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Patterns and trends of topsoil carbon in the UK: Complex interactions of land use change, climate and pollution

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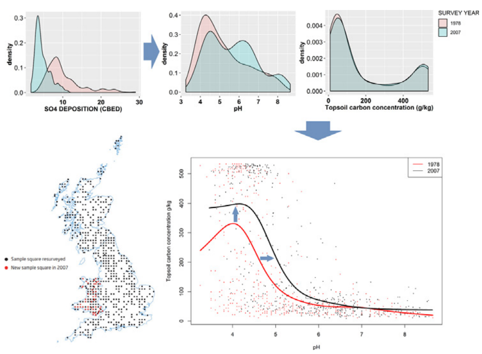
HIGHLIGHTS

- Land use change caused most carbon change in soils with high baseline concentration.
- The effect of change in woodland cover is also dependent on baseline carbon.
- UK soil pH distribution has changed dramatically, due to recovery from acidification.
- Models suggest this recovery has shifted the relationship between carbon and pH.
- SO_4 deposition helps to explain the spatial pattern of these shifts.

GRAPHICAL ABSTRACT



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ABSTRACT

The UK Countryside Survey (CS) is a national long-term survey of soils and vegetation that spans three decades (1978–2007). Past studies using CS data have identified clear contrasting trends in topsoil organic carbon (tSOC) concentrations (0–15 cm) related to differences between habitat types. Here we firstly examine changes in tSOC resulting from land use change, and secondly construct mixed models to describe the impact of indirect drivers where land use has been constant. Where it occurs, land use change is a strong driver of SOC change, with largest changes in tSOC for transitions involving SOC-rich soils in upland and bog systems. Afforestation did not always increase tSOC, and the effect of transitions involving woodland was dependent on the other vegetation type. The overall national spatial pattern of tSOC concentration where land use has been constant is most strongly related to vegetation type and topsoil pH, with contributions from climate variables, deposition and geology. Comparisons of models for tSOC across time periods suggest that declining SO_4 deposition has allowed recovery of topsoils from acidification, but that this has not resulted in the increased decomposition rates and loss of tSOC which might be expected. As a result, the relationship between pH and tSOC in UK topsoils has changed significantly between 1978 and 2007. The contributions of other indirect drivers in the models suggest negative relationships to seasonal temperature metrics and positive relationships to seasonal precipitation at the dry end of the scale. The results suggest that the CS approach of long-term collection of co-located vegetation and soil biophysical data provides essential tools both for identifying trends in tSOC at national and habitat levels, and for identifying areas of risk or areas with opportunities for managing topsoil SOC and vegetation change.

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1. Introduction

Soil carbon represents the largest terrestrial carbon pool and its fate has important implications for climate change mitigation (Batjes, 1996; Smith et al., 2008). The recent "4 pour mille" initiative, for instance, sets a global target for annual soil carbon increases of 0.4% through changes in land use and management to offset global C emissions (Rhodes, 2019), although the feasibility has been challenged (e.g. Poulton et al., 2018). Soil carbon is also a primary indicator of soil health (e.g. FAO and ITPS, 2015) and a strong determinant of global food and nutritional security (e.g. Lal, 2016). Within the soil carbon pool, topsoil or shallow soil carbon concentrations are generally the easiest to measure and most often used to identify trends in the soil carbon pool or provide indicators of soil health. Approximately 699 Pg of carbon are stored in the top 30 cm of soils globally (Hiederer and Köchy, 2011) and most decadal scale fluctuations in total soil carbon stocks occur in topsoil (e.g. Chen et al., 2013). Spatial and temporal patterns of topsoil carbon concentration are determined by both local drivers (e.g. land use, soil, geological parent material) and global drivers (e.g. climate, weather, pollutant deposition) which govern the balance between soil organic carbon (SOC) inputs from vegetation and losses from decomposition (Schmidt et al., 2011). The interplay and shifting balance amongst local and global drivers leads to complex patterns of change in topsoil carbon, making large-scale, long-term trends in soil carbon difficult to detect and interpret with current soil monitoring budgets which are frequently well below those for other natural capital assets such as water and biodiversity.

Large declines of topsoil carbon have been reported in China (Xie et al., 2007) and New Zealand (Schipper et al., 2007), and there are concerns that climate change will cause SOC loss, creating a positive feedback loop in the carbon cycle (Davidson and Janssens, 2006). However, in the UK, studies disagree over whether there is a trend of loss in tSOC which can be linked to changes in climate (Bellamy et al., 2005; Chapman et al., 2013; Reynolds et al., 2013). The pattern of change is also variable across Europe; increase has been shown for agricultural soils in the Netherlands (Reijneveld et al., 2009) whilst losses have been identified in the French mountains (Saby et al., 2008), and cropland soils in Norway, Belgium and Finland (Riley and Bakkegard, 2006, Sleutel et al., 2007, Heikkinen et al., 2013). In Belgium the direction of change has been shown to vary with land use (Goidts et al., 2009), and spatially varying fluctuations have also been recorded in Bavaria (Kühnel et al., 2019). Where trends have been recorded, there is uncertainty about the drivers of topsoil carbon change, with a range of possible explanatory factors proposed including: climate change (Bellamy et al., 2005; Davidson and Janssens, 2006); land use and land use change (Guo and Gifford, 2002; Dawson and Smith, 2007; Xie et al., 2007); land management practices (Schipper et al., 2007; Smith et al., 2007); increased atmospheric CO_2 concentrations (Jastrow et al., 2005); and atmospheric deposition of pollutants (de Vries et al., 2009; Janssens et al., 2010; Tipping et al., 2017). Many studies identify a combination of factors, for example, decline in SOC for Finnish croplands may be linked to climate warming and historic land use change (Heikkinen et al., 2013); similarly, losses in Belgium may be attributed to land use and management changes and climate warming (Sleutel et al., 2007).

Previous studies have identified statistically significant trends in topsoil organic carbon (tSOC) concentrations (0–15 cm) at national and habitat levels from the Countryside Survey (CS) dataset; a national long-term survey of soils and vegetation that spans three decades (1978–2007). At a national scale for GB there was a small increase in tSOC between 1978 and 1998, followed by a small decrease to 2007, resulting in no significant overall change between 1978 and 2007 (Reynolds et al., 2013). When considered for sites with no change in vegetation type, there are contrasting trends related to differences between habitat types. There was an increase for woodlands from 1978 to 1998 and from 1978 to 2007; an increase for heath and bog from 1978 to 1998; and a decrease for arable from 1998 to 2007 and 1978 to 2007 (Emmett et al., 2008). These changes may be related to management, e.g. reduced woodland management (Kirby et al., 2005; Smart et al., 2014) may explain the observed carbon increase, and increased intensity of arable management may explain the observed decrease (Smith et al., 2007). Whilst it is interesting to explore the impacts of changes in management based on habitat level assessments, these analyses do not help us to understand influence from long term large scale drivers such as climate change and air pollution.

