

## THE TEMPORAL DISTRIBUTION OF 'BOMB' $^{14}\text{C}$ IN A FOREST SOIL

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**ABSTRACT.** Patterns of  $^{14}\text{C}$  enrichment in the superficial plant debris and mineral soil horizons of an established woodland have been monitored at regular intervals during the past 15 years. These data are compared with a model evaluation of carbon turnover based on the recorded changes in atmospheric  $^{14}\text{C}$  concentration since AD 1900.

Leaf litter and decomposing plant debris are characterized by steady-state turnover values of ca 2 and ca 8 years, respectively. A two-component system of 'fast' ( $\leq 20$  yr) and 'slow' (ca 350 yr) cycling carbon is indicated for the surface (0–5cm) soil humus; below 10cm, the 'fast' component is rare ( $< 5\%$ ).

Selective microbial humification of leaf litter, branch, and root debris is proposed to explain a delay of several years in the peak transfer of 'bomb'  $^{14}\text{C}$  to the soil carbon pool.

### INTRODUCTION

As was first recognized by Jenkinson (1963), the transient enrichment of soil carbon due to nuclear weapon's testing offers unique opportunities in pedological study. In particular, the relatively rapid changes in  $^{14}\text{C}$  concentration induced in the surface horizons can afford a hitherto unobtainable precision in quantitative assessments of the flux of organic carbon through this biologically-crucial region of the soil profile. Detailed studies involving the use of 'bomb'  $^{14}\text{C}$  as a tracer for the input and turnover of organic carbon have been reported for various soil-plant systems (eg, Rafters & Stout, 1970; Martel & Paul, 1974; Jenkinson & Rayner, 1977; O'Brien & Stout, 1978; Ladyman & Harkness, 1980; Stout & Goh, 1980; O'Brien, 1984).

In this paper we discuss, in context of dynamic modeling, data collected over the past decade from a natural woodland in northwest England. The work described forms part of an on-going program of measurements at this and other sites, with the ultimate aim of developing appropriate models that can be used to 1) characterize carbon cycling in natural soil systems, and 2) predict the likely long-term impact of management on the physical characteristics and nutrient status of forest and agricultural soils.

### EXPERIMENTAL METHODS AND RESULTS

#### *Site Description*

Meathop Wood (Natl Grid Ref SD 435 795) is a *Quercus-Fraxinus-Betula* type woodland with a ground flora dominated by *Rubus fruticosus*, *Scilla non-scripta*, and *Mercurialis perennis*. The soil is a brown earth (mull humus) derived mainly from drift comprising Silurian non-calcareous sedimentary rocks with subordinate Carboniferous limestone overlying Carboniferous limestone. A variable non-compacted layer of leaf litter (Ol) and

decaying leaf, branch and other plant material (Of) at most 3cm deep overlies the mineral soil surface. The pH of the soil within the site ranges between 4.6 to 7.0.

### Sampling

Since the initiation of this study in 1972, the overlying litter (O1 and Of) layers and the soil humus to a depth of 15cm have been monitored routinely. Bulk soil samples were also available from collections in 1961, 1969, and 1970. As the work developed, it became clear that a knowledge of the temporal distribution of  $^{14}\text{C}$  in other parts of the ecosystem was necessary to clarify the pathways for carbon transfer. Consequently, sampling and  $^{14}\text{C}$  analyses have been extended at various times to include 1) soil humus to 35cm depth, 2) undecomposed plant debris within the soil horizons, 3) branch litter, and 4)  $\text{CO}_2$  in air at various heights within and above the tree canopy.

Atmospheric  $\text{CO}_2$  was collected by the ca 4-week exposure of fresh prepared 8M KOH solution saturated with  $\text{Ba}^{2+}$  (Harkness, 1970). Samples of the superficial plant debris [leaf litter (Ol), the fermentation layer (Of) and branch fragments] were taken directly from the ground surface. Soil horizons were sampled, in 5cm or 10cm depth increments relative to the mineral surface, from the profile exposed in a series of freshly dug pits.

### Analytical Procedures

Bulk density values for the soil profile were obtained from a comprehensive but independent site survey and agree, within the limits of site variability, with provisional estimates made during the current sampling program. Corresponding carbon contents (wt %) were determined by volumetric measurement of the  $\text{CO}_2$  recovered from quantitative high-pressure combustion of 150 to 300g aliquots of screened soil (see below).

Prior to  $^{14}\text{C}$  measurement, all organic materials were digested in 0.5M HCl at 80°C followed by washing to neutral pH. For soil samples, stones and undecomposed plant fragments were removed by sieving the acid-washed slurry through a 1mm mesh screen before drying to constant weight in a vacuum oven. It is important to note, therefore, that in the following discussion, 'soil humus' is the remaining organic matter, defined solely by this physical screening procedure.

TABLE 1  
Distribution of humus carbon in the Meathop soil profile

Horizon depth (cm)	Carbon content (wt %)	Bulk soil density ( $\text{g cm}^{-3}$ )	Mean wt carbon ( $\text{g cm}^{-2}$ )
0 to 5	6.4	0.782	$5.00 \times 10^{-2}$
5 to 10	2.3	0.906	$2.08 \times 10^{-2}$
10 to 15	1.5	0.906	$1.36 \times 10^{-2}$
15 to 25	1.1	0.906	$1.00 \times 10^{-2}$
25 to 35	0.8	0.906	$0.72 \times 10^{-2}$

TABLE 2  
Measured  $^{14}\text{C}$  enrichments (% modern) for leaf litter and mineral soil horizons

