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Jenkinson, D. S., Poulton, P. R. and Bryant, C. 2008. The turnover of organic carbon in subsoils. Part 1. Natural and bomb radiocarbon in soil profiles from the Rothamsted long-term field experiments. *European Journal of Soil Science*. 59 (2), pp. 391-399.

The publisher's version can be accessed at:

• https://dx.doi.org/10.1111/j.1365-2389.2008.01025.x

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# The turnover of organic carbon in subsoils. Part 1. Natural and bomb radiocarbon in soil profiles from the Rothamsted long-term field experiments

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### Summary

The Rothamsted long-term field experiments, started more than 150 years ago, provide unique material for the study of carbon turnover in subsoils. Total organic C, <sup>14</sup>C and <sup>13</sup>C were measured on soil profiles taken from these experiments, before and after the thermonuclear bomb tests of the mid-20th century. Four contrasting systems of land management were sampled: land cultivated every year for winter wheat; regenerating woodland on acid soil; regenerating woodland on calcareous soil; and old grassland. The mean radiocarbon ages of all the pre-bomb samples from cultivated land were 1210 years (0-23 cm), 2040 years (23–46 cm), 3610 years (46–69 cm) and 5520 years (69–92 cm). Bomb radiocarbon derived from thermonuclear tests was present throughout the profile in all the post-bomb samples, although below 23 cm the amounts were small and the pre- and post-bomb radiocarbon measurements were often not significantly different. Values of  $\delta^{13}$ C increased down the profile, from -26.3% (0–23 cm layer, mean of all measurements) to -25.2% for the 69–92 cm layer. The C/N ratios decreased with depth in virtually all of the profiles sampled. Excluding the surface (0-23 cm) soils from the old grassland, the hyperbola m = 152.1 - 2341/(1 + 0.264n) gave a close fit to the radiocarbon data from all depths, all sampling times and all sites, where n is the organic C content of the soil, in t ha<sup>-1</sup>, and m is the radiocarbon content of the soil, in  $\Delta^{14}$ C units, corrected for expansion or contraction of soil layers with time. The aberrant grassland soils almost certainly contained coal: one of them was shown by <sup>13</sup>C-NMR to contain 0.82% coal C. In Part 2 (this issue) of this pair of papers, these radiocarbon and total C measurements are used to develop and test a new model for the turnover of organic C in subsoils.

# Introduction

This paper presents radiocarbon and associated measurements made on soils taken before and after the thermonuclear bomb tests of the mid-20th century, tests which briefly doubled the radiocarbon content of the atmosphere. The soils came from plots on the Rothamsted long-term field experiments that have been under substantially the same management since the mid-19th century or from nearby areas of cultivated land that had been abandoned in the 1880s and have since reverted to woodland. Radiocarbon measurements in samples taken from the same place before and after the thermonuclear bomb tests provide a stringent test of any model for the turnover of organic carbon (C) in soil.

Correspondence: P. R. Poulton. E-mail: paul.poulton@bbsrc.ac.uk Received 26 September 2006; revised version accepted 2 January 2008 Apart from its intrinsic interest as a relatively unexplored part of soil science, subsoil C turnover is relevant to an important environmental issue: the effects of global warming on the stock of organic C held in soil. Of the 1600 Gt of organic C held in the top metre of the world's soils (Prentice, 2001), about half is in the 25–100 cm layer (Jobbágy & Jackson, 2000; Lal & Kimble, 2000).

Radiocarbon measurements indicate that there is a sharp increase in age down the profile (see for example, Scharpenseel & Becker-Heidmann, 1989; Pessenda *et al.*, 1996; Trumbore, 2000; Torn *et al.*, 2002). Furthermore, the proportion of soil organic C that is held as microbial biomass decreases with depth (Dictor *et al.*, 1998; Fierer *et al.*, 2003; Castellazzi *et al.*, 2004).

Global warming brought about by greenhouse gases will result in more rapid decomposition of soil organic matter, thus releasing more  $CO_2$  to the atmosphere. This positive feedback will add to the warming processes, to an extent that is under active debate (Jenkinson *et al.*, 1991; Kirschbaum, 2000; Friedlingstein *et al.*, 2001; Jones *et al.*, 2005; Knorr *et al.*, 2005). A realistic sub-model for the turnover of organic C in soil is an essential component of any fully-coupled model of the whole global carbon cycle. As a first step, it may be sufficient to treat the top metre of soil as a homogeneous unit when modelling the turnover of soil C on a global basis, as was done, for example, by Jenkinson *et al.* (1991). However, a more realistic sub-model, in which C turnover is not assumed to be the same down the profile, may well prove more useful to the global carbon cycle modellers.

For this work, four contrasting systems of land management were selected from the Rothamsted long-term experiments: land cultivated every year for winter wheat; regenerating woodland on acid soil; regenerating woodland on calcareous soil; and old grassland. Some of the soil samples described in this paper were also used by Poulton *et al.* (2003) in an earlier paper on the accumulation of C and N in land reverting to woodland.

The aim of this paper is straightforward: to present and discuss measurements of organic C and radiocarbon, made on soils sampled down the profile on selected plots from the Rothamsted long-term experiments, before and after thermonuclear testing. In Part 2 (this issue) we use these measurements and others taken from the literature to develop and test a dynamic model for the turnover of organic carbon in subsoils.

#### Materials and methods

#### Experimental sites

All of the sites were on Rothamsted Experimental Farm (51°49'N, 0°21'W). The old cultivated site (termed 'Broadbalk Arable' throughout this and the succeeding paper) was plot 08 on the Broadbalk Winter Wheat experiment, started in 1843, on land that was marked as under cultivation on a map of 1623. This plot receives N (144 kg N ha<sup>-1</sup>), P, K and Mg annually and is limed as needed to prevent acidification: for details of treatments and yields see Poulton (2006).

The two areas of regenerating woodland (subsequently termed 'Broadbalk Wilderness' and 'Geescroft Wilderness') have developed on two old cultivated areas that were fenced off and allowed to revert to woodland in the 1880s; for site maps and a detailed history see Poulton *et al.* (2003). Broadbalk Wilderness was and is calcareous; Geescroft Wilderness, although neutral when fenced off, is now strongly acid.

Broadbalk Wilderness is located at the top end of the field carrying the Broadbalk Winter Wheat experiment and was once part of this field. The starting Broadbalk samples were not taken from the actual site of the future Wilderness, but in 1881 from the unfertilized plot (3) on the Broadbalk Winter Wheat Experiment, which runs up to the edge of the Wilderness. However, historic measurements across the plots of the Broadbalk Winter Wheat Experiment (see the map in Poulton *et al.*, 2003) indicate that there is little variability in subsoil C across the plots of the Broadbalk Experiment: the mean organic C content of the 23–46 cm layer for the 1881 sampling of plots 3, 5, 8, 10, 14 and 16 was  $0.57 \pm 0.011\%$ ; the corresponding figure for the 46–69 cm layer was  $0.45 \pm 0.003\%$ .

