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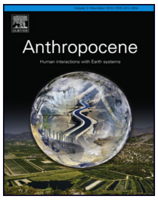

Changes in carbon storage since the pre-industrial era: A national scale analysis

Victoria Janes-Bassett[a,*], Richard Bassett[a], Ed C. Rowe[b], Edward Tipping[c], Dmitry Yumashev[a], Jessica Davies[a]

[a] Lancaster Environment Centre, Lancaster University, UK
[b] Centre for Ecology and Hydrology, Bangor, UK
[c] Centre for Ecology and Hydrology, Lancaster, UK

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ABSTRACT

Carbon stores in the terrestrial biosphere globally represent over 50 % of present-day organic carbon reservoirs and have significantly altered over the last three centuries owing to anthropogenic disturbances. Conversion of natural land to agricultural uses often results in a loss of soil carbon, whilst atmospheric deposition of pollutants such as nitrogen has increased carbon storage in both soil and biomass. Terrestrial carbon storage underpins a range of ecosystem services, including climate regulation, food production, and water services. This storage is crucial for sustainable land management. Quantification of terrestrial carbon cycling at regional and national scales, and understanding how human-induced drivers have impacted present-day carbon stores is therefore required to inform sustainable land use policy. This study applies the N14CP model, an integrated soil-plant biogeochemistry carbon-nitrogen-phosphorus model, across the United Kingdom to simulate changes in terrestrial carbon storage from 1700 to 2020. The analysis shows that change in anthropogenic terrestrial carbon storage is a complex picture comprising of gains in natural areas due to nitrogen deposition and afforestation, and losses in arable areas. We observed an overall net increase in total terrestrial carbon storage of 6.9 %. We note, however, that continued increases in carbon storage cannot be assumed due to (i) reduced influence of future nitrogen deposition as these systems become limited by other nutrients, (ii) the need to continue enhanced nitrogen inputs to maintain carbon sequestered, and (iii) carbon declines in arable areas continuing alongside diminishing gains in other land use types. This research provides a full picture of anthropogenic impacts on terrestrial organic carbon storage, accounting for changing nutrient cycles at a national scale.

1. Introduction

Carbon stored within the terrestrial biosphere plays a key role in the global carbon cycle, representing over 50 % of global organic carbon reservoirs, with relatively rapid turnover times of decades to millennia (Ciais et al., 2013). Biogeochemical cycling has significantly altered as a result of human activities due to the combined effects of land use changes and atmospheric pollution. Agricultural land use has been widely observed to deplete soil organic carbon stores as a result of decreased plant matter inputs and management practices such as tillage (Smith et al., 2016; Wei et al., 2014). Since 1700, global agricultural land has expanded by approximately 400 % and currently accounts for over a third of all land use (FAO, 2017; Goldewijk et al., 2017). Conversely, anthropogenically accelerated atmospheric deposition of nitrogen has been noted to increase both soil and biomass carbon stores (Phoenix et al., 2012; Thomas et al., 2009; Tipping et al., 2017). As a result of these human-induced changes, the cycling and storage of terrestrial carbon has significantly altered over the last three centuries. Despite the extent of these changes, a fairly static understanding of terrestrial carbon stores exists at regional and national scales, since this knowledge is often underpinned by survey evidence that is necessarily limited in spatial resolution and in the length of time-series. Understanding how extensive human-induced changes on Earth affect the current state of terrestrial carbon storage is therefore limited.

Estimation of changes to carbon storage in the terrestrial biosphere is important for mitigating climate change. As the largest store of organic carbon, soils play a key role in climate regulation, and their climate mitigation potential is reflected in

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* Corresponding author.
E-mail address: v.janes@lancaster.ac.uk (V. Janes-Bassett).

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global climate policy such as the ‘4 per mille’ initiative launched at COP21 (21st Conference Of Parties to the United Nations Framework Convention on Climate Change) (Lal, 2004; Minasny et al., 2017). Understanding the dynamics of carbon between vegetation and soil pools is also crucial as soil carbon storage is integral to a range of other ecosystem services (Adhikari and Hartemink, 2016). The physical structure of soils is influenced by soil carbon through aggregate formation, thereby affecting the water holding capacity of soils (Rawls et al., 2003) and flood and drought resilience. Soil carbon also influences nutrient retention capacity that is critical to food security (Lal, 2005). A need exists to quantify terrestrial carbon cycling to understand and manage long-term carbon storage, and ensure the sustainable provision of these multi-sectoral benefits (Davies, 2017).

Biogeochemical models can be used to estimate how terrestrial carbon stores have changed over time, in particular in a period of accelerating human influences on Earth. Yet, estimation is complex due to the coupled nature of carbon and other nutrient cycles, and simultaneous changes in multiple drivers relating to climate, nutrient pollution, land use and management. The close coupling between the cycling of carbon with nitrogen and phosphorus and the significant disturbances humans have made to nutrient cycles through atmospheric pollution and the application of fertiliser mean that integrated models of carbon-nitrogen-phosphorus (C-N-P) are required for accurate estimation of carbon change (Achat et al., 2016; Goll et al., 2012). Several process-based models of integrated C-N-P cycling exist (CENTURY, Cong et al., 2014; Metherell et al., 1993; CLM-CNP, Yang et al., 2014; ECOSYS, Grant et al., 2001; JSBACH, Goll et al., 2012; Roth-CNP, Muhammed et al., 2018). However, most of these models simulate either (semi-) natural or agricultural environments, not both. The lack of models including representation of both natural and agricultural ecosystems limits capabilities of estimating the impacts of land use changes, such as forest clearance for crop production. Furthermore, informing land use and climate policies requires national scale estimates of terrestrial carbon stores, yet many biogeochemical models have a site-specific calibrated focus, making them unsuitable for application at these scales. Whilst studies have applied some models at large spatial scales (Wang et al., 2010), these models are limited in their representation of detailed agricultural management practices (such as crop rotation options and tillage) and significant drivers of biogeochemical change.

This study addresses this research gap by providing a first national-scale insight into changes in carbon stock, that considers land use change between natural and agricultural land uses and nutrient interactions. Specifically, we focus on the following research questions:

1) To what extent can a biogeochemical model of integrated carbon-nitrogen-phosphorus (C-N-P) cycling that incorporates both semi-natural and agricultural land uses simulate present-day terrestrial organic carbon stocks at a national scale?
2) What is the magnitude of change in organic carbon storage in the terrestrial biosphere across the United Kingdom (UK) since the pre-industrial period (over the last 300 years), and how does this vary spatially?
3) What is the combined influence of key drivers, including land use change, management and nutrient pollution during this period?

To address these questions, we apply an integrated soil-plant biogeochemistry C-N-P model, the N14CP model originally developed by Davies et al. (2016a, 2016b) and Janes-Bassett et al. (2020), that includes both semi-natural and agricultural land uses, and represents a range of agricultural land management options. The model uses readily available input data and does not require site-specific calibration, enabling application at large spatial scales, and providing a baseline of change that contextualises current organic carbon stores.