To make sense of the apparently inconsistent trends reported for the UK, it helps to understand the expected effects of both local and global drivers on topsoil carbon, and to account for the (potentially co-varying) spatial patterns and temporal trends of these drivers. An analysis of change based only on repeated observations (surveys) of topsoil carbon is insufficient evidence of soil carbon risk (even discounting the scarcity of such long-term, national scale observations). It has been noted that such surveys face the challenge of simultaneously minimising sample variability to maximise change detection power, and maximising variability to maximise potential for understanding drivers of change (Goidts et al., 2009). We are also moving into unknown territory, with the potential for climate extremes beyond those experienced in recent history.

If drivers of change are varying in a compensatory manner, the net effect on topsoil carbon might be no change over several decades. It would be foolhardy to plan policy for sustainable soils based on such simple trend observations (the future is seldom identical to the past especially where land management policies are concerned). Robust decision support for managing soil health and contributing to global climate mitigation requires an integrated understanding of the drivers of change for soil carbon in order to anticipate future shifts in the balance of processes affecting topsoil carbon. Modelling approaches combining a range of climate and site factors can be used to improve understanding of spatial and temporal patterns of changes in tSOC (e.g. Kühnel et al., 2019).

Land use and land use change can potentially have the most significant impacts on topsoil carbon concentration due to the associated changes in carbon inputs from primary production and physico-chemical conditions related to management practices and rooting traits. In general, meta analyses have found conversion to grassland causes SOC increase, conversion to arable causes SOC decrease and conversion to forest produces mixed results (Guo and Gifford, 2002, Poeplau et al., 2011, Poeplau and Don, 2013). Impacts are generally nonlinear over time, and trends may reverse as soils approach a new equilibrium, which may take over a century (Poeplau et al., 2011, Poeplau and Don, 2013). Even without change in land use, changes in land management practices such as efficiency of removal of yields, depth of tillage and rates of manure application can also have significant impacts (Smith et al., 2007). Losses of topsoil carbon from arable soils reported by multiple UK surveys (Bellamy et al., 2005; Emmett et al., 2010; Chapman et al., 2013) may relate to ongoing or intensified management (Smith et al., 2007).

Climate driven changes in temperature and precipitation have varied impacts on SOC, due to nonlinear responses of decomposition and plant growth to both drivers and interactions between their effects. Rising global temperatures are expected to enhance SOC losses due to increased rates of microbial decomposition and enzyme activity, particularly in topsoil (Wiesmeier et al., 2019). Response of decomposition rates to warming is modulated by initial temperature, water stress, complexity of substrates, and substrate availability which is controlled by concentration, diffusion, and physical and chemical protection

(Knorr et al., 2005, Davidson and Janssens, 2006, Davidson et al., 2006). Thresholds may also be important, either for decomposition or for plant species composition and growth (e.g. Barraclough et al., 2015). Colder sites may experience more relative change in decomposition, since Q_{10} of decomposition reactions increases with decreasing temperature, and there may be more recalcitrant carbon at cold sites (Davidson and Janssens, 2006) resulting in higher risk for high SOC soils (Crowther et al., 2016).

The strong correlation of soil moisture and temperature makes it difficult to separate apparent and intrinsic sensitivity to either driver (Davidson and Janssens, 2006; Sierra et al., 2015). Soil moisture has a nonlinear relationship to decomposition, which peaks at intermediate moisture levels where oxygen supply and substrate diffusion through soil water are balanced (Skopp et al., 1990). For some sites, climatic changes may also increase or decrease net primary productivity (NPP) and associated organic carbon inputs to soil, affecting carbon balance (Gang et al., 2017). Modelling suggests that global impacts of increased temperatures may be more significant for accelerating decomposition than for increasing NPP (Kirschbaum, 2000). Subsequent work has shown the added importance of dry soil anomalies, which may reduce NPP more than wet soil anomalies increase it (Green et al., 2019).

Fundamental shifts in soil properties affecting soil carbon can also result from increasing frequency of climate extreme. Periods of drought can alter soil structure to prevent re-wetting, increasing aerobic conditions which increases decomposition losses, particularly for wet podsols (Robinson et al., 2016). Lowered water tables can also have significant impacts through accelerated decomposition in peats (Ise et al., 2008). Conversely, long periods of saturated water content in mineral soils have been shown to increase C losses through reduction of Fe and release of mineral bound SOC (Huang and Hall, 2017).

Soil pH is known to affect soil organic matter cycling and CO_2 efflux (Reth et al., 2005; Oulehle et al., 2006), and is commonly applied as a modifier for SOC decomposition rates in models (e.g. Smith et al., 2010). Direct impacts of pH on microbial decomposition rates have been demonstrated in lab studies (Andersson and Nilsson, 2001), and in China, an increase in SOC in croplands has recently been linked to acidification (Zhang et al., 2020). The relationship is nonlinear with greatest inhibition of decomposition in acidic soils and decomposition increasing as pH rises from 2 to 5 (Smith et al., 2010). Soil pH has changed significantly in Europe and North America over the last 80–120 years as soils have acidified in response to atmospheric sulphur emissions during the industrial revolution (Hedl et al., 2011), and subsequently recovered as acidic deposition has declined due to international air quality policies (Menz and Seip, 2004, Oulehle et al., 2006, Kirk et al., 2010). Increase in soil pH across all UK habitats was reported by several independent monitoring schemes: The National Soil Inventory (NSI); The Countryside Survey (CS); Environmental Change Network; International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests- Level II; and the resurvey of British Woodlands (Kirby et al., 2005; Emmett et al., 2010; Kirk et al., 2010; Vanguelova et al., 2013). Within CS data, the shift was an increase of 0.5 pH unit at a national scale from a mean of 5.39 to 5.87 which given the baseline distribution of pH moves many soils from sitting within the microbial pH-sensitive range (Reynolds et al., 2013).

Fertilisation by atmospheric deposition of inorganic nitrogen has been shown to increase NPP and associated organic carbon inputs to soil from vegetation in woodland, grassland, shrubland and heath (Magnani et al., 2007; de Vries et al., 2009; Tipping et al., 2017). Nitrogen deposition impacts may differ for oligotrophic bogs, which have been shown to shift in species composition from sphagnum and heather to more easily decomposable grasses such as Molina with subsequent SOC losses (Bobbink et al., 1998). Even where sphagnum remains, increased nitrogen deposition will reduce polyphenol content of litter, increasing decomposition rates (Bragazza and Freeman, 2007).

Geological parent material determines many soil properties that ultimately affect SOC cycling and retention such as soil mineralogy, texture, pH and cation concentration (Torn et al., 1997, Wiesmeier et al., 2019). Exchangeable soil calcium, for instance, is important for carbon stabilisation and has been shown to positively correlate with SOC across multiple studies, depending on other limiting factors (Wiesmeier et al., 2019). Parent material also affects drainage controlling rates of formation of soils, depth to water table and suitability for different types of vegetation (Jenny, 1941).

