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Long-term organic carbon turnover rates in natural and semi-natural topsoils

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Abstract

We combined published and new radiocarbon and ancillary data for uncultivated topsoils (typically 15 cm depth), to make two databases, one for the United Kingdom (133 sites), and one global (115 sites). Forest topsoils are significantly higher in radiocarbon than non-forest soils, indicating greater enrichment with "bomb carbon" and therefore faster C turnover, if steady-state conditions are assumed. Steady-state modelling, taking into account variations in atmospheric $^{14}CO_2$, including the effects of 20th century nuclear weapons testing and radioactive decay, was used to quantify soil carbon turnover rates. Application of a model with variable slow (20 yr mean residence time, MRT) and passive (1000 yr MRT) carbon pools partitioned the topsoil C approximately equally, on average, between the two pools when the entire data set was considered. However, the mean slow:passive ratio of 0.65:0.35 for forest soil was highly significantly different ($p < 0.001$) from the 0.40:0.60 ratio for non-forest soils. Values of the slow and passive fractions were normally distributed, but the non-forest fractions showed greater variation, with approximately twice the relative standard deviations of the forest values. Assuming a litter input of 500 g C m^{-2} a^{-1}, average global C fluxes (g C m^{-2} a^{-1}) of forest soils are estimated to be 298 (through a fast pool of MRT 1 yr), 200 (slow pool) and 2.0 (passive pool), while for non-forest soils, respective average fluxes of 347, 150 and 3.3 g C m^{-2} a^{-1} are obtained. The results highlight the widespread global phenomenon of topsoil C heterogeneity, and indicate key differences between forest and non-forest soils relevant for understanding and managing soil C.

Keywords Carbon, modelling, radiocarbon, soil, turnover

Introduction

Terrestrial soil organic matter (SOM) derived from dead biomass accounts for the largest global pool of organic carbon, totalling 2300 Pg (Jobbagy & Jackson, 2000) and therefore greater than both the oceanic pool of 1000 Pg, and the living terrestrial biomass pool of 600-1000 Pg (Falkowski et al., 2000). It comprises a range of organic material at various stages of decomposition and stabilisation, from recently-deposited labile plant material and senesced microbial biomass with fast turnover (seconds to years), to more stable material turning over on decadal to millennial timescales (Trumbore 2000, 2009; Amundson 2001). Quantification of SOM turnover with respect to environmental conditions and litter quality and quantity is crucial to understanding the resilience of soil C to perturbations such as climate change and land use conversion in the longer term and at the global scale (Jenkinson et al, 1991; Kirschbaum, 2000; Schlesinger & Adams, 2000; Smith et al., 2008; Schmidt et al, 2011). The response of topsoil SOM is of particular importance since it is in close contact with the atmosphere and the least stabilised against decomposition.

Much of the carbon entering soil is respired quickly, within months or a few years, and can be studied with relatively short term experiments and observations, leading to detailed understanding (Melillo et al., 1982; Berg and McClaugherty, 2008). However, the bulk of soil carbon is in slowly-cycling SOM pools and far less amenable to experimental investigation. The most generally-applicable approach at these longer timescales is the determination of soil radiocarbon and analysis of the data by modelling, usually assuming steady-state conditions to apply. Advantage is taken of the long half-life of naturally-formed ^{14}C (5730 years) to estimate centennial or millennial turnover, while the anthropogenic bomb-^{14}C spike of the mid 20th century provides estimates of C flow on decadal timescales (Gerasimov, 1974; O'Brien & Stout, 1978; Harkness et al., 1986; Harrison, 1996; Trumbore, 2000, 2009; Torn et al. 2009). The technique is insensitive to the fast passage of C through small, short-lived litter pools. According to Amundson (2001) and Torn et al. (2009), radiocarbon measurements show that considerable amounts of soil carbon are in quite stable pools, turning over much more slowly than is implied by simply taking the quotient of soil respiration and the total soil C pool, which gives soil C residence times of 10-90 yrs for all biomes except tundra and wetlands (Raich & Schelsinger, 1992). To understand and manage soil organic matter stability and nutrient cycling, and improve quantification of land-atmosphere CO_2 exchange, this heterogeneity of soil carbon with respect to turnover needs to be characterised at regional and global scales

A general, globally representative, description of SOM turnover based on radiocarbon is lacking, because most applications to date have been at the plot scale, to investigate turnover of different pools in different soil horizons (see e.g. O'Brien and Stout, 1978; Harkness et al., 1986; Richter et al., 1999; Gaudinski et al., 2000; Leifeld et al., 2009; Schulze et al, 2009; Tipping et al., 2010, 2012a; Baisden et al., 2013). The main attempt to synthesise data was by Harrison (1996), who amalgamated radiocarbon results from *c*. 50 studies on different soils, sampled during the period 1957 to 1991, and applied a steady-state model, representing SOM with a passive pool (turnover 6500 years) and an active pool (25 years). Further survey work includes studies on (a) soils under single vegetation types (Tipping et al., 2010; Fröberg et al., 2011; Harrison et al., 2000), (b) an altitudinal sequence (Leifeld et al., 2009), (c) a climatic transect (Frank et al., 2012), and (d) zonal soils of European Russia (Brovkin et al., 2008). However, in the last two studies only parts of the topsoil were analysed, mineral soil in the first case and isolated humic acid in the second.

Here, we report a systematic application of a two-pool, steady-state model of topsoil carbon turnover to data from c. 250 sites with soils from natural or semi-natural ecosystems. By "semi-natural" we mean that fertilisers have not been applied, and any land-use management such as grazing or forestry is minor, so that the characteristics of a natural ecosystem are maintained. The data were obtained from new measurements on UK soils, together with published results for global sites (including the UK). This generated separate data sets for the UK and global non-UK sites, which were also combined into a single data set for further analysis. We applied a form of meta-analysis to the data, which means that if it can be demonstrated that the sampling locations are unbiased and representative, it is justified to analyse the data by, for example, comparing mean values for sites with different attributes, or by regression of data or derived data against site characteristics such as mean annual temperature (MAT) or pH. This is a widely-accepted approach in environmental research; examples include analyses of N fixation (Cleveland et al, 1999), litter decomposition (Manzoni et al., 2008), carbon-nitrogen stoichiometry (Taylor & Townsend, 2010; Yang et al., 2011), and changes in surface water concentrations of dissolved organic carbon (Monteith et al., 2010).

The first aim was to establish representative topsoil turnover rates, and their ranges, to inform soil and ecosystem modelling at national or global scales. Secondly, we wanted to test the generality of reports that carbon turnover in soils developed under non-forest vegetation is slower than in forested soils (Bol et al., 2000; Tipping et al. 2010; Brovkin et al., 2008).

Finally we explored possible relationships of turnover parameters with the possible driving variables mean annual temperature, mean annual precipitation (MAP), pH, carbon concentration and soil type.

Methods

Soil samples from the United Kingdom

New topsoil samples were collected specifically for the present study from 37 field sites used for field experiments or other ecological research in the UK, none of which have experienced significant land-use change during their known histories (Mills, 2011). In addition, we randomly sub-sampled 59 archived soil samples from Countryside Survey 2007 (Emmett et al., 2010), with vegetation classified as semi-natural. To minimise the possibility of significant past land-use change, we compared contemporary land-use with that recorded at the same location on the 1930s UK land-use map (Stamp, 1932), which classified semi-natural land into three categories: (i) meadow and permanent pasture, (ii) forest, and (iii) heath moor and rough pasture. These classifications were compared with current descriptions, and if the common vegetation class from the two sources was the same, the results were included in the database.

Soil sample collection and analysis followed the protocol of the United Kingdom Countryside Survey conducted in 2007 (Emmett et al., 2008, 2010). In summary, samples were collected using PVC tubes with a length of 15 cm and an internal diameter of 3.8 cm, with one end bevelled to a finer edge for easier ground penetration. Surface vegetation was parted, and the tube placed on the soil surface after removal of any coarse loose litter. The tube was cut into the soil with a sharp knife and then hammered until the full 15 cm was filled with sample, or until impenetrable material was reached. The tube was removed from the soil using pliers, bagged and labelled, and sent to CEH Lancaster where samples were kept at 4°C until analysis. Samples were weighed and their depths measured after careful extrusion from the cores. The soil was then manually homogenised and sub-samples (10 g moist soil) were taken for determination of pH (in deionised water), and loss on ignition (LOI) by heating at 375°C for 24 hours. The remaining sample was air dried, sieved to 2 mm to remove large particles and roots, and weighed for the determination of bulk density (BD). Sub-samples of the sieved soil were ball-milled in preparation for analysis of C and N (Elementar Vario-EL elemental analyser) and radiocarbon. If a sample had a pH > 5.5 it was soaked overnight in 0.5 M hydrochloric acid at room temperature to remove carbonates, before being washed with deionised water and dried. This procedure solubilises little organic matter and so will not have caused significant losses of C. Carbon stocks were calculated from bulk density, %C and the soil layer thickness.

For radiocarbon analysis, sieved soil samples were combusted in a high-pressure bomb in the presence of high purity oxygen, and sample CO_2 cryogenically separated from other combustion products. Isotopically homogenous sub-samples of CO_2 were converted to an iron-graphite mix using iron/zinc reduction (Slota et al., 1987). Determination of ^{14}C was carried out at the Scottish Universities Environmental Research Centre (SUERC) by accelerator mass spectrometry (AMS) using the 0.25 MV Single Stage AMS (NEC, Wisconsin, US; Freeman et al., 2008) or 5 MV tandem accelerator (NEC, Wisconsin, US; Xu et al., 2004). The ^{14}C enrichment of a sample is measured as a percentage of the ^{14}C activity relative to a modern standard (oxalic acid provided by the US National Bureau of Standards, now National Institute of Standards & Technology), where 100% modern is defined as the theoretical atmospheric ^{14}C in AD 1950, in the absence of anthropogenic influence (the Suess Effect). The data are reported as absolute % modern, which involves a mathematical adjustment to account for ongoing radioactive decay of the international reference standard (oxalic acid) since AD 1950 (Stuiver and Polach, 1977). Stable carbon isotope ratios were measured on sub-samples of CO_2 using a dual-inlet mass spectrometer with a multiple ion beam collection facility (VG OPTIMA) to normalise ^{14}C data to –25 ‰ $\delta^{13}C_{VPDB}$.

The new UK data were combined with 40 previously published UK results (see Table S1) to create a UK database of topsoils (all the fine-earth material, including O-horizons) from 136 sites. For 25 sites, radiocarbon data were available for more than one sampling year (Table S1).