2. Methods

We applied the N14CP model (Davies et al., 2016a, 2016b; Janes-Bassett et al., 2020) across the UK. The model simulates fluxes of C, N and P and is driven by climate, atmospheric deposition of N, sulphur (S), and base cations (BC), the weathering of BC and P, and land use history. Phosphorus enters the system from an initial pool of weatherable $PWeath_0$, set as an initial condition based on a broad soil classification as per Davies et al. (2016a, 2016b). The pool of weatherable P declines over time through an annual release determined by a temperature dependent first-order rate constant. The model uses a quarterly timestep, and does not require site specific calibration of parameters, meaning it is capable of simulating long timescales (several centuries). The model N14CP includes both natural/semi-natural and agricultural land uses, and land use transitions. Natural/semi-natural Plant Functional Types (PFTs) include broad-leaved trees, coniferous trees, herbs, and shrubs. Agricultural PFTs include cereals, roots, oilseeds, legumes and improved grasslands. Agricultural land management practices including additions of N and P fertilisers, tillage and grazing are also represented within the model. Nutrient demand is determined by stoichiometric requirements of PFTs, with plant growth occurring in the second and third quarters of each year. The model simulates net primary productivity (NPP) following Leibig’s law of the minimum determined by one of four factors: temperature, precipitation, N availability or P availability.

The non-site-specific N14CP model has been rigorously tested against multi-variate C-N-P plant-soil data in natural and agricultural location across Northern Europe. Davies et al. (2016a, 2016b) calibrated and tested N14CP in across 88 sites in Northern Europe, showing statistically significant results when comparing observed and simulated soil C, N and P. The results also showed enhanced model performance when allowing the initial pool of weatherable phosphorus to vary on a site basis. The ability of the model to simulate agricultural land uses and a range of management options was explored by Janes-Bassett et al. (2020). The model was applied to 62 long-term experimental plots, and both spatial and temporal performance of the model was tested against soil biogeochemistry and crop yield data. The model indicated statistically significant correlation between observed and simulated soil biogeochemical variables but indicated a slight underestimation of crop yields, which was partly attributed to the quarterly time-step of the model. In Tipping et al., (2019, 2017), the model has also been blind-tested against 1000s of soil carbon and NPP observations in natural land uses across the UK, indicating comparable and statistically significant performance across semi-natural land use types.

In this study, we applied the model across the UK using a spatial 5 km x 5 km grid resolution. The model was run from the start of the Holocene (10,000 BCE) to 2020. Quarterly mean temperature and total precipitation were used as input data for the model. For the period 1901–2016 this was calculated using monthly CRU TS4.00 (Climatic Research Unit Time-series version 4.00) modelled data (Harris et al., 2014; Harris and Jones, 2017). Prior to 1901, mean annual temperature (MAT) was temporally varied using an anomaly based on Davis et al., 2003. We held mean annual precipitation (MAP) constant at the 30-year average for 1901–1931. For 2016–2020, we applied a 30-year mean quarterly data. We estimated inputs of N, S and BC deposition over the period 1800–2010, which accounted for vegetation type, similarly to Tipping et al., 2017. In order to estimate the impacts of human-induced changes on terrestrial carbon stores since the industrial revolution, in accordance with one definition of the "Anthropocene" (Lewis and Maslin, 2015) as signifying accelerating human interactions with the Earth system, we used model outputs from the year 1700 as a pre-industrial benchmark.

Contemporary land uses were derived using UK Land Cover Map 2007 data (LCM2007, Morton et al., 2011). We processed these data to provide a fraction of cover of each PFT within each grid cell. Historical sources provided land use histories for the period 1600–1931 (Stamp, 1931; Thirsk, 1989). Grid-cell fractions with a land use type of shrubs in the year 1600 were assumed to have the same vegetation prior to this date. Where grid-cell fraction land use was either rough grassland or arable in the year 1600, we assumed them as converted from broadleaf forest with a deforestation date based on data from Roberts (2014). Whilst uncertainty exists in the timing of forest clearance, Tipping et al. (2017) showed that model outputs are more sensitive to variation in contemporary N deposition than assumptions surrounding land use in the distant past. For all fractions with forest before 1600, natural succession of vegetation was assumed as herbaceous plant cover from 10,000 BCE to forest cover in 6000 BCE.

Literature sources provided information regarding typical arable rotations and their variation through time. Initial rotations included cereals legumes and ley grass, and root and oilseed crops were included in later years (Allen, 2004; Crop Protection Association, 1996; Paul et al., 1996; Turner et al., 2001, see supplementary information tables S1 for further details). We assumed harvesting to occur in the third quarter of each year, with 95 % of above-ground biomass removed, leaving the remainder as stubble (as per Janes-Bassett et al., 2020). Tillage was implemented in the third quarter (immediately after harvest) from 1850 onwards reflecting the widespread use of plough machinery around this time, and application rates of N and P fertilisers were estimated based on literature sources (Defra, 2018, 2017, 2016; Grigg, 1989; Muhammed et al., 2018; Turner et al., 2001; Wang et al., 2012, see supplementary information S2). Fallow land was introduced as an additional PFT and included within early arable rotations. No fertilisers were applied to fallow land. After 1850, fallow land is tilled and no plant growth was simulated. Where fallow land use occured before 1850 grass was grown, which was then assumed to be harvested and used for fodder. The model was then further developed to include C addition from manuring based on a conservative C:N estimate of 6 g g^{-1} for manures (Bhogal et al., 2010), with C from manures entering the litter pool. Prior to 1940, we assumed that additions of fertiliser were entirely from manures. Between 1940 and 1975 this declined to 50 %, and after 1975, all fertiliser additions were assumed to derive from mineral fertilisers (with no C addition). Grazing animals in improved grassland (defined as grassland where fertilisers are added) were assumed to consume 60 % of the above ground biomass, with 25 % of C returning to the soil and 75 % of N and P returned (similarly to Davies et al., 2016a, 2016b and Janes-Bassett et al., 2020). Improved grasslands were grazed in seasons 2 and 3. Grass leys (within arable rotations) were cut twice annually.

In this study, we also included a new module simulating peat. Similarly to the natural and agricultural components of the model, the peat module simulated a number of stoichiometrically linked pools of carbon, nitrogen and phosphorus representing plant biomass in coarse and fine tissues, plant litter, and soil organic matter in two layers. Instead of a topsoil and subsoil component, however, the two layers were conceptually compartmentalised into an acrotelm and a catotelm layer, similar to Clymo (1984). We defined the acrotelm as the plant accessible layer that is experiencing relatively aerobic conditions. Our conceptualisation of the catotelm, in contrast, is that it consists of buried material, inaccessible to plants, and experiencing constant anaerobic conditions due to persistent saturation. Literature-derived parameters used for the peat module were taken from (Wang et al., 2014). The catotelm layer in the model was accumulated from the burial of organic matter from the acrotelm. Once the acrotelm layer reached a threshold mass, any further organic matter became buried. In this way, the acrotelm remained a fixed depth/mass in the model, and the catotelm grew, simulating the development of peat depth. Similar to the agricultural and semi-natural modules, carbon and nitrogen lost in gaseous form is simulated. Further details of the peat module are found in the supplementary information (S3).