The present Geescroft Wilderness was part of an experimental field growing field beans between 1847 and 1878; the area that is now Geescroft Wilderness was marked as cultivated land on the 1623 map. Some of our Geescroft samples came from areas that had received P between 1847 and 1878, some from areas that had not. The starting Geescroft samples were taken in 1883 from two plots (3 and 4) of the beans experiment that had never received P.

The old grassland samples came from four subplots on the Park Grass Continuous Hay Experiment; they are referred to in this paper as from 'Park Grass'. Sir John Lawes stated that the field known as Park Grass had been under pasture for at least 150 years before he and Sir Henry Gilbert started the Continuous Hay Experiment in 1856. However, the field was once under the plough; the characteristic ridge-and-furrow marks of medieval ploughing are still faintly visible. The herbage on the Park Grass Experiment is cut and removed twice a year (see Poulton, 2006).

The four subplots we used were: unfertilized unlimed (subplot 3d); unfertilized limed (subplot 3b); fertilized unlimed (14/2d) and fertilized limed (14/2c). The fertilized areas receive N (96 kg N ha<sup>-1</sup> as NaNO<sub>3</sub>), P, K and Mg annually. Lime is added at intervals to subplot 3b to maintain topsoil pH at about 6 (Johnston, 1972). However, because no lime has yet been added to subplot 14/2c, it and 14/2d can be regarded as replicates.

## Soils

The soil on the old cultivated site and on both of the sites that have been allowed to revert to woodland is mapped as Batcombe Series. That part of the old grassland site from which our samples came is mapped as Hook Series, which differs from Batcombe only in that the depth to clay-with-flints is greater than 80 cm. The Batcombe Series soil is a moderately well-drained silty clay loam overlying clay-with-flints, which in turn overlies chalk at a depth of several metres (Avery & Catt, 1995). It is classified as a Stagnogleyic Paleo-Argillic Brown Earth by the Soil Survey of England and Wales; the USDA classification is an Aquic Paleudalf. Particle-size analysis shows that clay increases with depth in the Batcombe Series. The 0-23 cm layer of Geescroft Wilderness contained 21% clay, the 23-46 cm layer 43%, the 46-69 cm layer 55% and the 69–92 cm layer 63%; the corresponding figures for Broadbalk Wilderness are 23, 30, 50 and 49% (Jenkinson, 1971, a reference that gives the compete particle-size analysis for soil profiles from each Wilderness). In Part 2 (this issue) of this pair of papers, the Broadbalk Arable profile and the Park Grass profile are taken to have the same clay contents as Broadbalk Wilderness.

#### Soil sampling

Sampling in 1999 was as already described (Poulton *et al.*, 2003). Briefly, the samples were taken by power drill (see Poulton

et al., 2003), using a 34-cm diameter drill for the 0-23 cm layer and a 12.5-cm drill for the 23-46 and 46-69 cm layers. A 2.5cm diameter semi-cylindrical auger was used for the 69-92 cm layer. The soil was sieved and air-dried, to give the mass of 'fine soil' (i.e. dry soil passing a 0.635 cm sieve) and of dry stones that did not pass the 0.635 cm sieve. The mass of 'fine soil' per sampling layer, rather than bulk density, which was unduly influenced by the occasional large flint, was used to convert analytical measurements on a % dry soil basis to a per hectare basis. Living plants and roots remaining on the sieve were excluded, but dried and weighed. This living plant material was excluded from the all the measurements presented in the present paper. The Park Grass soils were treated differently, because of their dense root mat: the surface 5 cm, with its mass of roots, was separated from the remainder of the 0-23 cm layer. After removal of the roots and stones, the 'fine soil' from the 0-5 cm layer was combined with the 'fine soil' from the 5–23 cm layer for analysis.

The pre-bomb samples had been taken by the traditional Rothamsted procedure, using an open ended steel box 15  $\times$ 15 cm by 23 cm in depth (i.e.  $6 \times 6 \times 9$  inches), driven downwards into the soil, layer by layer (Dyer, 1902; Jenkinson, 1971). The soil was dried at 40°C, sieved (0.635 cm) to remove stones and the 'fine soil' then stored at room temperature for up to 130 years in sealed bottles. The mass of 'fine soil' in each of the sampling layers was obtained from the Rothamsted archives. Blake et al. (2000) were unable to detect any change in the C or N contents of soils from Park Grass that had been kept air-dry in bottles for 32 years, making it likely that changes in the total C and N contents of our stored samples were also negligible. All 'fine soil' samples, pre- and postbomb, were ground for 3 minutes in a disc mill (Tema Model T100, Tema Machinery Ltd., Woodford Halse, Northants, UK) before analysis.

Geescroft was the only site with appreciable litter cover in 1999; this was collected, sieved (< 0.635 cm) and weighed from 0.66 m<sup>2</sup> areas at each sampling position, before sampling the mineral soil. Prior to analysis (including radiocarbon dating), a quantity of the Tema-milled litter was returned to the 0–23 cm soil sample, in proportion to the area sampled.

The number of holes sampled at each site is given in Tables 1 and 2.

### Analytical methods

Total C and N were determined by combustion using a LECO CNS Analyser (LECO Corp., St. Joseph, MI, USA). Carbonate C was determined by manometry and organic C taken as the difference between total C and carbonate C. Soil pH was measured in water, using a ratio of 10 g soil to 25 ml water. All measurements (except radiocarbon and  $\delta^{13}$ C, see below) were made individually on samples from each layer of each sampling hole. Measurements are expressed on an oven-dry basis (24 hours at 105°C). If a row in Table 1 or 2 contains the pre-

fixes I- or NPL-, the analytical methods to obtain the data in that row are as described by Jenkinson *et al.* (1992).

# Radiocarbon and <sup>13</sup>C measurements

For reasons of economy, radiocarbon measurements were carried out on combined samples, containing equal masses of the contributing soils. Soils from the sampling holes across a site were allocated alternately to an 'odd' or 'even' sample, which were then analysed separately for radiocarbon. The soils for radiocarbon dating were pre-treated with acid to remove carbonate; for consistency this was done to all soils, whether carbonate was present or not. The soil samples were soaked in 0.1 M HCl for 10 hours, stirred occasionally and the pH measured to ensure that it remained < 3. They were then washed free of mineral acid with distilled water, dried and homogenized. About 3% of the organic C originally present in the soil is lost during decalcification (Jenkinson et al. 1992); the radiocarbon content of the C thus lost was not measured. The total C in a known weight of decalcified soil was recovered as CO<sub>2</sub> by heating with CuO in a sealed quartz tube (Boutton et al., 1983) and the CO<sub>2</sub> then converted to graphite by Fe/Zn reduction (Slota et al., 1987). The graphite samples were analysed for radiocarbon at the University of Arizona NSF-AMS facility (Donahue, 1995); these samples (Tables 1 and 2) are indicated by the prefix AA-. Tables 1 and 2 also contain a few earlier measurements by other radiocarbon dating laboratories, indicated by the appropriate laboratory prefix; some of these have been published before (Jenkinson & Rayner, 1977; Jenkinson et al., 1992). The <sup>13</sup>C isotope ratios were measured on a sub-sample of CO<sub>2</sub>, using a dual inlet mass spectrometer with a multiple ion beam collection facility (VG OPTIMA; Fisons Instruments, Middlewich, UK). The mass spectrometer was calibrated with international reference materials to a precision of  $\pm 0.1$ %. The  $\delta^{13}$ C measurements are ultimately related to the PDB belemnite standard and the radiocarbon data to 0.95 of the <sup>14</sup>C activity of the NBS standard oxalic acid.