Global data set

We collated data on soil carbon pools and radiocarbon from 114 sites, by searching peer-reviewed literature, with some additional values obtained from PhD theses and personal communications (Table S1). Data were only accepted for complete soil samples, i.e. all fine-earth material including O-horizons. Ideally, the following data were required to perform modelling; bulk density (BD), C content, depth of sample (from surface) and the measured ^{14}C content, along with the dates of sampling and analysis. Data were taken only from unfertilised sites with natural or semi-natural vegetation, and for which long-term land-use change was reported as insignificant, or where this could reasonably be assumed to be the case. If C was not reported we assumed that LOI was 55% C (Emmett et al., 2010). If BD was not reported we estimated it from the carbon concentration using the equation; BD = 1.29 exp (-0.206 %C) + 2.51 exp(-0.003 %C) – 2.057 (Emmett et al., 2010; Reynolds et al., 2013); this was necessary for 19 (17%) of the sites. Carbon stocks were calculated from BD, %C and the soil layer thickness. In many cases, data were reported for several soil layers, and the 15 cm (or nearest possible) ^{14}C value was calculated by weighting according to pool size. In a few cases linear interpolation was used to fill gaps in the profile (noted in the data base, Table S1).

Ancillary data

For each site, in both data sets, as many as possible of the following ancillary data were assembled; location (latitude, longitude), MAT and MAP, altitude, soil type, year of ^{14}C sample(s), depth of sample, and soil pH. If climate data were not reported in the source text, location data were used to obtain MAT and MAP from the Oak Ridge National Laboratory database (New et al., 2000). Location data were also used to assign a soil classification, by reference of location within the Harmonised World Soils Database (HWSD, 2012). Information regarding vegetation cover was obtained at either the plant functional type, common or species-name level, and from this information sites were categorised as forest, herb or shrub. We originally had planned to gather data on soil N content, texture, base cation content, and phosphorus, but these were available in relatively few cases, and so were not included in the final collation.

Modelling

Tipping et al. (2010) identified a family of steady-state soil turnover models as follows. Model I estimates the soil C residence time simply from the quotient of soil C pool and total litter C input, i.e. without the use of radiocarbon. In Model II, litter C that is not rapidly recycled enters a single homogeneous topsoil C pool, characterised by a mean residence time (MRT). In Model III, litter C that is not rapidly recycled enters either a slow or a passive pool, each with a defined MRT, fixed *a priori*. In steady-state, the input to the topsoil of C in litter and exudates is balanced principally by gaseous losses (CO_2, CH_4), leaching of dissolved and particulate organic carbon, and erosion. In the case of peats (histosols), the total soil may be accumulating, due to the burial of SOM in the anaerobic catotelm (see e.g. Clymo et al., 1998), but the more aerobic topsoil can still be considered to be in steady-state, with burial considered as an additional loss process.

A single radiocarbon value suffices to calculate the MRT in Model II, or the partitioning of the soil C between the slow and passive pools in Model III (the slow and passive fractions sum to unity). An equivalent to Model III was described, including the naming of the pools, and discussed by Amundson (2001). Harrison (1996) used a version of Model III to analyse topsoil radiocarbon data, referring to the slow pool as "active". Both Models II and III can be used to calculate temporal changes in soil radiocarbon content, making it possible to compare the C turnover characteristics of soils in steady state but sampled at different times.

Model III is more realistic than Model II in that it recognises the heterogeneity of soil C cycling rates. This has been demonstrated by density fractionation which reveals a substantial range in turnover rates even in a defined soil horizon (Swanston et al., 2005; Leifeld et al., 2009; Tipping et al., 2012a), while the different horizons that will often exist within the topsoil will add further variability. The models give somewhat different results, in terms of the simulated temporal variation of soil ^{14}C, and in cases with data at more than one time-point, Model III performs slightly better (Tipping et al., 2010). Therefore in this work we report results with Model III, although for completeness the data base (Table S1) also includes outputs from Model II.

As noted above, some litter C is assumed to enter a fast pool of recent litter which turns over rapidly. Much of the material comprising this pool will be removed during sampling or sample preparation, and so it can be assumed to be negligible in the fine soil analysed for radiocarbon and to determine the soil C pool (See Appendix 3). Calculations were performed using a Microsoft Excel spreadsheet, to track the amount of ^{14}C in the soil annually over the period 1000 AD to the present. A trial value of the steady-state input of C to the soil was chosen, and multiplied by the appropriate atmospheric ^{14}C value to obtain the input of ^{14}C. Atmospheric ^{14}C data for different global regions were obtained from Hua & Barbetti (2004), Levin & Kromer (2004) and Reimer et al. (2004), together with modest forward extrapolations to the year 2008. For both models, the input of ^{14}C is calculated on a yearly basis by taking the product of the fractional replacement of soil C, and the ^{14}C content of litter. The fractional replacement is found by trial-and error, to match the observations, while the litter ^{14}C content is taken to be that of the atmosphere in the current year for herbs, the previous year for shrubs, and for two years earlier for trees, to reflect the turnover of C in the different vegetation types. The loss of ^{14}C in each year is equal to the product of the steady-state C flux and the ^{14}C content of the soil. Then the new soil ^{14}C value is calculated from the change in ^{14}C after adjustment for radioactive decay. The modelled ^{14}C value(s) for the year(s) of sampling were subtracted from the observed values, and the differences squared and summed to obtain the error in prediction, which was minimised by improving the trial input, using the Microsoft Excel Solver function. Inspection of the plotted ^{14}C data enabled initial trial input values to be adjusted, thereby ensuring that steady-state was reached within the period of calculation. The fraction of the slow (or passive) pool can be calculated just from the observed soil ^{14}C content, i.e. knowledge of the soil C pool (e.g. in g C m^{-2}) is unnecessary, but knowledge of the C pool also permits calculations of the input and output fluxes of C, which are equal at steady-state.

Model III results depend upon the choice of turnover times for the slow and fast pools. In previous work (Tipping et al., 2010) we assumed rates of 15 and 350 years to describe soils under deciduous forest. However, for this wider application, greater flexibility in the chosen values was required, and we adopted turnover times of 20 years and 1000 years. Figure 1 shows two examples of the application of Model III, including the variation of atmospheric ^{14}C and the separate traces for the slow and passive pools. When the single-pool Model II is used to analyse soil ^{14}C data, there can be ambiguity in the input rate of C to the soil, i.e. two different MRT values can produce the same contemporary soil ^{14}C value, although invariably one of the possible MRTs has an unrealistically high carbon input rate and can be discarded. Ambiguity does not arise with Model III, although a few soil ^{14}C values give rise to physically-impossible negative pools and inputs (see Results).

Statistical analysis

Model output data from both modelling approaches were analysed using t-tests for differences between forested and non-forested sites after inspection of data for normality using quantile-quantile plotting. Non-normal data were transformed using either log or square root transformations prior to analysis. To explore possible relationships between ancillary variables and modelled output, regression analysis was used, following the same normality checking procedure as for between-groups tests. All statistics were carried out using the computing software R (R Development Core Team, 2010).

Results

The UK data set comprised 63 grassland sites, 38 shrub sites and 35 forest sites, the global set 63 forest, 48 grassland and 3 shrub. Preliminary analysis of the UK data showed no significant difference in turnover parameters between the grassland and shrub soils and so these were treated as a single class, i.e. non-forest, and the same was done for the global sites. The UK and global data sets each provide a broad geographical coverage (Table 1, Figure S1), with forest soils and soils under non-forest vegetation being similarly distributed. Therefore we can justify the assumptions that derived turnover parameters are representative, that data for the two vegetation types can be compared quantitatively, and that regression analysis can be applied to test for relationships between turnover and potential driving variables. At the outset, we considered the UK and global data sets separately, as well as the combined data, because if significant geographical variation in soil C turnover occurred, analysis only of the combined data set, in which more than half the sites are from a small area (i.e. UK), could cause biased results.

Table 2 summarises information about the soils, subdivided by data set and vegetation type. The mean soil depths are similar in all cases, the UK ones showing very little variation because they have been obtained largely through surveys, while global values are more variable because they were largely from site-specific studies. Soil %C is greater for non-forest than forest soils in the UK, but the opposite is true for the global soils, and the values are similar for the combined data set. Non-forest topsoil C pools are about 30% greater than the forest ones in the UK and combined data sets, but there is no significant difference in the global data set. The non-forest soil C pools show greater variability (SD values) than the forested C pools. Mean pH values and ranges are similar across the six categories. The average MAT values are 1-2^{o}C higher at the forested sites in all three data sets, but the difference is only significant for the UK and combined cases. Forested sites are wetter than non-forested ones globally and in the combined data set, but not in the UK. In all three data sets, at low %C (< 10%) the C pool was approximately proportional to %C, but above about 10% there was no dependence, owing to the compensatory effect of variation in bulk density.

We compared the global soils data with the much larger World Soils data set of Batjes (1996), which refers to a soil depth of 30 cm, by calculating the weighted average of the carbon pools of different soil types from the Batjes compilation. The Batjes weighted average was 9.5 kg C m^{-2}, which means that our c. 15 cm global average of 6.1 kg m^{-2} (forest and non-forest soils together) is 64% of the 30 cm value, and this seems reasonable, given the shallower sampling and the tendency of soil stocks to decrease with depth (Batjes, 1996; Jobbagy & Jackson, 2000). In other words, our global data set can be regarded as representative with respect to C stocks of the soil types sampled.

Figure 2 shows radiocarbon data plotted against sampling year, following the approach of Harrison (1996). Three UK sites, each non-forested, sampled in 2007 or 2008 with exceptionally low ^{14}C values (61 - 71% modern) were considered to be outliers, and were omitted from the modelling analysis, reducing the number of sites in the UK data set to 133. The great majority of the data fall between steady-state MRTs of 20 and 1000 years. There is a clear tendency for forest soils to be richer in ^{14}C than non-forest ones, and this is confirmed by comparisons of mean values for similar short time periods (over which inter-year differences can be neglected) shown in Table 3. For each of the 8 separate comparisons the average ^{14}C content is higher for forested than non-forested soils, and in the five cases with the greatest numbers of data, the difference is highly significant ($p<0.001$ or $p<0.02$). If steady-state conditions apply, as we assume in this analysis, the higher concentrations of bomb carbon in the forest soils indicate a faster rate of soil C turnover, independently of any modelling.