To provide an additional blind test of model performance over the region simulated, we compared the output against a national-scale dataset of topsoil organic carbon storage, assessing the accuracy of simulations in relation to magnitude and spatial variation. To enable comparison with modelled data, we transformed the datasets to provide total topsoil carbon at 15 cm depths across the model grid cells (see supplementary information S4). The LandIS NATMAP carbon product (Cranfield University, 2020), providing data throughout England and Wales derived from the National Soil Map, enabled validation of the model output.

3. Results

3.1. Model validation

The NATMAP carbon dataset indicated greater variability of soil carbon concentration across grid cells compared to the model output (see supplementary information S4 Fig. 1). Fig. 1A shows the topsoil organic carbon (0–15 cm depth) from the validation dataset and the model (simulated in the year 2000). Total topsoil organic carbon simulated by the model over the area of the dataset deviated by +12.5 % (see supplementary information S4 Table 6). Agreement between the NATMAP data and model output were highly significant ($R^2 = 0.435$, $p < 0.001$, see Fig. 1B). Noting that the model has not undergone a calibration procedure in this application, the performance supports a good level of confidence in the results. The model outputs were also compared with NATMAP data based on land use type to check for systematic error or bias, and all four land use groups showed statistically significant correlations between the NATMAP and simulated data; arable $R^2 = 0.063$ $p < 0.001$, improved grassland $R^2 = 0.357$ $p < 0.001$, Forested $R^2 = 0.439$ $p < 0.001$, Non-forested semi-natural $R^2 = 0.029$ $p < 0.001$ (see supplementary information S4 Fig. 2). Each land use group shows greater variation of topsoil carbon in the NATMAP dataset compared with model simulations (see supplementary information S4 Fig. 3). With the exception of outliers, model simulations and NATMAP data overlap for each land use. Given uncertainty associated with NATMAP data and the blind testing in this study, the results indicate a reasonable performance across all land use groups

3.2. Carbon storage – spatial analysis

Model outputs indicate that the total terrestrial organic carbon storage in the UK from 1700 has increased by 6.9 % to a present-day value of 3617 Mt. In this same period, carbon storage in the vegetation, topsoil, and subsoil pools increased by 13 %, 6%, and 3% respectively (see supplementary information S5 Table 7). These increases correspond to gains of 100, 95 and 38 Mt C respectively.

Whilst carbon storage increased overall throughout the UK since 1700, significant spatial variation existed in the magnitude and direction of total carbon changes (Fig. 2; see also supplementary information S6 Fig. 4). Changes in total carbon storage per grid cell varied between -0.59 to +0.40 Mt C (equivalent to -236 to +160 t C ha^{-1}). The largest absolute gains in total carbon storage simulated across south and western Scotland and central Wales are

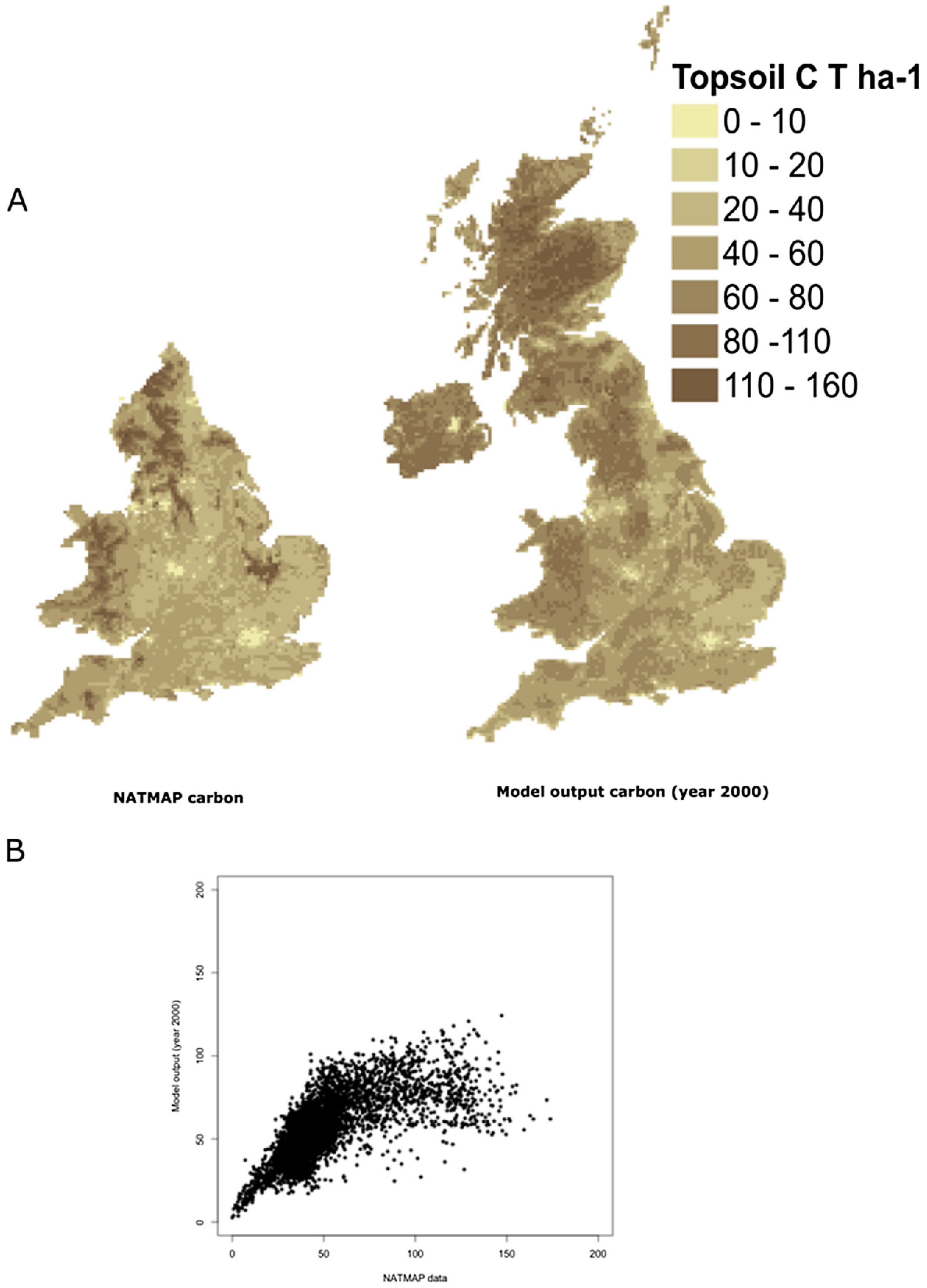

Fig. 1. Topsoil (0-15 cm) t C ha^{-1} per grid cell derived from the NATMAP carbon dataset, and model output for the year 2000. NATMAP carbon data © Cranfield University (NSRI) and for the Controller of HMSO (2020).

