond carbon, PC was observed to influence other soil properties. Benefits observed included, decreased bulk density, increased water holding capacity, and increased cation exchange capacity. While PC amendment did not significantly increase wheat (*Triticum aestivum*) crop yield, manifold benefits in terms of increased SOC, long-term carbon storage potential, and improved soil quality sustains PC as a beneficial soil conditioner.

1. *Introduction*

More than 200 million hectares of agricultural land worldwide have been acknowledged as dangerously degraded, where soil carbon stocks are reduced by >50% (Lal, 2001). Due to the shaping influence on soil physical, chemical, hydrological, and biological properties, soil carbon, or more broadly soil organic matter (SOM), constitutes one of the most important factors underpinning soil health, and by extension the maintenance/delivery of soil ecosystem services (Abiven et al., 2009; Bhogal et al., 2009; Keenor et al., 2021; Power, 2010; Powlson et al., 2012). Society relies upon these essential ecosystem services for the provision of goods (food and resources), environmental regulation (water filtration and flood mitigation), and to support environmental functions (carbon cycling and sequestration) (Dominati et al., 2010; Keenor et al., 2021; Latawiec et al., 2020). Soils rich in organic matter are often regarded as having greater resilience to the environmental pressures, for example, drought and erosion (Bhogal et al., 2009; Powlson et al., 2012).

Soil organic carbon (SOC) degradation linked to agriculture, forestry and land use change has underpinned a considerable emission of carbon to the atmosphere, contributing significantly to climate change (Smith, 2008). An estimated 135 Pg C have been lost over the past 150 years with a further 36 Pg C of projected loss by 2050 (IPBES, 2018; Lal, 2018). Furthermore, soil derived emissions from agriculture account for approximately 24% of the global annual GHG emission (Smith et al., 2014). SOC reductions have been linked to decreased crop yields (Follett, 2001; Gomiero, 2016; Ivits et al., 2018; Kimetu et al., 2008), loss of soil biodiversity (Lal, 2001; Tsiafouli et al., 2015), altered soil hydrology and nutrient provision (Kimetu et al., 2008; Lal, 2006), soil erosion (Lal, 2019; Olson et al., 2016), and impairment of soil ecosystem services (Power, 2010). Consequently, SOC loss and soil degradation have significant implications for present and future food security, resource sustainability and essential ecosystem service provision (Follett, 2001; Gomiero, 2016; Lal, 2006; Lal, 2016; Power, 2010).

To combat these issues, interventions are urgently needed to restore SOC, and mitigate the negative effects of this legacy loss. By adopting soil-centric land management practices, the effects of soil degradation may be arrested; thus, better protecting existing soil carbon stocks and stimulating additional carbon sequestration (Latawiec et al., 2020). Interventions that enable soils to sequester rather than emit C, such as, less aggressive tillage, reduced agrochemical input, cover-crop rotations, and ‘feeding’ soil with C-rich amendments, provide opportunity to rejuvenate soils and capture carbon (Keenor et al., 2021; Lal, 2004; Powlson et al., 2012; Soussana et al., 2019).

Direct intervention methods, such as augmenting soil with C-rich amendments (manures (Lal, 2004), composts and paper waste (Chantigny et al., 1999; Powlson et al., 2012) and biochar (Lehmann et al., 2006; Smith, 2016), afford a means of increasing SOC stocks over a short timeframe and without the need for a radical shift in land management practice.

Paper crumble (PC), the focus of this research, is a co-product of the paper and cardboard recycling process. Comprised of wood pulp fibre, PC contains high levels of carbon (up to ~37% dry weight dependant on feedstock), 20-30% of which may be considered recalcitrant (Powlson et al., 2012; Zibilske et al., 2000).

To date, several papers have evaluated the benefits of PC as a soil amendment in a variety of agricultural and environmental contexts (Abiven et al., 2009; Chantigny et al., 1999; Chow et al., 2003; Ea, 2005; Foley and Cooperband, 2002; Gallardo et al., 2012; Powlson et al., 2012; Powlson et al., 2011; Rasa et al., 2021; Zibilske et al., 2000). Previous publications have reported beneficial effects on soil physical, chemical, and hydrological properties. PC has been observed to minimise surface water runoff and associated soil erosion (Foley and Cooperband, 2002; Powlson et al., 2012; Rasa et al., 2021; Zibilske et al., 2000), and substantially increase SOM/SOC content. However, this evidence is fragmented with respect to different soils and contrasting PC materials. In addition, the permanence of SOC uplift following PC amendment has, to date, not been reported.

To consolidate evidence regarding the potential for PC to increase soil carbon stocks and its wider influence upon soil properties (physical, chemical, hydrological) and crop yield, a field experiment was undertaken using applications of PC between 50 and 200 t ha^{-1}. Soils were assessed to establish soil organic matter (SOM) / total carbon (TC) contents. Subsequently thermogravimetric analysis (TGA) was used to profile PC-carbon stability. This data was then used to inform a modified ROTH-C carbon fate model to evaluate the long-term carbon storage potential of PC amended soil. In complement, the influence of PC on soil strength, bulk density, water holding capacity, pH, cation exchange capacity (CEC) and major/trace element concentrations were assessed. Finally, a laboratory batch-equilibration study was undertaken to explore potential interactions between PC and N-fertiliser. Given the size of the PC resource (e.g., in the UK, ~1Mt produced p.a) (CPI, 2014), this research sought to evidence the opportunity for this resource to re-carbonise soil and to improve soil quality.

2. *Materials and methods*

2.1 Field Experiment

The field experiment was established at Estuary Farm (King’s Lynn, Norfolk, UK; 52˚46'46.0"N 0˚24'08.8"E). Soil was of the Wallasea Series; a palo-alluvial gley soil, with stoneless A-horizon of silt clay texture (Hodge et al., 1984). Field measurements and samples were collected in 2019: soil physical data (January/May), hydrological data (January), chemical data (January) and crop data (August).

PC was provided by Palm Paper Ltd (King’s Lynn, Norfolk, UK). PC was applied to fields using a Bunnings Spreader and then incorporated to a depth of C. 5cm by culti-pressing and flat-lifting the soil (September 2018). PC was applied at rates of 50, 100, 150 and 200 t ha^{-1} to 36 x 400 m strips of the same field. Soils were drilled with winter wheat (*Triticum aestivum*) (September 2018). The field margin was used to benchmark outcomes in PC amended soil. The properties of the PC are provided in Table 1.

2.2 Soil sampling

Soil samples (0-20 cm; N=4) were obtained using a Dutch Auger, sieved *in-situ* (1 cm) and further sieved in the laboratory (2 mm). Soil samples were sealed and retained in cold storage (≤ 4 ˚C) prior to laboratory analysis.

2.3 Soil organic matter, C & N content and thermal analysis

SOM content was measured by loss on ignition (ISO, 1995). Briefly, soil (10 g; N = 4) was dried (74 ˚C for 16 h) and then combusted (470 ˚C for 36 h). For Total Carbon (TC) and Total Nitrogen (TN), milled dry soil samples (5 mg; N = 4) were packed in tin capsules (8 × 5 mm). TC and TN were measured using an elemental analyser (Exeter CHNS analyser).

The thermal stability of PC was assessed using a Thermo-gravimetric analyser (Mettler Toledo TGA/DSC 1). Samples (N=3) were heated in a nitrogen atmosphere, at a rate of between 10 to 20 ˚C min^{-1} from 25 to 1000 ˚C. To benchmark the PC samples, cellulose and lignin (obtained from Sigma-Aldrich) were assessed using the same method.

2.4 Carbon fate modelling

The Rothamsted Carbon (RothC) Model (Coleman and Jenkinson, 1996) is a widely used model for assessing the turnover of soil organic carbon (SOC). The recent versions of the model include four active carbon pools and one inert carbon pool (inert organic matter; IOM). The model divides incoming organic inputs into decomposable plant material (DPM) and resistant plant material (RPM), both decomposing to form microbial biomass (BIO), humified organic matter (HUM) and CO_2. The standard model considers plant residues and farmyard manure as organic carbon (OC) inputs and uses a pre-defined ration of DPM/RPM. In order to be suitable for PC amendment, the RothC model was modified and propagated using TGA/DTG assigned organic carbon fractions (Section 3.2).

Soil carbon modelling was performed in RStudio. Within the model initial soil carbon level was set to zero (thus, only PC carbon was considered). The model runs considered PC applied to soil under the following two scenarios: i) a single application of 50 t ha^{-1} in year 0; and ii) a 50 t ha^{-1} application every 4 years from year 0 to year 49. The input parameters used to inform the model are summarised in the supporting information (Table S1).

2.5 Soil physical attributes

Penetration resistance (Eijkelkamp Hand-Penetrometer; 13mm diameter 30˚ cone, 10mm diameter rod (N = 16)) and shear resistance (Pilcon 19mm soil shear vane (N = 24)), were measured *in situ* (January/May). Soil core samples (N = 4) were obtained using a Dent soil corer (core sleeves 7.5 cm height and 8.8 cm diameter) (January). Cores were oven dried (74 ˚C) and soil bulk density calculated (N = 4).