We applied Model III to each individual site, and calculated the fractions of the topsoil organic carbon stock contained within the slow and passive pools. Since the slow and passive fractions must sum to unity, we report and analyse the results only in terms of the slow fraction. The greater is the slow fraction, the faster is the overall turnover rate. Taking all soils together, there are broadly equal amounts of modelled slow and passive soil carbon (Table 4), but comparison of the slow fractions for forest and non-forest soils reveals highly significant ($p<0.001$) differences in both the UK and global data sets. However, there is no significant difference ($p > 0.05$) between the average slow fractions of the UK and global forest soils, while the difference is only weakly significant ($p = 0.04$) between the non-forest soil UK and global average values. The higher average slow fraction for the entire global data set, compared to the UK data (Table 4), arises largely because the UK data set has a higher proportion of non-forest soils. Combination of the UK and global data to make the combined data set (Table 4) is justified. The slow fraction values for both forested and non-forested soils are normally distributed (Figure 3). In both data sets, the non-forest soils show a substantially greater spread of the slow fraction than the forest soils, with approximately double relative standard deviations (Table 4). The derived C fluxes through forest and non-forest soils also differ highly significantly (Table 4).

Twelve negative values of the slow fraction (Figure 3) arise because the chosen passive MRT (1000 yr) is too short to accommodate low ^{14}C values (see below). Nine of the negative values are for UK non-forested soils, one is for UK forested soil, and two are for non-forested soils in the USA. Considering the combined data set, if the forested negative value is omitted, there is essentially no change in the average and standard deviation of Table 4, while if the non-forested negative values are omitted, the mean slow fraction increases from 0.399 to 0.445. The difference in average slow fraction between the forested and non-forested soils remains highly significant ($p<0.001$).

We examined the results for systematic variation with soil type (Table S2). The only significant variation was that non-forested UK gleys and podsols had low average slow fractions (0.27 and 0.25 respectively), significantly different ($p<0.001$) from the other non-forest UK soils. However, even with the gley and podsol results removed, the remaining non-forested UK soils still have a significantly ($p<0.01$) smaller slow fraction than the forested soils.

The slow fraction tended to decrease with increasing soil C stock for both forest and non-forest soils. When all data from both data sets were amalgamated by normalising the slow fraction values to the mean values, a highly significant ($p<0.001$) decrease in the slow fraction with C pool was obtained, although only 7.4% of the variance was explained (Figure S2). To illustrate, the trend means that on average the slow fraction for a soil C pool of 12.5 kg m^{-2} is 64% of that for a pool of 2.5 kg m^{-2}.

Because the SOC pools in forested soils tend to be lower than in non-forested ones, we compared turnover rates for subsets of the two categories that had similar pools. By considering sites with SOC < 8 kg m^{-2}, we obtained nearly identical average SOC pools of 5.39 kg m^{-2} (78 forested soils) and 5.33 kg m^{-2} (88 non-forested soils). The average slow fractions of 0.66 (forested) and 0.48 (non-forested) differed significantly ($p < 0.001$), which means that the significant difference found for the full data sets is not an artefact arising from the different ranges of SOC pools.

We carried out regression analyses to attempt to establish relationships between the derived slow fraction and C flux values, and four possible drivers of soil C cycling, i.e. MAP, MAT, and pH. No relationships to MAP were found, and no relationships within the forest soils data at all. The following weak relationships were found for non-forest soils.

$$\text{slow fraction} = 0.051\text{pH} + 0.11 \qquad r^2 = 0.03 \qquad p < 0.05 \qquad n = 119 \qquad (1)$$

passive C flux $= -1.28\text{pH} + 12.2$ $r^2 = 0.12$ $p < 0.001$ $n = 119$ (2)

slow fraction $= 0.011\text{MAT} + 0.31$ $r^2 = 0.03$ $p < 0.05$ $n = 149$ (3)

passive C flux $= -0.16\text{MAT} + 6.44$ $r^2 = 0.04$ $p < 0.02$ $n = 149$ (4)

Thus the slow fraction tends to increase, and the passive C flux to decrease, with pH and MAT, which is consistent with faster SOC turnover at higher pH and temperature. We also tested for relationships between the slow fraction and %SOC, since the latter reflects the mineral content of soils, and therefore might be related to sorptive stabilisation. However, no significant variations were found.

To put the results from Table 4 into context, we constructed steady-state soil organic carbon cycling diagrams from the global results, by including a fast pool with an MRT of one year (Figure 4). The C passing through this fast pool includes, as well as rapidly recycled litter, grazed material, large wood fragments, and other forms of C that do not become part of the soil organic matter. To obtain a representative input rate of litter C, we combined estimates of global terrestrial net primary productivity of c. 60 Pg C a^{-1} (Ajtay et al., 1979; Saugier et al.; 2000) and a terrestrial area of c. 1.5 x 10^8 km^2. This yielded a value of c. 400 g C m^{-2} a^{-1}, which we increased to 500 g C m^{-2} a^{-1} to account for the likelihood that the soils in our database are biased towards higher NPP, since they include no deserts few high-latitude sites. The results (Figure 4) are only intended to be illustrative for the average cases, and it is acknowledged that a wide range of circumstances (due to variations in NPP and edaphic conditions) actually occurs. Most litter C passes through the fast pool, followed by the slow and then passive pools. For forest soils, the fluxes are in the proportions 0.596:0.400:0.004, while for non-forest soils they are 0.694:0.300:0.007. The approximately two-fold difference in the small passive C fluxes plays a major role in producing the different passive pools. More than 99% of C input to soils passes through the fast and slow pools, i.e. within a few decades at most.

Reliability of the modelling approach

The assumption that the soil carbon is in steady state is obviously an approximation. Past disturbances will have perturbed any steady state, notably changes in land-use or management (Wutzler & Reichstein, 2007), wildfires (Parker et al., 2001), the relatively recent fertilisation of some terrestrial ecosystems by atmospheric N deposition (Tipping et al., 2012b), and climate change. If historical information about such changes could be obtained, sites could be excluded from the data compilation and analysis, but usually only general information is available. The steady state assumption might also be invalidated by modification of the radiocarbon content of the soil, due to the presence of material low in or devoid of radiocarbon, such as charcoal and “black carbon”, or contaminant “hot” material enriched in ^{14}C. We explored these issues through sensitivity analyses (Appendix 1). For most of the identified effects, we modified Model III to impose inputs and losses of C and ^{14}C to the soil, and thereby simulate the C pool and radiocarbon content in the year 2000. The effects of the perturbing factors were evaluated by modelling the simulated data, with the assumption of steady state, to obtain the apparent values of the slow fraction. The results were then compared with the slow fraction obtained for default soils in true steady state.

In many of the cases examined, simulations of young soils, soils affected by plausible historical land management practices (but not increased grazing pressure), and soil receiving enhanced N deposition, create conditions in the year 2000, which, when analysed assuming steady state, produce a greater slow fraction, i.e. apparently faster turnover. The more recent were the management changes, the greater is the effect. The changes in non-forest soils are relatively greater than those in forest soils, owing to carbon losses during the hypothesised land managements, and then the relatively greater subsequent uptake of C (and bomb carbon) into the slow pool. This means that the differences in turnover between forest and non-forest soils are if anything greater than shown by the results in Table 4. Apparently faster turnover would also be caused by the incorporation into the soil of “hot” material, enriched in radiocarbon, but this is presumed unlikely to be general, and only to be serious at sites close to emission sources. Contamination by charcoal and black carbon is likely more diffuse, and has the opposite effect, by reducing the soil ^{14}C level compared to the value without contamination. A management practice that can cause an apparent decrease in soil C turnover, i.e. reduction of the slow fraction, is recently increased grazing pressure, which would have decreased the input of bomb carbon to the soil.

Bioturbation might mix significant quantities of C between topsoil and deeper soil, and this is explored in Appendix 2. The results suggest that if significant bioturbation is occurring, then the inputs to, and outputs from, the topsoil would be greater than found with the non-exchanging model. For the highest assumed exchange rate due to bioturbation (5% per annum), the slow C flux is estimated to be about 50% greater, and the passive C flux about 100% greater, than those required in the absence of bioturbation. The reported data on biotubation are likely biased towards sites where it is demonstrable, and it will be less important in nutrient-poor acid soils.

Model assumptions about plant C residence times, the fast (litter) C pool, and the choice of MRTs for the slow and passive pools, are explored in Appendix 3. The choices of plant residence times have modest effects. They influence absolute estimates of the turnover variables but not relative behaviours, and certainly do not affect any conclusions about differences in carbon turnover between forested and non-forested soils. Neglect of the fast pool has minimal effect on the model outputs, especially since it is unlikely to be fully represented owing to the removal of surface and root litter before analysis of soil for ^{14}C. The choices of 20 and 1000 yr in Model III are somewhat arbitrary, although they have the advantage of bracketing the observations (Figure 2) so that the great majority of soils can be described. The results detailed in Appendix 3 show that setting the slow MRT to 10 yr leads to unrealistic C fluxes, as discussed above, while setting it to values > 20 yr would generate more physically unrealistic negative soil pools. Moreover, referring to Figure 4, if 10 yr were chosen for the slow MRT, the C flux through the slow pool would be higher, 300-400 g C m^{-2} a^{-1} (Appendix 3), unreasonably close to the total input flux. Model analysis of data sets with repeated measurements of soil ^{14}C over extended time periods (up to 43 yr in one case) provides some additional support for the 20 year value (Appendix 3).

The choice of 1000 yr for the MRT of passive topsoil carbon leads to a few anomalous results when the ^{14}C content of a soil is low, producing negative slow fractions (Figure 3). This might be resolved by increasing the passive MRT, to say 2000 yr, but that would imply a doubling of the time required for a soil to reach steady state. For example, with an MRT of 1000 yr, 3000 yr are needed to achieve 95% of the steady-state passive C pool, whereas 6000 yr are required with an MRT of 2000 yr. Therefore the modelling advantage gained by increasing the MRT to 2000 yr would be offset by the greater uncertainty associated with the assumption of approximately constant conditions over a much longer period. The 1000 yr MRT chosen for the passive fraction is best regarded as an order-of-magnitude value, suitable for representing the most stable topsoil C. Refining the value to accommodate a small number of anomalous results would not be justified.

Discussion

The model-derived results provide information about topsoil C turnover at two large scales; national for the UK, and global (land area ratio c. 1:600). The mean slow fraction values for the two vegetation types that we consider hardly differ at these two scales, suggesting that the estimates are robust and widely applicable, and that the UK and global datasets can be combined. On average, the bulk of topsoil carbon can be partitioned into two similarly-sized pools (slow and passive), one with a decadal turnover rate, the other much longer-lived, with a residence time of the order of 1000 years. However, there is appreciable variation in the slow and passive fractions amongst soils, indicating a range of carbon turnover characteristics. An intriguing finding is that forest soils are richer in the slow pool, while those under non-forest vegetation have more passive carbon (Figure 2, Table 3). In other words, on average, forest topsoil C turns over faster than non-forest topsoil C.