largely due to the expansion of forest during the 1900s. The largest losses are confined to the east of England, notably the East Anglia region owing to forest clearance and the expansion of agriculture (as shown in the land use component of Fig. 2, see supplementary information S6). Changes in topsoil carbon between 1700 and 2020 varied from -0.08 to +0.06 Mt C per grid cell (equivalent to -32 to +24 t C ha^{-1}) with the highest gains in Wales and western England, and greatest losses in the east and north-east of England. Vegetation carbon changes varied from -0.56 to +0.38 Mt C (with largest decreases due to forest clearance and increases due to forest expansion). This variation shows a similar spatial pattern to total C change. Change in subsoil carbon throughout the studied time period varied from -0.41 to +0.03 Mt per grid cell.

3.3. Carbon storage – analysis by land use

Arable and improved grassland area has increased by 181 % and 53 % accounting for a present-day land area of 6.3 × 10^6 ha and 6.5 × 10^6 ha (25 % and 26 % of the total UK land area, see Fig. 3A) respectively. Fig. 3B shows simulated total organic carbon storage within each land use classification, again illustrating the overall increase in total carbon storage from 1700 to 2020. Overall forested area in the UK increased throughout the studied time period by 24 % (accounting for a present-day land area of 2.8 × 10^6 ha; 12 % of total UK land area) due to expansion of coniferous plantations. Due to the 50 % reduction of non-forested semi-natural land (incorporating rough grassland, marsh, heath and bog), total carbon stored within these land use types has decreased by 35 % throughout the studied time period, whereas carbon stored (and proportion of UK land area) for other land use groups increased by 37 %, 45 % and 911 % in forest, improved grassland and arable land uses, respectively (see supplementary information S7 Table 8).

Fig. 4 shows the average carbon stored per unit area by each land use group across the UK, and the proportion of carbon storage in topsoil, subsoil and vegetation pools. Both forested and non-forested semi-natural land use groups show increasing total carbon stored per unit area (+11 % and +30 % respectively) throughout the studied time period. This increase coincides with inputs of N from atmospheric deposition since 1800. The total increase in carbon throughout the studied time period is consistent across the topsoil, subsoil and vegetation pools with the exception of forest biomass which shows a slight overall decrease (-1%). This decrease is due to young vegetation becoming established during forest expansion in the 1900s. Topsoil carbon storage per unit area by land use group also shows considerable spatial variation, shown in Fig. 5. Non-forested semi-natural land use shows a slight drop in the median values of carbon stored per unit area around 1800, resulting from the conversion of carbon rich areas of this non-forested semi-natural land to arable use (see the east of England in Fig. 2). Conversely, forested land shows increases in both the

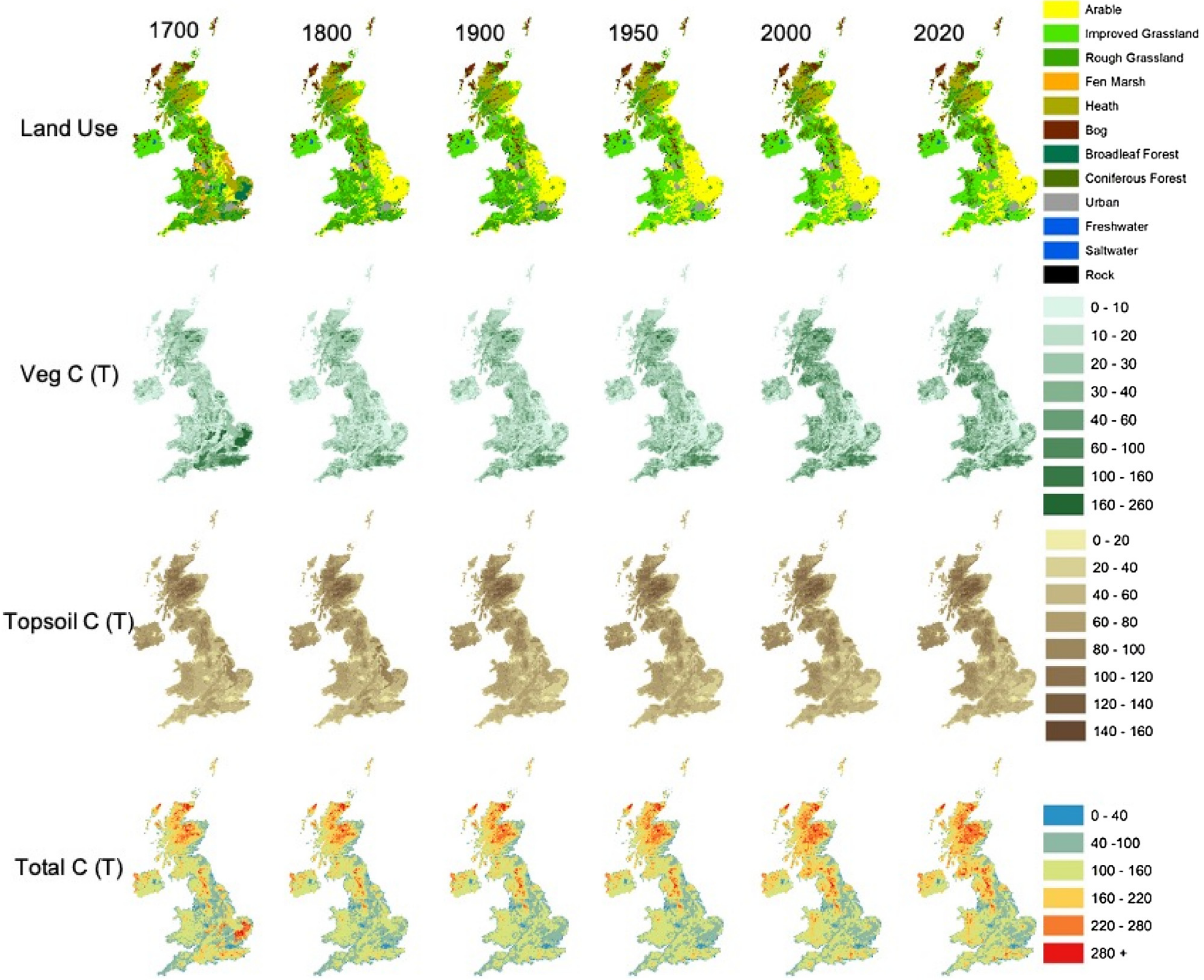

Fig. 2. Land use model input (showing majority land use per grid cell) and simulated magnitude of terrestrial carbon pools (t C ha^{-1}) across the UK from 1700 to present day.