2.6 Soil hydrological attributes

Water holding capacity (WHC; N=4) was assessed by placing soil (~ 20 g) in a filter funnel (Whatman No.1 filter paper) and saturating the soil with distilled water. Samples were allowed to drain until gravity release of water had stopped. Moisture content of the soil was determined (drying at 74 ˚C for 16 h).

2.7 Soil chemical attributes

Soil pH (N = 4) was measured (ISO, 1994) in 1:10 soil/water suspension using a pH electrode (Mettler Toledo Pro pH) and pH meter (Mettler Toledo 5 Easy).

Cation Exchange Capacity (CEC) (N=4) was assessed by sodium acetate exchange method (USEPA, 1986). In brief, soil (5 g) was mixed with 1M sodium acetate (30 ml), shaken (16 h), centrifuged and the supernatant discarded (repeated two further times, with 1 h shake times). Thereafter, acetone (30 ml) was used to rinse the soil pellet (1 h shake, centrifuged and supernatant discarded; repeated three times). Finally, 1M ammonium acetate (30 ml) was added to the sample, followed by agitation (16 h) and centrifugation. The supernatant was decanted into a volumetric flask (100 ml) through a No.1 filter paper. This procedure was repeated two further times and the samples were made up to volume with ammonium acetate (1M). Sodium (Na) content was measured by ICP-AES (Varian Vista Pro CCD Simultaneous).

Major and trace elements concentrations (N=4) were measured following soil/PC extraction with 0.01 M $CaCl_2$ solution (Quevauviller, 1998). Samples (50g) were mixed with 0.01 M $CaCl_2$ solution (500 ml), shaken for 3h and allowed to settle (30 mins). Aliquots (50ml) of the extract were then centrifuged and filtered (0.45 µm). Major and trace element concentrations were measured using ICP-AES (see above).

2.8 *Nitrogen species interaction with soil and PC*

A fertiliser solution was prepared by dissolving 0.357g of ammonium nitrate in 1 L Milli-Q water (18.2 MΩ.cm). Fertiliser solution (40ml) was added to Wallasea soil or soil/PC mixtures (equivalent to a PC application of 50 t ha^{-1}) (4 g, N =4). The fertiliser addition represented 200 kg N ha^{-1} (i.e. assumes a mixing depth of 1.5 cm and soil bulk density of 1.03 g cm^{-3}). The samples were shaken (18h), centrifuged and filtered (0.45 µm). In parallel, Milli-Q water (40 ml) was added to soil or soil/PC mixtures (4g, N = 4) and the same process was repeated. Ammonium and nitrate concentrations in the resultant solutions were measured using a Skalar San++ Flow Analyser.

2.9 *Crop Sampling*

Seed heads (quadrat, 0.25m^2; N=4) were collected prior to harvest in August. Samples were dried (74 ˚C for 24 h) and 100 undamaged seed heads separated and threshed. Total yield was then calculated using threshed sample mass and an average number of plants per m^2 of 460 per m^2 (Wheat Growth Guide, 2018)); and scaled to t ha^{-1}.

2.10 Statistical analysis

On-way analysis of variance (One-way ANOVA) was used to test the variable addition of PC on soil carbon, physical, hydrological and chemical attributes, nitrogen species interaction and crop yields in field margin and PC amended soils. Significance level was set to 95% ($P < 0.05$) and determined by a *post hoc* test with Tukey's HSD comparison. This procedure was completed using IBM SPSS 25. Statistical analysis results are displayed in bar charts along with mean values and standard deviation (SD).

3. Results and discussion

3.1 SOM, TC, TN and C:N ratio

SOM (measured as LOI) increased significantly ($P < 0.05$) in all PC treatments ≥ 100 t ha^{-1}, relative to the field margin soil (Figure 1A). SOM in the field margin soil was 7.5%, while in the 50, 100, 150 and 200 t ha^{-1} treatments, SOM contents were 10.5%, 12.8%, 15.4%, and 14.7%, respectively (Figure 1A). The maximum increase in SOM, observed in the 150 t ha^{-1} treatment, was 2.1-fold higher than the field margin benchmark; with 150 and 200 t ha^{-1} treatments showing no significant difference ($P > 0.05$) to each other (Figure 1A).

Considering TC (measured by elemental analysis), a similar trend was observed to that of the SOM (Figure 1B). All PC treatments increased TC content, with these increases being significant ($P < 0.05$) in treatment ≥ 100 t ha^{-1} (Figure 1B). TC in the field margin soil was 5.0%, while in the 50, 100, 150 and 200 t ha^{-1} treatments, TC contents were 7.4%, 8.4%, 9.9%, and 9.7%, respectively (Figure 1B). As was the case with SOM, TC increases plateaued (with no further significant increase ($P > 0.05$)) at PC amendment levels of 150 and 200 t ha^{-1} (Figure 1A&B). Increases in TC for each 50 t ha^{-1} increment up to this plateau were 1.25 % (i.e. 12.5 g_C kg^{-1}$_{soil}$). Given that the PC used in this investigation had a carbon content of 244 g kg^{-1} (Table 1) an amendment of 50 t ha^{-1} could theoretically deliver 1.22×10^7 g_C ha^{-1}. Assuming incorporation of PC to a depth of 5 cm and a soil bulk density of 0.98 g cm^{-3} (calculated for the Wallasea series soil using empirical-pedogenic method (see section S3)) the predicted uplift in TC per 50 t PC applied to 1 ha would be 14.9 g_C kg^{-1}. Thus, the observed TC uplift was in keeping with the expected outcome. Small discrepancies (noted for applications up to 150 t ha^{-1}) may be attributed to some decomposition of the PC over the intervening 5-month period between incorporation and sampling. In the high amount (200 t ha^{-1}) treatment, the observed plateau in SOM/TC (and divergence for the expected uplift), suggests incomplete incorporation of the amendment with subsequent loss by wind action.

TC contents were converted to C stocks per unit area (assuming an incorporation depth of 5 cm and soil bulk density of 0.98 g cm^{-3}). C uplifts, (above the C stock in field margin (24.5 t C ha^{-1})), were: +11.8 t C ha^{-1} (50 t ha^{-1} PC treatment), +16.7 t C ha^{-1} (100 t ha^{-1} PC treatment), +24.0 t C ha^{-1} (150 t ha^{-1} PC treatment) and +23.0 t C ha^{-1} (200 t ha^{-1} PC treatment).

Total nitrogen (TN) content followed a similar trend to TC (Figure 1C). Like TC, TN was observed to increase in all PC amendment treatments, with 100, 150 and 200 t ha^{-1} treatments reaching a plateau where no significant difference ($P > 0.05$) between these treatments was observed (Figure 1C). TN in the field margin was 0.31%, increasing to 0.40% (in the 50 t ha^{-1} PC treatment) and to a maximum of 0.47-0.48% (in the 100 to 200 t ha^{-1} PC treatments) (Figure 1C).

The C:N ratio was higher in all PC treated soils when compared to the field margin (Figure 1D). However, increases were only significant ($P < 0.05$) in treatments ≥150 t ha^{-1} (Figure 1D). C:N in the field margin was 16:1, increasing to a maximum value of 21:1 in the 150 t ha^{-1} treatment. C:N in other PC products previously studied have been reported to range between >70:1 to <20:1 (Foley and Cooperband, 2002), the C:N (45:1) of PC was central within this range.

3.2 TGA carbon stability profiling and long-term carbon storage

Cellulose and lignin were used to benchmark the TGA profiles. These components of biomass represent relatively degradable and recalcitrant carbon, respectively (Mckendry, 2002). Cellulose is an unbranched glucose polymer, while lignin is highly branched phenolic polymer; as such these different chemical structures influence their relative stability (Lu et al., 2017; Yang et al., 2007). Thermal stability of materials has been related to the biodegradation of different organic matter or organic carbon pools to determine labile carbon (such as cellulose-C) that quickly degrades and stable carbon (such as lignin-C) that decomposes slowly (Capel et al., 2006; Plante et al., 2005).

By benchmarking temperature zones for attrition of cellulose and lignin, carbon fractions in PC were distinguished in terms of this relative stability. Following initial moisture loss from 25 to 125 ˚C (Raveendran et al., 1996; Yang et al., 2007), the cellulose sample remained stable until a temperature of 210 ˚C was reached. Thereafter rapid attrition of cellulose was observed between 210 – 400 ˚C (Figure 2). C attrition in this temperature range was ~81.8%. Above a temperature of 400 ˚C but below 600 ˚C, a smaller amount of cellulose residue (~13%) was pyrolyzed. All cellulose was pyrolyzed by a temperature of 600 ˚C. Attrition of lignin was protracted over a wider temperature range (220 – 750 ˚C) (Figure 2). As observed with the cellulose sample, initial moisture loss from the lignin occurred between 25 and 125

˚C, thereafter the sample remained stable until a temperature of 220 ˚C was reached. Between 220 - 450 ˚C steady attrition of lignin was observed, with the maximum mass loss rate (0.29 %/˚C) observed at ~390 ˚C. Thereafter mass loss accelerated slightly between 450 and 750 ˚C, with the maximum mass loss rate (0.25 %/˚C) observed at ~530 ˚C (Figure 2). All lignin was pyrolyzed by a temperature of 750 ˚C. These results conform with previous studies (Rao and Sharma, 1998; Yang et al., 2007; Yaras et al., 2021) and confirm greater thermal stability for lignin over cellulose.