The radiocarbon data are presented in two ways: as conventional years Before Present (BP, relative to 1950), unless so much bomb-derived radiocarbon was present that an age could not be calculated, and as % modern absolute. The % modern absolute values were obtained by multiplying the % modern values (as measured) by  $exp(-\lambda v)$ , where  $\lambda$  is the decay constant (based on the true radiocarbon half-life of 5730 years) and v is the number of years between 1950 and the year of measurement. This correction allows for the ongoing radioactive decay of the NBS oxalic acid reference standard since 1950 (Stuiver & Polach, 1977). For samples collected in 1999 and analysed in 2001 (Tables 1 and 2), this factor was 0.9939. It was taken to be negligibly different from 1 for the non-AA- measurements, which were done in the 1960s or early 1970s. The radiocarbon ages in Tables 1 and 2 were calculated from % modern (not % modern absolute) as -8033 ln(% modern/100), using the conventional Libby half-life of 5568 years. To allow for isotopic fractionation, all <sup>14</sup>C measurements are corrected (Stuiver & Polach, 1977) to  $\delta^{13}C_{PDB\%} = -25$ , using the appropriate <sup>13</sup>C values in Tables 1 and 2.

# Calculation of equivalent depths

The mass of 'fine soil' in the 0-23 cm layer of both Geescroft and Broadbalk Wildernesses was markedly less in the 1999 samplings than in the corresponding layers of soil taken before reversion to woodland commenced (Table 1). We therefore calculated 'equivalent depths', in order to allow for changes with time in bulk density, organic matter content, etc. (Dyer, 1902; Jenkinson, 1971; Gifford & Roderick, 2003). For the top layer, equivalent depth is here defined as 'the depth to which the top layer should have been sampled at time t, so that it contained the same mass of organic-matter-free and calcium-carbonatefree 'fine soil' as at  $t_0$ , when the first sample was taken to a depth of 23 cm'. Similar definitions hold for deeper layers. Equivalent depths and mass of organic C in equivalent layers were calculated as in Poulton et al. (2003). In essence, equivalent depth corrections were made by adding or taking away a slice of material with the composition of the layer below, assuming that transitions between layers were continuous and smooth. In making these corrections, we also assume that surface erosion was negligible over the sampling period in all of our (near-level) sites.

The % radiocarbon absolute (r'') in a specified equivalent layer (for example, the 23–46 cm layer) at time *t* was calculated from the expression

$$r'' = [f''x'' + f'''x'''(H'' - D'')/L - f''x''(H' - D')/L]/(c''),$$
(1)

where primes indicate layer; for example, x'' is the measured mass of organic C (in t ha<sup>-1</sup>) in the 23–46 cm layer at time t, x' the corresponding quantity in the 0-23 cm layer and x''' that in the 46-69 cm layer. D'' (in cm) is the measured depth to the bottom of the specified layer (i.e. 46 cm in this case), each layer being of thickness L cm; H'' (also in cm) is the equivalent depth at time t; c" is the calculated mass of organic C in the 23-46 cm equivalent layer at time t and f' is % modern absolute, but adjusted to the value it would have been had the soil been analysed immediately after sampling. This correction (calculated using the true radiocarbon half-life of 5730 years, corresponding to a decay constant of 0.0001210 years<sup>-1</sup>) was necessary because some of the radiocarbon measurements were made more than a century after sampling. To make the correction, the values of % modern absolute, as given in Tables 1 and 2, were divided by exp(-0.000121w), where w is the period, in years, between sampling and analysis. Finally, the values of  $\Delta^{14}C_e$  in Tables 1 and 2 are given by (10r''-1000), where  $\Delta^{14}C_e$ is  $\Delta^{14}$ C corrected to equivalent depth.

# NMR spectroscopy

Two 0–23 cm soils from Park Grass were analysed for coal by solid-state <sup>13</sup>C NMR, one of which we thought (from radiocarbon measurements; see Results and Discussion section) contained coal as contaminant and the other not. The <sup>13</sup>C cross-polarization magic-angle spinning (CPMAS) NMR spectra were measured on a Bruker MSL 300 spectrometer (Bruker Analytik GmbH, Rheinstetten, Germany). The NMR measurements were done by N. Mahieu and E. W.Randall (private communication, 2003).

The following experimental parameters were used: spectrometer frequency 75.5 MHz, contact time 1 ms, relaxation time 1 s, and spinning speed around 4.8 kHz, elimination of spinning side-bands using the TOSS (total suppression of sidebands) sequence (Dixon, 1982) and line broadening 50 Hz.

Three runs were done on the putative coal-free sample; by itself, with 1% of coal picked from the Park Grass site and with 2% coal. A fourth run was done on the coal alone. A fifth run was done on the contaminated sample. Before analysis, the soils had been ground for 3 minutes in a Tema disc mill, with added coal where appropriate. The coal sample was ground likewise. The ground samples were packed into cylindrical zirconia rotors  $(5.6 \times 17.0 \text{ mm internal dimensions})$  sealed with Kel-F caps (3M Company, Minneapolis, MN, USA). We accumulated 601 433 scans (7 days) for the putative coal-free sample alone, 479 148 (5.5 days) for it ground with 1% coal, 346 936 (4 days) for it with 2% coal, 226 591 (2.6 days) for the contaminated sample and 56 998 (0.7 days) for coal by itself.

### **Results and discussion**

#### Non-isotopic soil measurements

Organic C and total N decreased down the profile, in all except one instance (the 69–92 cm layer sampled from plot 3d on Park Grass in 1999; Table 2). Taking all the data in Tables 1 and 2 together, there was a close ( $R^2 = 0.98$ ) linear relationship between percentage N and C in the soils, given by the equation:

$$C = 13.0N - 0.324.$$
(2)

Except for two measurements in the 69–92 cm layer, C/N ratios decreased with depth in all of the profiles sampled. This decrease is caused, at least in part, by the increasing proportion of the soil N held as fixed ammonium with depth. Bremner (1959), working with soils from plot 07 on Broadbalk, a plot next to that used in the present paper (08), showed that the 0–23 cm layer contained 5.6% of its N as fixed NH<sub>4</sub>, rising to 18.4% in the 46–69 cm layer and 22.6% in the 114–137 cm layer. This fixed ammonium was held on clay, not on organic matter.