Here we examine spatial and temporal trends in tSOC concentrations across the UK, using Countryside Survey (CS) data on soils and vegetation, and explicitly accounting for spatial and temporal changes in the drivers discussed above. This work differs from previous analyses of the CS soils data in exploring the influence of potential drivers of change, rather than looking for changes in mean at national or habitat level (as per Reynolds et al., 2013) or using the data in process based modelling to explore the importance of N deposition in semi-natural habitats (e.g. Tipping et al., 2017). We use tSOC data from 783 soil plots sampled in both 1978 and 2007 along with national datasets for the drivers downscaled to the soil sample plots. Replicated changes in vegetation cover occurred on 36% of these re-sampled soil plots between the two survey years providing robust estimates of the direction and magnitude of tSOC change for 21 different land use transitions. For the 64% of re-sampled soil plots with no land use change, we developed mixed models of the spatial patterns of tSOC in 1978 and 2007 using as local and global drivers: topsoil properties (pH); geology (parent material attributes); atmospheric deposition (nitrogen and sulphur); climate (mean and extreme precipitation, growing degree days, mean temperature and range); and land use (aggregate vegetation class, broad habitat type).

Using the plots with no land use change, we also attempted to develop a mixed model of the temporal changes in tSOC between the two years but did not achieve a significant result. Despite the robustly significant models of tSOC spatial pattern in each of the years, it was not possible to derive a model based on the observed changes between the years. However, the changes in relative importance of the drivers between the 1978 and 2007 spatial models provides evidence and insight into the covariation and shifting balance of the drivers of tSOC change, and re-enforces the assertion that no detectable change in tSOC does not necessarily mean no significant dynamics affecting tSOC (further discussion of the change model is restricted to the supplementary material).

2. Materials and methods

2.1. Data sources

The tSOC data analysed here are from the Countryside Survey (CS), a unique audit of the natural resources of the countryside of Great Britain (http://www.countrysidesurvey.org.uk). The CS dataset provides a stratified random sample of topsoil, vegetation and ecosystem properties across England, Scotland and Wales. Samples were collected from 5 randomly selected locations within 1 by 1 km squares across GB in three years: 1978 (256 squares), 1998 (569 squares) and 2007 (591 squares) resulting in an increase from 1280 to 2955 sampling locations over time, whilst re-sampling in the same locations as the previous surveys. The survey squares were stratified within each of 45 land classes derived from major environmental gradients based on topography, climate and human infrastructure (Firbank et al., 2003) (see supplementary material for more detail). The use of a stratified random sample enables estimation of means and trends at a national scale. Full details of the sampling design and methodology are described elsewhere (Carey et al., 2008). Soil analysis included measurement of pH in water and soil carbon by loss on ignition (LOI); detailed description of the topsoil (0–15 cm) sampling and analysis is available elsewhere (Emmett et al., 2008, 2010) and summarised below.

Sampling of topsoil (0–15 cm) was carried out on 2 m × 2 m "soil plots" located at the centre of randomly placed 200 m^2 X-plots at 5 locations within each survey square (Wood et al., 2017). Vegetation and biophysical data associated with each soil plot were collected over the area of the associated X-plot. In 2007, the soil plots from the 256 squares sampled in 1978 were relocated using maps and/or markers placed in the 1978 survey and the topsoil resampled. The repeat soil plots are within 2–3 m of the original survey locations, and detailed vegetation and biophysical measurements were taken at the same X-plot locations for both surveys. Loose leaf litter was brushed from the soil surface before sampling in both years. In 1978 soil samples were taken from soil pits, whereas soil cores were sampled in 2007. Cross comparisons between pit and coring methods indicated similar bulk densities were achieved (Emmett et al., 2008) however, since bulk density was not measured in 1978, we were not able to analyse change in stock. Rigorous cross-comparison of laboratory analytical methods employed in the two survey years was also carried out (Emmett et al., 2010). In some cases the original soil plots could not be located or the data from one of the surveys was incomplete, but the resampling effort nonetheless produced 783 "repeat soil plots" with complete observations. The tSOC values from these 783 sites form the core of our analyses here.

Land use was determined using the Aggregated Vegetation Class (AVC) scheme evaluated for each soil plot in each survey year, based on species composition as recorded in the field (Bunce et al., 1999a, 1999b). The AVC classifies floristically well-defined vegetation types where differences in species composition between classes are maximised. On this basis they constitute powerful strata for analyses of plot data, increasing the chances of detecting meaningful land use change. Shifts of individual plots within aggregate classes imply subtle changes due to management and/or gradual environmental change or succession. Transitions between aggregate classes imply major changes in land use, land management or environmental drivers (Bunce et al., 1999a). The eight AVC classes are: 1) crops and weeds; 2) tall grass and herb; 3) fertile grassland; 4) infertile grassland; 5) lowland wooded; 6) upland wooded; 7) moorland grass and mosaic; 8) heath and bog.

All 8 AVC classes appear in the 783 soil plots analysed here. AVC class was used to define land use for each soil plot in a given survey year. Transitions from one AVC to a different AVC defined land use change between survey years. If land use change occurred for a given plot, the exact timing of AVC transitions within the 29 years between surveys is not known. Of the 783 soil plots analysed, 504 plots recorded no land use change (same AVC in both years) and 279 plots recorded AVC transitions, implying major land use changes had occurred (Supplementary Table S4). There are 56 possible pairwise AVC transitions that can occur. To provide replication in our analyses of land use change, we only included AVC transitions that occurred on three or more soil plots. This resulted in 263 soil plots representing 21 different AVC transitions for our analysis of land use change.

Climate data at 5 km resolution were processed for the five years preceding each survey year (Met Office, 2014). Mean seasonal temperatures (°C) were derived from monthly data, to account for impacts on plant growth and tSOC decomposition. Annual average growing degree days (GDD), were calculated to best reflect impacts on NPP. Mean seasonal precipitation values (mm) were included as surrogates for soil moisture content and associated controls on decomposition rates and moisture limitations on vegetation growth. Change was calculated as the difference between the two periods.

Atmospheric deposition data at 5 km resolution, to account for potential soil acidification and fertilisation impacts on tSOC decomposition and plant growth, were obtained from Concentration Based Estimated Deposition (CBED) (Dore et al., 2015). Due to CBED data limitations the deposition values associated with the 1978, 1998 and 2007 surveys are from 1987, 1997 and 2005 respectively. Average annual deposition of sulphur and inorganic nitrogen (kg ha^{-1} yr^{-1}) were calculated for each countryside survey 1 km × 1 km square. To account for the varying deposition rates over different land use types (arable, forest, moorland and grassland) countryside survey squares were assigned the deposition rate for the land category which best matched the dominant land use type for that square (i.e. a square which was dominated by coniferous woodland was assigned the deposition estimate associated with forest).

Soil parent material data was obtained from British Geological Survey (BGS), and key descriptors of soil group and $CaCO_3$ rank were included as terms for model selection, to account for soil texture and mineralogy (British Geological Survey, 2010).