TGA profiling of PC, revealed three phases (Figure 2). The first phase occurred between 25 – 125 ˚C. This phase was assigned to moisture evaporation (Mendez et al., 2009; Yaras et al., 2021). The second phase occurred between 125 – 700˚C. This second phase was assigned to pyrolysis of organic matter (Mendez et al., 2011). The final phase between 700 - 820˚C was assigned to attrition of inorganic carbonates (Marouani et al., 2019; Mendez et al., 2009). Benchmarking the PC TGA profile against profiles for cellulose and lignin, the organic matter attrition between 150 – 375˚C was assigned to less stable (*labile*) components, while organic matter attrition between 375 – 700 ˚C was assigned to components of greater recalcitrance (*resistant*). Resulting TGA profiles from this investigation conform with the previous findings of TGA profiles for paper mill wastes and de-inking paper sludge (Mendez et al., 2011; Mendez et al., 2009).

PC had a moisture content of 40% and TC content of 24.4% (Table 1). Of this TC, 42.5% was associated with the OC fraction (125-700 ºC) and 57.5% with the inorganic carbon fraction (700-1000 ºC) (Figure 2). The amounts of labile and resistant carbon were evaluated to be 28.3 kg ton^{-1} and 33.9 kg ton^{-1} (on a bulk weight basis) (Figure 3).

Using these quotients of relatively degradable and relatively recalcitrant carbon, the ROTH-C carbon fate model (Coleman and Jenkinson, 1996) was used to predict long term carbon storage for PC amended to the Wallasea soil series of the field trial (input parameters are provided in Table S1).

The RothC model was modified to predict the carbon storage potential of high carbon soil amendments in previous studies. These modifications introduce additional exogenous organic matter pools, thus allowing the model to accept a range of exogenous organic inputs (e.g. compost, argo-industrial waste and digestate) (Mondini et al., 2017; Peltre et al., 2012; Yokozawa et al., 2010) . Laboratory experiments were conducted to define the size and decomposition rates of addition entry pools in the additional exogenous organic matter pools (Mondini et al., 2017). In this research, the RothC model was refined to take entry OC pools ascribed by the TGA/DSC profiling as either labile or resistant fractions. The RothC model was subsequently run to provide a prognosis on the longevity of carbon storage under PC amendment scenarios.

Two scenarios were modelled. Scenario 1 considered a single application of 50 t ha^{-1} PC in year 0 and the fate of its carbon over the following 50 years. This scenario established short- medium- and long-term organic carbon uplifts (at 10, 25 and 50 years) to be 0.82, 0. 48 and 0.36 t ha^{-1}, respectively (Table 2). The residual proportion of PC carbon at 50 years was 12% of the OC mass initially amended. Scenario 2 assumed PC deployment to soil at an application rate of 50 t ha^{-1} on a rotational basis (i.e. every 4 years). Using this scenario, the model established the short- medium- and long-term OC uplifts (at 10, 25 and 50 years) to be 2.85, 5.08 and 6.65 t ha^{-1}, respectively (Table 2).

3.3 Soil strength, penetration resistance, and shear resistance

Soil penetration and soil shear resistances were measured *in situ* in both January and May. Management practices that influence soil structure and aggregation facilitate root growth and penetration and air and water storage in soil pores this supporting crop success (Pagliai et al., 2004). Soils with high penetration/shear resistance (and soil bulk density (Section 3.4)) may limit water infiltration, reduce water availability inhibit the growth of plants due to compaction (Nawaz et al., 2013; Taylor and Brar, 1991).

In January, penetration resistance in the field margin was 2.2 MPa (Figure 4A). At this time penetration resistance in all of the PC amended soil treatments was significantly lower ($P < 0.05$); with values ranging from 1.0 to 1.4 MPa (Figure 4A). Similarly, soil shear resistance, in January, was much higher in the field margin (79.0 kPa) and significantly different ($P < 0.05$) to the values observed in all of the PC amended treatments; wherein soil shear resistance varied from 24.9 to 29.4 kPa (Figure 4C). No significant differences ($P > 0.05$) were observed between the different PC treatments (Figure 4C). It is likely that the substantial differences observed between the PC treatments and the FM at this stage in the season reflect the effects of soil tillage.

In May, ground conditions were much drier, and both penetration and shear resistance were higher (compared to January) (Figure 4A/B *vs.* 4C/D). A small (non-significant ($P > 0.05$)) increase in penetration resistance was observed in the 50 t ha^{-1} treatment (3.3 MPa) relative to the FM soil (3.0 MPa) (Figure 4B). Reductions (not significant ($P > 0.05$)) in penetration resistance were observed in treatments of ≥100 t ha^{-1} relative to the FM soil (Figure 4B). A stepwise decrease in soil shear resistance was observed with increased quantities of PC (Figure 4D). Significant decrease ($P < 0.05$) in shear resistance was observed between the FM soil (89.9 kPa) and the 200 t ha^{-1} PC treatment (67.6 kPa). No significant differences ($P > 0.05$) were observed between the PC treatments (Figure 4D).

Of the penetration resistances measured in January (Figure 4A), no soil was observed to exceed the soil compaction threshold for heavy texture soils (2.1-2.5 MPa), suggesting no impediment to propagation of plant roots (Mckenzie et al., 2002; Stirzaker et al., 1996). In contrast, all soils measured in May were found to meet or exceed this limit. It is suggested that this increased soil strength was linked to PC-aggregate binding along with seasonal soil settlement, consolidation, and aggregation (Chantigny et al., 1999; Rajaram and Erbach, 1999).

3.4 Soil structure, bulk density, and hydrology

Soil structure, porosity and hydrological properties are underpinned by a variety of interconnecting factors; soil texture, structure, organic matter content, biological activity, moisture content and land management practice (Bormann and Klaassen, 2008; Zibilske et al., 2000). Changes in any of these soil attributes thus exhibit commensurate changes in the physical and hydrological properties of soil. Enhancing and altering soil physical properties through direct SOM inputs, often leads to soil bulk density (SBD) reductions , in turn providing benefits to water infiltration and WHC (Franzluebbers, 2002). Increasing SOC/SOM contents is an effective method of enhancing soil hydrological properties, where WHC can be increased by 1 – 10 g per 1g SOM (Lal, 2006).

SBD was observed to decrease significantly ($P < 0.05$) in all PC treated soils relative to the field margin (Figure 5A). SBD in the field margin was 1.21 g cm^{-3} and decreased to range between 1.03 g cm^{-3} (50 t ha^{-1}) and 0.89 g cm^{-3} (150 t ha^{-1}) (Figure 5A). No significant differences ($P > 0.05$) were observed between different PC treatments (Figure 5A). The reductions in SBD in the PC treatments are likely due to the direct influence of the low-density of PC (0.39 g cm^{-3}) "diluting" the denser soil. Aligning SBD observations with soil strength observations (Section 3.3), supports proposed interaction mechanism, wherein lower amounts of PC amendment assisted soil particle cohesion (acting as a "glue") this promoting soil aggregation, enhancing soil porosity and decreasing SBD. In PC treatments exceeding 100 t ha^{-1} there were discrete PC zones (visually evident in soil samples), the presence of such zones (varying in diameter ~0.1 mm to ~10 mm) may have resulted in a trade-off between PC-facilitated soil-aggregate cohesion and pockets of low-density PC offsetting gains in soil strength but offering zones for enhanced water holding capacity (see below).

PC amendment to soil was observed to significantly increase ($P < 0.05$) soil WHC in all treatments relative to the field margin benchmark. WHC in the field margin was 38.6 % while WHC in the PC amended soils ranged between 48.6% and 57.3% (Figure 5B). No significant difference ($P > 0.05$) was observed between the different PC treatments (Figure 5B). Changes in soil WHC linked closely to SBD changes associated with PC addition, following the same overall stepwise decrease (from 50 to 150 t ha^{-1}) and plateau (at 150 and 200 t ha^{-1}) (Figure 5). Such changes may have arisen due to a combination of increased soil aggregation (*in low PC treatments*) and discrete PC zones (PC WHC of 131.4%; Table 1), these providing pockets where water could be adsorbed within the PC-soil matrix.

These results are consistent with previous publications linking enhanced soil aggregate stability and increased water holding capacity (Chantigny et al., 1999; Chow et al., 2003; Foley and Cooperband, 2002; Gallardo et al., 2012; Powlson et al., 2012; Zibilske et al., 2000).