There was little change in pH over the last century in the Broadbalk soils, nearly all being in the 7–8 range, as would be expected for soils containing free  $CaCO_3$  throughout the experimental period. The  $CaCO_3$  content did, however,

decrease with time, as generally observed in the Rothamsted field experiments (Bolton, 1977). Soil pH fell slightly with time in the unlimed Park Grass unfertilized soils and rose by about a unit in the limed unfertilized soils; it stayed just under 6 in the fertilized unlimed treatments. In contrast, pH fell sharply in Geescroft Wilderness, from 7.1 in 1883 to a mean of 4.4 in 1999 (see Poulton *et al.*, 2003).

#### Radiocarbon measurements

Apart from some surface soils from Park Grass, which will be considered later, radiocarbon age increased down the soil profile (Tables 1 and 2). For soil taken in 1893 from the Broadbalk Winter Wheat Experiment, the radiocarbon age increased from 1192 years in the 0–23 cm layer to 12 090 years in the 206–229 cm layer. Taking all the pre-bomb cultivated soil samples together (i.e. all the samples listed in Table 1, except those collected in 1999), there was a linear relationship ( $R^2 = 0.94$ ) between the radiocarbon age (*x*-axis) and negative depth to the middle of each sampling layer (*y*-axis), given by:

$$y = -0.0162x + 5.28. \tag{3}$$

Radiocarbon 'ages', as given in Tables 1 and 2, must not be taken at their face values, as representing the depositional age of the organic matter in a particular layer of a particular profile. The age of a particular sample is really the age of a homogeneous sample that has the same  ${}^{14}C/{}^{12}C$  ratio as that of the heterogeneous soil sample analysed. Unacceptable errors will be introduced if the ages in Tables 1 or 2 are taken as those of a homogeneous pool of C. This can be seen by considering the 1893 sample of the 0-23 cm layer from Broadbalk Arable, which contained 29.4 t organic C ha<sup>-1</sup>, with a radiocarbon age of 1192 years. To a close approximation, this site is under steady state conditions, with the annual input of organic C equal to the annual output. If we treat the organic C in this soil as a homogeneous pool, of average age 1192 years, then the annual steady-state input of organic C is given by 29.4/1192, or 25 kg C ha<sup>-1</sup> year<sup>-1</sup>. This is a ludicrously small value. The average annual offtake of C in grain and straw from this plot over the period 1990-2005 was 3500 kg C ha<sup>-1</sup>. The return of C in stubble to the soil was about 200 kg C ha<sup>-1</sup> year<sup>-1</sup>, and this figure does not include C returned in roots and chaff. In reality this calculation is misleading because soil organic C contains fractions of vastly different stabilities (see Part 2, this issue).

Taking the radiocarbon data from all the depths and all the sites together, but excluding the Park Grass 0–23 cm soils, there was a hyperbolic relationship (Figure 1) between the radiocarbon content of a soil (*m*, expressed as  $\Delta^{14}C_e$ ) and its organic C content (*n*, in t ha<sup>-1</sup>) given by

$$m = 152.1 - 2341/(1 + 0.264n). \tag{4}$$

Paul et al. (2001) fitted a polynomial to similar data for soils from the US Midwest; their polynomial and our hyperbola



**Figure 1** Plot of the amount of organic C in a soil layer, against  $\Delta^{14}C_e$  for the same layer, for all the layers and all the sites listed in Tables 1 and 2. The line shows the hyperbola m = 152.1 - 2341/(1 + 0.264 n) fitted ( $R^2 = 0.88$ ) to all the data except the Park Grass 0–23 cm soils, where *m* is the radiocarbon content of the soil, expressed as  $\Delta^{14}C_e$ , and *n* its organic C content, in t ha<sup>-1</sup>. The arrow shows how allowing for coal shifts the point for one of the Park Grass surface soils (the 0–23 cm layer sampled in 1906 from the unlimed fertilized treatment, with a coal C content of 0.82%).

give broadly similar radiocarbon predictions for soil C contents of between 10 and 80 t C  $ha^{-1}$ .

It is worth pointing out that the points on the lower left of Figure 1 are from the deepest subsoils, which contain the least C, although this C is of great age. Those at the top right are from surface soils with much C, although this C is relatively young, again except for the Park Grass surface soils. The principal value of an empirical relationship like Equation (4) is that it immediately draws attention to aberrant data like the Park Grass surface soils.

# Measurements of $\delta^{13}C$

Values of  $\delta^{13}$ C increased down the profile, from  $-26.3_{00}^{\prime} \pm 0.14$  (SE) in the 0–23 cm layer (mean of all measurements, excluding one outlier) to  $-25.8_{00}^{\prime} \pm 0.17$  for the 23–46 cm layer,  $-25.5_{00}^{\prime} \pm 0.11$  for the 46–69 cm layer (excluding another outlier) and  $-25.2_{00}^{\prime} \pm 0.09$  for the 69–92 cm layer. Such increases with depth are common (Boutton, 1996; Torn *et al.*, 2002) but not universal (Krull & Skjemstad, 2003). There was a decrease in  $\delta^{13}$ C as organic matter accumulated in the 0–23 cm layer of the Wildernesses, from a starting value in the 1880s of  $-25.3_{00}^{\prime} \pm 0.08$ , decreasing to  $-26.7_{00}^{\prime} \pm 0.16$  in 1999 (means for both Wildernesses, again omitting one outlier). Excluding the same  $\delta^{13}$ C outliers, there was a weak relationship ( $R^2 = 0.46$ ) between  $\delta^{13}$ C and  $\Delta^{14}$ Ce, given by:

$$\delta^{13}C = -0.00303(\Delta^{14}C_e) - 26.43.$$
 (5)

Thus soil organic C gets older with depth and the discrimination against <sup>13</sup>C becomes less. Presumably the influence of recentlyadded plant debris (with strongly negative  $\delta^{13}$ C values) becomes increasingly diluted by older soil organic matter (with less negative  $\delta^{13}$ C values) down the profile.

#### Carbonized material in the soils

Soils often contain carbonized material such as charcoal, coal or lignite (Krull & Skjemstad, 2003; Rumpel *et al.*, 2003). Carbonized C had earlier (Jenkinson *et al.*, 1992) been measured in some of the soils listed in Tables 1 and 2. The 0–23 cm layers of the samples taken from Broadbalk in 1881 and from Geescroft in 1883 each contained 0.4 t ha<sup>-1</sup> of carbonized C, roughly 1% of the total soil organic C. There was no detectable carbonized C in the corresponding 23–46 and 46–69 cm layers from either site (detection limit; 0.1 t ha<sup>-1</sup> carbonized C).