Modelling

To apply a consistent method of interpretation of the available data, we were obliged to employ a simple modelling approach, involving both the assumption of steady-state and the assignment of *a priori* turnover rates. Implications of the steady-state assumption were explored (see Results and Appendix 1), and it can be concluded that errors due to past land use change (except recent increased grazing pressure), or contamination with materials rich in ^{14}C, will tend to make rates appear faster (increase the apparent slow fraction) as will bioturbation (Appendix 2), whereas the presence of black carbon, coal or charcoal, and increased recent grazing pressure, would operate in the opposite direction. Given that the uncertainties can lead to errors in both directions, systematic bias in the derived average turnover variables can be considered unlikely. The numerical results are not unique, and different MRT choices would lead to different absolute values. However, the trends and patterns would be the same, and the difference between forest and non-forest soils would persist, principally because the model parameters have to account for the greater enrichment of forest soils with bomb carbon (Table 3).

It is important to recognise that the slow and passive pools are simply model partitions, and do not imply that all soils have the same physical, chemical or biological types of material, e.g. the passive pool could be stable due to either molecular recalcitrance or physico-chemical stabilisation (cf. von Lutzow et al., 2006; Schmidt et al., 2011; Kleber, 2010) or both, and this need not be the same in all soils. However the structure of Model III accords with the idea that litter contains materials with different susceptibilities to decomposition, i.e. variations in molecular recalcitrance cause differences in C turnover. To fit better with the idea that physico-chemical stabilisation controls soil C turnover, an alternative model could be constructed in which the fractional inputs of litter to the slow and passive pools, and the slow pool MRT, were fixed *a priori*, and variations in soil ^{14}C produced by adjusting the MRT of the passive pool. This would generate an average passive pool MRT for forest soils of about 600 yr (see Appendix 3).

Forest vs non-forest soils

The finding that forest topsoil OM is on average significantly richer in radiocarbon than non-forest OM (Figure 2, Table 3) implies faster cycling, a conclusion that does not rely on modelling if steady-state conditions are approximated. This result confirms previous suggestions by Bol et al. (1999) and Tipping et al. (2010) which were based on results for only a few sites. Similarly, Brovkin et al. (2008) derived turnover rates from ^{14}C data for humic acids extracted from soils of European Russia, and their results correspond to MRT values of 128-625 years for forest soils, considerably less than the range of 313-5000 years for grasslands. Their MRT values are generally greater than values derived from the present data set (Table S1), which presumably arises because humic acid is more stable than SOM as a whole.

Referring to Figure 4, there are two differences between forest and non-forest soils in the idealised, average, cases. Firstly, in the non-forest system, more carbon passes through the fast pool, and therefore less through the slow, than in the forest system. The results produce a greater fast litter flux in non-forest soils, which indicates that forest litter contains material that decomposes too slowly to appear in the fast pool, and so enlarges the slow pool; this might be largely due to lignin. However the difference between the soil classes is relatively small and depends upon the assumption that litter inputs are equal for average forest and non-forest ecosystems. More quantitatively significant is the greater rate of C input to the passive pool in non-forest soils, which is about twice that in forest soils and leads to the larger passive fraction (Table 4).

If steady-state conditions are well-approximated for soils in both vegetation classes, possible explanations of the forest / non-forest difference include variation in the intrinsic decomposability of litter (molecular recalcitrance), possibly affected by grazing (more prevalent in non-forest systems), and differences in edaphic conditions, including microbial decomposer communities, physico-chemical stabilisation, root architecture, and microclimate. If non-steady state conditions apply, increased grazing pressure at non-forest sites in the 20th century may have restricted the accumulation of soil radiocarbon during the period of atmospheric enrichment, causing them to appear to have lower steady-state turnover (Appendix 1). However, quite severe reductions in litter C input would have been required to achieve this, and it seems unlikely that overgrazing can fully explain the observed differences.

A further distinction on the basis of vegetation type is that the relative standard deviation in the slow fraction is appreciably greater (about 2-fold) for non-forest soils than forests in the UK, global and combined datasets (Table 4). The differences may arise because non-forest soils or ecosystems are a less homogeneous group than forested ones, in terms of either litter quality variation or soil conditions or both, although the standard deviations of basic soil and climatic variables are not consistently greater for the non-forest soils (Table 2). The sensitivity analysis (Appendix 1) suggests that non-steady-state influences tend to be greater on soils presently under non-forest vegetation, which may have led to the greater contemporary variability.

Controlling factors

Apart from the forest / non-forest distinction, we found little explanation of turnover rate variance from soil type, MAT, MAP, pH, C concentration, or total soil C pool, any significant relationships being weak (equations 1-4). A possible effect of soil type is suggested by the small slow fractions in UK gleys and podsols (Table S2), but otherwise no patterns were evident. It may be that the historical and site-specific factors considered in the sensitivity analysis, i.e. non-steady state, contamination and bioturbation, are the most important controls on soil carbon turnover, at the scale of our analysis. This being so, the variations amongst the soils demonstrated by the radiocarbon-based analysis suggest that caution should be exercised when drawing general conclusions about C turnover from plot-scale observations or experiments.

Wider relevance

Our results give a broad picture of topsoil C turnover, provide new constraints to conceptual or quantitative models of soil C turnover, and complement detailed site-specific investigations that combine fractionation of the soil C with radiocarbon measurements (Tipping et al., 2010; Leifeld et al., 2009; Koarashi et al, 2012). The compiled radiocarbon data, used individually or as averages or distributions, are a resource for other modelling work (not necessarily steady-state), for example soil C cycling with RothC (Jenkinson, 1990), ecosystem models such as Century (Parton et al., 1987) and N14C (Tipping et al., 2012b), and dynamic global vegetation models, notably the LPJ (Sitch et al., 2003) which already includes a version of Model III. The modelled turnover parameters demonstrate that heterogeneity of topsoil carbon is a widespread global phenomenon, which should be taken into account in assessing the stability of soil organic matter. This is significant not only for understanding the soil C cycle and its variation in space and time, but is also relevant to the development of policy with regard to the protection and management of soil carbon. For example, the finding that organic matter turns over more quickly in forested topsoils raises questions about the efficacy of afforestation as a means to promote carbon storage. In considering sequestration by soils, not only is the amount of carbon important, but also its range of residence times.

Conclusions

1. Topsoils under forests have significantly higher ^{14}C contents than those under non-forest vegetation, owing to greater enrichment with "bomb carbon", which indicates a faster rate of soil C turnover in the forest soils, if steady-state conditions are approximated.
2. Application of a two-pool steady-state soil C cycling model to 133 UK soils divides topsoil C 0.61:0.39 between a slow pool (MRT 20 yr) and a passive pool (MRT 1000 yr) for forest soils, while for non-forested soils the division is 0.36:0.64. Corresponding ratios for 115 global soils are 0.68:0.32 and 0.47:0.53, and for the combined data set 0.65:0.35 and 0.40:0.60.
3. The non-forest soils are more variable in their contents of the two SOC fractions, having a relative standard deviation of the slow fraction about twice that of the forest soils both in the UK and globally.
4. Considering the combined data set, the mean flux of C through the slow pool of forest topsoils is 195 g C m^{-2} a^{-1}, while for the non-forest soils it is 141 g C m^{-2} a^{-1}. Fluxes through the passive pool are much lower, with values of 2.2 and 5.1 g C m^{-2} a^{-1} for forest and non-forest soils respectively.
5. None of the derived variables (slow:passive fractionation, C fluxes) shows a strong association with on the possible driving variables MAT, MAP, pH or soil type, although in some cases there are statistically significant relationships with MAT and pH, while UK non-forested gleys and podsols have significantly smaller slow fractions
6. than other UK non-forested soils.Assuming a fast soil carbon pool with an MRT of one year, and an average litter input of 500 g C m^{-2} a^{-1} to the topsoil, on average the global soil carbon fluxes are partitioned among the fast, slow and passive pools in the ratio 0.606:0.390:0.004 in forest soils, and 0.693:0.300:0.007 in non-forest soils.

Supplementary material

Table S1	Database including references (Microsoft Excel file)
Table S2	Trends with soil type
Figure S1.	Geographical locations of sites.
Figure S2	Normalised slow fraction for all soils plotted against topsoil C pool
Appendix 1	Sensitivity analyses; land management, contamination
Appendix 2	Sensitivity analysis; bioturbation
Appendix 3	Model parameter choices

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Table 1. Geographical distribution of sites in the global dataset.

	Forest	Non-forest	Total
Africa	1	2	3
Asia	7	11	18
Australasia	2	3	5
Europe[a]	28	8	36
North America	14	23	37
South & Central America	11	4	15
latitude ranges (deg)[b]			
0 - 22.5	15	6	21
22.5 - 45	21	29	50
45 - 67.5	26	14	40
67.5 - 90	1	2	3

[a] Does not include UK sites.

[b] The latitude values are absolute, i.e. N and S are combined.

Table 2. Summary data for topsoils and their sites. Key: n number of samples, SD standard deviation, MAT mean annual temperature, MAP mean annual precipitation The p values show the significances of differences between forest and non-forest means.