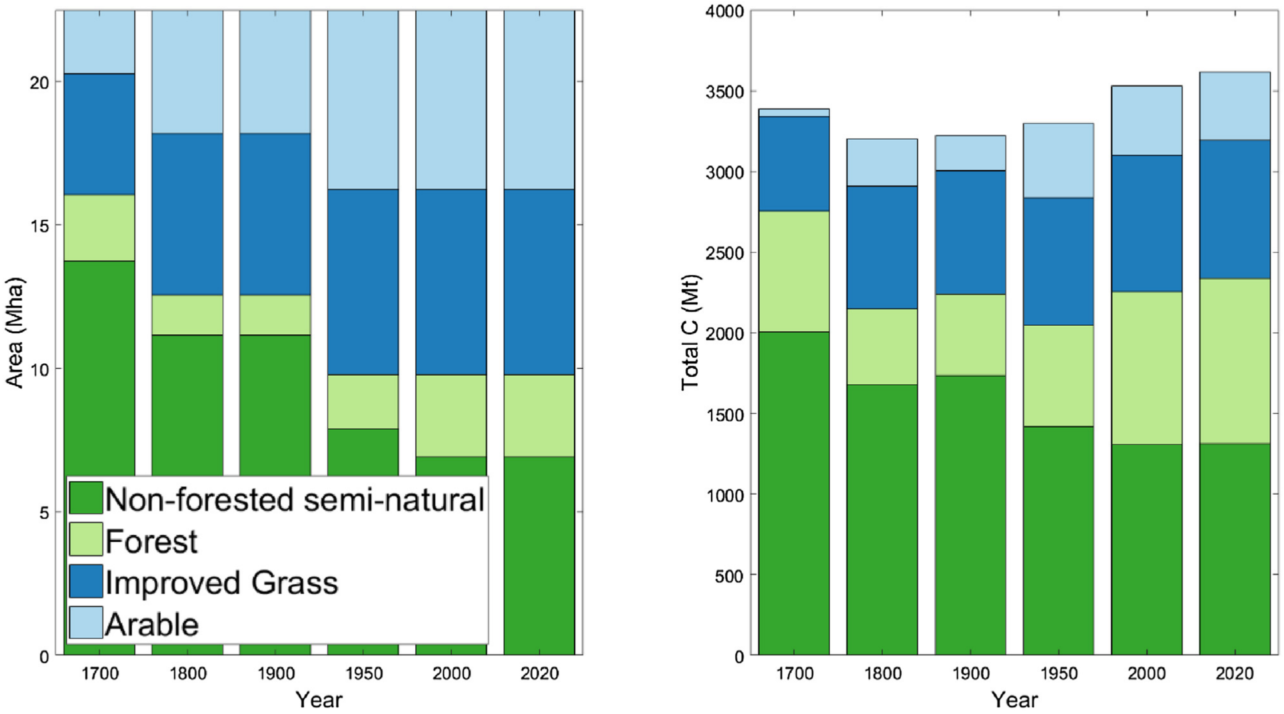

Fig. 3. A - Total area of each land use group throughout the model simulation. Forest includes both coniferous and broadleaf forest PFTs. B - Total simulated carbon storage (topsoil, subsoil and vegetation) by land use throughout the studied time period. Non-forested semi-natural land includes rough grassland, heath, bog and fen marsh PFTs.

median and upper range of topsoil carbon values, due to the combination of atmospheric N deposition and the expansion of coniferous forest to carbon rich areas such as peatlands during the 1900s.

Arable land shows an overall increase in carbon stored per unit of land area throughout the studied time period. This increase was due to an expansion in both topsoil and subsoil pools over this period. Carbon stored per unit area of arable land increased in 1800 and 1950 (see Fig. 5 and supplementary information S7 Table 8), corresponding with a conversion of carbon rich land (previously forest or non-forested semi-natural) to arable use. Once converted to arable usage, however, topsoil carbon storage rapidly decreased. Fig. 5 shows the range of topsoil carbon per unit area for all arable land with notable increases in upper values corresponding to land use conversion. These areas of carbon rich soil show an initial rapid decline in soil carbon after conversion, with the rate of carbon loss

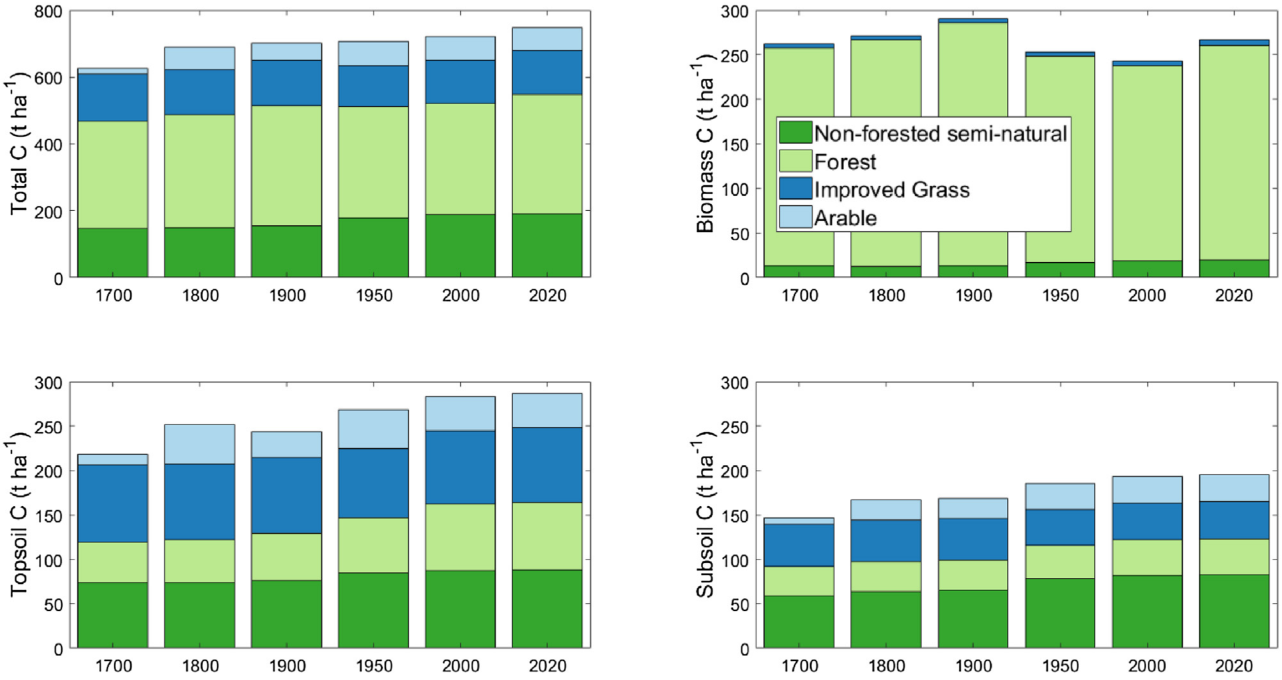

Fig. 4. Simulated average carbon stored per unit area (t ha^{-1}) for each land use group.

