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Long-term bare fallow experiments offer new opportunities for the quantification and the study of stable carbon in soil

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Abstract

The stability of soil carbon is a major source of uncertainty for the prediction of atmospheric CO_2 concentration during the 21st century. Isolating experimentally the stable soil carbon from other, more vulnerable, pools is of prime importance for calibrating soil C models, and gaining insights on the mechanisms leading to soil organic carbon (SOC) stability. Long-term bare fallow experiments, in which the decay of SOC is monitored for decades after inputs from plant material have stopped, represent a unique opportunity to assess the stable organic carbon. We synthesized data from 6 bare fallow experiments of long-duration, covering a range of soil types and climate conditions, at Askov (Denmark), Grignon and Versailles (France), Kursk (Russia), Rothamsted (UK), and Ultuna (Sweden). The conceptual model of SOC being divided into three pools with increasing turnover times, a labile pool (~ years), an intermediate pool (~ decades) and a stable pool (~ several centuries or more) fits well with the long term SOC decays observed in bare fallow soils. The modeled stable pool estimates ranged from 2.7 gC kg^{-1} at Rothamsted to 6.8 gC kg^{-1} at Grignon. The uncertainty over the identification of the stable pool is large due to the short length of the fallow records relative to the time scales involved in the decay of soil C. At Versailles, where there is least uncertainty associated with the determination of a stable pool, the soil contains predominantly stable C after 80 years of continuous bare fallow. Such a site represents a unique research platform for future experimentation addressing the characteristics of stable SOC and its vulnerability to global change.

1 Introduction

Soils contain about three times more carbon than the atmosphere (Jobbagy and Jackson, 2000) and it is recognized that small variations in soil carbon may have important consequences for atmospheric CO_2 concentrations (Cox et al., 2000). We know that soil organic matter (SOM) levels in soils are governed by soil texture, climate, the input

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of organic material and its rate of decomposition and the rate at which existing SOM decomposes (Johnston et al., 2009). However, Heimann and Reichstein (2008), contend that SOC carbon dynamics are poorly understood from the point of view of their sensitivity to climate (Davidson and Janssens, 2006), and to ecosystem productivity changes (Jones et al., 2005). This leads to a wide range in the global model predictions of soil carbon storage in the future (Friedlingstein et al., 2006; Sitch et al., 2008). A better understanding of SOC stability and vulnerability to land use change and to climate change as well as improved modeling of the soil carbon cycle are therefore crucial to providing more accurate predictions of future atmospheric CO_2 concentration.

Using ^{14}C measurements (Trumbore et al., 1997; Jenkinson et al., 2008) or using long-term C_3/C_4 vegetation changes (Balesdent et al., 1988), a fraction of SOC has been shown to be hundreds to thousands of years old. In soil C models such compounds are represented by a C pool with a high turnover time (several centuries) or even by an inert C pool (Falloon and Smith, 2000). However, in many respects, this stable soil carbon remains terra incognita. Indeed, the mechanisms explaining its stability are poorly understood (Fontaine et al., 2007) and its vulnerability to climate change is highly debated (Knorr et al., 2005; Reichstein et al., 2005; Fang et al., 2006). Moreover, as the stable C pool has not been isolated or characterized experimentally, it has never been properly quantified and its size has only been estimated through model outputs, or extrapolated from brief experiments 2 or 3 orders of magnitude shorter than the typical turnover time of the slowest C pools. A better understanding of this C pool is particularly crucial as it represents a potentially long lasting carbon sink. Elliott et al. (1996) and Christensen (1996) challenged soil scientists and modelers to "measure the modelable or model the measurable". We aim here to take up this challenge by showing that it is possible to isolate the stable C pool from other C pools, by finding soils that seemingly contain only stable C or at least soils with a very large proportion of stable C.

The easiest, if not the only way, to be sure that only old C is present in a soil, is to stop C inputs and wait patiently for the more labile compounds to decompose. Long-term

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bare fallow experiments (LTBF) provide such conditions. Bare fallow plots, which were designed as reference plots in several long-term agronomical experiments, are plots that have been kept free of plants through manual or chemical treatments for several decades. Although C inputs are not technically zero in bare fallow plots, due to atmospheric deposition, or occasional weeds, such inputs can be considered as negligible. Consequently, if bare fallow plots are sampled regularly, one can measure directly the decay of the SOC initially present and estimate empirically through first order kinetics assumptions the concentration of the stable carbon pool and the uncertainty of that estimate. Until now, existing bare fallow plots have seldom been investigated in this respect and have not been compared with each other. Yet, they represent a unique opportunity for quantifying and characterizing stable soil C in different soils.

The present work had several objectives. These are to: (i) initiate a network of existing LTBF experiments and synthesize the data from several sites; (ii) compare SOC decays with time at the LTBF plots; (iii) determine whether soil C decay in LTBF plots can be explained by compartmental models with first order kinetics; (iv) estimate the stable C pool concentration and determine if it has been reached.

2 Materials and methods

2.1 Descriptions of the sites included in the LTBF network

Bare fallow experiments included in the network needed to meet the following criteria: (i) plots kept bare for more than 25 years; (ii) regular measurements of % organic C in the soil of ploughed layer. Six sites met the required criteria and were included in the network.