		UK sites			Global sites			Combined sites		
		forest	non-forest	p	forest	non-forest	p	forest	non-forest	p
depth	n	35	101		64	51		99	152	
cm	mean	15.0	14.9	>0.05	15.5	15.8	>0.05	15.3	15.2	>0.05
	SD	0.0	1.1		5.0	4.1		4.0	2.5	
%C	n	35	101		41	36		76	137	
	mean	10.3	17.6	<0.05	17.0	9.0	<0.05	13.9	15.4	>0.05
	SD	3.9	16.8		14.9	9.9		11.7	15.7	
pH	n	35	96		30	26		65	122	
	mean	5.6	5.1	<0.05	5.2	5.7	>0.05	5.4	5.2	>0.05
	SD	1.1	0.9		1.3	1.1		1.2	1.0	
C pool	n	35	101		64	51		99	152	
kg m^{-2}	mean	6.97	8.91	<0.01	5.73	6.29	>0.05	6.17	8.03	<0.001
	SD	1.43	3.94		2.80	3.76		2.47	4.06	
MAT	n	35	101		64	51		99	152	
^{o}C	mean	8.9	7.8	<0.001	11.5	8.9	>0.05	10.6	8.2	<0.01
	SD	0.9	1.4		7.2	8.2		5.9	4.9	
MAP	n	35	101		64	51		99	152	
mm a^{-1}	mean	1101	1145	<0.05	1304	880	<0.01	1232	1056	<0.05
	SD	652	386		581	624		611	490	

Table 3. Comparison of average ^{14}C values (% modern absolute) for forest and non-forest soils, over equal time periods. The p values show the significance of the differences between forest and non-forest ^{14}C values.

	forest			non-forest			
	n	^{14}C	SD	n	^{14}C	SD	p
UK sites							
2002-2008	35	107.8	1.1	94	100.2	0.8	<0.001
Global sites							
1947-1962*	6	94.7	1.1	6	91.5	1.7	>0.05
1991-1998	26	111.5	0.9	18	107.4	2.0	>0.05
2000-2006	31	110.8	0.7	30	102.1	0.9	<0.001
Combined sites							
1947-1962*	6	94.7	1.1	6	91.5	1.7	>0.05
1970-1978	28	111.3	1.2	6	106.1	3.4	>0.05
1990-1998	27	111.5	0.9	21	104.8	2.2	<0.02
2000-2004	43	110.2	0.8	26	100.8	1.3	<0.001
2005-2008	22	107.4	1.3	98	100.7	0.8	<0.001

* Duplicate results.

Table 4. Summary of SOC fractionation and C fluxes (g m^{-2} a^{-1}) derived with Model III. Key: n number of samples, SD standard deviation, RSD relative standard deviation (%). Note that the passive fraction = (1 – slow fraction). The p values show the significance of the differences between forest and non-forest values.

			all sites	forest	non-forest	p
UK		n	133	35	98	
	slow fraction	mean	0.43	0.61	0.36	<0.001
		SD	0.29	0.20	0.29	
		RSD	0.68	0.33	0.8	
	slow C flux	mean	161	209	144	<0.001
		SD	129	73	141	
	passive C flux	mean	5.1	2.7	5.9	<0.001
		SD	3.9	1.6	4.1	
Global		n	114	63	51	
	slow fraction	mean	0.58	0.67	0.47	<0.001
		SD	0.21	0.14	0.24	
		RSD	0.37	0.21	0.5	
	slow C flux	mean	170	198	134	<0.002
		SD	111	107	107	
	passive C flux	mean	2.7	2	3.6	<0.002
		SD	2.3	1.4	2.9	
Combined		n	247	98	149	
	slow fraction	mean	0.5	0.65	0.4	<0.001
		SD	0.27	0.16	0.28	
		RSD	0.54	0.25	0.7	
	slow C flux	mean	165	202	141	<0.001
		SD	121	96	130	
	passive C flux	mean	3.9	2.3	5.1	<0.001
		SD	3.5	1.5	3.9	

Figure captions

Figure 1. Example applications of Model III. Upper panel: forest soil, site 84, soil C pool 5.97 kg C m^{-2}. Lower panel: non-forest soil, site 221, 4.02 kg C m^{-2}. The symbols show the observations of bulk topsoil ^{14}C, the bold lines indicate the fitted model. The slow fraction in the forest example is 0.71, that in the non-forest example is 0.23. Other details are in Table S1. Note that the examples include multiple dates, but for the majority (85%) of sites only a single soil radiocarbon value is available for fitting.

Figure 2. Topsoil radiocarbon contents plotted against sampling date for the combined data. Open symbols represent forested sites, closed symbols non-forested sites. The upper and lower curves are bulk topsoil ^{14}C calculated for steady-state mean residence times of 20 and 1000 years respectively. For clarity, one sample taken in 1900 is not plotted (site 184, non-forest soil, 93.3 % modern absolute).

Figure 3. Cumulative distributions of the slow fraction for forest and non-forest topsoils. The lines are the fitted normal distributions.

Figure 4. Mean carbon pools (g C m^{-2}, normal text) and fluxes (g C m^{-2} a^{-1}, italics) derived from the global data set using Model III. Values in brackets are the assumed mean residence times (yr). Inputs from the left are C in litter, outputs to the right comprise CO_2, dissolved and particulate organic C, CH_4 etc.

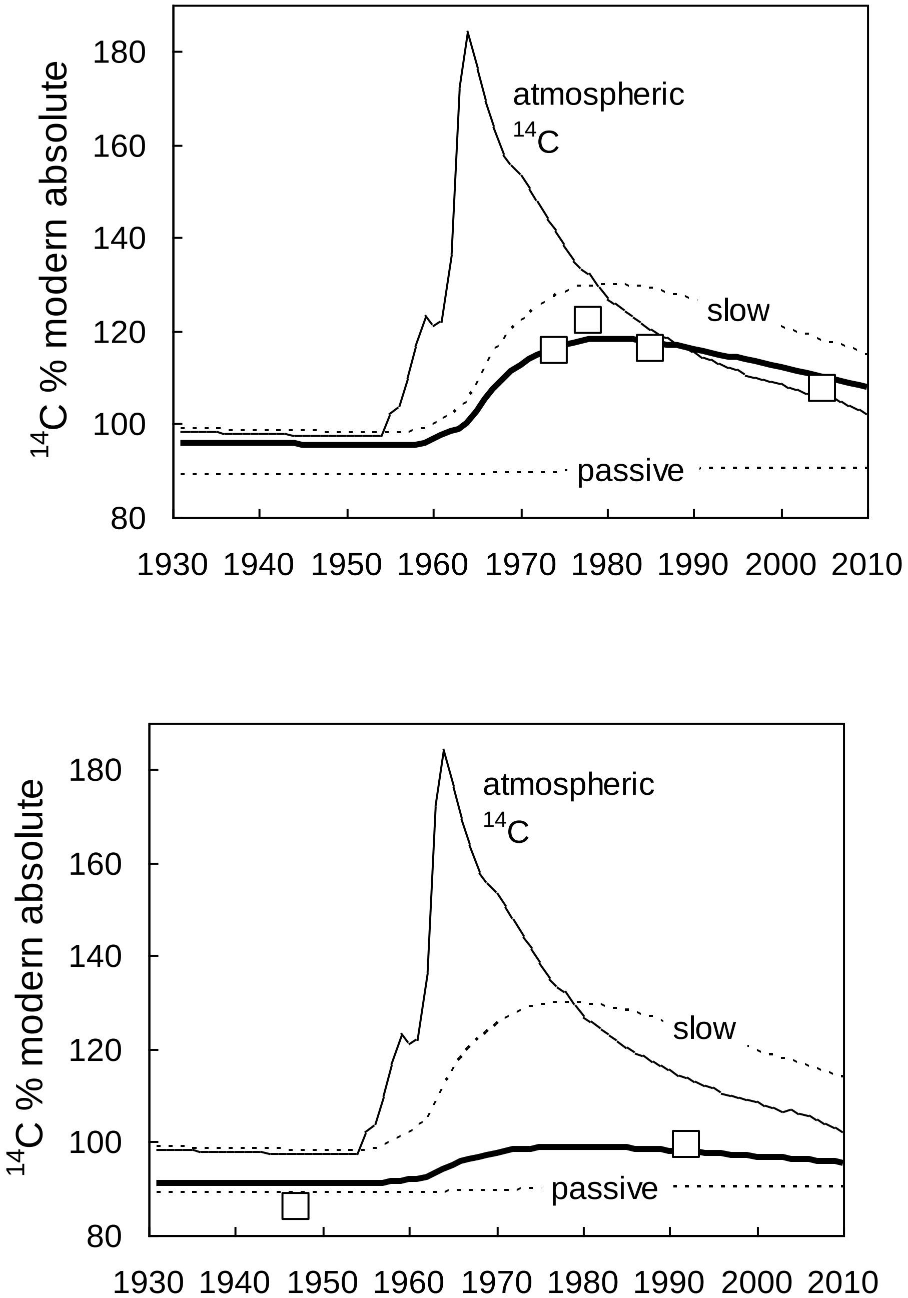

180
160
140
120
100
80
^{14}C % modern absolute
1930 1940 1950 1960 1970 1980 1990 2000 2010
atmospheric
^{14}C
slow
passive
180
160
140
120
100
80
^{14}C % modern absolute
1930 1940 1950 1960 1970 1980 1990 2000 2010
atmospheric
^{14}C
slow
passive

Figure 1.