2.2. Modelling approaches

Models of spatial patterns of tSOC as a function of local and global drivers were constructed for each survey year, and a combined spatial model was constructed using data from both survey years. A further model was constructed for 2007 using only drivers available at national scale, to allow construction of a predictive map (see Supplementary Fig. S4). Of the 504 soil sites with no land use change, 472 had full data for all required variables, and were used for these spatial models. An additional model of temporal changes in tSOC between the 1978 and 2007 surveys was also fitted to the drivers using the 472 constant land use soil sites. To account for random factors, we used a mixed model structure (Generalised Additive Mixed Model, GAMM). The countryside survey square (each of which contained up to 5 soil plots) was included as the random factor in the separate year models, and survey square nested within survey year in the combined spatial model. Importance of selected discrete variables was then assessed against residuals of this model, using Kruskal-Wallis rank sum tests, and those with a significant relationship were put into the main model. All models were constructed and tested in R (R Core Team, 2013).The full list of input variables for the spatial model is listed in Table S1. Due to the bimodal distribution of tSOC, Gaussian models were not appropriate, and a Tweedie distribution was used for model fitting.

The distribution of the data is important for interpretation of the models as well as statistical fit. The two peaks in the data represent organic soils and mineral soils, and datapoints in between could be split into humus-mineral and organo-mineral, as per previous reporting (e.g. Emmett et al., 2008). Although soils exist as a continuum across these categories, the groupings are instructive conceptually due to varying processes controlling measured tSOC change. Since organo-mineral and humus mineral soils generally consist of an organic later over mineral soil, the measured tSOC will reflect spatial or temporal variation in the depth of the organic layer, rather than variation in concentration per se. Whilst organo-mineral soils make up a relatively small proportion of the dataset, humus-mineral are more significant (6–7% and 35–38% respectively, across CS as a whole). Short range variation is of particular importance for monitoring these soils, but impacts on data should not contain bias, as discussed in supplementary material. It is also important to consider that organic soils, which will include deep peats, are often subject to loss of depth rather than C concentration, which will not be identified by modelling or monitoring tSOC. Because of these conceptual differences, splitting the soils into these categories for model construction was considered, however the impacts on sample size outweighed any potential for improvement in the ease of interpretation of the model. This issue also applies for any other potential subsetting of the data, e.g. by vegetation type; although the dimensionality of the problem is reduced, the reduction in sample size reduces the statistical power of findings. Furthermore, this work seeks to identify influence of large scale drivers across habitat and soil types.

As part of model fitting, non-linear smoothly varying functions of covariates, henceforth termed "smooths", were fitted to all model terms and deviations away from a constant zero effect were tested. Smooths allow the model to vary the coefficient applied to the covariate of interest in a non-linear manner. Smooths were applied via the gamm function in the 'mgcv' library (Wood, 2011). In order to test the importance of our variables as predictors, we applied a double penalty smoother approach, which allows the penalized regression routine that selects for the "wiggliness" of the smooths to also shrink spurious covariates out of the model entirely (as demonstrated by Marra and Wood, 2011). To do this, we applied shrinkage smoothers using cubic regression splines for each term. Two dimensional tensor product smooth interactions (Wood, 2006) were applied to the spatial variables (combined latitude and longitude) to account for large scale spatial variation in the data. Variables which were not important (F = 0 and no statistically significant effect) were removed when the discrete variables were added to produce the final model, to enable the models to converge. We did not include interaction terms in the models, in order to assess the marginal influence of individual covariates.

It's important to note that there were statistically significant correlations between most of the variables tested here (Supplementary Table S2). The underlying spatial co-variance driving these co-correlations between variables has been minimised and mitigated by inclusion of the purely spatial variables within the model. The use of double penalized smooths allows the model to fit nonlinear partial responses for each variable, and thus to only include variables so far as they improve the model. This approach should create the model which best fits the data, but nevertheless, as with all correlative studies care should be taken with interpretation.

For model validation, we applied the 2007 model generated on the CS data to predict topsoil C concentration for the independent LUCAS (Toth et al., 2013) dataset (sampled in 2009). An additional spatial model for topsoil C concentration was created for mapping across GB at a 1 km grid resolution, using only variables that were available at a GB scale (Supplementary Table S3, Fig. S4).

3. Results and discussion

3.1. Land use change effects

Replicated (n ≥ 3) land use change occurred at 263 of the re-sampled CS soil plots between 1978 and 2007. These plots were used to provide estimates of the direction and magnitude of tSOC change for 21 different land use transitions (Fig. 1, Table S5). The pattern of change in tSOC with change in AVC becomes more apparent if the observed land use changes in Fig. 1 are aggregated into broader directions of land use transition (Fig. 2, Table S5).

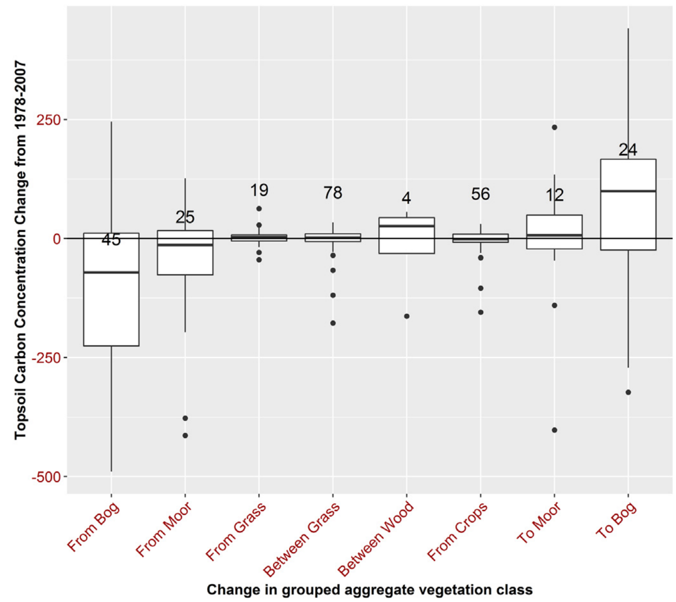

Fig. 2. Impacts of land use change on tSOC concentration at 263 CS soil plots grouped according to directional changes in aggregate vegetation class (AVC) that occurred between 1978 and 2007 (box: 25th (lower), 50th (median), 75th (upper) percentiles; whiskers extend to highest value within ±1.5 interquartile range of these; outliers plotted as points). AVC is assigned from ordination of vegetation characteristics. Moorland grass mosaics are typically upland habitats, consisting of grassy vegetation and dwarf shrubs. Only those AVC transitions that occurred on 3 or more soil plots are included. These data have been aggregated from Fig. 1 as indicated in Supplementary Table S5.

Fig. 1. Impacts of land use change on tSOC concentration at 263 CS soil plots grouped according to pairwise changes in aggregate vegetation class (AVC) that occurred between 1978 and 2007 (box: 25th (lower), 50th (median), 75th (upper) percentiles; whiskers extend to highest value within ±1.5 interquartile range of these; outliers plotted as points). AVC is assigned from ordination of vegetation characteristics, hence species changes can create a transition from upland to lowland woodland. Moorland grass mosaics are typically upland habitats, consisting of grassy vegetation and dwarf shrubs. Only those AVC transitions that occurred on 3 or more soil plots are included, giving 21 different land use changes in the analysis.