2.1.1 Askov

The Askov bare fallow experiment was initiated in 1956 on the Askov Lermarken site. The experiment was adjacent to the B3- and B4-fields (blocks) of the Askov Long-Term

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Experiments on Animal Manure and Mineral Fertilizers (Christensen et al., 2006). The Lermarken site was first brought into cultivation around year 1800. According to Land Register maps from 1793, the site was at that time covered in open, mixed heath- and grassland with scattered deciduous scrubs. Most likely the area had historically been used for occasional haymaking and light grazing. The soil originates from morainal deposits and classifies as a Typical Hapludalf (USDA Soil Taxonomy). The texture qualifies as a coarse sandy loam, the clay fraction being dominated by illite, smectite and kaolinite. The pH of the topsoil is kept at 5.5 to 6.5 by liming every 4 to 5 years. Soil bulk density is 1.50 g cm^{-3}. Mean annual precipitation and temperature (1960–1990) is 862 mm and 7.7 °C. The plot size of the Askov LTBF was 11.7 m × 9.4 m (110 m^2), with four replicate plots in both the B3- and the B4-field. The plots were kept free of vegetation by frequent tillage but received annual dressing of mineral NPK fertilizers. Bulk soil (composed of 9 to 11 cores) was sampled every year (spring) from the 0 to 20 cm depth of each replicate plot and analyzed for total-C content by dry combustion. The experiment was terminated in 1985, and further details on the experiment can be found elsewhere (Christensen, 1990; Christensen and Johnston, 1997; Bruun et al., 2003; Christensen et al., 2006).

2.1.2 Grignon

The Grignon LTBF experiment forms part of the larger 36-plot experiment, created in 1959 in the gardens of the "Chateau de Grignon". In this experiment, 6 treatments (bare fallow, bare fallow + nitrogen, straw, straw + nitrogen, composted straw and composted straw + nitrogen) were replicated 6 times in 6 blocks. Only the 6 bare fallow plots were used in the present study. Previous land use was unmanaged grassland. The soil is a silty loam textured Agrudalf (USDA Soil Taxonomy), developed within a colluvial carbonated deposit, with pH (in water) of 8.0–8.3. The size of the plots is 3.2 × 3.2 m^2; all plots are dug by hand twice a year to 25 cm depth, and are kept free from vegetation by hand weeding and chemical treatment. OC concentrations were measured by dry

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combustion. Further details on the experiment can be found elsewhere (Morel et al., 1984; Houot et al., 1989).

2.1.3 Kursk

The 45-year old Kursk continuous fallow field is located within the long-term field experiment of the Kursk Institute of Agro-Industrial Production in the Kursk Region of Russia. Formerly it was part of Kursk State Agriculture Experimental Station, Central Branch the site was brought into cultivation more than 200 years before start of the experiment in 1964. The experiment (355 × 605 m) was designed to compare dynamics of crop yield and soil parameters in 5-year rotations and continuous crops (corn, alfalfa, potatoes, winter wheat, peas) given two different background treatments – control and application of N200P250K150 and 20 t ha^{-1} FYM per rotation. The site is located in the forest steppe climatic zone – temperate, moderately cold. The soil is a haplic chernozem (typical deep heavy loam chernozem on loess according to Russian classification). Initial soil pH (KCl) was 6.5 and exchangeable Ca 42 $cmol^{+}$ kg^{-1}. The large (150 × 14 m) bare fallow plot has been continuously plowed to 22–25 cm twice a year. Weed growth has been controlled by periodic cultivation (usually up to 5 times per vegetation period) to a depth of 10 cm. Initial mixed core samples were collected in 1965 and 1970 from 10 × 10 m plots within a 18 × 14 m part of continuous fallow plot, followed by individual sampling in 1978, 1983, 1988 and 2001: ten samples from 0–25 and 5 from 25–40 cm. C was analyzed by dry combustion.

2.1.4 Rothamsted

The Rothamsted Bare Fallow was started in December 1959 on a site which had been in managed grassland since 1838. It is adjacent to a Ley-arable experiment which had started 11 years earlier and its purpose was to provide complementary data on changes in soil organic C. The soil is a flinty silty clay loam (Chromic Luvisol; F.A.O., 1990). The area of the site is c. 50 × 7 m; this is divided into four sub-plots,

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each 12.5 × 7 m, for sampling purposes. Soils were sampled in December 1959 (immediately before the site was first ploughed) using semi-cylindrical augers and sampling to a depth of 23 cm. It has been sampled on six occasions subsequently, most recently in September 2008. On two occasions, 1978 and 2000, soils were taken from the Bare Fallow and adjacent areas of permanent grass, with a sampling box (c. 15 × 15 × 23 cm deep) to determine bulk density. In December 1959, organic C in the soil was 29.0 gC kg^{-1} and pH (in water) was 6.3; in 2008, organic C had declined to 10.0 gC $kg^{-1}$and pH to 5.2. The site is kept free from weeds by plowing/cultivating 2–4 times per year; herbicides are used occasionally. Meteorological data is from a weather station c. 200 m from the Bare Fallow site.

2.1.5 Ultuna

The "Ultuna continuous soil organic matter experiment" was started in 1956 on a field at the Swedish University of Agricultural Sciences that has probably been used for agriculture for several hundred years. The main aim of the experiment is to test the long-term effect of mineral fertilizers and organic amendments on crop yields and soil characteristics such as carbon content. The experiment consists of 15 treatments replicated in four blocks, 60 plots in total. The size of each plot is 2 × 2 m. The plots are separated with wooden frames that extend initially about 30 cm into the soil and about 10 cm above the soil surface. Since 1956, all cultivation has been performed by hand, with a spade. The soil was sampled using a soil corer at five random locations in each plot that were combined to one composite sample before analysis. C was analyzed using the Walkley-Black method from 1956 to 1983 and by dry combustion thereafter. Basic information and results for the first 35 years of the experiment were presented by Kirchmann et al. (1994). Earlier time series of C data from the bare fallow as well as other treatment have been used for calibrating and validating different soil C models (Andrén and Kätterer, 1997; Hyvönen et al., 1996; Paustian et al., 1992; Petersen et al., 2005).