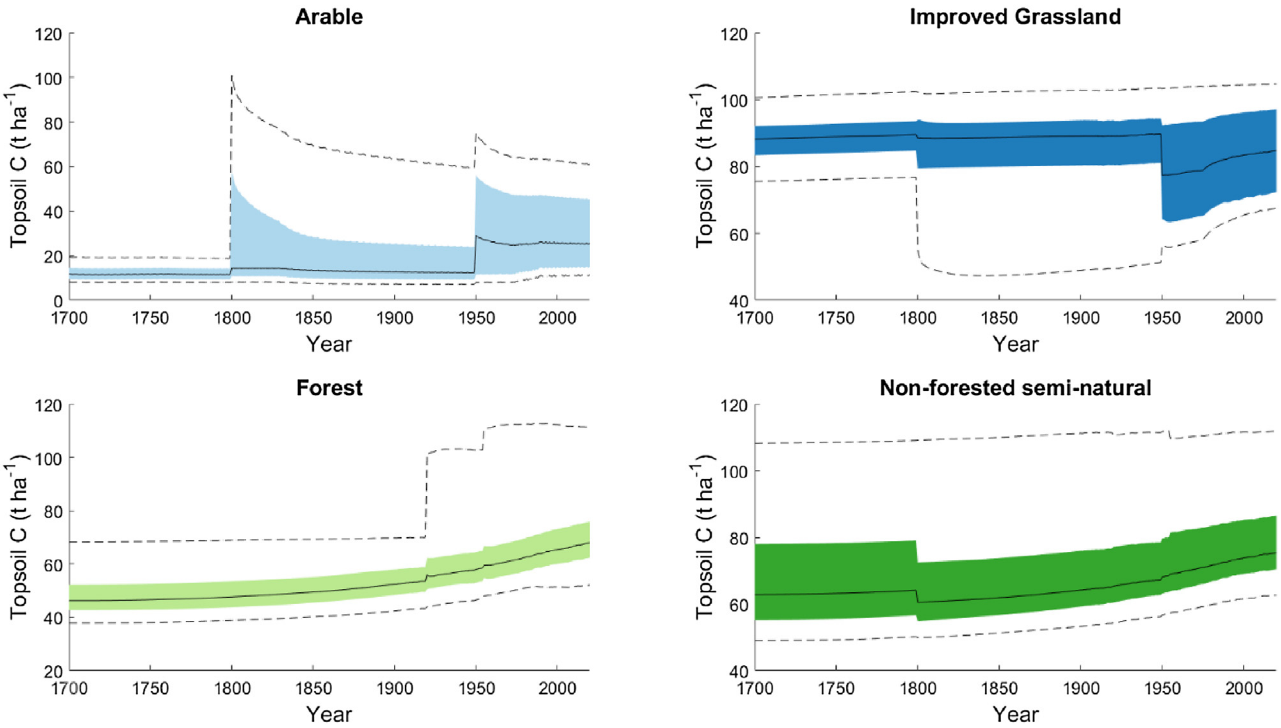

Fig. 5. Range of topsoil carbon stored per unit area by land use type across the UK throughout the studied time period. Coloured area shows the interquartile range, solid line shows the median, and dotted lines show 5th and 95th percentiles.

decreasing over time. This decline indicates that present-day values of topsoil carbon stored per unit area in arable land are very dependent on the length of time since conversion to arable usage (see supplementary information S7 Fig. 4). Topsoil carbon per unit area in land converted to arable use in 1800 and 1950 decreased between 35–58 % and 24–40 % respectively. This decrease represents a total loss of 74 and 47 Mt of topsoil carbon in these land areas since conversion to arable use.

Improved grassland shows a slight overall decrease in carbon stored per unit area throughout the studied time period, due to a decrease in both topsoil and subsoil carbon storage and an increase in biomass storage. Fig. 5 illustrates that land converted to improved grassland in both 1800 and 1950 had a lower topsoil carbon content than existing improved grassland (converse to agricultural conversions during this period), and over time grassland management has resulted in carbon sequestration within these areas (see Fig. 2 and supplementary information S6 Fig. 4). The topsoil carbon pool of land converted to improved grassland in 1800 and 1950 has increased to present day levels by 2.9 and 27 Mt respectively.

The model simulated a total loss of carbon from arable land in the UK of 326 Mt throughout the studied time period, whereas for

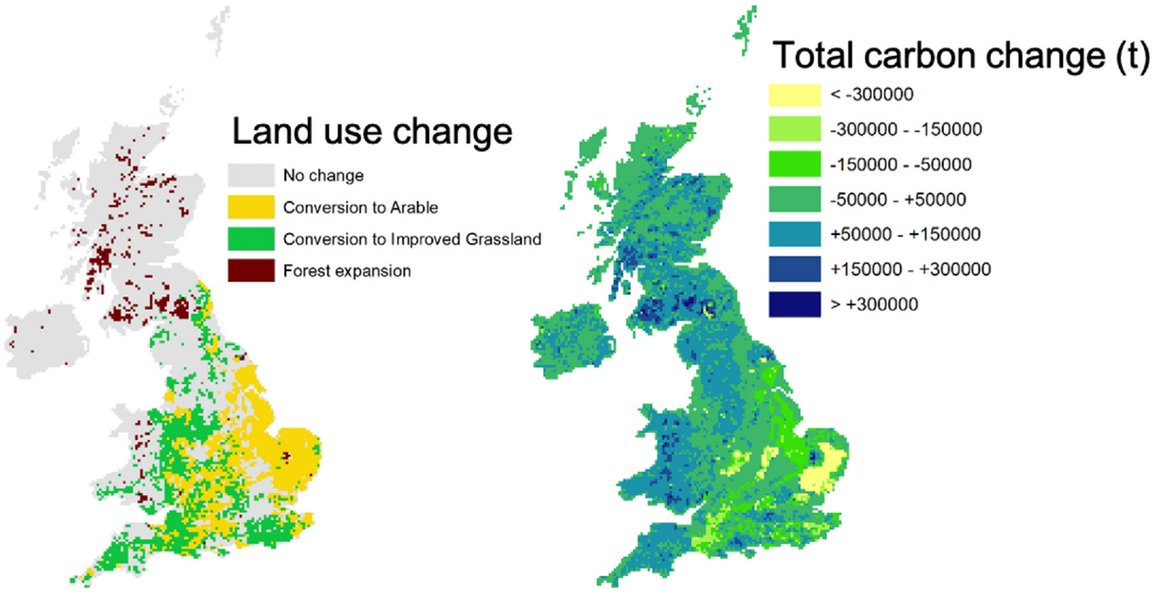

Fig. 6. A: UK land use change and B: Simulated change in total (topsoil, subsoil and vegetation) carbon stores throughout from 1700 to 2020. Carbon values are in t per 5 × 5 km grid cell.

improved grassland, forest, and non-forested semi-natural land increases of 96 Mt, 345 Mt and 119 Mt respectively are found. Fig. 6 shows the total carbon change across the UK throughout the studied time period and the simultaneous land use changes. The largest carbon losses are located in areas of arable expansion, particularly where this land was previously forested (see Fig. 2, and supplementary information S6 Fig. 4) and the largest carbon gains are found in areas of forest expansion.