Some of our Park Grass surface soils contain much more carbonized C than the corresponding layers on Geescroft or Broadbalk. Earlier work (Jenkinson et al., 1992) showed that samples of 0-23 cm soil from plot 3 on Park Grass (samples taken at various times, none corresponding to any of the samples in Table 2) contained 4.5–6.0 t  $ha^{-1}$  carbonized C, accounting for some 5% of the total soil organic C. Stones larger than 0.635 cm in diameter were excluded when preparing our samples for analysis and several of the Park Grass plots sampled in 1999 contained discrete fragments of bituminous coal in the larger than 0.635 cm stone fraction. The coalcontaminated plots are on the end of the experiment closest to Rothamsted Manor and it is possible the coal was present in ash from domestic fires, probably spread during the late 18th or early 19th centuries. Several of the Park Grass surface soils deviated considerably from Equation (4). We decided to use <sup>13</sup>C-NMR to see if these deviations were caused by coal contamination, using two Park Grass surface soils. Data from the first (taken in 1999 from the limed section of the unfertilized treatment, containing 3.54% C; see Table 2) fell close to the line on Figure 1, suggesting it contained little if any coal, whereas the other (taken in 1906 from the fertilized unlimed treatment, containing 3.25% C) fell far below this line, suggesting that it contained a considerable amount of coal. From the height of the aromatic band at ca. 127 p.p.m. (after normalising all the runs to give the same ratio (height of carbonyl band at c. 173 p.p.m.)/(% soil organic C)) we calculated that the contaminated sample contained 0.82% coal C. The arrow on Figure 1 shows the data for the putative contaminated soil before and after deducting coal C. This observation, that deducting coal C in this way returned the sample point to just below the line on Figure 1, confirms our view that coal is the reason why some of the Park Grass 0-23 cm samples fell markedly below this line.

A rough estimate of the amount of coal-free organic carbon in the 0–23 cm layer of the Park Grass samples was made as follows. The difference between the value of  $\Delta^{14}$ C as measured for each individual sample and as given by Equation (4) was calculated. The concentration of coal was calculated for each sample of topsoil by relating this difference to that in the contaminated sample containing 0.82% coal. The 0–23 cm layer of all the pre-bomb Park Grass samples contained 69.0 t coalfree organic C ha<sup>-1</sup>: the corresponding mean for all of the 1999 samples was 75.2 t ha<sup>-1</sup> (Table 3: in square brackets).

#### Data aggregation

Table 3 presents a summary of pre- and post-bomb organic C and radiocarbon for these Rothamsted Long-Term Experiments, calculated from the mass of data in Tables 1 and 2. Data from the part of Geescroft Wilderness that once received P have been meaned with the corresponding data from the part that never received P; no differences have yet been observed between the two parts (Jenkinson, 1971; Poulton *et al.*, 2003).

The 'pre-bomb' data in Table 3 are means of measurements on samples taken between 1881 and 1906. This was done to obtain some measure of sampling variation, assuming that all belonged to a single population. The individual measurements in Table 1 suggest that this is not an unreasonable assumption, bearing in mind the paucity of data. However, it should be noted that many of the standard errors for  $\Delta^{14}C_e$  in Table 3 are based on only two replicates and are therefore themselves subject to large error.

The samples taken in 1999 from Geescroft Wilderness, Broadbalk Wilderness and Broadbalk Arable contained more organic C than the corresponding pre-bomb sampling, all the way down the profile (Table 3). However, t tests comparing the C content of each layer sampled in 1999 with the C content of the corresponding pre-bomb layer showed that these differences were seldom significant (P < 0.05, here and subsequently), apart from the surface layers. Carbon gains in the woodland sites are discussed in detail by Poulton et al. (2003). The 1999 samples from Geescroft Wilderness, Broadbalk Wilderness and Broadbalk Arable all contained more radiocarbon than the corresponding pre-bomb sample (with one exception, the 23-46 cm layer from Broadbalk Arable), although, apart from the surface layer, none of the differences reached significance (Table 3). One of the two 0-23 cm samples taken in 1999 from Broadbalk Arable was much older than the other  $(\Delta^{14}C_e)$  $-116.7\%_{00}$ , compared with  $-38.2\%_{00}$ ; Table 1), making the corresponding standard errors in Table 3 very large. The great age of this sample (957 years) is out of line with the 1977 sampling of the same plot (Jenkinson et al., 1992). We have no explanation for this discrepancy.

The 'pre-bomb' data in Table 3 for the old grassland (Park Grass) are means of measurements made in 1870, 1876 and 1906. All the data from the 1999 sampling have also been aggregated, partly because of the problem with coal (see above) and partly because too few samples were taken on the different plots for individual treatment. If the effects of coal are disregarded, there was little change in organic C in the 0-23 cm layer over the 100 or

Table 2 Organic C and D	Ce III oOI		villation poing-1 of	The putting of the	הור מזוח מורה ווור ווו		IC313		
		Organic C in equivalent layer	$\Delta^{14}C_e$ in equivalent layer	Organic C in equivalent layer	$\Delta^{14} C_e in$ equivalent layer	Organic C in equivalent layer	$\Delta^{14} C_e in$ equivalent layer	Organic C in equivalent layer	$\Delta^{14}C_e$ in equivalent layer
	I	$/t ha^{-1}$	/%	$/t ha^{-1}$	/%	$/t ha^{-1}$	/00	$/t ha^{-1}$	/%
	I	0–23	cm	23-4(	5 cm	46-6	9 cm	6-69	2 cm
All pre-bomb arable <sup>a</sup>	Mean	29.0	-141	17.1	-225	14.6	-362	11.7	-498
	SE (n)	1.08 (4)	4.5 (4)	0.45 (4)	17.1 (4)	0.51 (4)	28.3 (4)	0.78 (2)	37.4 (2)
Broadbalk arable (1999)	Mean	33.4 <sup>c</sup>	-77°	19.3°	-229°	15.4	-312	13.9	-425
	SE (n)	1.67 (6)	39.3 (2)	0.55 (6)	32.8 (2)	0.58 (6)	21.5 (2)	0.93 (6)	14.1 (2)
Regenerating woodland,	Mean	63.1	27	23.5	-178	18.6	-265	11.7	-397
Geescroft (1999) <sup>b</sup>	SE (n)	3.15 (8)	8.2 (4)	1.43 (8)	11.6 (4)	0.87 (8)	19.4 (4)	0.68 (8)	33.3 (4)
Regenerating woodland,	Mean	78.0	44	25.6	-153	19.6	-267	17.3	-329
Broadbalk (1999)	SE (n)	4.53 (4)	5.8 (2)	1.18 (4)	27.0 (2)	1.76 (4)	44.3 (2)	1.96 (4)	42.2 (2)
All pre-bomb grassland	Mean	81.6 [69.0] <sup>f</sup>	-201 [-55] <sup>f</sup>	22.5	-171	12.3	-365	9.5	-508
plots <sup>d</sup> (Park Grass)	SE (n)	1.91 (6)	12.9 (6)	1.12 (6)	8.9 (6)	0.27 (6)	15.4 (6)	0.37 (6)	23.8 (6)
All grassland plots <sup>e</sup>	Mean	79.7 [75.2] <sup>f</sup>	-79 [-23] <sup>f</sup>	25.7	-136	16.2	-250	12.8	-397
(Park Grass: 1999)	SE (n)	1.16 (16)	21.0 (6)	1.60 (16)	11.7 (6)	0.66 (16)	11.6 (6)	0.89 (16)	25.9 (6)
<sup>a</sup> Pre-bomb sample data ar- <sup>b</sup> Means of the part of Gee: <sup>c</sup> There may have been som <sup>d</sup> Means of 1870 (plots 3 an	e means c scroft tha e mixing d 14), 18'	of 1881 (Broadbalk), t received P before 1 between these layers 76 (plots 3 and 14) a	1883 (Geescroft), 1: 880 and the part thi (see text). nd 1906 (plots 3UL	893 (Broadbalk) and at did not. and 14UL).	d 1904 (Broadbalk)	samplings.			