3.5 pH and Cation Exchange Capacity

Soil pH and CEC are key to regulating chemical functions and thus have a direct effect upon soil fertility, biological activity, and productivity (some essential and trace elements are more or less available in different pH ranges) (Alam et al., 1999; Hazelton and Murphy, 2007; Kemmitt et al., 2006). Soil CEC provides a direct measure of the soils ability to absorb, hold and exchange cations within the soil matrix; these ions support crop growth, and may also assist in buffering soil pH (Mccauley et al., 2009; Méndez et al., 2015).

PC had a circumneutral pH (6.94) (Table 1). Following PC amendment, soil pH generally decreased comparing to the field margin (8.63) but no significant changes ($P > 0.05$) were observed, with all soils remaining alkaline in a range between 8.53 – 8.56 (Figure 6A). Previous studies have reported contrasting changes to soil pH following PC amendment; these contrasting outcomes, in part, are likely related to different pH values for the PC materials (dependant on the process and initial feedstock). Alkaline PC amendments (pH ~8.0) have been reported to increased soil pH (Chantigny et al., 1999; Ea, 2005; Rasa et al., 2021), while circumneutral PC amendments (~7.0) (Gallardo et al., 2012), and acidic PC amendments (~6.7), have been reported to slightly decrease soil pH (Foley and Cooperband, 2002; Méndez et al., 2015). The results reported here (for circumneutral PC), are consistent with these previous reports for similar pH value PC products. Thus, circumneutral PC has the potential to beneficially reduce the soil pH in alkaline soil; however, long term experiments would be needed to support.

PC addition increased the CEC of all PC amended soils relative to the field margin (86 me/100g); with increase in CEC being significant ($P < 0.05$) in the 150 and 200 t ha^{-1} treatments (Figure 6B). CEC in PC amended soil ranged from 90 me/100g (50t ha^{-1} treatment) to 103 me/100g (150 t ha^{-1} treatment). Significant differences ($P < 0.05$) in CEC were observed in the 50 and 150 t ha^{-1} treatments while no significant differences ($P > 0.05$) were observed between other PC treatments (Figure 6B). Relative to the field margin increases in CEC ranged between 1.0- and 1.2-fold. PC enhanced CEC was consistent with other studies that have reported increases in the CEC in PC amended soils (Fierro et al., 1999).

3.6 Elemental analysis

Soils should contain a variety of essential elements in sufficient concentrations to ensure effective uptake and use by plants in a variety of processes (Grusak, 2001). For an element to be considered essential it must be required for completion of the plant life cycle (i.e., underpinning a key metabolic process or function) and may not be entirely replaceable by another element (Grusak, 2001; Kirkby, 2012). 17 essential elements are required for plant growth, with 14 derived from the soil, including the major elements K, Mg P, and trace elements, B, Cu, Mo, Ni, Zn (those measured in this investigation) (Jones and Jacobsen, 2005; Mahler, 2004). Na is essential for some plants and being chemically similar to K it is beneficial when K is limited (Pilon-Smits et al., 2009). The major elements (macronutrients) are often observed in plants in concentrations >0.1% dry tissue weight, while trace element (micronutrients) concentrations are generally <0.025% (Grusak, 2001; Jones and Jacobsen, 2005). Alongside the measured major and trace elements, several non-essential and potentially toxic elements (Cr, Cd, Hg, Pb) were measured.

Of the essential major elements only Na and P were significantly influenced ($P < 0.05$) by PC amendment.

Stepwise increases in available Na were observed with successive PC amendment, ranging from 1.7 - 2.5-fold (Figure 7C). Available Na concentrations in the field margin was 19.7 mg kg^{-1}, while in the 50 t ha^{-1} and 200 t ha^{-1} treatments it was 34.0 mg kg^{-1} and 48.4 mg kg^{-1}, respectively. Due to the high concentration of Na in the PC (781 mg kg^{-1}) it is likely that the increases observed in the soils are a function of amendment incorporation. Low concentrations of Na in soil can improve the yield of cereal crops, however at high concentrations Na may exhibit plant toxicity (Kronzucker et al., 2013; Rawlins et al., 2012). Soil structure decline and soil permeability decrease occur when the Na concentration exceeds the critical level 5% (Clancy, 2009; Horneck et al., 2007). The highest Na level observed in PC amended soils was 0.005% (48.4 mg kg^{-1}; Figure 7C) and is unlikely to cause plant Na stress.

PC had an available P content of 5.61 mg kg^{-1} (Table 1), while the available P in PC treated soils ranged from 1.93 – 0.87 mg kg^{-1} (Figure 7D). A small (non-significant ($P > 0.05$)) increase in available P was observed in the 50 t ha^{-1} PC treatment (1.93 mg kg^{-1}; field margin

1.83 mg kg^{-1}). For all PC supplied available P this was not translated into increased available P in the treated soils (Figure 7D). Available P decreased in PC treatments ≥100 t ha^{-1}. This decrease was significant ($P < 0.05$) in the 200 t ha^{-1} treatment (0.88 mg kg^{-1}) (Figure 7D). Maximum P-availability occurs between pH of 5.5-7.5 (Fernández and Hoeft, 2009). Thus, available P delivered in the PC amendment may have been repartitioned due to soil pH being alkaline (8.5 – 8.7 in amended soils); this leading to the P-complexation by calcium ions (Fernández and Hoeft, 2009; Siddique and Robinson, 2003). P is essential to plant growth and plays an important role in energy transfer (Grusak, 2001; Jones and Jacobsen, 2005). While deficiency in P can lead to slow and stunted growth with yield losses (Shenoy and Kalagudi, 2005) in the present research no significant differences were observed in the crop yields (Section 3.8).

Of the essential trace elements only B and Mo were significantly influenced ($P < 0.05$) by PC amendment (Figure 8A&E).

Available B concentrations significantly increased ($P < 0.05$) in all PC treated soils relative to the field margin soil (353 µg kg^{-1}) and ranged between 582 – 731 µg kg^{-1}, a 1.6- to -2.1-fold increase (Figure 8A). Uplift in available B concentration was broadly equivalent across all treatments, with the exception of the 150 t ha^{-1} amendment rate where the largest available B concentration (731 µg kg^{-1}) was observed (Figure 8A). No significant differences ($P > 0.05$) were observed between PC treatments (Figure 8A). B is an essential micronutrient and plays a vital role in creation and maintenance of plant cell walls (Koshiba et al., 2009; Rerkasem and Jamjod, 2004). Deficiency of B is the most wide-spread and frequent micronutrient deficiency (Gupta, 1980; Koshiba et al., 2009). There is a risk of deficiency in B when the concentrations are lower than 150 – 500 µg kg^{-1} (Ahmad et al., 2012). Available B in PC treated soils were noted to be above this threshold (Figure 8A). Available Mo concentrations increased in all PC treatments relative to the field margin benchmark soil, however a significant increase ($P < 0.05$) was only observed in the 100 t ha^{-1} treatment (Figure 8E).

Non-essential elements were below the limit of detection and thus were not significantly influenced ($P > 0.05$) by PC amendment, suggesting the PC amendment does not represent a risk with respect to introducing potentially toxic elements to land.

3.7 PC Interactions with fertiliser N-species

Organic fertiliser can release nitrogen from the time it was applied to as much as several years after application but to be useful to plants nitrogen (N) must be present as either ammonium ion or nitrate ions (Hue and Silva, 2000). Available concentrations of ammonium in unfertilised soils and PC amended soils were below the limit of detection (Figure S1.A). Thus, no changes in ammonium availability were observed in the presence of PC. Low concentrations of nitrate were observed in the unfertilised control soils (22 mg kg^{-1}) and unfertilised PC amended soils (20 mg kg^{-1}) (Figure S1.B). No significant difference ($P > 0.05$) was observed where soil only and soil with PC were compared in unfertilised tests. Where soils were equilibrated with fertiliser solution, no significant difference ($P > 0.05$) in ammonium availability was observed between the control soils (409 mg kg^{-1}) and 50 t ha^{-1} PC treatments (444 mg kg^{-1}) (Figure S1.A). The application of PC (50 t ha^{-1}) significantly increased ($P < 0.05$) available nitrate (1.1-fold) in the fertilised treatments.

Previous reports have suggested PC to be a nitrogen deficient soil amendment with the potential to “lock-up” nitrogen (Fierro et al., 1999; Foley and Cooperband, 2002; Powlson et al., 2012). In contrast to these publications, the present results suggest, for this PC and the Wallasea soil, that PC amendment (50 t h^{-1}) was of no detriment to ammonium nor nitrate species added to soil as N-fertilisers.

3.8 Crop Yields

PC addition was found to have no significant ($P > 0.05$) effect upon the yield of wheat (Figure S2). Total grain yields varied from 5.69 t ha^{-1} (in the 150 t ha^{-1} treatment) to 6.28 t ha^{-1} (in the 100 t ha^{-1} treatment) (Figure S2). This outcome was likely underpinned by the use of agrochemicals throughout the crop cycle to optimise nutrients and suppress pests. These yields are notably low when contextualised, with average UK wheat yield 2000-2020 (Defra, 2020a), that range from 6.7 to 9.0 t ha^{-1}. It has been reported that optimal wheat grows occur where soil pH is between 6.0 and 7.0 (Vitosh, 1994). Thus, the lower yields observed may in part be attributable to the alkaline pH of the soil used in this experiment.