Past studies have suggested soils with larger carbon stocks have greater potential for SOC loss (Crowther et al. (2016)). That pattern is also observed here with larger losses of tSOC at CS sites converted from heath and bog (ranging from losses of 209 g/kg to 53 g/kg) (Figs. 1 and 2). Comparatively, transitions at cropland and grassland sites where carbon stocks are generally lower were associated with much smaller changes in tSOC (ranging from 22 g/kg loss for crops and weeds to tall grass and herbs to 6 g/kg gain for tall grass and herbs to fertile grassland).

Losses of tSOC are generally seen in organic-rich soils where land use change has led to a greater abundance of grasses i.e. along the gradient from bogs to moorland grass mosaics (less boggy habitats consisting of grassy vegetation and dwarf shrubs) and infertile grasses. These land use transitions may reflect management changes in e.g. grazing intensity, drainage or changes in atmospheric N deposition or temperature, both of which have been shown to shift the species composition of bog towards grassier species (Bobbink et al., 1998; Barraclough et al., 2015). The observed tSOC changes are in line with expectations based on soil nutrient status and decomposability of OM inputs from grasses to soil. Both transitions are therefore likely to represent both an increase in proportion of C inputs from readily decomposable grasses and an increase in soil oxygen and nutrient status.

Surprisingly, transitions from woodland to heath and bog had tSOC increase of similar magnitude (74 g/kg) to the transition from moorland grass mosaics to bog, highlighting that woodlands do not necessarily increase SOC in these surface layers as is often assumed. The shift in hydrological conditions associated with bog formation or loss over-rides any increase in SOC driven by the presence of trees and higher rates of primary productivity. However transitions in both directions between woodland and the moorland grass mosaic habitat always lost tSOC perhaps indicating management practices with these transitions enhanced decomposition or loss of litter or topsoil.

Although grassland to forest transitions tends to result in C loss initially (Poeplau et al., 2011), or no change for broadleaved (Guo and Gifford, 2002), our data show a trend for increased tSOC. This may reflect sampling depth, since afforestation of grassland has been shown to generally lose aggregated C from subsoil (30–80 cm, which is not captured here), but gain tSOC in surface layers from increased organic matter (OM) input (Poeplau et al., 2011,Poeplau and Don, 2013). SOC loss from deeper layers on afforestation may also be significant, since the N balance of forests has been shown to suggest mining of soil N (Emmett et al., 1997). For afforestation, forest type and management are also important (Guo and Gifford, 2002; Pérez-Cruzado et al., 2011). These issues likely contribute to variance in our data and disagreements in the literature over influence of afforestation on soil C stocks for grasslands.

Change in tSOC with transition from upland wooded to lowland wooded species had a large variance which reflects diverse species composition within both classes. (It should be noted that the reported change of upland to lowland woodland vegetation class is based on the increased presence of ground flora species more commonly associated with lowland habitat types). Crops and weeds to lowland wooded was accompanied by a small increase in tSOC, in line with findings from meta-analyses (Guo and Gifford, 2002). Evidence from (Poeplau and Don, 2013) suggests there may be no increase in SOC for horizons deeper than the sample for this transition.

Potential influences on topsoil C from land use which cannot be accounted for by this analysis include; changes taking place before the first survey which could have significant impact on changes during the survey as soil pools move to a new equilibrium (as also noted for the NSI data Smith et al., 2007), annual or other rotational changes which mean a consistent land use should not really be assigned, or changes in management such as reductions in manure inputs or incorporation of crop residues (Smith et al., 2007).

It is important to note that changes below the top 15 cm of soil which are not measured here may be significant, and in some cases might reverse the direction of observed change (Chapman et al., 2013). Erosion associated with land use change, or changes in topsoil density may also create spurious results (Smith et al., 2007). Future surveys are including changes in bulk density and visual evidence of erosion which will enable these issues to be explored.

3.2. Climate and pollution drivers of tSOC

For 472 of the re-sampled CS soil plots there were no land use changes recorded between 1978 and 2007, and all required variables were available for modelling. The distribution of tSOC on these plots was largely unchanged between the two years (Fig. 3a), with a slight decrease in tSOC at sites with very low carbon and slight increase in tSOC at sites with very high carbon. However, from visual comparison, there was some change in the distribution of a number of global (climate, pollution) and local (soil pH) drivers of tSOC between 1978 and 2007 (Fig. 3b–h).

SO_4 deposition has reduced significantly across the UK (RoTAP, 2012) and for our sample (Fig. 3b) between 1978 and 2007. Declining deposition allows recovery from acidification, with pH rising at rates dependent on soil buffering capacity, rates of weathering and other site factors. This recovery from acidification due to high deposition in the 70s explains the drastic change in pH distribution from a peak in acidic topsoils in 1978 to a broader distribution with a greater number of neutral topsoils in 2007 (Fig. 3c). This is reflected in the statistically significant increase in mean pH previously reported across the countryside survey data (Reynolds et al., 2013). Similar pH recovery patterns have been observed elsewhere in Europe, and the US over recent decades (e.g. Driscoll et al., 2001; Menz and Seip, 2004; Reinds et al., 2009; Lawrence et al., 2012). Associated change in SOC has only been shown for some sites (e.g. Lawrence et al., 2012). For peat, recovery from acidification leads to desorption and increased solubility of dissolved organic carbon (DOC) (Evans et al., 2006; Kipton et al., 1992) which may explain large increases in export to streams in Europe and North America (Monteith et al., 2007).

There has also been some decline in NO_3 and NH_4 deposition at CS sites over the study period (Fig. 2d–e) in line with the national trend; combined N deposition decreased by 17% between 1970 and 2005 in the UK, according to outputs from FRAME, as published in RoTAP (2012). This may have led to some reduction in C inputs associated with the N fertilisation effect increasing NPP (Magnani et al., 2007; de Vries et al., 2009; Tipping et al., 2017), but may also have led to vegetation changes and reduction in decomposability of OM inputs at oligotrophic bogs (Bobbink et al., 1998; Bragazza and Freeman, 2007).

Metrics of temperature (seasonal means and GDD) have increased between 1978 and 2007 (Fig. 3f–j). The change for precipitation (Fig. 3k–n) is less dramatic, but indicates some shift towards higher spring and summer precipitation for our sample. The impacts of these changes on soil carbon will be dependent on the balance of the effects on decomposition and NPP (Kirschbaum, 2000), and site factors including baseline precipitation regime which affect soil moisture status in relation to the response curve (Skopp et al., 1990).