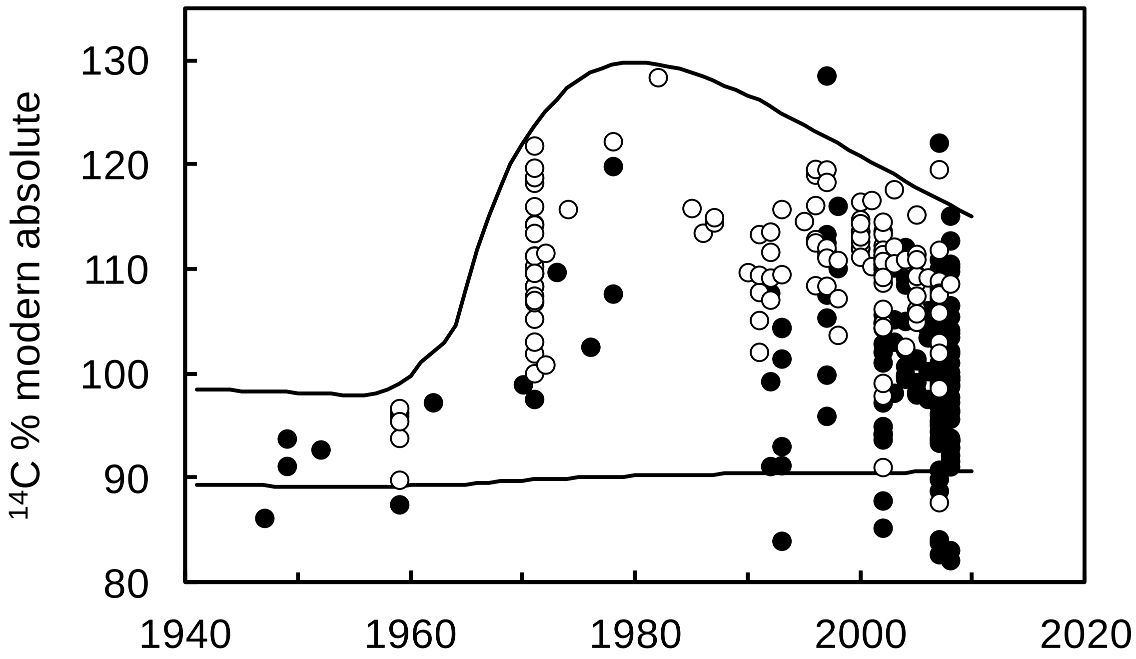
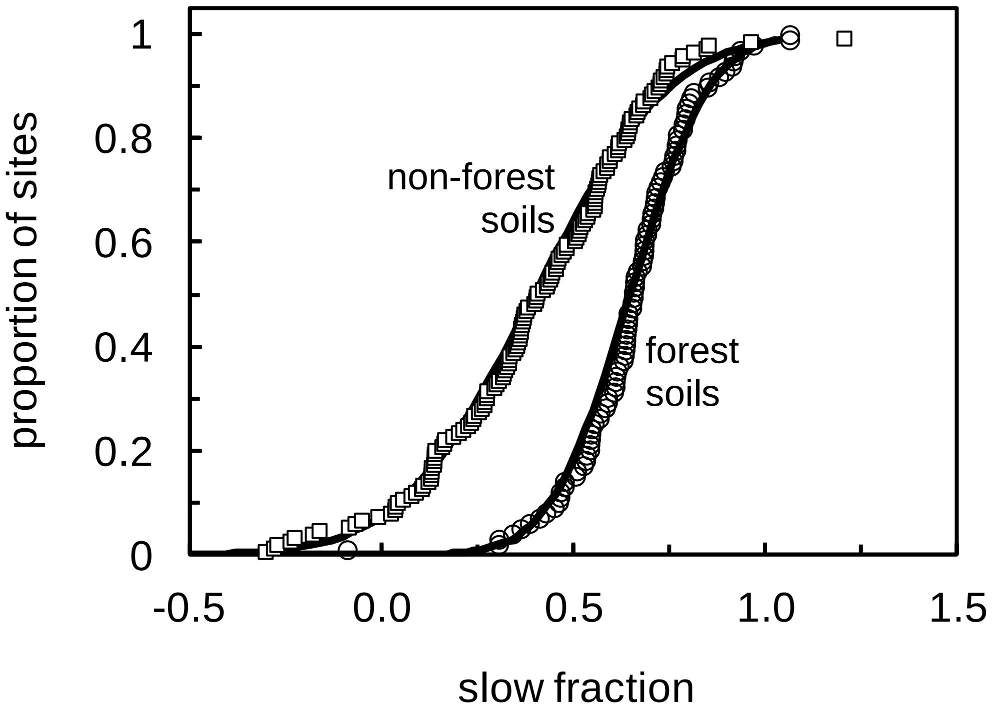

130
120
110
100
90
80
1940
1960
1980
2000
2020
14C % modern absolute

Figure 2.



1
0.8
0.6
0.4
0.2
0
proportion of sites
-0.5
0.0
0.5
1.0
1.5
slow fraction
non-forest
soils
forest
soils

Figure 3.

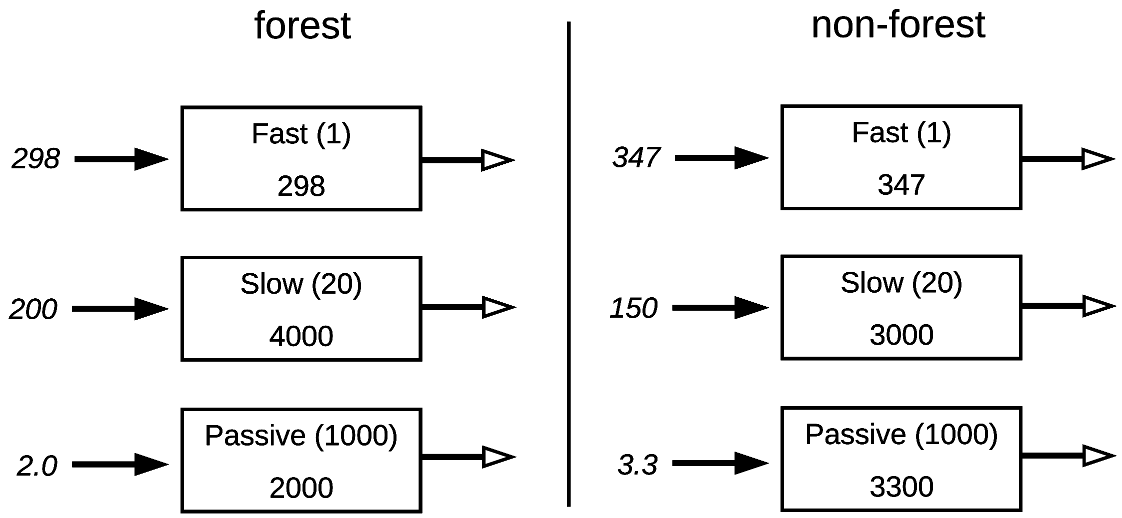

forest
non-forest
Fast (1)
298
298
Slow (20)
4000
200
Passive (1000)
2000
2.0
Fast (1)
347
347
Slow (20)
3000
150
Passive (1000)
3300
3.3

Figure 4.

Long-term organic carbon turnover rates in natural and semi-natural topsoils

R.T.E.Mills, E.Tipping, C.L.Bryant, B.A.Emmett

Table S2. Mean slow fractions and standard deviations (sd) for different soil types with 8 or more occurrences in the data sets.

		UK			global		
		n	mean	sd	n	mean	sd
forest	acrisol	-	-	-	13	0.62	0.16
	cambisol	13	0.61	0.12	12	0.66	0.08
	leptosol	8	0.68	0.16	8	0.77	0.07
	podsol	-	-	-	12	0.63	0.11
non-forest	cambisol	19	0.42	0.29	9	0.43	0.11
	gleysol	22	0.27	0.21	-	-	-
	histosol	17	0.42	0.35	-	-	-
	leptosol	-	-	-	8	0.50	0.15
	luvisol	12	0.44	0.30	16	0.45	0.23
	podsol	21	0.25	0.26	-	-	-

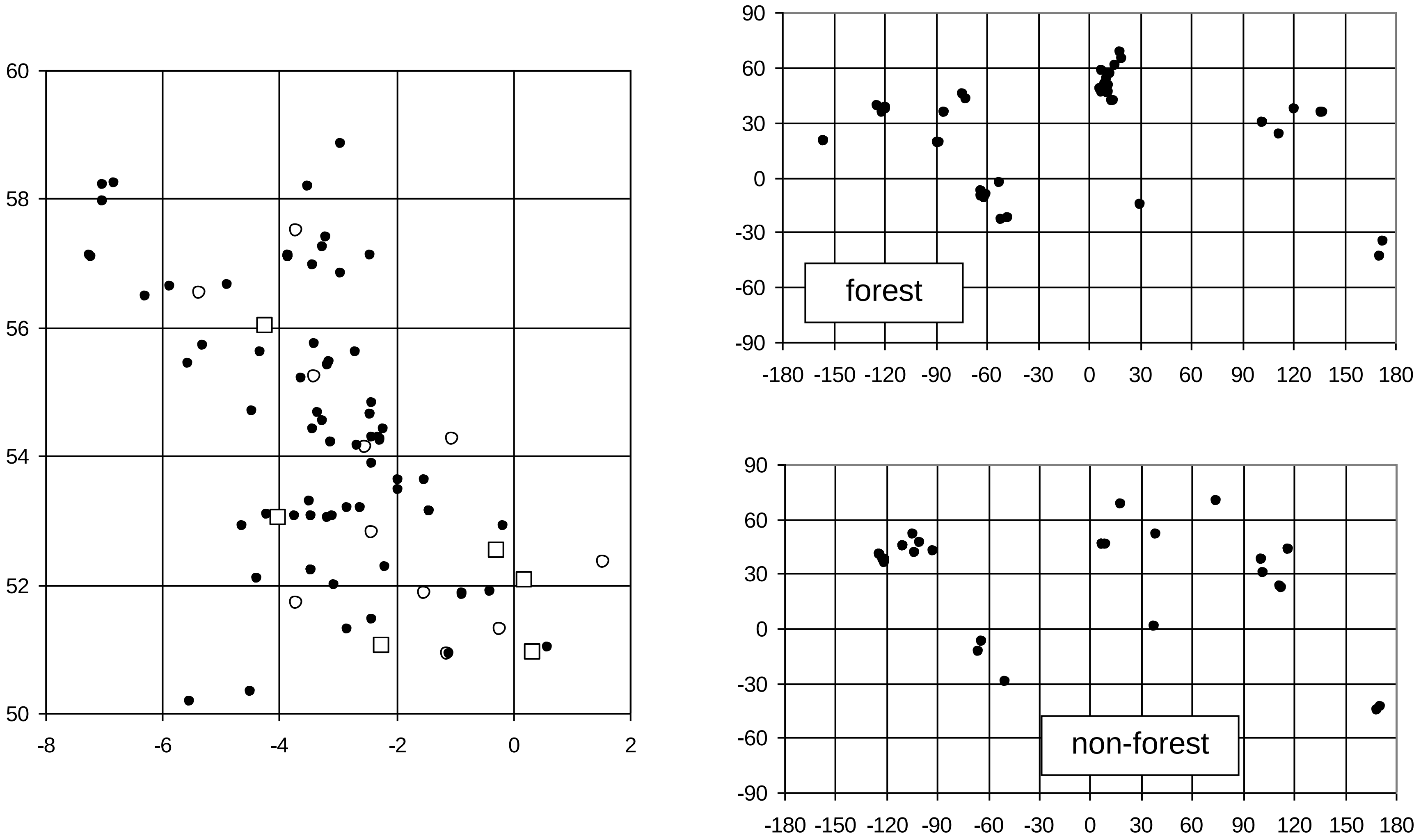


Figure S1. Geographical locations plotted as latitude and longitude. Left panel: UK forest (open symbols) and non-forest (closed symbols) sites; the squares each indicate a location at which four separate forest sites are close together. Right panels: global sites (UK sites omitted).