3.9 Soil carbon uplift, policy and carbon off-setting

Restoring soil C stocks within agricultural soils, has moved to the forefront of the climate policy agenda in recent years. Championed by programmes such as the ‘4p1000’ and UNFAO RECSOIL initiatives efforts to re-sequester C in soil are being integrated in national policy (Smith et al., 2020; Soussana et al., 2019; Unfao, 2020). For example, in the UK, the 2020 Agriculture Act (Defra, 2020a) that seeks to lever increases in soil carbon stocks through a new environmental land management scheme (ELMs). This scheme proposes the use of public money to pay for public goods, including carbon storage, biodiversity net-gains, flood mitigation and climate change adaptation (Defra, 2020b; Defra, 2020c; Klaar et al., 2020). Simultaneously, supporting soils to deliver greater C storage will assist in meeting societal obligations under the Paris Agreement, sustainable development goals and Net Zero aspirations (Latawiec et al., 2020; Ncc, 2020; Soussana et al., 2019).

At a global scale soil recarbonisation offers technical potential for re-sequestration of up to ~5Gt C yearly (Smith et al., 2020; Soussana et al., 2019; Unfao, 2020). It is salient to attaining short term goals that, management interventions such as no/minimum tillage, cover cropping, use of ground cover, land use change avoidance and increased use/application of organic amendments to soil are fully adopted to deliver recarbonisation at low cost and in short timeframes (Keenor et al., 2021; Lal, 2004; Powlson et al., 2012; Smith et al., 2020; Soussana et al., 2019; Unfao, 2020).

The research presented herein highlights the significant potential for soil amendments, such as PC, to align with soil recarbonisation aspirations. Herein, modelling the fate of PC carbon, based on its stability profile on a rotational basis (re-application ever 4 years) returned an accrue a total carbon uplift of 6.65 t_{OC} ha^{-1} over a 50-year period (Table 2). In the context of '4p1000' initiative, where an annual uplift of 4 g_{OC} kg^{-1} (0.4%) is aspired to, PC accrued carbon over 50 years would be ~200 g_{OC} kg^{-1}; using the same assumptions as those presented in section 3.1, this uplift would equate to 0.9 t_C ha^{-1}. Comparison 6.65 *vs* 0.9 t_C ha^{-1}, highlights the significant recarbonisation potential of PC amendment.

It is highlighted that the carbon stability profiling coupled to the modelling approach defined long-term stable C (stable to 50 years) associated with a 50t ha^{-1} PC amendment to be 0.36 t_C ha^{-1} (Table 2). This is a small portion (11.5%) of the total carbon (3.11 t_C) entrained in a 50t ha^{-1} PC amendment at time of application. It is highlighted that while this amount of carbon is small, the approach used to define it ensured the quantification of carbon stored with "permanence" (here 50 years). The evaluation of C storage "permanence" is fundamentally important to appraising soil recarbonisation strategies. There is little merit in claiming CO_2 removal from the atmosphere to soil if the prospect of long-term carbon storage is wanting.

Results presented indicate that an average farm, in the East of England of 120 ha (with 79 % as arable) (Defra, 2021), would achieve an OC uplift (associated with a 4-year rotational application of PC; 50 t ha^{-1}) equate to 630 t C (equivalent to 2310 t CO_2e). At time of writing the unit price of soil C has not been equilibrated in the formal carbon market, however, other carbon off-sets have been tested under market forces for many years (i.e., the EU emissions trading scheme (EU ETS) (Europeancomission, 2015)). At the time of writing the EU ETS market price for 1 t CO2e was EUR €95 (February 2022; having increased from EUR €40 per 1 t CO_2e the same time of the previous year) (EU Carbon Permits, 2022). Applying this carbon price of €95 per 1 t CO_2e to the calculated uplift of 2310 t CO_2e, the value of carbon sequestration could be €219,450 (equivalent to €46 ha^{-1} y^{-1}). Conflating the estimated PC resource in the UK ~1Mt (CPI, 2014) with its long-term stable carbon quotient (0.0072 t_C t_{PC}^{-1}), yields 7200 t_C; equivalent to 26,400 t CO_2e of permanent storage. At EUR €95 t CO_2e this long-term carbon storage could potentially leverage an off-set value of €2.5m p.a.

4. Conclusion

Protection and recovery of soil carbon stocks is of paramount importance to sustaining productive agriculture, improving food security, and more broadly to improve the delivery of ecosystem services (Adhikari and Hartemink, 2016; Doran, 2002; Latawiec et al., 2020; Power, 2010). PC, as detailed herein, has the potential to make a significant contribution to soil recarbonisation in terms of both uplift per unit area and, given the amount of PC resource available (e.g. in the UK ~1Mt (CPI, 2014)), at a meaningful scale. While PC was not observed to increase crop yield in this research (likely due to high levels of agricultural intervention) it may afford benefits to crops where soil fertility is lower and agricultural inputs more restricted. Results reported herein support such an premiss, in so much as they indicate significant benefits to soil nutrient concentrations, CEC, soil bulk density and WHC. Further research to explore the influence of PC on soil fertility in low management intensity systems, regenerative agriculture systems and across a range of soil types and geographies is recommended to broaden understanding of PC influence on soils.

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Tables

Table 1. PC properties including: OM, TC, TN, C:N, Water Holding Capacity, Bulk Density, Cation Exchange Capacity, pH, Essential and Non-essential Elements (N=4; mean ± std dev).

Table 2. Rothamsted carbon (RothC) modelling outputs: organic carbon uplift at 10, 25 and 50 years under two modelling scenarios (single application and 4 yearly application of 50 t ha^{-1} PC).

Figures

Figure 1. Soil organic matter (A), total carbon (B), total nitrogen (C), C:N ration (D) in field margin soil (FM) and PC treated soil (50, 100, 150 and 200 t ha^{-1}). Error bars represent SD of the mean (N=4). Bars that share a lower-case letter are not significantly different (P >0.05).

Figure 2. Thermogravimetric analysis (TGA) curves (A) and derivative thermogravimetric (DTG) curves (B) of paper crumble, cellulose, and lignin (N=3).

Figure 3. Paper crumble composition in terms of: moisture, non-carbon, inorganic-carbon, organic-carbon; and labile-carbon/resistant-carbon. Annotated values are % of C in undried “bulk” PC amendment.

Figure 4. Soil penetration, January (A), May (B) and soil shear January (C), May (D) in field margin soil (FM) and PC treated soil (50, 100, 150 and 200 t ha^{-1}). The top and bottom of the box indicates the upper and lower quartiles, the horizontal line indicates the median, the symbol (x) indicates the mean (N = 16 for penetration & N = 24 for shear-vane). Error bars represent SD of the mean. Bars that share a lower-case letter are not significantly different (P > 0.05).

Figure 5. Soil bulk density (A) and water holding capacity (B) in field margin soil (FM) and PC treated soil (50, 100, 150 and 200 t ha^{-1}). Error bars represent SD of the mean (N=4). Bars that share a lower-case letter are not significantly different (P > 0.05).

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Figure 8. Essential trace elements B (A), Zn (B), Cu (C), Ni (D) and Mo (E) in field margin soil (FM) and PC treated soil (50, 100, 150 and 200 t ha-1). Error bars represent SD of the mean (N=4). Bars that share a lower-case letter are not significantly different (P > 0.05).

Table 1. PC properties including: OM, TC, TN, C:N, Water Holding Capacity, Bulk Density, Cation Exchange Capacity, pH, Essential and Non-essential Elements (N=4; mean ± std dev).

Parameter	Unit	Value
OM[a]	$\%_{\text{dry mass}}$	29.9 ± 0.3
Total C	$\%_{\text{dry mass}}$	24.4 ± 1.6
Total N	$\%_{\text{dry mass}}$	0.55 ± 0.06
C:N	dimensionless	45:1
WHC[b]	%	131 ± 10.81
Bulk density	g cm^{-3}	0.39 ± 0.01
CEC[c]	me/100g	89.4 ± 2.7
pH	dimensionless	6.94 ± 0.04
Essential major elements		
K	$\text{mg kg}^{-1}{}_{\text{dry mass}}$	67.4 ± 3.8
Mg	$\text{mg kg}^{-1}{}_{\text{dry mass}}$	142 ± 3.9
Na	$\text{mg kg}^{-1}{}_{\text{dry mass}}$	781 ± 18
P	$\text{mg kg}^{-1}{}_{\text{dry mass}}$	5.61 ± 0.84
Essential trace elements		
B	$\text{mg kg}^{-1}{}_{\text{dry mass}}$	0.37 ± 0.01
Zn	$\mu\text{g kg}^{-1}{}_{\text{dry mass}}$	BDL
Cu	$\mu\text{g kg}^{-1}{}_{\text{dry mass}}$	0.26 ± 0.01
Ni	$\mu\text{g kg}^{-1}{}_{\text{dry mass}}$	0.11 ± 0.01
Mo	$\mu\text{g kg}^{-1}{}_{\text{dry mass}}$	0.14 ± 0.01
Non-essential elements		
Cr	$\mu\text{g kg}^{-1}{}_{\text{dry mass}}$	0.01 ± 0.002
Cd	$\mu\text{g kg}^{-1}{}_{\text{dry mass}}$	BDL
Hg	$\mu\text{g kg}^{-1}{}_{\text{dry mass}}$	BDL
Pb	$\mu\text{g kg}^{-1}{}_{\text{dry mass}}$	BDL

Note. In several instances available concentrations of elements were below the detection limit for the method; where this is the case values have been annotated “BDL”.