4. Discussion

The N14CP model was applied at a national scale, simulating integrated C-N-P cycling, enabling estimation of change in terrestrial carbon stocks (across topsoil, subsoil and vegetation pools) throughout the studied time period. Validation with available national-scale data indicates that the model performs well in relation to the magnitude of topsoil carbon stocks. As the model was not initialised with measured soil C data, nor was it calibrated before application, the results are a strong validation of the model accuracy, and that assumptions for parameterisation are reasonable. Spatial performance of the model when compared to the NATMAP dataset shows a statistically significant correlation. Where previous studies have applied process-based integrated C-N-P models at national to global scales (Muhammed et al., 2018; Wang et al., 2010), this study is the first such model, to our knowledge, that incorporates representation of both semi-natural and agricultural land uses and agricultural management practices (such as tillage, fertiliser application and crop rotations) at these scales. This study is novel as it enables simulation of the impacts of land use changes on soil carbon stocks over timescales of several centuries, and assessment of the impacts of land management throughout the studied time period, without prior model initialisation.

The 6.9 % increase in simulated terrestrial C storage of the UK throughout the studied time period results from an increase in carbon stored within semi-natural land (both forested and non-forested) as a result of atmospheric N deposition being greater than the decrease within arable land. Since 1700, the model simulated an increase in both soil and vegetation carbon stored in semi-natural land of 111 Mt and 352 Mt respectively. Studies considering semi-natural land over similar time periods have estimated an increase of NPP of up to 90 % (Tipping et al., 2019). This increase is due to these systems being predominantly N limited, meaning that the increased input of N into the system since 1800 (around the start of the industrial revolution) enabled greater biomass accumulation of C, which also resulted in increased litter inputs and increased soil C (Tipping et al., 2017). Similar increases in NPP and C storage as a result of atmospheric N deposition have been observed previously (Field et al., 2017; Fleischer et al., 2019; Maaroufi et al., 2015; Schulte-Uebbing, 2018; Wang et al., 2019; Yue et al., 2016). We improved on these earlier studies by providing estimation of the combined effects of agricultural expansion, afforestation and atmospheric N deposition at a national scale, indicating that increases to date in terrestrial C storage throughout the UK as a result of N deposition, expansion of forest, and conversion of land to improved grassland are 234 Mt greater than C losses from the expansion of arable land.

The simulated C losses of 86 Mt in the model from arable lands are due to removal of biomass vegetation C stores (on conversion from semi-natural land, and when harvesting), and greater C offtake than inputs from plant residues and/or manures. Muhammed et al., 2018 estimated a change in net topsoil C in the UK arable land from 1800 to 1950, 1950–1970 and 1970–2010 as -0.18, -0.25 and -0.08 t C $ha^{-1}yr^{-1}$ respectively, which is comparable to estimates in this study (−0.12, −0.29 and −0.01 t C $ha^{-1}yr^{-1}$). Similar losses from topsoil C in arable land across the UK have been observed in the Countryside Survey of England and Wales (Reynolds et al., 2013), and more widely loss of soil C as a result of land conversion from semi-natural to arable use has been observed (Deng et al., 2016; Guo and Gifford, 2002; Murty et al., 2002; Smith et al., 2016). As shown in Fig. 5, the model simulated an initial rapid decrease of topsoil C after conversion of land to arable, consistent with previous observations of the majority of C losses occurring in the first years following conversion (Wei et al., 2014).

Improved grasslands show a net C gain with slight losses to the biomass C pool but an increase of 108 Mt in soil C throughout the studied time period. Within the model representation, improved grasslands are fertilised, and N fixation due to the presence of leguminous species within grassland mixes is also represented. This resulted in greater NPP and litter/grazing returns, meaning that inputs of C from both plant and animal sources simulated are greater than losses through decomposition and offtake, resulting in a net C gain. Similar values of soil C gain were estimated by Muhammed et al. (2018); land from 1800 to 1950, 1950–1970 and 1970–2010 as +0.20, +0.47 and +0.25 t C ha^{-1} yr^{-1} respectively which is comparable to estimates from this study (+0.01, +0.04 and +0.14 t C ha^{-1} yr^{-1}). The soil C gains simulated in improved grasslands are consistent with observations from previous studies (Conant et al., 2017; Deng et al., 2016; Guo and Gifford, 2002).

Whilst this research suggests that current levels of increased C sequestration as a result of N deposition are greater than C losses from agricultural expansion at this national level, continued increase of terrestrial C stocks cannot be assumed for several reasons. Firstly, the increase of C stocks in response to N deposition to date is due to widespread N limitation of semi-natural UK ecosystems, yet other factors may limit future responses to N inputs. Productivity of ecosystems shows a non-linear response to N deposition, with lesser/no growth response at higher levels of N deposition (Flechard et al., 2020). With increasing N availability, the NPP of ecosystems is likely to become limited by other factors, such as temperature, water and availability of other nutrients, predominantly phosphorus. As noted by Tipping et al. (2019), N14CP is rarely limited by P in semi-natural soils across the UK, due to the relatively young age of these soils, and therefore the significant weatherable P is still available. However, during the study period of 1700–2020, the model indicated increases of 0–3 % and 9–16 %, respectively, for the proportion of forested and non-forested semi-natural land where NPP is limited by phosphorus. Arable and improved grasslands were not limited by P (or N) throughout the study period due to fertiliser additions. Fleischer et al. (2019) simulated decreased future global C uptake per unit N deposition as a result of P limitation, and in areas of the UK with high rates of N deposition P and N/P co-limitation has been observed (Taylor et al., 2020). Secondly, to maintain C sequestered as a result of N deposition, enhanced N inputs would need to be maintained. Tipping et al. (2017) also noted the majority of soil organic carbon sequestered in semi-natural land resulting from N deposition is predicted to have a decadal turnover time, making it sensitive to future decreases in N deposition. Thirdly, the model outputs indicated that arable soil C per unit area has not reached an equilibrium and is still decreasing (see Fig. 5), whilst C sequestration in semi-natural land uses in response to N deposition is slowing. Non-forested semi-natural land total C storage increased on average by 0.49 t C $ha^{-1}yr^{-1}$ from 1900 to 1950 compared to 0.18 and 0.07 t C $ha^{-1}yr^{-1}$ from 1950 to 2000 and 2000–2020 respectively. If these trends continue, the small gains seen in terrestrial C storage at the UK scale could be reversed. Therefore, future land use and management strategies should be considered carefully to ensure long-term sustainability of C storage. This study further highlights the need for models of integrated C-N-P cycling for such evaluations (Achat et al., 2016; Goll et al., 2012).