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2.1.6 Versailles

The Versailles LTBF experiment forms part of the larger 42-plot experiment, created in 1928 in Versailles at the Central Agronomy Station, becoming INRA in 1946. Located in the gardens of the "Chateau de Versailles", this experiment was designed to study the effects of long-term application of major fertilizers and amendments on the composition and physical properties of loess soils (Burgevin and Hénin, 1939). Previous land use was unmanaged grassland. Ten of the 42 plots were used as bare fallow reference plots. The six plots sampled the most regularly since 1929 have been analyzed for C content in 2009 and are included in this study. The soil is a silty loam textured Luvisol, developed in aeolian loess covers, characteristic of the Paris Basin. In 1929 the soil had a pH of 6.4 and a CEC of 15.3 $\mathrm{cmol^+\,kg^{-1}}$ (Cobaltihexamine method). Seventy years later (1999), the pH of the reference plots had decreased to 5.6 and the CEC to 8.7 (Pernes-Debuyser and Tessier, 2002). The size of the plots is $2 \times 2.5\,\mathrm{m}^2$, all plots are dug by hand twice a year to 25 cm depth, and are kept free from vegetation by hand weeding and chemical treatment. OC concentrations were measured by dry combustion.

2.2 Soil C stock measurements

Initial and final soil stocks were calculated on an equivalent soil mass basis. When no bulk density changes were reported (Ultuna) or assumed (Kursk and Askov), soil stock was calculated by multiplying the soil mass in the sampled horizon by the C concentration in the same layer. For sites where bare fallows were established on grassland soil, bulk density was observed to increase with time (Rothamsted and Versailles) or was assumed to increase (Grignon). At Grignon, where bulk density has never been measured, bulk density values were determined according to the following relationship proposed by Kätterer et al. (2006):

$$\text{Bulk density } (\mathrm{g\,cm^{-3}}) = 1.6384 - 0.0945 \times \text{SOC } (\%)$$

Due to increased bulk density, the sampled horizons contained a higher soil mass at the last sampling date than at the initial sampling date. To calculate C stocks on an equivalent soil mass basis, the C mass contained in the soil that was below the sampling depth initially but in the sampling horizon eventually was added to the C mass contained in the sampled horizon at the first sampling date. The C concentration of the extra soil mass was assumed to be 12, 11 and 10 gC kg^{-1} at Rothamsted in 1959, 1963 and from 1971 to 2009 respectively and 10 gC kg^{-1} throughout the experiments at Versailles and Grignon. These values were estimated from C concentrations measured in soil horizons just below the sampling depth (23-25cm at Rothamsted and 25–40 cm at Grignon). For Rothamsted, where changes in bulk density with time are available (0.94 g cm^{-3} in 1959, 1.01 g cm^{-3} in 1963, 1.13 g cm^{-3} in 1971 and 1.25 g cm^{-3} since 1978), changes in the amount of C (t ha^{-1}) with time were calculated. Due to this bulk density increase, C stock values correspond to 0–30.7 cm in 1959, 0–28.6 cm in 1963, 0–25.3 cm in 1971 and 0–23 cm from 1978 to 2008. For the other sites, only initial and final C stock values were calculated.

2.3 SOC decrease fits

Linear ($y = a\ t + b$), mono-exponential ($y = a\ \exp(-b\ t)$), mono-exponential + constant ($y = a\ \exp(-b\ t) + c$), bi-exponential ($y = a\ \exp(-b\ t) + c\ \exp(-d\ t)$) and bi-exponential + constant ($y = a\ \exp(-b\ t) + c\ \exp(-d\ t) + e$) models, where t represents time under bare fallow and a, b, c, d, e parameters, were fitted to each replicate using a Bayesian curve fitting method described by Tarantola (1987). We aimed to find a parameter set that minimized the difference between model outputs and the corresponding observations, considering model and data uncertainties, and prior information on parameters (see below). With the assumption of Gaussian errors for both the observations and the prior parameters, the optimal parameter set corresponds to the minimum of the cost function $J(x)$:

$$J(\boldsymbol{x}) = 0.5[(\boldsymbol{y} - H(\boldsymbol{x}))^t \mathbf{R}^{-1} (\boldsymbol{y} - H(\boldsymbol{x})) + (\boldsymbol{x} - \boldsymbol{x}_b)^t \mathbf{P_b}^{-1} (\boldsymbol{x} - \boldsymbol{x}_b)] \tag{1}$$

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that contains both the mismatch between modeled and observed C content and the mismatch between a priori and optimized parameters, $\boldsymbol{x}$ is the vector of unknown parameters, x_b the a priori value, $H()$ the model and $\boldsymbol{y}$ the vector of observations. The covariance matrices $\mathbf{P_b}$ and $\mathbf{R}$ describe a priori uncertainties on parameters, and observations, respectively. Both matrices are diagonal as we suppose the observation uncertainties and the parameter uncertainties to be independent. A very large a priori error on parameters was considered in order not to influence the fitting procedure except that it was assumed that the decays cannot be positive by defining a lognormal distribution for these parameters. For the data errors, we used the standard deviation estimated from 15 measured values for C in soil samples taken from the same plot at Grignon in 1959. The measured standard deviation was 0.3 gC kg^{-1}. As the C content was determined on composite samples from the same plot in the different sites, it was considered that the error on measurements should be less than 0.5 gC kg^{-1}. This error on the measurement was considered a priori for every site. However, it appeared that the values of the cost function at the minimum (expressed as chi-2 values) were too big for Askov, Kursk and Versailles, given the hypothesis of Gaussian errors we made. This suggests that data or parameter errors were too small. We thus increased the 0.5 gC kg^{-1} data error to 0.75 gC kg^{-1} for these sites, keeping large parameter errors not to nudge the solution. The new normalized chi-2 values were all lower than 1 (a statistical criteria) and consequently considered as acceptable. Errors on measurements were therefore considered to be equal to 0.5 gC kg^{-1} for Grignon, Rothamsted and Ultuna and equal to 0.75 gC kg^{-1} for Askov, Kursk and Versailles. To determine an optimal set of parameters which minimizes $J(x)$, we used a gradient-based algorithm (1987). We typically converged to a minimum of $J(x)$ within less than 5 iterations.

Once each replicate was fitted, parameters values were averaged at the site scale and the error covariance matrix of the parameters was estimated using the linearity assumption at the minimum of $J(x)$. This parameter error covariance matrix was also propagated on the model outputs, i.e. the predicted SOC content (95% confidence interval in Fig. 1). For each site, best models were selected according to the Akaike's

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information criterion (AIC) developed by Akaike (1974). This criterion considers both the goodness of fit and the number of free parameters:

$$AIC = 2k + n[\ln(RSS/n)] \tag{2}$$

where k corresponds to the number of free parameters, RSS is the residual sum of squares and n the number of observations.