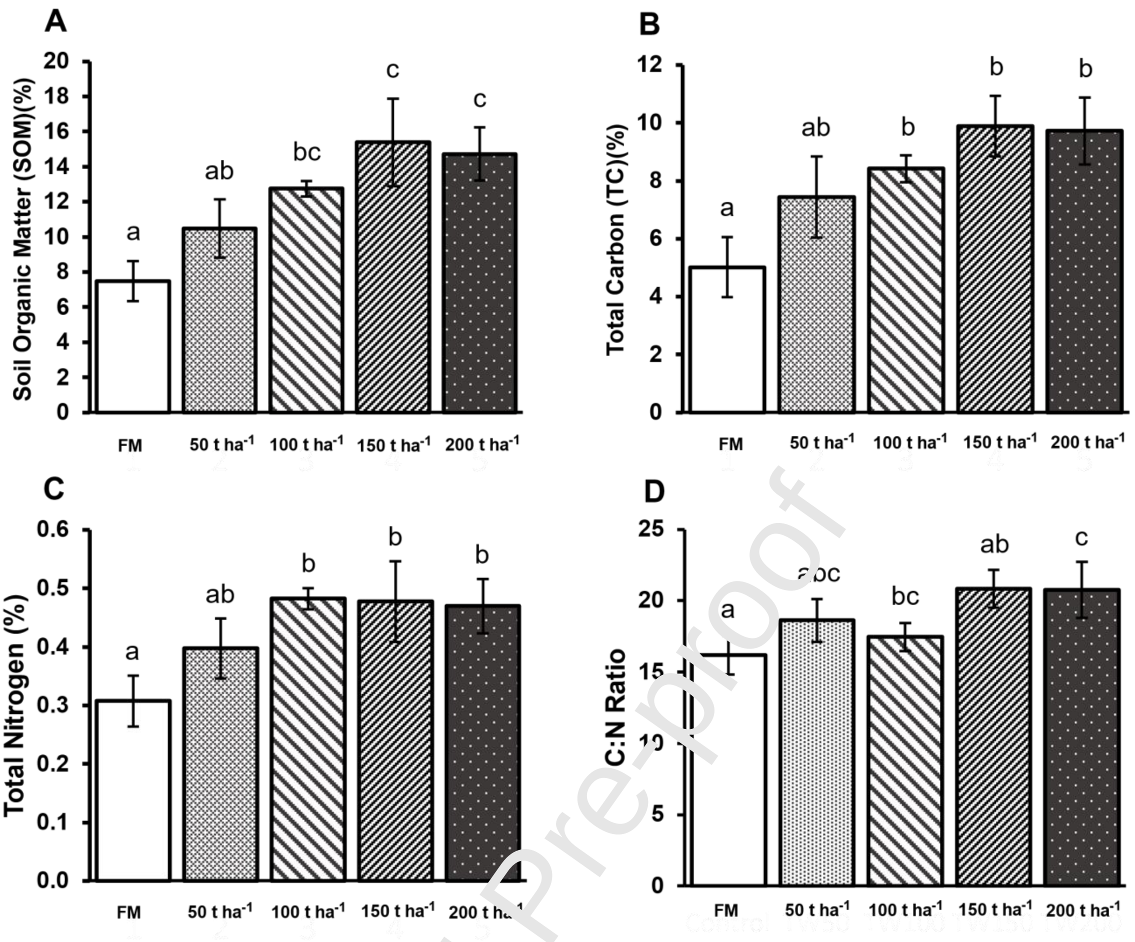
[a] OM: organic matter.

[b] WHC: water holding capacity.

[c] CEC: cation exchange capacity.

Table 2. Rothamsted carbon (RothC) modelling outputs: organic carbon uplift at 10, 25 and 50 years under two modelling scenarios (single application and 4 yearly application of 50 t ha^{-1} PC).

Scenario	Application rate	OC per 50 t ha^{-1} amendment	Application scheme	OC uplift		
				10 year-	25 year-	50 year-
	t ha^{-1}	t ha^{-1}		t ha^{-1}	t ha^{-1}	t ha^{-1}
Scenario 1	50	3.11	Single application	0.82	0.48	0.36
Scenario 2	50	3.11	Quadrennial application	2.85	5.08	6.65



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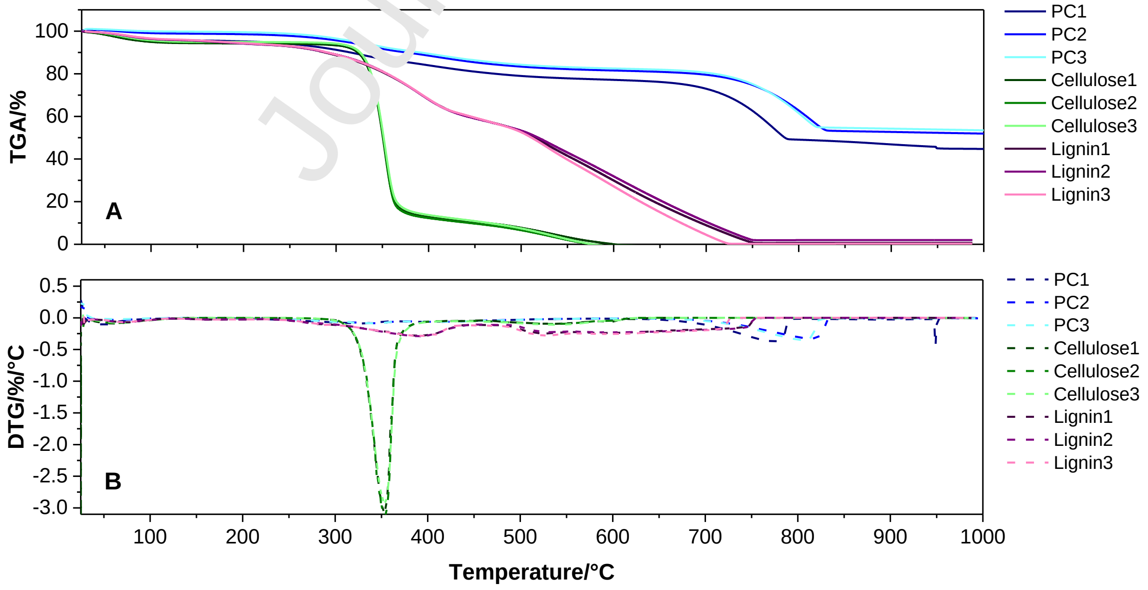

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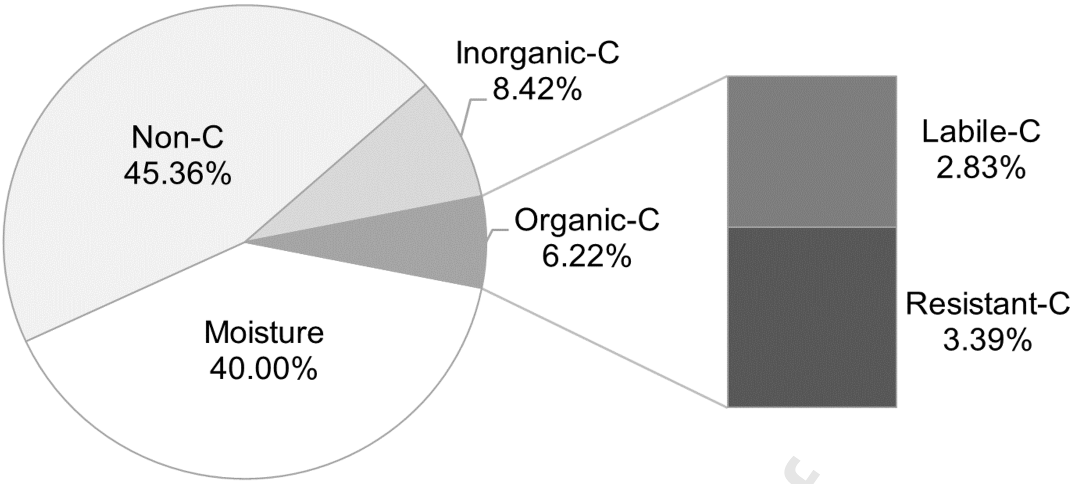