**Table 3** Organic C and  $\Lambda^{14}$ C in soils taken from the Bothamsted I ono-Term Experiments before and after the thermonuclear bomb tests

<sup>f</sup>Data in square brackets corrected for coal (see text). <sup>e</sup>Plots 3b, 3d, 14/2c and 14/2d.

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so years in Park Grass. However, if changes in coal content are factored in, there was a small C gain in the 0–23 cm layer over the experimental period. Each of the lower layers contained a little more organic C when sampled in 1999 (Table 3) than in the corresponding layer of the pre-bomb profile, although only a few of the gains in the deeper layers reached significance (P < 0.05). It is possible that soil organic C on Park Grass had not yet reached equilibrium by the end of the 19th century – the site had once been cultivated and was not under continuous grass until 1700 – or earlier. The aggregated 1999 samples from Park Grass contained significantly more radiocarbon than the corresponding pre-bomb samples, all the way down the profile (Table 3).

#### Acknowledgements

We thank N. Mahieu and E. W. Randall for the <sup>13</sup>C-NMR measurements, D. D Harkness for help and advice on the radiocarbon measurements and A. Todd for statistical advice. M. V. Hewitt, G. Denton and S. Francis helped with sampling and analysis. Rothamsted Research receives grant-aided support from the UK Biotechnology and Biological Sciences Research Council; the Lawes Agricultural Trust also provided support. The Radiocarbon Laboratory at East Kilbride is supported by the Natural Environment Research Council.

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				Sampling layer	Mass OD F fine soil	3quivalent depth	ч рН С	03 Orga C C	anic Tot	tal C/N	Organic C equivalen layer	in t Radiocarbon dating	8 <sup>13</sup> C	Radiocarbon	Radiocarbon age e	$\Delta^{14}C_{e}$ in quivalent layer
Field	Treatment and plot code	Sampling year	Holes combined	/cm	/Mkg ha <sup>-1</sup>	/cm		0	%		/t ha <sup>-1</sup>	laboratory code	/%	/% modern absolute	/years BP	/%00
Geescroft	Arable without P (plots 3 & 4)	1883	444	0–23 23–46 46–69	2.73 3.01 3.10	22.9 45.7 68.6	7.1 0. 7.1 0. 7.1 0.	00 1.0 00 0.5 00 0.4	<ul> <li>0.1</li> <li>0.1</li> <li>0.0</li> <li>0.0</li> <li>0.0</li> </ul>	18 8.8 84 6.90 71 6.90	1 28.4 0 17.5 0 15.2	I-3399 NPL-214 NPL-215	-25.1 -27.2 -25.4	85.3 79.2 67.2	1280 1870 3180	-138.2 -199.8 -321.1
Geescroft	Regenerating woodland without P	6661	0000	0–23 <sup>a</sup> 23–46 46–69 69–92	2.17 2.99 3.54 3.54	27.6 49.9 70.0 89.9	4.3 0. 5.0 0. 5.6 0. 5.9 0.	00 2.8 00 0.7 00 0.6	82 0.1 75 0.0 80 0.0 82 0.0	94 14.5 85 8.8( 77 7.8( 47 6.8(	1 65.9 0 21.6 0 18.1 5 10.0	AA-38845 AA-38849 AA-38853 AA-38857	-27.1 -25.5 -26.2 -25.4	103.19 81.44 68.51 51.97	post-bomb 1600 2989 5208	16.8 -208.7 -321.0 -480.3
Geescroft	Regenerating woodland without P	1999	0000	0–23 <sup>a</sup> 23–46 46–69 69–92	2.20 3.12 3.16 3.16	27.2 49.2 71.7 94.0	4.4 0. 4.9 0. 5.7 0. 6.2 0.	00 2.5 00 0.7 00 0.6	54 0.1 77 0.0 52 0.0 87 0.0	75 14.5. 86 8.88 79 7.82 58 6.32	5 60.4 8 22.4 1 18.2 1.4	AA-38846 AA-38850 AA-38854 AA-38858	-26.8 -25.4 -25.5 -25.3	106.86 83.33 75.39 58.07	post-bomb 141 5 2220 431 7	51.6 -177.2 -260.9 -419.6
Geescroft	Regenerating woodland with P	6661	0000	0–23 <sup>a</sup> 23–46 46–69 69–92	2.38 3.06 3.88 3.88	26.1 48.0 66.4 84.5	4.7 0. 5.1 0. 5.6 0. 5.6 0.	00 0.5 00 0.6 00 0.6	75 0.1 06 0.0 50 0.0 39 0.0	92 14.30 90 10.68 74 8.11 50 7.75	0 69.3 8 27.5 1 19.3 5 12.1	AA-38847 AA-38851 AA-38855 AA-38859	-26.8 -25.3 -25.4 -25.4	102.89 83.15 75.74 65.50	post-bomb 1433 2183 3350	17.6 