In a second step, it was considered that SOC decrease should reach a positive plateau (corresponding to the stable pool). For this second series, we considered a lognormal distribution for the plateau parameter and the optimization was redone. For each site, we assumed a priori that the plateau was comprised in a 95% interval confidence that was [0.6 gC kg^{-1}; 35.6 gC kg^{-1}].

3 Results

3.1 Presentation of the LTBF network

A survey of Long Term Bare Fallow (LTBF) experiments identified several sites, all located in Europe. A network of six LTBF sites was initiated including: Askov (Denmark), Grignon and Versailles (France), Kursk (Russia), Rothamsted (UK) and Ultuna (Sweden). Selected characteristic data of these sites are presented in Table 1.

The age of the bare fallow experiments ranges from 30 years (Askov) to 81 years (Versailles). At Askov, the bare fallow experiment was conducted on two blocks with four replicates per block. The two blocks have been analyzed separately in the present work. The soils are clayey and/or silty textured, except for Askov which is a sandy soil. Kursk and Ultuna are colder than the other sites; Grignon and Versailles are the warmest. Askov is wetter than Rothamsted which is wetter than the other four sites. The LTBF plots were initially established on arable land at Askov, Kursk and Ultuna and on grassland at Rothamsted, Grignon and Versailles. The history of the Rothamsted, Versailles and Grignon sites are different. At Rothamsted, the bare fallow was established on land that had been in managed grassland since 1838 and the first samples were taken from the grassland immediately before ploughing and conversion to bare fallow. In contrast, at Versailles and Grignon, the bare fallows were established on unmanaged grassland but the first samples were taken several months after ploughing and conversion to bare fallow.

Bulk density has been regularly determined at Rothamsted and Ultuna. At Rothamsted, the soil became more compact with time under bare fallow, whereas the bulk density did not vary significantly at Ultuna. At Versailles the void ratio and bulk densities were measured in 1929 and 2008 respectively. Assuming a 2.60 g cm^{-3} density for the solid phase of the soil, bulk density evolved from 1.30 g cm^{-3} in 1928 to 1.44 g cm^{-3} in 2008. At Askov, the bulk density has been considered by Bruun et al. (2003) to be constant and equal to 1.50 g cm^{-3}. At Kursk, a 1.13 g cm^{-3} bulk density was measured in 1983 (Lazarev, 2007). Bulk density has never been determined at Grignon and was assumed to increase from 1.50 g cm^{-3} in 1959 to 1.56 g cm^{-3} in 2007 according to the pedotransfer function proposed by Kätterer et al. (2006). Initial and final C stock values calculated using measured or estimated bulk density are given in Table 1. However, due to the sparse information on soil bulk density, this work mostly discusses changes in C concentrations. However, as bulk density variations are relatively small, except at Rothamsted (where the amount of C present in the plow layer as well as C concentrations will be discussed) the conclusions based on C concentration data are probably also valid for C stocks.

3.2 SOC losses in bare fallow soils

Changes in SOC concentrations with time in the plowed layer at all sites are presented in Fig. 1. Initial SOC concentrations ranged from 13 to 20 gC kg^{-1} at Askov, Grignon, Ultuna and Versailles. Initial SOC concentration was larger at Rothamsted and even larger at Kursk. SOC decreased with time at all sites. However, at Grignon, Versailles and more markedly Rothamsted, where bare fallows were established on grassland

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soils, SOC exhibited a much steeper initial decrease with time compared to the other sites. In contrast, sites previously in arable cropping did not show a steep SOC decrease during the first years following the start of bare fallowing. During the 10 first years, SOC decrease rates were on average lower than 1.6 gC kg^{-1} per year for sites formerly under arable cropping and greater than 3.5 gC kg^{-1} per year for sites formerly under grassland. The steep decrease observed after grassland conversion to bare fallow is likely due to the quick exhaustion of labile organic material.

Figure 1h shows the decrease in the amount of SOC for equivalent soil masses at Rothamsted (soil mass in the 0–23 cm horizon at the last sampling date was considered as the reference soil mass). The shape of the decay curve for the amount of C is similar to that for C concentration. However, as bulk density increased rapidly during the first 19 years under bare fallow at Rothamsted, the initial SOC decrease is less steep when working with C masses in equivalent soil masses. Indeed, after 19 years, the soil had lost 49% of its C when using concentration data but only 40% when considering the amount of C.

3.3 Testing of simple compartmental models of SOC decay

Linear model and first order models with up to three pools were successively tested to represent SOC concentration decays. The different models were fitted to the data using a least square minimization approach, assuming Gaussian errors for the data and the model. When a model was over-parameterized (the last pool concentration was not different from 0) or did not provide a better Akaike's Information Criterion (AIC) (this criteria balances the goodness of fit and the degrees of freedom of the model; Aikaike, 1974) than the previous one, the models with more parameters were not investigated. The best fit was considered to be the fit with the lower AIC having all its parameters significantly different from 0. The best fits for each site are plotted on Figure 1 and the AIC of all tested models are reported on Table 2.

Two-pool models performed better at sites formerly under grassland. For Grignon it is a mono-exponential + constant, i.e. a bi-exponential model with the second exponential

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parameter equal to 0. The best fit at Grignon suggests the existence of a SOC pool with a 33-year turnover time and a SOC plateau. The existence of this plateau could reflect a pool that has not decomposed at all since the beginning of the LTBF, or a pool that is decomposing so slowly that it can be considered as inert over the duration of the experiment. Best fits at Rothamsted and Versailles do not have a plateau, but a second exponential decay with a longer turnover time instead (150 and 350 years respectively). The first exponential decay has a very short turnover time at Rothamsted (c. 7.5 years) whereas it is longer and comparable to Grignon at Versailles (c. 20 years). At Rothamsted, the bi-exponential model also performed best when working with SOC stocks. Contrary to Rothamsted, the initial steep SOC decay at Grignon and Versailles was not described by a short-lived C pool. This suggests that, at these two sites, the quantity of labile OM was too small to introduce an individualized labile carbon pool to account for its decay, in the fitting process.