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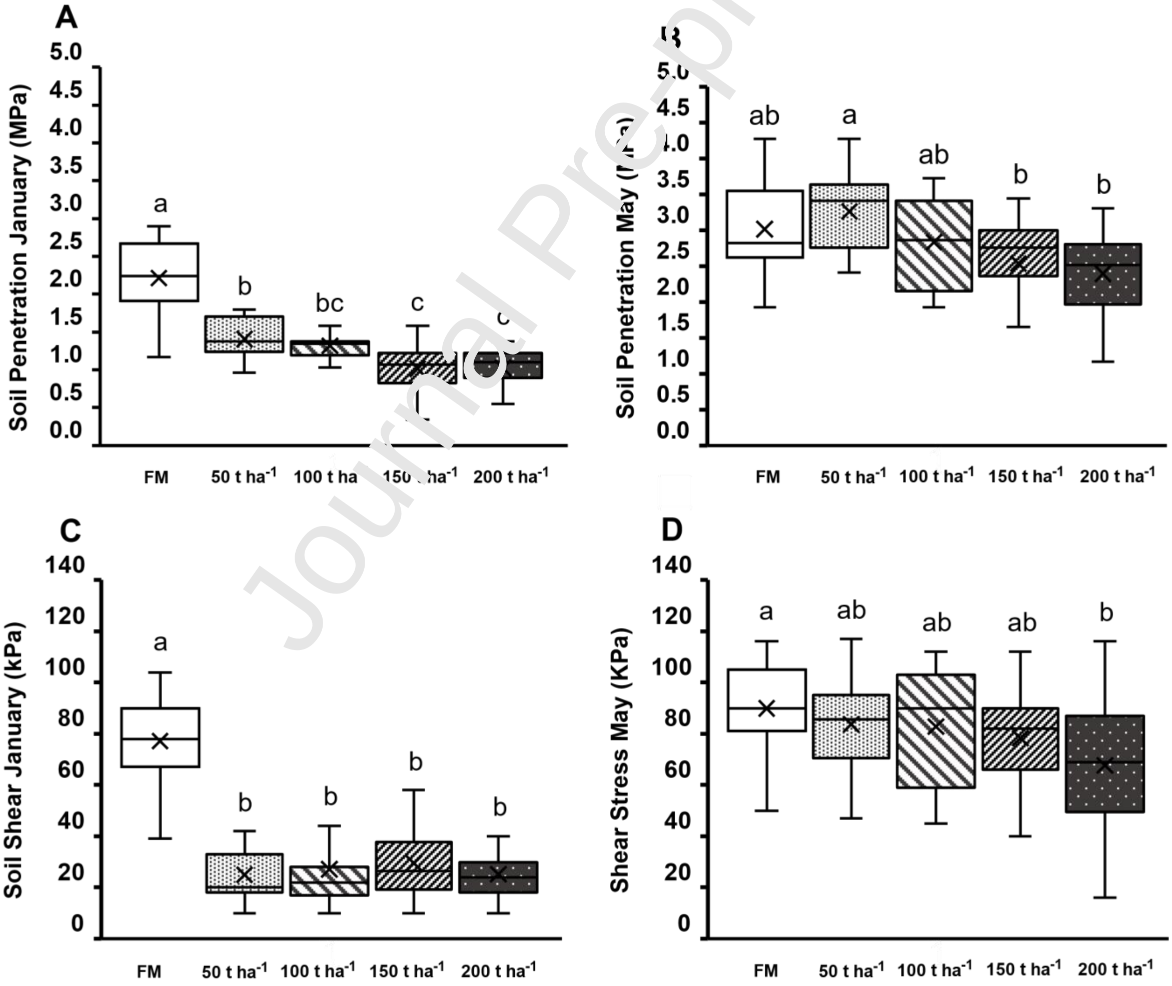

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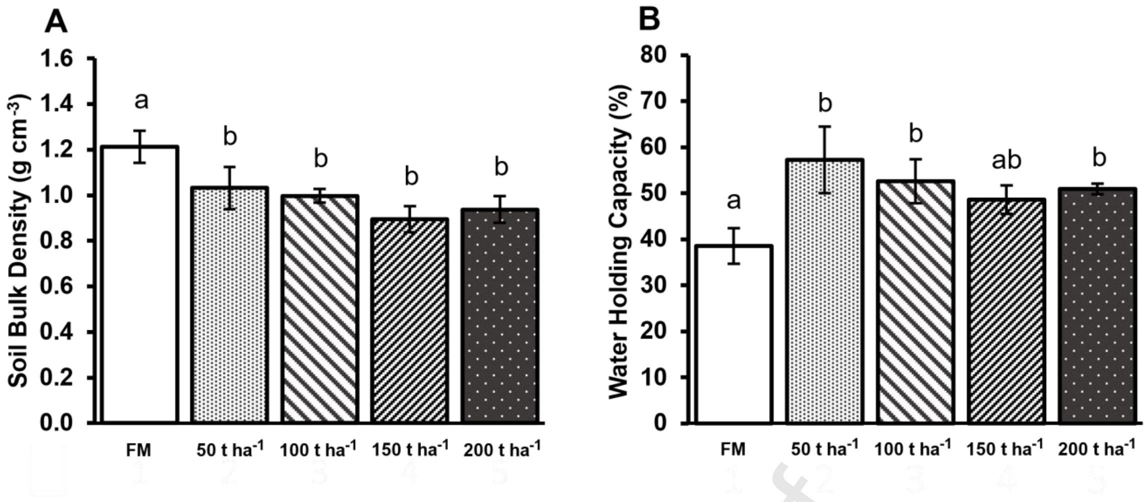

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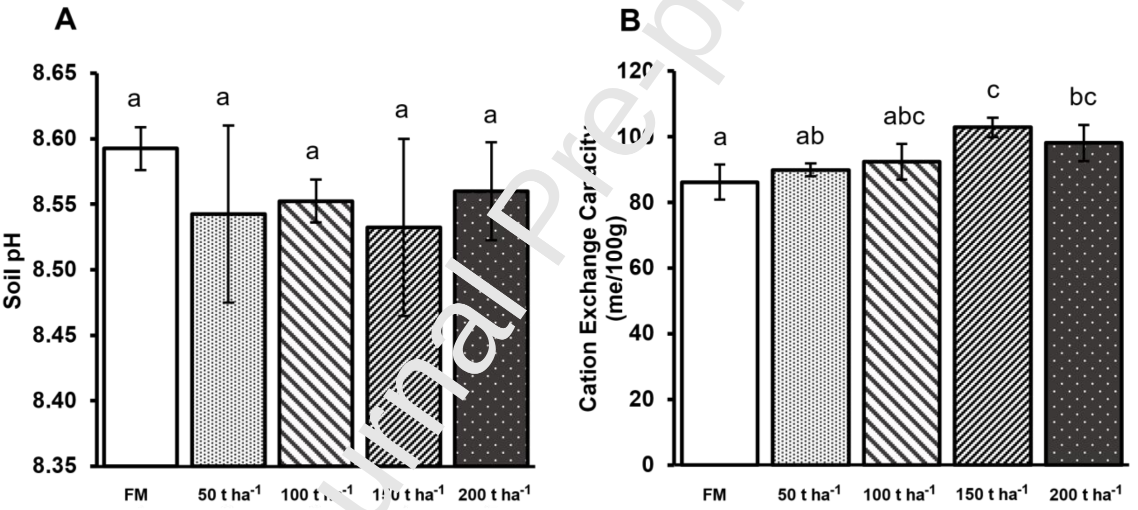