Currently, as the N14CP model does not simulate urban environments, it did not account for land use classified as urban at present-day. Previous studies have observed loss of C from topsoil after urban sealing (Pereira et al., 2021; Vasenev and Kuzyakov, 2018) whereas Edmondson et al. (2012) noted significant SOC stores in both urban greenspace and sealed soils. Given the uncertainty associated with soil processes and C change in urban environments, we are not aware of any biogeochemical models (of C-N-P cycling) that include representation of urban areas, indicating a key research gap.

Inherent uncertainties exist in relation to simulating a 300-year time period, associated with the climate input data, land use history, land management history, and atmospheric deposition. Throughout the time period studied, the model indicates that NPP is predominantly limited by N, meaning that uncertainty associated with mean quarterly temperature and precipitation will unlikely affect the accumulation of biomass significantly. Whilst compiling land use history over such long periods inevitably includes errors, Tipping et al. (2017) noted that the model is more sensitive to contemporary N deposition than historical land use. The management history represented in the model assumes spatially consistent management of agricultural land (with separate practices for arable and grassland) throughout the UK, which is unrealistic. Due to limited information in the literature, however, insufficient evidence is available to support spatial variation of management throughout the historical time period.

The model results simulates NPP is predominately limited by N availability, indicating that future increases in temperature may not result in significant increased biomass or topsoil C accumulation. Conversely, increasing temperatures will likely enhance rates of microbial decomposition and release of C from soil organic matter, resulting in a decline of topsoil C stores (Koven et al., 2017; Wiesmeier et al., 2019). The N14CP model applied in this study does not currently include concentrations of carbon dioxide (CO_2) as a limiting factor on NPP. Recent studies, however, suggest that the effects of elevated CO_2 may now be declining as systems are limited by other factors such as N and P availability (Wang et al., 2020).

This research indicates that key mechanisms for increasing C sequestration at a national scale are to avoid and/or reverse the conversion of land to arable usage. This suggestion follows findings by Sanderman et al. (2018) that the global C debt in the top 2 m of soil of 133 Pg C results from agricultural expansion over the last 12,000 years. Given projected increasing population over the next century, and hence demand for food and competition for urban land, afforestation of significant areas of arable land in the UK is unlikely to be a viable option. Therefore, improved management of arable soils to reduce soil C declines and promote sequestration should be a priority, whilst acknowledging limitations to long-term sequestration over large areas (Poulton et al., 2018).

As the model results indicate significant spatial variation of changes to terrestrial C stores throughout the UK, with a mixture of both net losses and gains, the impacts of these changes to C stocks will therefore also be highly variable. Loss of C is linked to negative impacts such as decreased drought resilience and crop yields (Ankenbauer and Loheide, 2017; Lal, 2005; Oldfield et al., 2019). Conversely, terrestrial C gains, such as those simulated in areas of afforestation and improved grassland, have been linked to flood resilience (Li and Shao, 2006). Increases in biomass and productivity in natural areas in response to N pollution, however, are a key driver of losses in biodiversity (Hautier et al., 2009; Stevens et al., 2004) and increased N loss to waterways resulting in damage to water quality and aquatic ecosystems. The overall net C gain simulated in the UK throughout the studied time period indicates only a minor contribution to mitigation of climate change. The 233 Mt of C gain simulated is equivalent to 855 Mt of CO_2, making up just 1.1 % of the estimated total UK emissions from 1751–2017. This study, however, only considered changes to C storage. Several studies have indicated the impact of N deposition enhancing N losses as N_2O to the atmosphere through denitrification (Flechard et al., 2020; Zaehle, 2013) offsetting climate mitigating effects of C sequestration. Therefore, climate-related benefits simulated here should be interpreted with caution. This study, however, shows the relevance of considering long-term and large-scale effects when considering intensification, rewilding, afforestation and other strategic approaches to land use.

5. Conclusions

In conclusion, this modelling study has provided a first national-scale insight into changes in human-induced terrestrial carbon stock since the onset of industrialisation. It takes into account manipulation of nutrient cycles and changes in land use and management. Answers to the research questions addressed in this paper are as follows:

1) When compared with a national topsoil carbon dataset (NATMAP), the N14CP model indicated a slight over-estimation across the area covered by the dataset (+12.5 %), and statistically significant spatial performance. This result indicates a good level of confidence in the ability of the model to simulate present-day organic carbon stocks.
2) Terrestrial organic carbon storage throughout the UK has increased by 6.9 % since 1700, to a present-day value of 3617 Mt.
3) The significant spatial variation in the magnitude and direction of total carbon changes is indicative of the variation in drivers associated with change. In arable areas, and areas of arable expansion, terrestrial carbon storage has decreased due to land clearance and management practices. Semi-natural land, however, increased in carbon storage due to atmospheric deposition of nitrogen.

This analysis has shown that change in terrestrial carbon storage over the studied time period across the UK is a complex picture driven by widespread changes in land use and addition of nutrients. To date, losses of terrestrial C from agriculture throughout the studied time period have been offset by increased C in semi-natural land as a result of increased atmospheric N deposition from human activities. Since 1700, combined soil and vegetation carbon stores across natural land have increased by 464 Mt due to N deposition and forest planting. This study provides an estimation of the combined effects of agricultural expansion, afforestation and N deposition at a national scale, indicating an overall increase in total C of 6.9 % occurs which is made up of greater gains in natural land and losses of C in arable settings. Changes in terrestrial soil C stocks throughout the UK during the studied time period show significant spatial variation, largely matching simultaneous changes in land use. Semi-natural and improved grassland show increases in C whilst arable areas decrease.

It is important to note that continued increase of terrestrial C stocks cannot be assumed for three key reasons; the potential for future atmospheric N deposition to have a reduced influence on NPP in natural areas; the need to maintain current levels of N deposition to retain carbon that has been sequestered as a result of this nutrient input; and the observation that areas where land use has changed have not reached an equilibrium. The rate of recent carbon gains in semi-natural and improved grasslands is slowing, whilst losses from arable land continue. Therefore, the relatively small gains in terrestrial C storage observed over the last three centuries could be reversed, highlighting the need for future land use strategies that enable long-term sustainability of C storage. The research has implications for future C sequestration targets, indicating land use change is a key mechanism for C sequestration, and that policies to increase C sequestration at a national scale should avoid and/or reverse the conversion of land to arable usage. Given the limited feasibility of this option future studies should therefore evaluate arable land management options and strategies to reduce and reverse the decline of C in arable soils.

Declaration of Competing Interest

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.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.ancene.2021.100289.

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