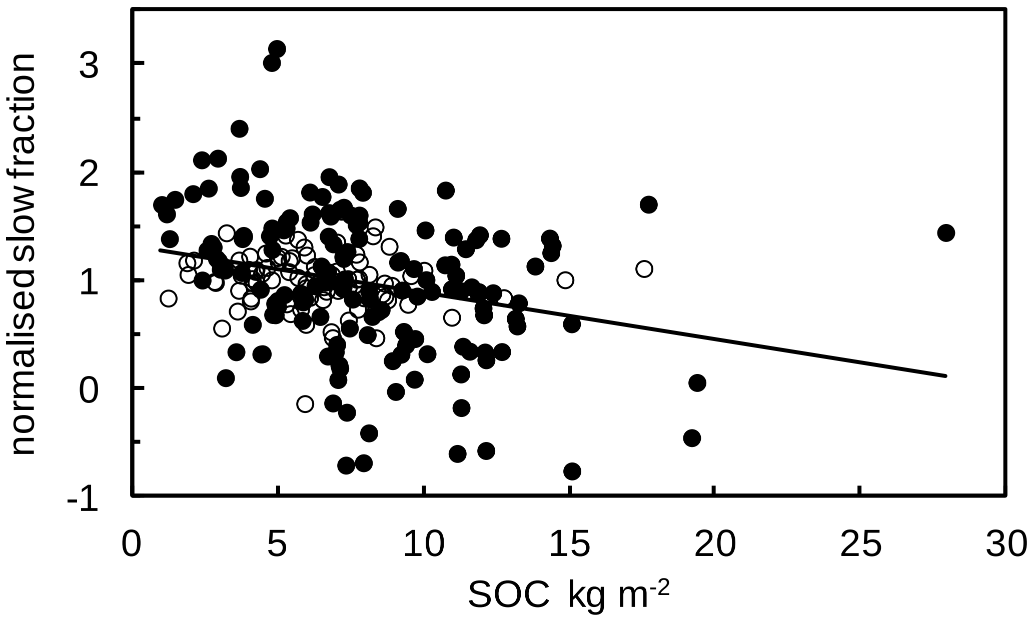


Figure S2. Normalised slow fraction (derived value / mean) as a function of topsoil C pool for the combined data set. Open circles represent forest sites, closed circles non-forest sites. The linear regression of all data is shown (r^2 =0.077, $p < 0.001$).

Long-term organic carbon turnover rates in natural and semi-natural topsoils

R.T.E.Mills, E.Tipping, C.L.Bryant, B.A.Emmett

APPENDIX 1: Sensitivity analyses; contamination and land management

We tested the sensitivity of the modelling approach by imposing plausible past, non-steady-state, effects, and using Model III to calculate soil C turnover and thereby the soil C pool and ^{14}C content in the year 2000. The data so-generated were then analysed with Model III, assuming that steady-state conditions apply, to obtain the *apparent* fraction of slow soil C. The results are summarised in Table A1 (below).

"Black carbon" and coal

By this we mean contaminating materials of low or zero ^{14}C content (Schmidt and Noack, 2000). This problem was encountered during turnover studies by Jenkinson et al. (1992) in soil samples collected at Rothamsted, UK, which contained coal particles. For nine samples of soils from two sites they measured carbonized organic matter and found between 20 to 50 gC m-2 (to a depth of 23 cm) which would correspond to around 1% of the total C pool. The atmospheric deposition of Spheroidal Carbonaceous Particles (SCPs), derived mainly from coal burning and used for dating in lake sediments (Rose, 2001), have not exceeded 0.01 g m^{-2} a^{-1} in the UK (Professor Neil Rose pers. comm.). If SCPs were 100% C and had been deposited for 200 years (both extreme assumptions) they would contribute only 2 gC m^{-2} of soil, i.e. < 0.1%. For soils used for maize cultivation in Germany, Rethemeyer et al. (2007) estimated, from levels of benzene polycarboxylic acids, that c. 15% of topsoil C was due to black carbon in a region of lignite mining and processing, and c. 4% in a region far from industry and major roads. These values are probably relatively high compared to soils of natural or semi-natural ecosystems, because of the lower inputs of carbon from vegetation as a consequence of cropping. Black carbon may be highly significant locally (e.g. Schmidt et al., 1999; Rumpel et al 2003), but is unlikely to be a widespread factor.

With 1% contamination by ^{14}C-free carbon, the derived slow fractions are decreased, by 6% for forested soils and 8% by non-forested soils. We looked into the UK data for evidence of such contamination, and indeed the lowest ^{14}C of c. 60% modern (which would require c. 40% of the soil C to be ^{14}C-free) was found for a soil sample collected from between the industrialised conurbations of Manchester and Sheffield (site 33 in Table S1). However, other low-^{14}C sites (< 90% modern) were not obviously close to industrial areas, several being in remote parts of Scotland. Contamination can be highly localised. For example, whereas most of their soils were only slightly contaminated, Jenkinson et al. (1992) found three Rothamsted soils to have c. 500 g carbonised C m^{-2}, 10 times greater than the majority. Leifeld (2008) carried out simulations with the Roth-C 26.3 model (Coleman and Jenkinson 1999) of the effects of black carbon on apparent soil C turnover, and reported under- or over-estimation of C turnover rates by up to 30%, i.e. a similar magnitude to the results reported here.

Charcoal

The presence of plant-derived charcoal interferes with the estimation of the turnover rate of the active soil. Charcoal will generally contain radiocarbon, depending on when the burning responsible for its formation occurred, but the most serious effects on MRT estimation will arise when the charcoal is old. Rodionov et al. (2010) determined "black carbon" in the chernozem or mollisol soils of 28 globally-distributed grassland ecosystems, but the material was charcoal as opposed to the radiocarbon-free black carbon referred to above. They found that 11 to 15 % of the SOC was due to charcoal, mostly derived from local plants, accumulated since the mid-Holocene, with the highest concentrations in deeper parts of the A horizon. Ohlson et al. (2009) reported a charcoal content of 77 gC m^{-2} (1 to 2%) with a mean age of 652 years in boreal forest soils. Based on these findings we calculated the effects on model outputs assuming 5 and 10% of the topsoil C was due to charcoal of mean age 2000 years. If 10% of the soil is comprised of such charcoal, the slow fractions calculated without taking this into account are decreased by 16 and 19% for forested and non-forested soils respectively, but these are likely more extreme than average.

Contamination by radiocarbon

Local contamination by material high in radiocarbon, for example from the incineration of waste, was actually exploited to investigate ecosystem carbon turnover in one study (Trumbore et al., 2002; Hanson et al., 2005; Swanston et al., 2005). The presence of “hot” material has the opposite effect to black carbon, i.e. the slow fractions appear higher, C turnover appears faster.

Land management

The clearance of forest for pasture would have affected the turnover of soil C. The more recently it was done, the greater would be the effects (Table A1). Thus, clearance 500 years ago would make the present-day grassland soil have a slow fraction 31% higher (i.e. apparent turnover would be faster) than in the default case. Biomass removal would decrease the soil C pool, but have only minor effects on the apparent turnover rates. Land that was cropped and ploughed for a period in the past will appear to have a greater slow fraction, i.e. faster turnover. For this calculation we also assumed that the ploughing would increase decomposition rates by allowing more efficient oxygenation. The greatest effects are on soils that currently have non-forest vegetation, because the higher rate of input of slow litter and the acquisition of bomb carbon override the slower responses.

Grazing

According to Model III, for a constant grazing pressure, the slow and passive fractions would not be affected, although the total soil C pool would be lower the greater the pressure. However, if grazing pressure increased during the 20th century, in particular during the period of increased atmospheric ^{14}C due to weapons testing, the slow fraction would apparently decrease. Removal of 50% of the C input to the soil due to recent grazing would reduce the slow fraction by 40%. Severe over-grazing, causing an 80% decrease in C input, would reduce it by 80%. The apparent turnover rate of soil carbon could thus be substantially reduced if severe overgrazing had taken place.

Young soil

If the soil was formed only recently, then it will not have built up so much carbon, and this will apply especially to the passive pool. Therefore it will appear to have a higher slow fraction, and faster turnover. Obviously the younger is the soil the greater this effect will be.

Fertilisation by nitrogen deposition

This is a relatively recent phenomenon which will have increased the input of litter to the soil over the past one or two centuries. This will have resulted in disproportionately more bomb carbon and consequently an apparently greater slow fraction (faster turnover).

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Table A1. Summary of simulations. NF = non-forested, F = forested.

	^{14}C		soil C		fraction slow		fractional change	
	NF	F	NF	F	NF	F	NF	F
Default								
Annual inputs (gC m-2 a-1): slow pool; 160 (NF), 190 (F); passive pool 4.3 (NF), 2.2 (F)	103.2	110.1	7500	6000	0.440	0.650	na	na
Black carbon								
Addition of 1% coal (^{14}C = 0)	102.2	109.0	7575	6060	0.406	0.614	-0.08	-0.06
Charcoal								
Soil mass is 5% 2000-year old charcoal, which does not decompose, nor is it eroded	101.9	108.5	7500	6000	0.397	0.598	-0.10	-0.08
Soil mass is 10% 2000-year old charcoal, which does not decompose, nor is it eroded	100.7	106.9	7500	6000	0.355	0.545	-0.19	-0.16
Contamination by radiocarbon								
Local contamination by radiocarbon, 10% above atmospheric 1980-2000	106.2	114.2	7500	6000	0.542	0.784	0.23	0.21
Land management								
Present-day grassland that was forest until 1500	107.2	na	6227	na	0.576	na	0.31	na
Present-day grassland that was forest until 1000	106.0	na	6728	na	0.536	na	0.22	na
Present-day grassland that was forest until 500	105.1	na	7032	na	0.506	na	0.15	na
Present-day forest that was forest until 1500, then cleared for pasture, reforested in 1900	na	107.8	na	6227	na	0.574	na	-0.12
Present-day forest that was forest until 1800, then cleared for pasture, reforested in 1900	na	108.7	na	5781	na	0.604	na	-0.07
Litter inputs reduced by 50% 1500-1900, due to biomass removal	103.8	110.9	6864	5675	0.461	0.675	0.05	0.04
Ploughing & cropping: litter inputs (gC m^{-2} a^{-1}); slow 80, passive 2.15, turnover times halved :								
Previously forested land, ploughed & cropped from 1500 to 1900, then turned to grass or forest	110.7	112.5	5061	5458	0.696	0.727	0.58	0.12
Previously forested land, ploughed & cropped from 1800 to 1900, then turned to grass or forest	110.7	112.5	5061	5458	0.696	0.727	0.58	0.12
Previously grassland, ploughed & cropped from 1500 to 1900, then turned to grass or forest	106.8	108.6	5914	6311	0.563	0.6	0.28	-0.08
Previously grassland, ploughed & cropped from 1800 to 1900, then turned to grass or forest	106.2	107.9	6265	6661	0.543	0.578	0.23	-0.11
Grazing pressure, 50% of input removed from 1900	98.0	na	5657	na	0.265	na	-0.40	na
Grazing pressure, 80% of input removed from 1900	92.9	na	4551	na	0.092	na	-0.79	na

Young soil								
Soil formation began in year 0 AD (2010 BP)	105.9	112.1	6937	5718	0.533	0.715	0.21	0.10
Fertilisation by N deposition								
Increases in annual litter inputs by 33% during 1900-1950, by 67% 1950-2000	107.2	113.6	9811	8593	0.577	0.764	0.31	0.18
Loss of C by burning								
Burning every 200 years to 1800, 20% loss of slow and passive C	110.1	115.3	5556	5028	0.676	0.819	0.54	0.26
Burning every 200 years to 1800, 20% loss of slow C only	103.2	110.1	7500	6000	0.440	0.650	0.00	0.00

APPENDIX 2: Bioturbation

This is the movement of soil by organisms (Schaetzl & Anderson, 2005; Paton et al., 1995). Paton et al. (1995) compiled data on mound formation by earthworms, ants and invertebrates. The results show that the most effective are earthworms, which can deposit 18 kg soil m^{-2} a^{-1} at the 90 percentile, the median of the data being 3.7 kg soil m^{-2} a^{-1}. If we regard the soil as two boxes each of depth 15 cm and bulk density (BD) 1 g cm^{-3} then each box will contain 150 kg m^{-2}, and so if movement were to exchange soil between these two boxes the 90 percentile value would correspond to 12% of the soil, and the median to 2%. But some of the movement will be within the boxes and so these are likely overestimates. We therefore assumed values of 1% and 5% for simulation modelling.