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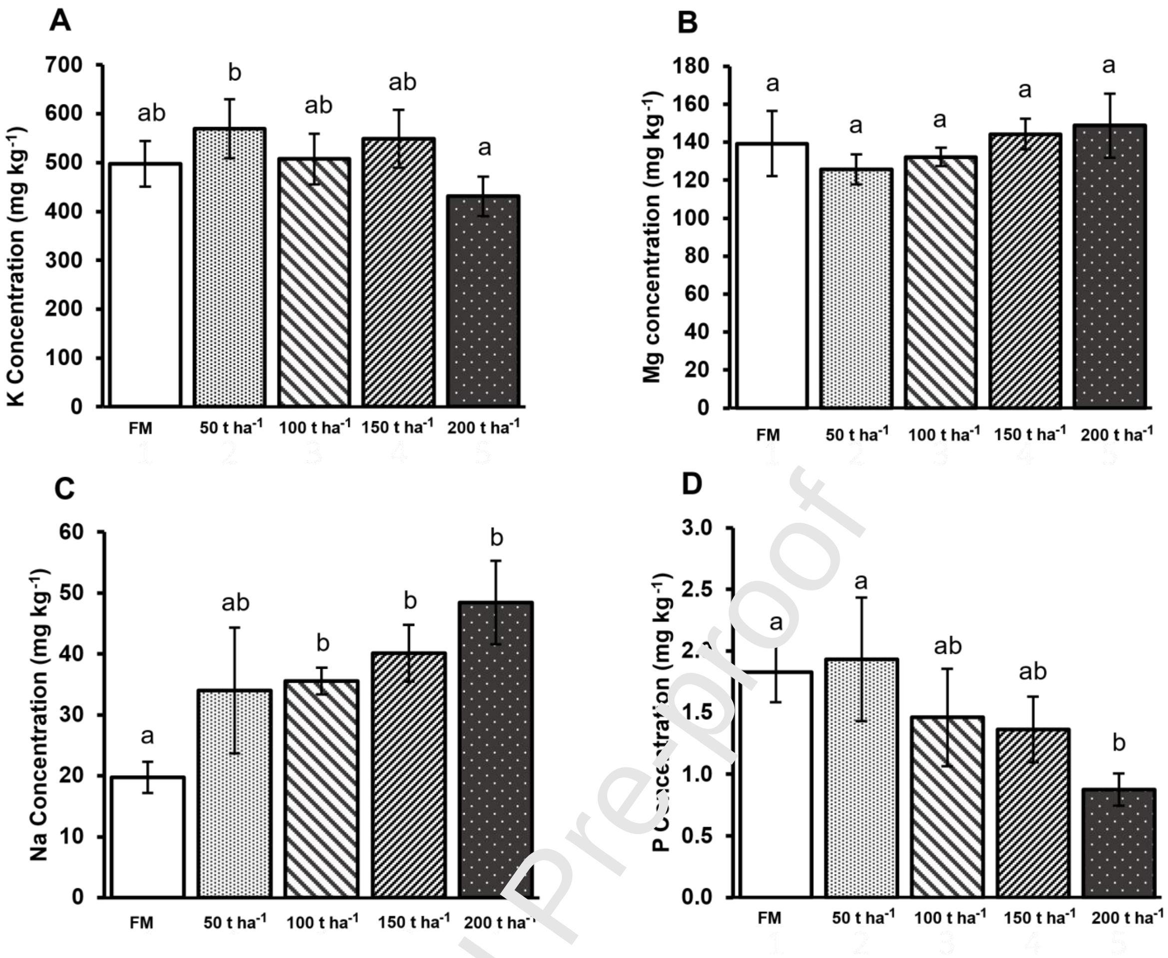

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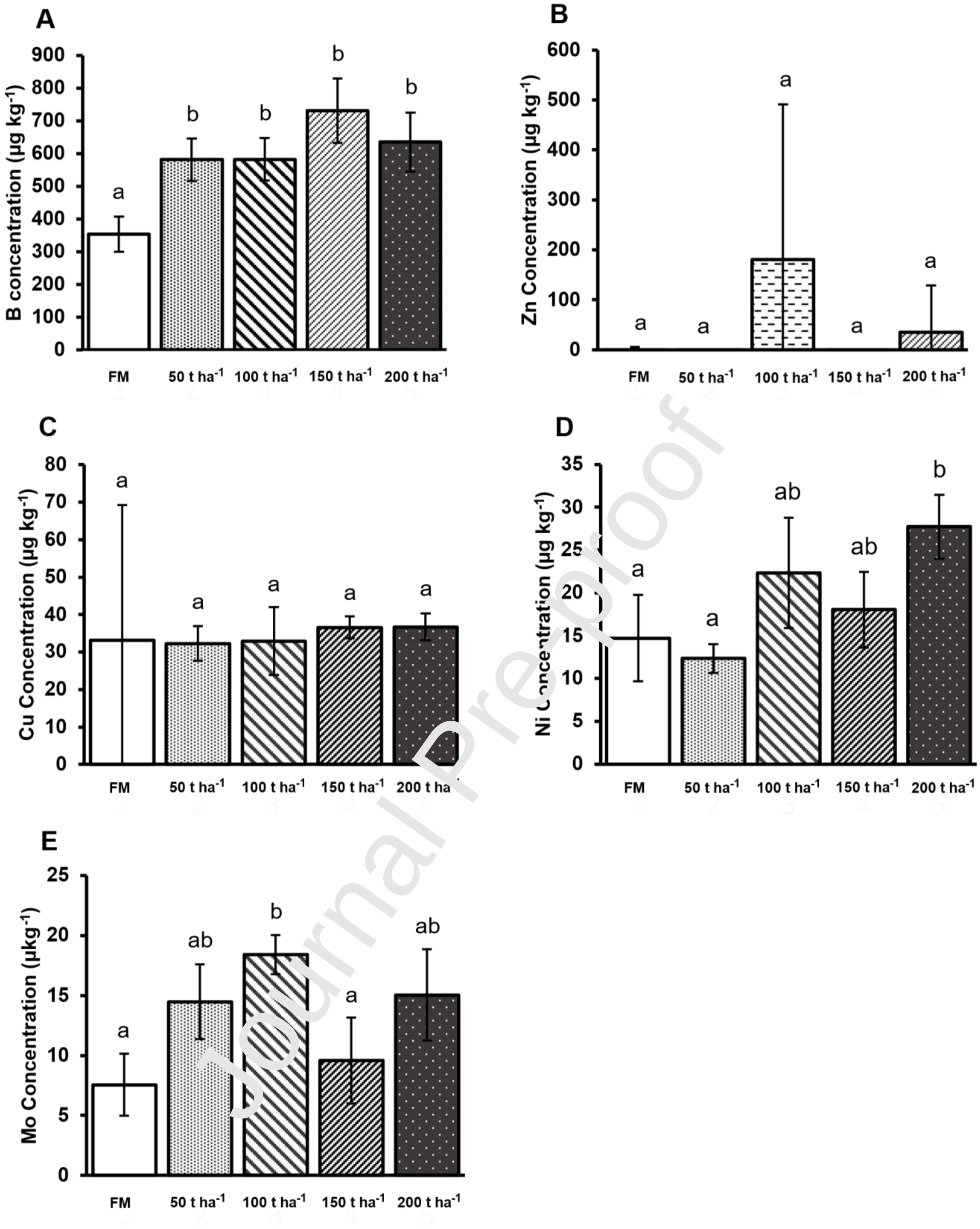

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Author Contributions Statement

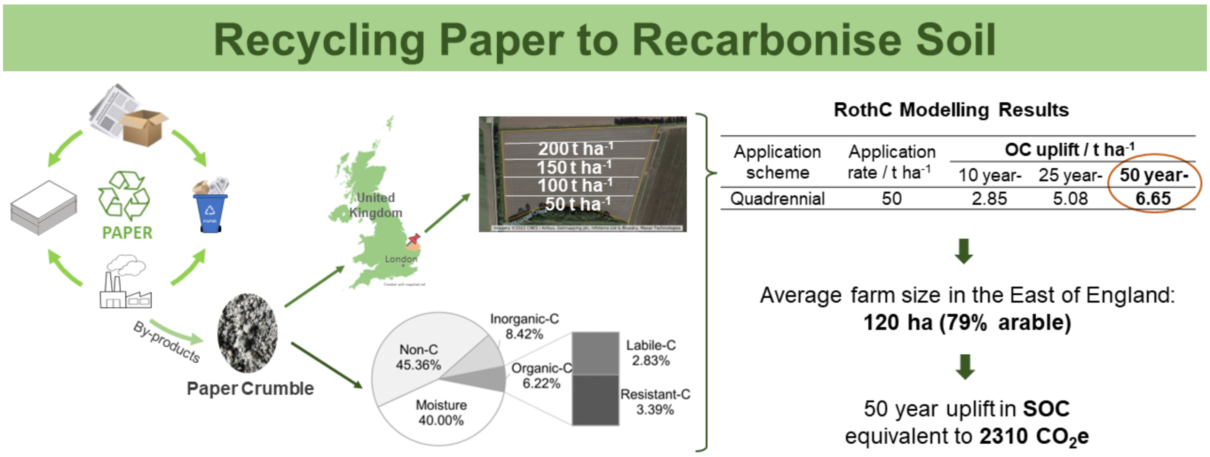
Reid was the Principal Investigator and Mao the Senior Researcher for this research. Kilham and Murfitt coordinated the field trials being established. Together Mao and Keenor undertook the fieldwork, laboratory work and data analysis, generating the data herein and led with drafting of the manuscript. Mao led the carbon modelling. All authors were involved in review and editing to deliver the final manuscript.

Declaration of interests

☐The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☒The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Brian Reid reports financial support was provided by Research England.



Recycling Paper to Recarbonise Soil

RothC Modelling Results

Application scheme	Application rate / t ha^{-1}	**OC uplift / t ha^{-1}**		
		10 year-	25 year-	**50 year-**
Quadrennial	50	2.85	5.08	**6.65**

Average farm size in the East of England:
120 ha (79% arable)

50 year uplift in **SOC**
equivalent to **2310 CO_2e**

Graphical abstract

Highlights:

- Paper crumble (PC) amendment significantly increased SOC
- PC amendment affords opportunity for long-term carbon storage
- PC amendment improved soil bulk density and water holding capacity
- PC did not decrease nitrogen availability