By contrast, mono-exponential or linear fits performed better on sites formerly under arable cropping. The fits, with lower AIC, also give SOC plateaus at Askov B4 and Ultuna, but the plateau values are not significantly different from zero. At Askov B3 and Kursk, the data do not indicate the existence of a plateau which is probably due to the shorter duration of these bare fallow experiments.

3.4 Comparing SOC decays at the different sites

It is necessary to fit the same, or at least comparable, models to all data to compare SOC decays between sites. The mono-exponential + constant model is the most consensual. It is the best model at Grignon, it would be the best at Askov B4 and Ultuna if the plateau were significant and it performed reasonably well at other sites apart from Rothamsted. However, it can easily be compared with a bi-exponential + constant model which did work quite well on Rothamsted. Therefore, a mono-exponential + constant fit was adjusted to all dataset except Rothamsted, but in these fits the constant was forced to a significant positive value using a lognormal transformation of that parameter in the fitting process. Within the conceptual framework of three C pools,

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the exponential decay can be interpreted as the decay of the intermediate pool or a mixed pool including labile material and the constant as the concentration of the stable pool (truly inert or so slowly decomposing that it appears inert at the time scale of the experiment). Similarly for Rothamsted, a bi-exponential + constant model (a labile, an intermediate and a stable pool) with a forced positive constant was adjusted. Doing so the initial steep decay observed at Versailles and Grignon, probably corresponding to labile organic matter, is incorporated into the intermediate pool. To test the influence of labile organic C on the decay rate of the intermediate pool and the concentration of the stable pool, mono-exponential + constant fits were also applied to Versailles and Grignon sequences after removing the data from the first 10 or 12 years respectively (bare fallow plots were sampled after 10 years at Versailles and 12 years at Grignon).

Table 3 reports the turnover time of the intermediate pool (years), the concentration of the stable pool (gC kg^{-1} soil), its size in the sampled horizon (tC ha^{-1}) and the AIC values of each fit. The differences between sites are not striking. The 95% confidence intervals are very wide for every parameter. This suggests that one will have to wait for a while to have better constraints on these parameters. The turnover time of the intermediate pools ranged from c. 21 years at Versailles to 133 years at Kursk. The turnover time of the intermediate pool was significantly higher at Kursk than elsewhere except Ultuna, Rothamsted and Grignon truncated. It is also significantly lower at Versailles than at other sites except Askov B4 and Grignon. There is no significant difference between the estimates of the stable carbon pool concentration across the sites. At this stage, the fits predict approximately 3–4 g of stable C per kg of soil for all sites but Versailles and Grignon. It is worth noting that the uncertainty on the plateau at Kursk is the same as the a priori uncertainty (0.66 gC kg^{-1}-35.60 gC kg^{-1}), meaning that the data does not yet give any constraint to the plateau value. The removal of the first 10 or 12 years increased the turnover time of the intermediate pool and lowered the concentration and size of the stable pool at Grignon, but did not change anything at Versailles.

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Figure 2 shows the modeled concentration of stable pools (i.e. the constant parameter of the chosen model) and the C concentration measured at the last sampling date. Strikingly, C concentration measured in bare fallow soils in 2008 at Versailles are very close to the modeled stable pool size values estimated from both truncated and non-truncated data. This indicates that the SOC remaining in the Versailles bare fallow plots is mostly if not exclusively stable carbon. At the other sites, the stable C pool has not yet been reached.

4 Discussion

4.1 Bare fallow data supports the three pool concept used in most soil C models

Most soil C models divide SOC into three carbon pools: a labile one (~ years), an intermediate one (~ decades) and a stable one (several centuries or more). Successfully observing a short-lived and small pool like the labile pool using long-term soil C data is very unlikely as the labile pool considered in compartmental models is very small compared to the others (e.g. ≈1% of total SOC in the Century model (Kelly et al., 1997)) and by definition it is quickly exhausted. The optimizing algorithms revealed that SOC of the LTBF soils can be described by one pool with a turnover of several decades and one stable pool for all sites, except Rothamsted where three pools can be observed, thanks to the sharp initial decline of SOC which equates to a quickly disappearing labile pool. Considering that we have probably missed the decay of a similar labile pool at all sites except Rothamsted, we conclude that our LTBF dataset supports the "classical" three pool structure and first order decay kinetics shared by most soil models (Parton et al., 1987; Coleman et al., 1996).

Bare fallows established following grassland exhibited a steeper SOC decrease during the earlier years compared to bare fallows established after arable cropping. This

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suggests that there is more labile organic matter in grassland than in arable soils. Two complementary explanations can be proposed: (i) the tillage of the grassland soils prior to bare fallow implementation and during the bare fallow experiment has de-protected labile SOC components that were quickly mineralized; (ii) the grasslands have more roots than arable crops in surface soil horizons (Jackson et al., 1996) and the steeper initial SOC decrease at the former grassland site, represents the mineralization of the larger amount of organic material present in grassland soils when turned into bare fallow. Larger roots and other debris are usually removed when soil is sampled to determine C content, and, although root exudates and smaller root debris will have contributed to soil C content, it is unlikely that stopping such C inputs to soil can account for all of the 12 $gC\,kg^{-1}$ (approximately 25 $tC\,ha^{-1}$) lost in c. 12 years at Rothamsted in the 0–23 cm horizon. Consequently, we suggest that the deprotection of labile organic matter induced by the periodic tillage of previously untilled soils mostly explains the steeper SOC decrease observed at Grignon, Rothamsted and Versailles. The fitting routine did not propose a proper decay for labile pools at Versailles and Grignon which suggests that the labile pool was much larger at Rothamsted that at the other sites. This is explained by the fact that Rothamsted was the only site where the first samples were taken *before* the grassland was ploughed.