3.3. Models of spatial patterns of tSOC

To explore the shift in the importance of the local and global drivers on spatial models of tSOC, we compared spatial models of tSOC across time periods for the 472 re-sampled soil plots with consistent land use and all available data. Models were constructed using the data for each of the years (1978, 2007) individually and also combined as a single dataset (Table 1). The drivers are all co-correlated, which complicates the process of interpretation (Table S2). The 2007 model was validated against the independent LUCAS (Toth et al., 2013) topsoil dataset (sampled in 2009) achieving r2 of 0.45, in spite of differences in sample depth between the surveys (LUCAS is 0–20 cm, compared to the CS 0–15 cm).

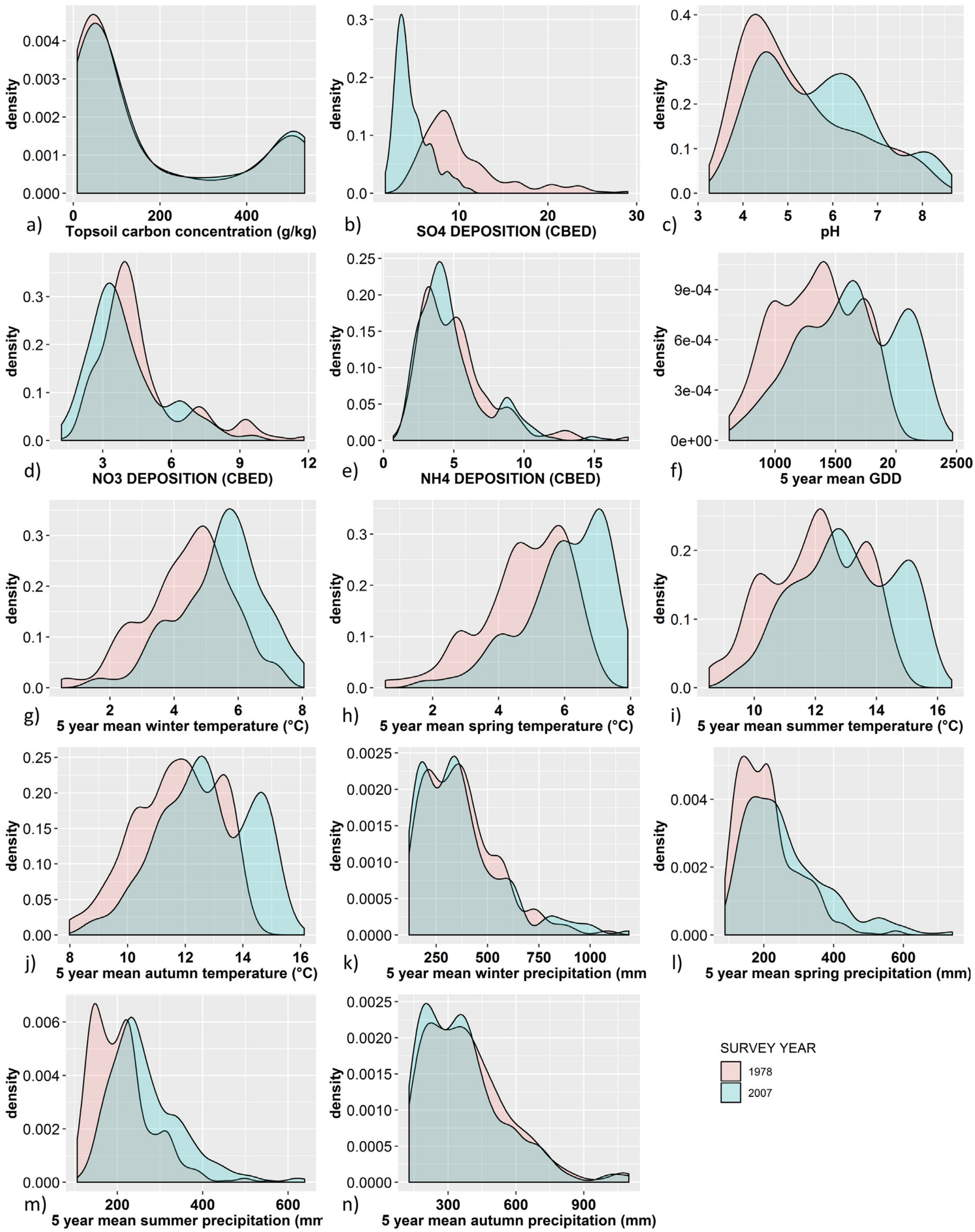

Fig. 3. Distributions of topsoil carbon and local and global drivers of tSOC change at 472 CS soil plots sampled in 1978 and 2007 for which no land use change was recorded.

The model results indicate that Broad Habitat and pH had the strongest relationships to tSOC in all three models. Climate metrics were important in all three models, however there was variation in the variables which best explained tSOC; in the combined years model summer temperature and precipitation and winter precipitation were all important, whereas in the 1978 model autumn temperature was selected, and in the 2007 model summer temperature was the only statistically significant climate metric. Growing degree days metric was not selected for any of the models, indicating that the seasonal climate metrics better explain the spatial and temporal pattern of tSOC. In the combined years model, SO_4 deposition was the second most important variable, but was not significant in either the 1978 or 2007 models. NH_4 deposition was significant in both the combined years model and the 2007 model. Spatial location was important in all three models.

3.3.1. Shifted relationship between soil pH and tSOC

The spatial relationship between pH and tSOC changed between 1978 and 2007 (Fig. 4). The relationship shifts upwards in 2007, particularly in acid soils. This reflects an increase in pH at low tSOC soils (reducing the number of these soils in the acidic range) as well as the

Table 1
Spatial models of topsoil carbon in 472 CS re-sampled soil plots where no land use change was recorded between 1978 and 2007. Variables are listed in order of descending F statistic (whilst direct comparison of F statistics is not statically robust due to differences in the estimated degrees of freedom for each variable, it is adopted here purely to serve as a guide to relative importance rather than for formal testing or comparison).

Model	1978		2007		Combined	
Goodness-of-fit	$r^2 = 0.67$, n = 472		$r^2 = 0.779$, n = 472 Validation against LUCAS: $r^2 = 0.45$		$r^2 = 0.711$, n = 944	
Parameter	F	p	F	p	F	p
pH	4.380	1.02e-08***	13.556	<2e-16***	11.955	<2e-16***
SO_4 deposition					2.775	1.02e-06***
Spatial location (easting, northing)	1.748	4.04e-08***	1.020	6.09e-06***	1.742	3.09e-08***
NH_4 deposition	1.101	0.004114**			1.413	0.000408***
Summer temperature (5 year mean)			6.521	3.79e-15***	0.831	0.003212**
Summer precipitation (5 year mean)	0.000	0.526366	0.000	0.5491	0.829	0.016122*
Winter precipitation (5 year mean)					0.774	0.006402**
Spring precipitation (5 year mean)					0.000	0.281051
Winter temperature (5 year mean)	0.081	0.102278	0.441	0.0145*	0.000	0.577801
Autumn temperature (5 year mean)	1.247	0.000496***				
Autumn precipitation (5 year mean)			0.051	0.2287		
Parametric terms (complete inclusion or exclusion based on significance against residuals)						
Broad habitat	16.01	<2e-16***	14.78	<2e-16***	16.346	<2e-16***
$CACO_3$ rank	Not significant against model residuals		Not significant against model residuals		3.104	0.00516**
Soil group	Not significant against model residuals		Not significant against model residuals		1.884	0.01624*

Significance codes: <0.001=***; 0.001 - 0.01=**; 0.01 -0.05 =*.

small increase in sites with high tSOC soils noted in Fig. 3a. The inflection point of the graph also shifts towards higher pH in 2007. There was a stronger relationship between SOC and pH in 2007 ($r^2 = 0.57$) than in 1978 ($r^2 = 0.39$). At pH 7, the relationships converge and tSOC for a given pH was broadly similar between 1978 and 2007.