Geescroft	Regenerating woodland with P	6661	0 0 0 0	0–23 <sup>a</sup> 23–46 46–69 69–92	2.30 3.02 3.63 3.63	26.5 48.7 68.3 87.7	4.4 0. 5.3 0. 5.9 0. 6.1 0.	00 2.3 00 0.7 00 0.6	81 0.1 77 0.0 60 0.0 13 0.0	75 13.2 92 8.3 79 7.5 61 7.1	3         56.8           8         22.4           8         18.7           9         13.5	AA-38848 AA-38852 AA-38856 AA-38860	-26.7 -25.3 -25.4 -25.3	103.43 86.10 75.41 65.95	post-bomb 1153 2218 3296	23.4 
Broadbalk	Arable (plot 3)	1881	Q Q	0–23 23–46 46–69	2.87 <sup>b</sup> 3.04 <sup>b</sup> 3.10 <sup>b</sup>	22.9 45.7 68.6	8.1 0. 7.7 0. 7.4 0.	52 0.5 01 0.5 00 0.4	0.1 0.1 59 0.0 46 0.0	04 8.8 78 7.56 66 6.9	5 26.4 5 17.9 7 14.3	I-4296+NPL149 NPL-154 NPL-153	-25.4 -26.0 -22.2	83.80 78.40 63.30	1418 1950 3670	-153.1 -207.7 -360.3
Broadbalk	Regenerating woodland	1999	0000	0–23 23–46 46–69 69–92	2.18 1.96 3.16 3.16	31.2 58.9 81.4 103.8	7.8 0. 8.0 0. 7.9 0. 7.5 0.	31 2.5 14 0.5 02 0.7 01 0.5	7 0.2 94 0.0 70 0.0 51 0.0	64 11.2 <sup>4</sup> 99 9.55 78 8.96 63 7.99	4 71.5 8 24.6 8 18.4 0 15.7	AA-41096 AA-41098 AA-41100 AA-41102	-26.1 -25.4 -26.4 -25.2	104.99 89.36 75.14 62.83	post-bomb 854 2247 3683	35.6 
Broadbalk	Regenerating woodland	1999	0000	0–23 23–46 46–69 69–92	1.92 2.50 3.04 3.04	31.8 57.4 80.9 104.2	7.6 0. 8.0 0. 7.9 0. 7.2 0.	18 3.8 24 1.0 06 0.7 00 0.6	<ul> <li>89 0.3</li> <li>90 0.1</li> <li>73 0.0</li> <li>51 0.0</li> </ul>	23 12.0 02 9.7 81 8.95 71 8.50	4         84.5           7         26.6           2         20.7           5         18.9	AA-41097 AA-41099 AA-41101 AA-41103	-30.9 -25.3 -25.3 -25.1	107.03 90.45 83.08 71.26	post-bomb 757 1440 2672	51.6 
Broadbalk	Arable (plot 8)	1893	6 % N 4 4 4 %	0–23 23–46 46–69 69–92 114–137 206–229	2.97 <sup>b</sup> 2.88 <sup>b</sup> 3.11 <sup>b</sup> 3.12 <sup>b</sup> 3.18 <sup>b</sup> 3.36 <sup>b</sup>	22.9 45.7 68.6 91.4 137.2 228.6	8.0 0. ND <sup>6</sup> 0. ND 0. 7.2 0. 6.3 0.	33     0.5       01     0.5       00     0.4       00     0.4       00     0.4       00     0.2       00     0.2       00     0.2	9     0.1       55     0.0       40     0.0       24     0.0       20     0.0	11     8.92       70     7.86       58     7.47       57     7.05       48     5.00       48     5.00       43     4.65	2 29.4 5 15.8 1 13.4 1 13.4 7.6 6.7	AA-41152 AA-41153 AA-41154 AA-41155 L-4918 L-4919	-25.4 -25.0 -25.3 -25.3 -23.7 -24.1	85.68 71.54 55.01 45.89 29.30 22.20	1192 2642 4752 6208 9861 12090	- 131.9 - 275.2 - 442.6 - 535.1 - 703.3 - 776.0
Broadbalk	Arable (plot 8)	1904	4444	0–23 23–46 46–69 69–92	3.00 <sup>d</sup> 3.17 <sup>d</sup> 3.36 <sup>d</sup> 3.36 <sup>d</sup>	22.5 43.4 64.6 85.8	ND 0. ND 0. ND 0. ND 0. ND 0. ND 0.	26 1.0 01 0.5 00 0.4 00 0.3	06 0.1 58 0.0 48 0.0 55 0.0	17 9.00 77 7.5 65 7.38 55 6.30	5 31.6 5 17.0 8 15.7 5 10.9	AA-41156 AA-41157 AA-41158 AA-41159	-25.2 -24.9 -25.0 -24.8	85.02 76.42 65.28 53.34	1255 2112 3377 4998	-139.0 -216.0 -323.8 -460.3
Broadbalk	Arable (plot 08)	6661	n n n n n n n n n n n n n	0–23 23–46 46–69 69–92	3.11 3.39 3.46 3.49	21.5 41.0 61.7 82.1	7.1 0. 7.3 0. 7.3 0. 7.2 0.	01 1.0 01 0.6 01 0.5 00 0.4	<ul> <li>99</li> <li>0.1</li> <li>52</li> <li>0.0</li> <li>50</li> <li>0.0</li> <li>14</li> <li>0.0</li> </ul>	11 9.85 74 8.39 65 7.64 58 7.56	32.6 18.8 116.0 13.8	AA-38861 AA-38863 AA-38865 AA-38867	-25.8 -25.1 -23.7 -23.1	95.27 72.88 68.22 58.87	340 2491 3023 4207	-38.2 -262.2 -290.5 -411.2
Broadbalk	Arable (plot 08)	1999		0–23 23–46 46–69 69–92	2.95 3.24 3.51 3.49	22.6 43.1 63.4 83.8	7.3 0. 7.5 0. 7.5 0. 7.4 0.	03 1.1 02 0.6 01 0.4 00 0.4	(6 0.1 56 0.0 47 0.0 45 0.0	15 10.13 81 8.14 67 7.03 63 7.20	2 34.0 4 19.8 1 14.8 1 14.1	AA-38862 AA-38864 AA-38866 AA-38868	-25.6 -25.4 -25.2 -25.3	88.23 78.98 64.58 56.06	957 1847 3463 4600	-116.7 -196.6 -333.5 -439.3