We modified Model III to include a deeper soil box, that could exchange soil (and associated carbon) with the topsoil. For simplicity, the BD was assumed to be the same in each layer, so that C transfer is proportional to soil transfer. The slow and passive pools in the deeper soil were assumed to have the same mean residence times in the deeper soil as in the topsoil. The difference between the two layers is thus that the topsoil receives new litter, but the deeper layer does not, gaining (and losing) C only by exchange due to bioturbation, and decomposition. The result is that the deeper soil becomes relatively depleted in slow C, and has lower ^{14}C values. Exchange with the deeper soil therefore reduces topsoil ^{14}C compared to the situation without exchange, which means that higher C inputs are required to cause the topsoil ^{14}C to have the same value that is predicted for the case of no exchange.

The model was fitted by adjusting the input rates of slow and passive litter in order to reproduce the observed ^{14}C and total soil C, while ensuring that both topsoil and deep soil C pools were in steady state. The results for three fractional exchange rates (0, 0.01 and 0.05) are summarised in Table A2. We find that at an exchanged fraction of 1%, the required passive input is about twice that in the situation without bioturbation, with only a modest increase in the slow input. But when the exchange is 5%, the slow input must be increased by about 60% to achieve the observed topsoil C pool and ^{14}C values.

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Table A2. Bioturbation effects. Results from fitting the two-box model (topsoil and deeper soil).

		topsoil							deep soil			
	fractional exchange	slow input	passive input	slow pool	passive pool	slow fraction	total soil C	^{14}C	slow pool	passive pool	total soil C	^{14}C
		$gC\ m^{-2}\ a^{-1}$	$gC\ m^{-2}\ a^{-1}$	$gC\ m^{-2}$	$gC\ m^{-2}$		$gC\ m^{-2}$	% mod	$gC\ m^{-2}$	$gC\ m^{-2}$	$gC\ m^{-2}$	% mod
non-forest	0	165	4.2	3300	4200	0.44	7500	103.2	na	na	na	na
	0.01	186	8.2	3200	4300	0.43	7500	103.2	530	3910	4440	92.6
	0.05	260	8.0	3470	4030	0.46	7500	103.2	1740	3950	5690	98.3
forest	0	195	2.1	3900	2100	0.65	6000	110.1	na	na	na	na
	0.01	226	4.0	3880	2120	0.65	6000	110.1	650	1930	2680	96.7
	0.05	307	3.8	4090	1910	0.68	6000	110.1	2050	1870	3920	104.8

APPENDIX 3: Model parameter choices

Plant residence times

Plant residence times of zero, one and two years were assumed for herbs, shrubs and trees respectively, and these choices have some effect on the derived data. To illustrate, consider first the default forest soil (Appendix 1). With the assumption of a two-year plant residence time for C, the slow fraction from the data in 2000 is 0.65, but if the residence time is assumed to be zero the slow fraction is 0.68, and for a four-year residence time it is 0.63. Had the analysis been done on a soil sample taken in 1970, i.e. nearer to the peak of "bomb carbon", the default slow fraction would still be 0.63, but for zero and four-year plant residence times, the values are 0.59 and 0.75. For the default grassland soil (Appendix 1) with a plant residence time of zero, the slow fraction is 0.43, whereas if a two-year residence time is adopted the slow fraction is 0.42 in 2000, and 0.48 in 1970. These variations are fairly modest, and will affect absolute estimates of turnover characteristics without changing the relative behaviours. They certainly will not affect any conclusions about differences in carbon turnover between forested and non-forested soils.

Neglect of the fast pool

The fast pool in Models II and III is rapidly cycling recent litter. Its rate of C input can be estimated as the difference between total litter production and the fluxes of C entering the slow and passive pools. Assuming a typical NPP of 500 gC m^{-2} a^{-1} which corresponds to the litter production rate at steady state, and using the overall average slow + passive flux of 195 gC m^{-2} a^{-1} for forest soils (Table 4), we obtain a typical fast flux of 305 gC m^{-2} a^{-1}. If a residence time of one year is assumed, the fast pool is 305 gC m^{-2}, which is about 5% of the average forest topsoil C pool. Now consider the implications for the default case of Table A1 if *all* of the fast carbon in forest soil is assumed to be present in the soil analysed for ^{14}C. From a mass-balance, the default forest soil, sampled in 2000, would have a slow + passive pool of 5.7 kg m^{-2} instead of 6.0 kg m^{-2} and the slow + passive C would have a radiocarbon content of 110.0% instead of 110.1%. The corresponding changes for non-forest soil would be a reduction in the slow + passive pool from 7.5 to 7.2 kg m^{-2} and a change from 103.2% to 103.0% for the slow + passive ^{14}C content. Thus the model outputs would hardly be affected in terms of the division of SOC into slow and passive pools, and the computed C fluxes would be reduced by about 5%. However, it is highly unlikely that the soil samples analysed for radiocarbon will have contained all of the fast pool, much of which will have been removed, either as above-ground relatively coarse material, by picking out dead roots, or by sieving. Therefore, neglect of the fast pool is justifiable.

Choice of fixed mean residence times

The choices of 20 and 1000 years in Model III are somewhat arbitrary, although they have the advantage of bracketing the observations (Figure 1) so that the great majority of soils can be described. We ran Model III in steady state with different MRTs for the slow fraction, to fit representative data (Table A3a). If an attempt is made to contain more data by reducing the slow MRT to 10 years (cf. Figure 1) unrealistic results are obtained, since the required inputs of C to the slow pool become very high (> 400 g m^{-2} a^{-1} for forest soil), and too similar to net primary production (= litter input) to be realistic. For the default case, if the value is increased from 20 to 30 or 40 years, the results remain physically reasonable. However, using higher residence times would mean losing more sites from the available data (cf. Figure 1), and so there is a definite constraint if we require a consistent model that account for most observations. Therefore the original choice of 20 years can be justified.

The passive MRT is set at 1000 yr in order to represent the most stable topsoil organic matter. An interesting result starting with the default non-forest soil (Table A1) and then keeping the C fluxes and slow pool MRT at the default values, but adjusting the passive pool MRT in order to attempt to simulate the default forest soil. The closest agreement is obtained if the passive MRT is set to 600 yr, which yields a total topsoil C pool of 5.8 kgC m^{-2} and a ^{14}C of 109.0%, which are quite close to the default forest values of 6.0 kgC m^{-2} and 110.1% respectively. This suggests that an alternative modelling approach could be based on different fixed MRT values for forest and non-forest, or on the adjustment of the passive MRT for different soils.

Further insight comes from application of Model III to data sets with ^{14}C observations made at the same site at different times. The most-studied site in this regard is a fertilised (and therefore not included in our database) grassland at Judgeford in New Zealand (O'Brien & Stout, 1978; Baisden et al., 2013) for which topsoil radiocarbon content has been measured on 10 occasions, between the years 1959 and 2002. Model applications with different assumptions about the MRTs of the slow and passive pools (Table A4) show only modest variations in the goodness-of-fit, and do not permit a definitive choice of turnover rates, although the smallest errors are achieved with a slow MRT of 20 years, our chosen value. Similarly, results for Meathop Wood (4 data points; see Figure 1 of the main paper) do not show sufficient differences in goodness-of-fit (Table A4) to decide upon exact MRT values.

References

Baisden WT, Parfitt RL, Ross C, Schipper LA & Canessa S (2013) Evaluating 50 years of time-series soil radiocarbon data : towards routine calculation of robust C residence times. Biogeochem 112: 129-137

O'Brien BJ & Stout JD (1978) Movement and turnover of soil organic matter as indicated by carbon isotope measurements Soil Biol Biochem 10: 309-317

Table A3. Results of simulations with Model III using different assumed MRT values for the slow pool. The passive pool rate was set to 1000 years in all cases. Default soils (Table A1) were used as the basis for simulations.

	MRT slow yr	slow fraction	slow input gC m^{-2} a^{-1}	passive input gC m^{-2} a^{-1}
forest	10	0.73	438	1.6
	15	0.66	263	2.1
	20	0.65	195	2.1
	30	0.67	134	2.0
	40	0.73	110	1.6
non-forest	10	0.51	379	3.7
	15	0.46	229	4.1
	20	0.44	165	4.2
	30	0.46	114	4.1
	40	0.49	91	3.9

Table A4. Fitting results from the application of Model III to two multi-point data sets. Goodness-of-fit is evaluated by the root-mean-squared-deviation (rmsd) between observed and simulated ^{14}C.

		Judgeford		Meathop Wood	
slow MRT yr	passive MRT yr	slow fraction	rmsd in ^{14}C	slow fraction	rmsd in ^{14}C
10	500	0.41	1.1	0.69	2.6
20	500	0.63	1.1	0.81	2.6
30	500	0.82	1.3	0.87	2.6
10	1000	0.50	0.9	0.55	2.6
20	1000	0.71	0.8	0.71	2.6
30	1000	0.87	1.1	0.79	2.6
10	2000	0.59	1.4	0.44	2.5
20	2000	0.78	0.8	0.61	2.5
30	2000	0.90	1.0	0.70	2.5