3.3.2. Climate and air pollution effects on tSOC

Fig. 3 shows that over the study period SO_4 deposition has declined significantly, which may have led to the observed shift in pH towards neutral soils. The distribution of tSOC has remained relatively constant, and as a result, the relationship between pH and SOC in UK topsoils has shifted between 1978 and 2007, as shown in Fig. 4. This shift may represent hysteresis creating a lag in the response of tSOC to acidification or recovery, or alternatively may indicate a shift between stable relationships in response to changes in other drivers.

In the models (Table 1), it was expected that SO_4 deposition would have a positive relationship to tSOC, reflecting the mechanism of acidic deposition lowering topsoil pH and slowing decomposition resulting in increased tSOC. However, SO_4 deposition was not important in either

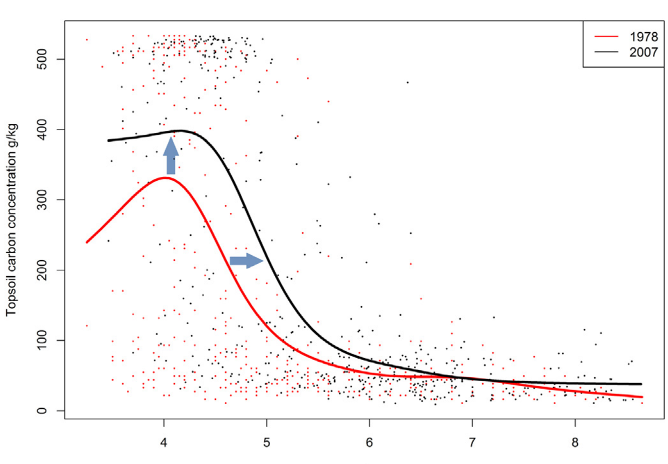

Fig. 4. Comparison of the relationships between soil pH and tSOC for 1978 and 2007 for 472 sites with no land use change between the years. Relationships plotted are for a model of tSOC from pH, fit with smooth to account for nonlinear relationship: (r^2 1978 = 0.39, 2007 = 0.57). Arrows indicate the shift of the pH tSOC relationship between 1978 and 2007 towards higher pH, and an increase in SOC relative to pH.

the 1978 or 2007 models, whilst in the combined model there was a negative relationship to tSOC. Therefore, the importance of SO_4 in the combined model suggests that the spatial pattern of SO_4 deposition accounts for some of the shift from 1978 to 2007 in the relationship between tSOC and soil pH (since year is accounted for as a random factor). There is a steep portion of the partial response curve at the high end of the SO_4 deposition, which only occurred in the lead up to 1978 (Supplementary Fig. S3f). We infer that these areas had been acidified due to high deposition in previous decades, and had lower carbon relative to inherently acidic sites (resulting from underlying geological parent material) in 1978. This is also seen in Fig. 4; the trend line in acidic soils was much lower in 1978, which may reflect recent acidification of these soils through high SO_4 deposition, without associated tSOC accumulation; this supports our interpretation of the combined model. A shift towards higher pH at these sites by 2007, with little change in tSOC suggests recovery from acidification has returned them to a more natural relationship between pH and tSOC. Fig. 4 also shows some soils with high tSOC are experiencing an increase in pH, and these may be vulnerable to SOC loss due to reduced pH suppression of decomposition. Thus it appears the response of UK soils to acidification (and subsequent recovery) may be more complex than the C sequestration recently reported for acidified croplands in China (Zhang et al., 2020).

The strong relationship between pH and tSOC may in part reflect the shared scale of measurement, and include influence of drivers which affect both pH and SOC. pH has strong negative correlations to precipitation and deposition metrics, and positive correlation to temperature metrics (see Supplementary Table S2). These co-correlations between drivers make it more difficult to identify direct causal relationships. In our dataset, acid topsoils tended to have higher precipitation (anoxia slows decomposition), and were colder (low temperature slows decomposition), with generally higher deposition (N inputs increase NPP and associated C inputs). Therefore the observed strong relationship between pH and tSOC may reflect suppression of decomposition in acid soils, but may also represent influence of these other variables.

Climate variables were less important than pH (lower F value) but were statistically significant in spatial models of tSOC for 1978 and 2007 and the combined years model. Previous work by Kühnel et al. (2019) explored relationships between seasonal climate variables and change in SOC, and found evidence suggesting that rising winter temperature and autumn precipitation increased decomposition, whilst increased temperature and precipitation in spring and summer increased

C inputs from vegetation, leading to tSOC accumulation. For our data sample, temperature and precipitation have similarly increased in spring, summer and winter, however autumn precipitation has decreased, leading to warmer but dryer soils during this period. Unlike the study by Kühnel et al. (2019), we found negative spatial relationships between summer temperatures and tSOC, suggesting that this metric is more important for decomposition than NPP. This has previously been seen in a review of laboratory and field experiments (Kirschbaum, 2000) and for annual temperature metrics in several studies across Europe (e.g. Riley and Bakkegard, 2006, Sleutel et al., 2007, Heikkinen et al., 2013). By allowing nonlinear relationships in our models, we can identify thresholds in these patterns for our sites.

The partial response curve (supplementary Fig. S3l) shows that in the 1978 model, the coefficient for autumn temperature decreases following a sigmoid curve, which is steepest between 10 and 12 °C, indicating greater change in the relationship to tSOC in this region, and flattens out above 12 °C where further warming does not affect the relationship to tSOC. Fig. S3q shows that in the 2007 model the coefficient for summer temperature followed a similar decreasing sigmoid curve, which flattens out above 14 °C, again indicating that above this temperature, further warming did not affect the relationship to tSOC. This may reflect the fact that the shift towards higher summer temperature between 1978 and 2007 for our sample (as seen from the large increase in sites >14 °C in Fig. 3i) has not been accompanied by reduction in tSOC at these sites. However, in the combined years model, the partial response curve for summer temperature (Fig. S3d) shows a linear negative relationship to tSOC, which does suggest that across the dataset, warmer sites do in general have lower tSOC. It is important to note that our data may reflect hysteresis in response to warming, which may create artificial apparent thresholds in the 2007 model, and that much larger changes in temperature metrics are forecast (IPCC Climate Change, 2014) which may lead to greater response in the future.