doi: 10.1111/j.1365-2389.2008.01025.x

 Table 1
 Sampling and analyses of arable and woodland soils before and after the thermonuclear bomb tests

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<sup>a</sup>Geescroft 1999 0–23 cm layer includes litter. <sup>b</sup>Broadbalk soil masses for 1881 and 1893 from Dyer (1902). <sup>c</sup>Composite samples from plots 5, 6 and 7. <sup>d</sup>Means of 1893 and 1999 measurements. <sup>e</sup>Not determined.

				Sampling layer	Mass OD fine soil	Equivalent depth	D Hq	၀ ဂို ပု	rganic 7 C	Fotal 6 N r	Z/N (atio eq	Drganic C in luivalent layer	Radiocarbon	δ <sup>13</sup> C F	tadiocarbon	Radiocarbon age e	$\Delta^{14}C_e$ in equivalent layer
Field	Treatment and plot code	Sampling year	Holes	/cm	/Mkg ha <sup>-1</sup>	/cm			0%			/t ha <sup>-1</sup>	laboratory code	///	/% modern absolute	/years BP	/00
Park Grass	Unfertilized grassland unlimed (plot 3)	1870		0–23 23–46 46–69 69–92	2.44 3.03 3.20 3.34	22.9 45.7 68.6 91.4		0.0	3.50 ( 0.73 ( 0.338 ( 0.33 (	).282 1 ).078 ).051 ).049	2.40 9.34 7.55 6.81	86.6 22.0 12.3 11.0	AA-41128 AA-41129 AA-41130 AA-41131	-26.5 -25.8 -25.3 -25.1	80.16 77.82 57.73 51.69	1726 1965 4363 5252	-185.6 -209.4 -413.5 -474.8
Park Grass	Unfertilized grassland unlimed (plot 3)	1876	<i>ო ო ო ო</i>	0–23 23–46 46–69 69–92	2.43 2.97 3.41 3.40	22.9 46.2 67.7 90.0	5.3 6.1 ND ND	0.0	3.31 ( 0.70 ( 0.38 ( 0.28 (	).272 1 ).074 ).048 ).042	2.19 9.45 7.87 6.59	80.4 20.9 9.2	AA-41136 AA-41137 AA-41138 AA-41139	-26.4 -25.8 -25.1 -26.0	81.61 84.17 66.26 56.71	1583 1335 3257 4508	-171.4 -147.7 -324.3 -424.3
Park Grass	Unfertilized grassland unlimed (plot 3UL)	1906		0–23 23–46 46–69 69–92	$2.43^{a}$ $3.00^{a}$ $3.31^{a}$ $3.37^{a}$	22.8 45.8 68.0 90.6		0.0	3.12 ( 0.68 ( 0.35 ( 0.27 (	).248 1 ).073 ).044 ).042	2.59 9.31 7.91 6.34	75.8 20.5 11.2 8.9	AA-41144 AA-41145 AA-41146 AA-41147	-26.3 -25.2 -25.3 -25.1	78.01 81.50 65.39 51.86	1946 1594 3363 5226	-210.9 -176.2 -335.9 -475.4
Park Grass	Unfertilized grassland limed (plot 3b)	1999	0000	0–23 23–46 46–69 69–92	2.33 3.48 3.35 3.47	23.6 43.4 65.4 87.3	6.2 6.2 6.2 6.2 6.2	0.0	3.54 ( 0.74 ( 0.43 ( 0.30 (	).297 1 ).074 1 ).051 ).039	1.94 0.00 8.50 7.53	83.4 23.4 14.6 9.9	AA-41104 AA-41110 AA-41116 AA-41122	-27.1 -26.0 -25.8 -25.5	91.40 82.51 77.00 68.14	673 1495 2050 3032	-87.0 -173.7 -220.9 -318.4
Park Grass	Unfertilized grassland limed (plot 3b)	1999	0 0 0 0	0–23 23–46 46–69 69–92	2.28 3.60 3.59 3.47	23.9 43.1 63.4 85.4	6.5 6.4 6.4 6.3	0.0	3.65 ( 0.82 ( 0.46 ( 0.32 (	).306 1 ).080 1 ).056 ).039	1.93 0.19 8.27 8.18	84.6 26.3 16.0 10.7	AA-41105 AA-41111 AA-41117 AA-41117 AA-41123	-27.2 -26.0 -26.0 -25.5	92.76 82.69 74.14 66.76	555 1478 2354 3196	-73.8 -153.2 -246.2 -332.3
Park Grass	Unfertilized grassland unlimed (plot 3d)	1999	0 0 0 0	0–23 23–46 46–69 69–92	2.28 3.43 3.36 3.39	23.8 44.0 65.9 88.4	5.2 5.4 5.4 5.4 6	0.0	3.26 ( 0.90 ( 0.46 ( 0.63 (	).267 1 ).080 1 ).059 ).071	2.21 1.20 7.83 8.90	75.5 28.5 15.0 14.3	AA-41106 AA-41112 AA-41118 AA-41124	-27.2 -28.8 -25.6 -25.4	99.46 86.29 69.35 56.39	6 1135 2890 4552	-6.7 -127.4 -290.2 -435.9
Park Grass	Unfertilized grassland unlimed (plot 3d)	1999	0000	0–23 23–46 46–69 69–92	2.38 3.68 3.42 3.39	23.2 41.8 63.3 85.7	5.1 5.3 5.4 5.2 6.2	0.0	3.36 ( 0.85 ( 0.52 ( 0.45 (	).275 1 ).082 1 ).060 ).055	2.22 0.43 8.64 8.22	80.3 27.9 17.3 15.1	AA-41107 AA-41113 AA-41119 AA-41125	-27.2 -26.2 -25.6 -25.2	96.30 87.01 69.61 56.93	253 1068 2861 4477	-38.4 -88.2 -279.6 -430.6
Park Grass	Fertilized grassland unlimed (plot 14)	1870		0–23 23–46 46–69 69–92	2.52 3.05 3.34 3.43	22.9 45.7 68.6 91.4		0.0	3.45 ( 0.92 ( 0.38 ( 0.28 (	).282 1 ).088 1 ).049 ).045	2.26 0.34 7.71 6.14	87.1 27.9 12.7 9.5	AA-41132 AA-41133 AA-41134 AA-41135	-26.1 -25.8 -25.3 -25.3	78.66 82.81 63.84 44.03	1879 1465 3555 6541	-200.8 -158.7 -351.4 -552.7
Park Grass	Fertilized grassland unlimed (plot 14)	1876	$\omega$ $\omega$ $\omega$ $\omega$	0–23 23–46 46–69 69–92	2.54 3.31 3.45 3.52	22.6 43.6 65.9 88.1	5.9 6.3 ND ND	0.0	3.05 ( 0.69 ( 0.36 ( 0.25 (	).271 1 ).080 ).051 ).040	1.27 8.61 7.00 6.15	77.3 22.0 12.4 8.4	AA-41140 AA-41141 AA-41142 AA-41143	-26.4 -25.7 -25.2 -23.6	80.85 81.83 62.16 44.78	1658 1561 3771 6404	-179.2 -159.2 -354.5 -545.4
Park Grass	Fertilized grassland unlimed (plot 14UL)	1906		0–23 23–46 46–69 69–92	$2.53^{a}$ 3.18 <sup>a</sup> 3.40 <sup>a</sup> 3.47 <sup>a</sup>	22.8 44.6 67.2 89.7		0.0	3.25 ( 0.69 ( 0.39 ( 0.29 (	).272 1 ).076 ).057 ).048	1.97 9.07 6.79 5.96	82.2 21.4 13.2 9.8	AA-41148 AA-41149 AA-41150 AA-41151	-26.0 -25.9 -25.6 -25.1	73.30 80.63 57.70 42.01	2446 1680 4367 6917	-258.7 -177.6 -408.9 -575.0
Park Grass	Fertilized grassland unlimed (plots 14/2c & 14/2d)	1999	4 4 4 4	0–23 23–46 46–69 69–92	2.85 3.13 3.57 3.65	20.3 42.9 64.5 85.9	5.9 6.1 5.9 5.8	0.0	2.76 ( 0.76 ( 0.49 ( 0.37 (	).243 1 ).077 ).056 ).048	1.36 9.80 8.77 7.62	76.0 24.1 17.3 12.6	AA-41108 AA-41114 AA-41120 AA-41126	-26.4 -26.3 -26.2 -25.6	85.44 85.49 73.92 51.93	1214 1209 2377 5215	-145.4 -134.3 -236.8 -480.2
Park Grass	Fertilized grassland unlimed (plots 14/2c & 14/2d)	1999	4444	0–23 23–46 46–69 69–92	2.72 3.38 3.72 3.65	21.5 42.4 62.9 84.3	5.8 6.2 6.0 5.9	0.0 0.0 0.0	2.91 ( 0.74 ( 0.49 ( 0.41 (	).254 1 ).078 ).055 ).052	1.44 9.61 8.85 7.81	78.2 24.1 17.1 14.0	AA-41109 AA-41115 AA-41121 AA-41127	-26.5 -26.1 -25.8 -25.6	87.77 85.20 74.59 61.52	999 1236 2305 3853	-122.5 -140.6 -226.7 -384.5
<sup>a</sup> Means of <sup>b</sup> Not deter:	1870 and 1876 measu mined.	trements.															

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