Turnover times inferred from the LTBF experiments for the labile (i.e. 5.2 years at Rothamsted) and intermediate (i.e. 18–133 years) C pools are in good agreement with those of the “active” and “slow” pools of the Century model (Parton et al., 1987) and those of the “microbial biomass” and “humified organic matter” of Roth-C (Coleman et al., 1996). In these models, pedo-climatic factors influence the decay rate of SOC pools, using empirical relationship difficult to discuss in the light of LTBF data. Indeed, no obvious or statistical relationship between estimated turnover times of the intermediate C pool and the pedo-climatic factors could be observed (Fig. 3). This can be explained by the large errors on the fit parameters and the small number of sites. It is unfortunate that more bare fallow experiments have not been established on different soil types, especially in warmer locations, in order to have a more contrasted range of

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climatic factors. Nonetheless, the relatively slower turnover observed at Kursk can be qualitatively explained, as Kursk is a cold and dry site with a very low sand content.

4.2 Estimation of the stable pool concentration through LTBF data

One outcome of the fitting procedure is an estimation of the concentration of C in the stable pool. It is approximately of 6 gC kg^{-1} for Versailles (truncated or not) and Grignon and around 3–4 gC kg^{-1} (near the a priori value) at Kursk, Askov, Ultuna, Rothamsted and Grignon truncated, but with large uncertainties. The shape of SOC decays at Grignon, Rothamsted and Ultuna suggests that the plateau value will probably be higher than the average value predicted. Altogether these results are in good agreement with those of Balesdent et al. (1988) who found a stable pool concentration of c. 7 gC kg^{-1} using a C_3/C_4 vegetation chronosequence.

The proportion of the stable pool concentration inferred from the fits is in accordance with the "passive" pool concentration commonly found using the Century model (Kelly et al., 1997). It is also in accordance with other estimates of the size of the compartment having the higher turnover times in other models such as CN-SIM (Petersen et al., 2005) or Daisy (Bruun et al., 2003). The unique body of data provided by the LTBF network therefore supports experimentally the concentration of the stable pool modeled by compartment models simulating the long-term dynamics of C in soil. The size of the stable C pool inferred from the LTBF data is however approximately 2 to 7 times larger than the "inert organic material" pool defined in the Roth-C model (Coleman and Jenkinson, 1996; Coleman et al., 1997; Falloon et al., 1998), a difference that could have a strong impact on the simulated terrestrial carbon balance in the long-term under climate and land-use change.

C inputs can increase or reduce SOC mineralization (Kuzyakov et al., 2000; Fontaine et al., 2003). These positive or negative priming effects are unlikely to have occurred in the LTBF experiments due to the absence of inputs. If they have occurred, priming effects may have changed the stable pool concentration. However, predicting the sign and the importance of the potential modification induced by priming effects is not easy

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as both positive and negative effects of fresh C inputs on SOC mineralization have been reported (Hamer and Marschner, 2005) and the quantified impact of priming effects on SOC stocks under field conditions has not yet been determined. However, one should keep in mind that LTBF data might need to be reconsidered once priming effects are more clearly understood. Indeed, LTBF sites, where no priming has occurred for a long time, could provide a suitable environment for experimental and modeling studies on this topic.

4.3 Soil containing only stable C: new opportunities

SOC concentrations measured in 2008 in the Versailles bare fallow soil are, on average, $6.30 \pm 0.93\,\mathrm{gC\,kg^{-1}}$. If we compare this with the estimated concentration of stable C (assessed from our model-data fit), we can deduce that at least 73% of the SOC of this soil is composed of stable carbon. At Grignon, the present SOC concentration would be close to the stable pool estimate when working with the whole dataset. In the other sites, estimating that proportion of SOC which is stable C is difficult, due to the large uncertainties associated with the estimates of C concentrations in the stable pool. Nonetheless, according to previous discussion on the possible under-estimation of the plateau value at Grignon, Rothamsted and Ultuna, we can consider that stable soil C will also be reached at these sites during the coming decades.

The long overlooked bare fallow experiments thus provide valuable data with which we can address the formidable challenge posed by Elliott et al. (1996) and Christensen (1996) to soil C scientists to “measure the modelable or model the measurable”. Most of the organic carbon in the surface layer of the bare fallow soil at Versailles is shown to be stable carbon. The comparison of this soil with stored samples from the same experiment or soil from adjacent plots under different land uses will allow us to try to characterize “the modelable”. Furthermore, isolating the stable C pool from the other C pools should allow us to take a step forward in the identification of the mechanisms explaining the long-term stabilization of SOC, which is a widely recognized limitation in the field. Moreover, this soil could also be used to test experimentally the vulnerability

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of stable soil C to global change which should bring important insights to the highly debated question of the comparative vulnerability of labile, intermediate and stable SOC components to increased temperature (Giardina and Ryan, 2000; Knorr et al., 2005; Fang et al., 2005, 2006). The stable pool will also likely be isolated in some other sites in the coming years (Ultuna, Rothamsted, Grignon) which will allow investigating the mechanisms leading to SOC stability, and assessing SOC vulnerability to warming on stable C developed under various pedoclimatic conditions.

5 Conclusions

Modeling of the data from the LTBF plots at Versailles suggests that these soils contain predominantly "stable" OC whereas soils of other LTBF sites are enriched in "stable" OC but still contain a mixture of "intermediate" and "stable" C. For theses reasons, long-term bare fallows open many exciting and unexplored opportunities to address the many unknowns and uncertainties concerning stable soil C and thus soil organic C in general. These sites, implemented decades ago, when the role of soils in the global C cycle was not a subject of interest, provide a unique opportunity to study stable soil C under contrasting pedo-climatic conditions. Due to the long time needed to get crucial data on stable C in bare fallow soils, it is unlikely that many new experiments will be started. Moreover, the results that new experiments could provide would, unfortunately, come too late to be used to mitigate atmospheric CO_2 concentration. We should consequently be grateful to those people who designed and maintained bare fallow experiments for such a long time without knowing that they would be used in such a way.