Precipitation is known to be one of the most important controls on SOC (Jenny, 1980) and has been shown to be effective in modelling SOC and other soil properties (Gray et al., 2009; Kühnel et al., 2019). However, control is via soil moisture, which is affected by many other factors such as evapotranspiration, geology and topography, which will reduce the strength of the relationship. Additionally, soil saturation has nonlinear effects on vegetation growth and decomposition rates, hence impacts of change are dependent on the initial saturation of soil (Skopp et al., 1990). This may explain why precipitation metrics were not statistically significant in the model for 1978 or 2007. Summer and winter precipitation were statistically significant in the combined spatial model; the partial response curves initially increased with increasing precipitation, levelling out at higher winter precipitation, and following a hump shaped curve to decrease at higher summer precipitation (Supplementary Fig. S3b–c). This may suggest that the small increase in sites with >800 mm winter precipitation (Fig. 3k) has not led to increase in tSOC. The increase in sites with wetter summers (Fig. 3m) observed for our sample correlates with lower than expected tSOC, with the partial function falling in this region (>350 mm), which suggests that the increase in sites with very wet summers may have led to tSOC losses. The partial response plot (Fig. S3c) indicates few data points in this region, which combined with possible hysteresis in soil response to the changing conditions may limit potential to draw conclusions.

Precipitation is strongly co-correlated with pH, which is sampled at the same spatial scale as tSOC, and therefore may account for the apparent unimportance of precipitation metrics in the individual year models. Precipitation also has positive co-correlation with deposition variables and negative relationship to temperature variables, which may all affect the observed relationships in the models.

NH_4 deposition was significant in 1978 and combined year models, with similar partial response functions, showing positive trends at low and mid-range of deposition. This could suggest that the spatial pattern of NH_4 deposition may have contributed to SOC accumulation in 1978 through increased NPP (as. per. Magnani et al., 2007, de Vries et al., 2009, Tipping et al., 2017), which might explain some of the variation between the surveys. However in the 1978 model the coefficient decreased in the high deposition range above 10 kg ha^{-1} yr^{-1}, whereas in the combined years model the coefficient levelled out in this data range. This suggests that sites with recent higher deposition in 1978 had not accumulated tSOC, and the slight shift towards lower deposition in 2007 does not appear to have caused any loss of tSOC. Positive correlation with SO_4 deposition, precipitation and mean temperature may mean that, in the models, these other variables could account for some of the influence of N deposition. Equally, co-correlation with SO_4 deposition may explain why the partial correlation coefficient for NH_4 decreases at high deposition in the 1978 model. Negative co-correlation with pH may mean that the observed relationship in the combined years model reflects a balancing out of lower than expected tSOC at acidic sites in 1978. The effect of N deposition was estimated to be ca. 10% SOC increase over 160 years in the UK data set compiled by Tipping et al. (2017) which included a subset of CS sampling locations. Therefore declining deposition will affect NPP and as well as pH and decomposition but at a far slower rate (0.06% pa) than promoted under the 4 pour mille initiative, which may not be easily detected in models or indeed by monitoring programmes such as CS.

3.4. Modelling topsoil carbon change

The process of identifying potential drivers of topsoil C change may be complicated by interactions between drivers, and nonlinearity of relationships, as well as the issue that many drivers affect both inputs and losses of C from topsoil. Land management changes have been shown to be useful in understanding and modelling SOC change elsewhere in Europe (e.g. Goidts et al., 2009; Kühnel et al., 2019). For the UK, changes in land management which are known to have occurred nationally over the study period, for which site level data are not available, further complicate the process of identifying influence from large scale drivers, but may be partially accounted for by the inclusion of habitat type in the models (Smith et al., 2007). Modelling by Manning et al. (2015) showed that impacts of cutting, grazing and fertiliser regimes did not help to explain national level variation in SOC, which were better explained by vegetation indices such as those collected here.

Understanding of site level change is further complicated by small scale variation, meaning that repeat samples will capture variation within a field or between horizons (particularly if erosion or tillage has occurred) in addition to change over time. A viable tSOC change model relating changes at the soil core scale to changes in drivers at 1 km scale is reliant on a change signal which is greater than the noise associated with resampling error. Although the validation of the 2007 spatial model against LUCAS data showed good performance, it would not be sufficient to pick up the 0.4% per year level of change in SOC of interest for the 4 pour mille initiative for site level changes (Rhodes, 2019). Indeed, Smith et al. (2010) note that for National Soil Inventory Scotland sites without land-use change, the uncertainty in current measurements exceeded recorded change over a 19–31 year period. However, site level errors can be expected to average out across a sufficiently large sample, hence the use of statistical approaches across a large dataset like CS, which are designed to detect national and habitat level trends in soil properties for national-scale reporting. At this level they have proved useful in demonstrating changes such as a 0.4% loss a year from well-mixed arable soils between 1978 and 2007 (Reynolds et al., 2013). However for site level changes, short range variability for many unimproved soils, combined with the current slow rates of change over our study period, limits the power of this type of dataset to use change models to identify potential drivers without major additional investment in sampling intensity. There is potential for rapid changes in upland soils driven by climate change extremes, which may result in a larger signal more easily detected in the future.

Nonetheless, we tested whether site level change and co-located driver data can be used to identify a model of tSOC change, assuming that error is equally distributed about the mean, and the large number of soil plots could enable identification of statistically significant effects. Using the 472 re-sampled soil plots with consistent land use and all required metrics, we attempted to develop a mixed model of the temporal changes in tSOC between the 1978 and 2007 (see supplementary material). Despite the robustly significant models of tSOC spatial pattern in each of the individual years, it was not possible to derive a useful model based on the observed changes between the years. However, the changes in relative importance of the drivers between the 1978 and 2007 spatial models provides evidence and insight into the covariation and shifting balance of the drivers of tSOC change, and re-enforces the assertion that no detectable change in tSOC does not necessarily mean no significant dynamics affecting tSOC.

It is critical to improve understanding of the impacts of the key drivers of change in SOC given projected global changes. Findings will support development of new mechanistic models in terms of process representation (e.g. work by the International Soil Modelling Consortium (Vereecken et al., 2016), or the MEMS model (Robertson et al., 2019)), as well as helping to identify possible risk factors for soil carbon loss, which will enable targeting of more data and time intensive assessments. Improved understanding of drivers, controls and areas at risk of soil C loss can also improve sustainability of soil management (Nisbet, 2007) and may help to meet the UN State of Soils ambition for more sustainable management of soils, the UN Sustainable Development Goal 15.3 to avoid land degradation and the 4 pour mille initiative (Rhodes, 2019).

CRediT authorship contribution statement

A. Thomas: Conceptualization, Methodology, Validation, Formal analysis, Writing - original draft, Writing - review & editing. **B.J. Cosby:** Writing - review & editing, Supervision, Project administration, Funding acquisition. **P. Henrys:** Methodology, Supervision. **B. Emmett:** Conceptualization, Methodology, Writing - review & editing, Supervision, Project administration, Funding acquisition.

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 data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.138330.

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