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Table 1. Selected site characteristics. Potential Evaporation (EP) was calculated over the bare fallow duration using Penman equation. The ratio annual rainfall/EP (P/EP) is a proxy for soil humidity. Soil masses used for C stock calculations correspond to the mass of soil in the sampled horizon at the last sampling date. Final C stocks refer to C mass in sampled horizon at the last sampling date. Initial C stocks refer to C mass in sampled horizon at the first sampling date plus the C mass that was in the soil below the sampling depth initially but in the sampling date eventually due to soil compaction. The C concentration of this extra soil mass was considered to be 12 $gC\,kg^{-1}$ at Rothamsted and 10 $gC\,kg^{-1}$ at Versailles and Grignon (see Methods).

Site	Kursk	Ultuna	Askov B3	Askov B4	Grignon	Versailles	Rothamsted
Longitude/ Latitude	51°73 N 36°19 E	59°49 N 17°38 E	55°28 N 9°07 E	55°28 N 9°07 E	48°51 N 1°55 E	48°48 N 2°08 E	51°82 N 0°35 E
Dates	1965–	1956–	1956–1985	1956–1985	1959–	1928–	1959–
Last sampling date	2001	2007	1985	1985	2007	2008	2008
History	Arable	Arable	Arable	Arable	Grassland	Grassland	Grassland
Size of plots (mxm)	10 × 10	2 × 2	11.7 × 9.4	11.7 × 9.4	3.2 × 3.2	2 × 2.5	7 × 12.5
Replication	1 rep	4 reps	4 reps	4 reps	6 reps	6 reps	4 reps
Sampling depth (cm)	25	20	20	20	25	25	23
Clay/Silt/Sand (%)	30/65/5	36/41/23	7/11/82	7/11/82	30/54/16	17/57/26	25/62/13
Mean annual temperature (°C)	5.4	5.5	7.8	7.8	10.7	10.7	9.5
Annual rain (mm)	574	533	862	862	649	628	712
P/EP (mm/mm)	0.40	0.45	0.73	0.73	0.43	0.43	0.61
Initial bulk density ($kg\,dm^{-3}$)	1.13[b]	1.44[a]	1.50[b]	1.50[b]	1.50[b]	1.30[a]	0.94[a]
Final bulk density ($kg\,dm^{-3}$)	1.13[b]	1.43[a]	1.50[b]	1.50[b]	1.56[b]	1.44[a]	1.43[a]
Soil mass used for C stock calculations ($t\,ha^{-1}$)	2825	2860	3000	3000	3900	3600	3289
Initial C stock ($tC\,ha^{-1}$)	100.3	42.5±2.4	52.1±5.9	47.7±1.5	53.6±3.5	65.5±4.3	71.7±2.0
Final C stock ($tC\,ha^{-1}$)	79.4	26.9±0.6	36.4±2.5	33.0±1.3	31.9±1.9	22.7±3.3	28.6±3.1

Bulk density values refer to measured (quoted by [a]) or estimated (quoted by [b]) values.

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Table 2. Akaike's Information Criterion values (AIC) of all tested models. Best fits (in bold) are the fits with the lowest AIC and all parameters significant. C = ns means that the constant value is not different from 0.

Site	Model				
	Linear	Mono-exponential	Mono-exponential + constant	Bi-exponential	Bi-exponential + constant
Kursk	**−2.8**	−2.2	−1.2 (C = ns)	/	/
Ultuna	−128.7	**−144.3**	−150.6 (C = ns)	/	/
Askov B3	**−156.4**	−147.3	−146.3 (C = ns)	/	/
Askov B4	−114.5	**−126.0**	−133.0 (C = ns)	/	/
Grignon	−93.7	−128.5	**−200.2**	−198.4	/
Versailles	64.8	20.4	−76.1	**−82.3**	−81.0 (C = ns)
Rothamsted	63.4	44.2	−10.2	**−63.4**	−62.9 (C = ns)
Rothamsted stock	118.4	94.2	45.2	**21.7**	8.2 (C = ns)

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Table 3. Akaike's information criterion values (AIC) of the mono-exponential + forced positive constant fits, current estimation of the turnover times and concentration of the C in the stable pool and size of the stable pool in the sampled horizon. 95% confidence intervals are associated to the estimation of the stable pool concentration and size and turnover times of labile and intermediate pools.

Site	AIC	Labile turnover time (years)	Intermediate turnover time (years)	Stable pool concentration ($gC\,kg^{-1}$)	Stable pool size ($tC\,ha^{-1}$)
Kursk	−1.2	/	133.3 (84.0–204.1)	2.97 (0.66–35.60)	8.4 (1.9–100.6)
Ultuna	−146	/	66.2 (44.8–99.0)	3.90 (1.93–9.17)	11.2 (5.6–26.4)
Askov B3	−143	/	65.4 (51.8–81.3)	2.48 (1.05–7.46)	7.4 (3.2–22.4)
Askov B4	−132	/	38.0 (27.2–51.9)	4.39 (2.10–10.84)	13.2 (6.3–32.5)
Grignon	−93	/	30.3 (17.5–50.0)	6.80 (4.86–9.82)	22.6 (16.2–32.7)
Grignon truncated	−104	/	74.6 (46.7–113.6)	3.26 (1.59–7.83)	10.8 (5.3–26.0)
Versailles	−76	/	24.4 (20.4–28.6)	6.12 (5.47–6.87)	22.0 (19.7–24.7)
Versailles truncated	−88	/	18.2 (7.2–38.3)	5.87 (4.56–7.68)	21.1 (16.4–27.6)
Rothamsted	−61.5	5.2 (4.1–6.7)	60.9 (38.9–95.6)	2.72 (1.05–7.03)	8.9 (3.5–23.1)

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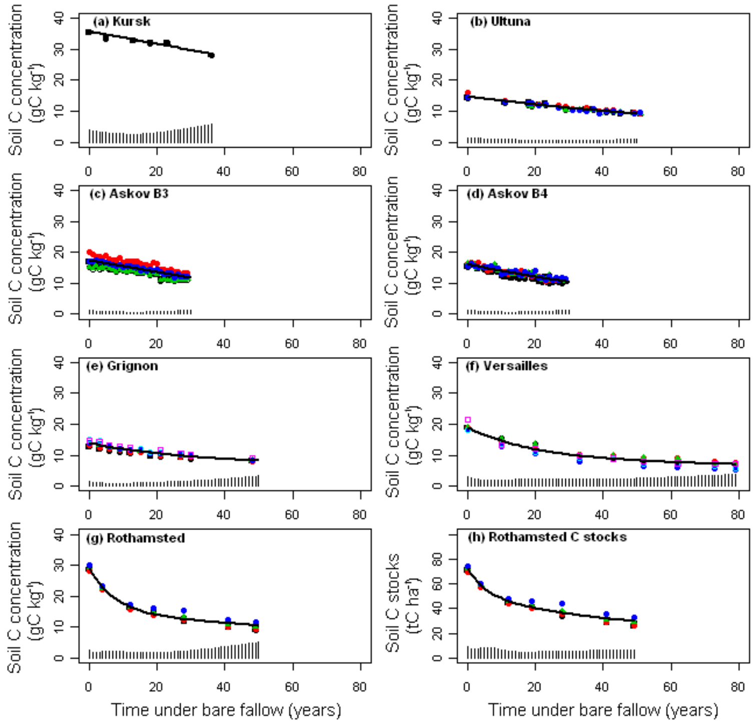

Fig. 1. Data and best fits. Different colors refer to different field replicates. The best fits are linear for Kursk and Askov B3; mono-exponential for Ultuna and Askov B4; mono-exponential + constant for Grignon; bi-exponential for Versailles and Rothamsted and bi-exponential + constant for Rothamsted when working with C stocks. The residuals are equally distributed (mean of the residues lower than 0.01 gC kg^{-1} excepted at Kursk where it is equal to 0.085 gC kg^{-1}) and did not present any trend with time. Vertical black bars correspond to the standard deviation multiplied by 5.

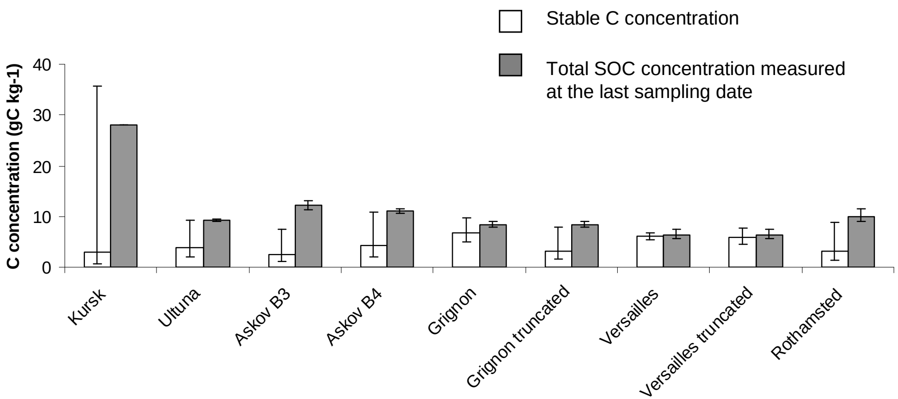

Fig. 2. Estimated stable pool concentration and soil C concentration measured at the last sampling date. Error bars refer to the 95% confidence interval for the stable C concentration and to minimal and maximal replicate values for the C concentration measured at the last sampling date.

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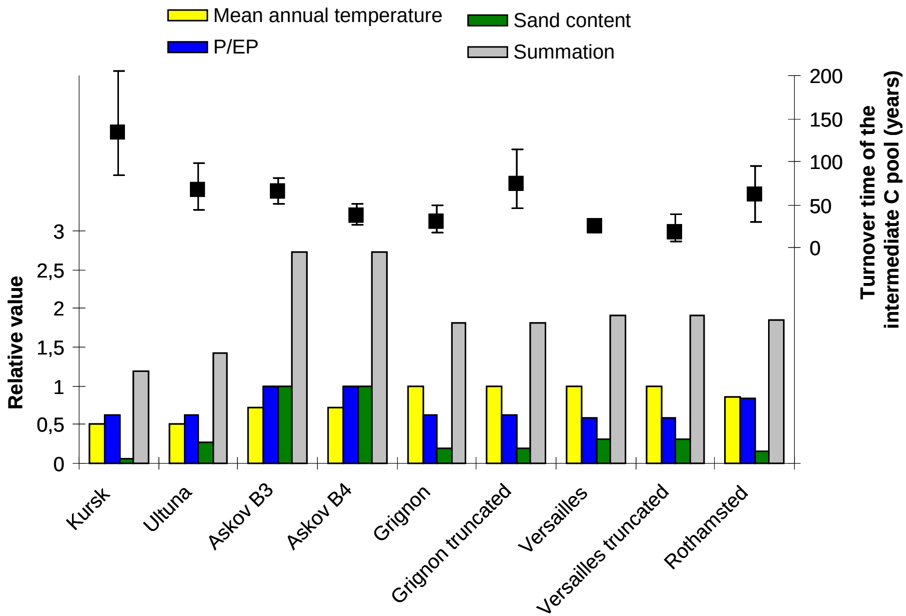

Fig. 3. Turnover time of the intermediate pools and pedo-climatical characteristics of the sites. Each mean annual temperature, humidity and sand content values were normalized by the highest value. The grey bars correspond to the summation of relative temperature, humidity and sand values. For each site the higher the grey bar, the quicker the turnover is expected (SOM is supposed to decompose more quickly in sandy, wet and warm soils). Black squares correspond to turnover time values of the intermediate pool with 95% confidence intervals.

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