Colln date	Leaf litter		Soil humus (Depth horizon)			Soil debris† (0–5cm)
	OI	Of	(0 to 5cm)	(5–10cm)	(10–15cm)	
1961	—	—	97.0	—	—	—
Summer '69	—	—	106.0	—	97.0	—
Autumn '69	—	—	109.5	—	96.0	—
Spring '70	—	—	109.5	—	97.0	—
Autumn '72	154.5	146.0	110.0	—	—	—
Autumn '73	150.0	145.5	116.0	101.0	95.0	—
Autumn '75	146.5	145.0	122.0	101.5	—	—
*Autumn '77	141.0	141.5	131.0	101.5	96.0	—
Autumn '82	—	—	126.5	—	108.5	—
Autumn '83	—	—	115.5	—	—	125.5
Winter '83	—	—	115.5	—	—	128.0
**Spring '84	126.0	134.5	117.0	109.0	105.0	126.5

\* 'Soil humus' at 15–25cm depth = 95.0; at 25–35cm depth = 93.0

\*\* 'Soil debris' at 5 to 10cm depth = 113.5; at 10–15cm depth = 110.0

† 'Soil debris' defined as all organic material (rootlets plus other decomposed plant remains) retained on wet sieving through 1mm mesh

$^{14}\text{C}$  measurement was via the liquid scintillation method (Harkness & Wilson, 1972) and  $^{14}\text{C}$  concentrations calculated as % modern after normalization of the measured radiometric enrichment of each sample relative to  $\delta^{13}\text{C}_{\text{PDB}} = -25\text{‰}$ .

### Results

Mean values for the bulk density and humus carbon content of the mineral soil horizons are given in Table 1. Over the period monitored, the carbon content, as measured for the individual soil horizons, has remained constant within  $\pm 3\%$  of the values quoted.

Measured  $^{14}\text{C}$  enrichment values are in Tables 2 and 3. These data are expressed at a limiting precision of  $\pm 1.0\%$  ( $1\sigma$  analytical confidence) as determined by an initial (Fall 1973) series of replicate analyses for soil samples taken at four random points within the site.

$\text{CO}_2$  samples collected within and over the woodland canopy during September 1984 showed no significant variation in  $^{14}\text{C}$  concentration relative to a mean recorded value of  $125.6 \pm 0.3\%$  modern.

TABLE 3  
Measured  $^{14}\text{C}$  enrichments for branch debris decomposing on the soil surface in April 1984

Branch diameter	% modern $\pm 1\sigma$
0.2 to 0.5cm	126.2 $\pm$ 0.8
0.5 to 1.0cm	131.7 $\pm$ 0.8
1.0 to 2.0cm	152.2 $\pm$ 1.0
2.0 to 5.0cm	117.9 $\pm$ 0.7

## INTERPRETATION AND DISCUSSION

As a first stage in interpretation, the temporal trends in  $^{14}\text{C}$  activity anticipated for different rates of carbon turnover were computed to allow a direct comparison with the data measured from the soil horizons of the Meathop ecosystem. A fundamental assumption in this approach is that the woodland exists in steady state, *ie*, there is a fixed and constant balance in the accretion, transport, and respiratory loss of organic carbon. This premise is consistent with the woodlands' known history for over 300 years (Satchell, 1983, 1984) and supported by the constant carbon content monitored for the soil horizons since 1961.

*Model Construction*

Modeling the transient isotopic enrichment due to 'bomb'  $^{14}\text{C}$  depends primarily on the photosynthetic fixation of contemporary atmospheric  $\text{CO}_2$  as the input function. Initial measurements of  $^{14}\text{CO}_2$  concentrations at different heights (0 to 10m) within the canopy of Meathop wood are not significantly different from that in the atmosphere above the canopy, in contrast to the situation in a barley field where a significant proportion of photosynthetically-fixed  $\text{CO}_2$  is derived from soil respiration (Monteith, Szeicz & Yabuki, 1964). However, two causes of  $^{14}\text{C}$  depletion may also be reflected in the soil carbon inventory *viz*, 1) the pre-1950 dilution of atmospheric  $^{14}\text{CO}_2$  by fossil fuel emissions (Suess effect), 2) the radioactive decay of  $^{14}\text{C}$ .

In the absence of anthropogenic effects, *ie*, before AD 1900, the  $^{14}\text{C}$  activity of the carbon in a uniformly mixed reservoir at year 't' can be expressed as

$$A_t = A_0 e^{-\lambda T} \quad (1)$$

where  $A_0$  is the natural  $^{14}\text{C}$  concentration in the atmosphere, T the mean age or turnover time ( $\tau$ ) of the carbon reservoir, and  $\lambda$  the  $^{14}\text{C}$  decay constant ( $1.245 \times 10^{-4} \text{ yr}^{-1}$ ). For the present century, a steady-state reservoir response to the progressive annual change induced in the atmospheric  $^{14}\text{C}$  concentration is described by the relationship

$$A_t = \alpha A_{(t-1)} + (1 - \alpha) A_i - A_{(t-1)} \lambda \quad (2)$$

where  $A_{(t-1)}$  is the reservoir  $^{14}\text{C}$  activity established for the previous year;  $A_i$  the (input)  $^{14}\text{C}$  activity prevalent in atmospheric  $\text{CO}_2$  during the preceding growth season; and  $(1 - \alpha)$  represents that fraction of the total carbon pool exchanged each year. For steady-state mass transfer the coefficient ' $\alpha$ ' is defined solely by the particular residence half-time ' $\tau_{1/2}$ ' for exchangeable carbon *viz*,  $\alpha = e^{-\ln 2 / \tau_{1/2}}$ . It can be expressed therefore in terms of a mean residence time ' $\tau$ ' and/or the corresponding exchange rate constant ' $k$ ', *ie*,  $\ln 2 / \tau_{1/2} = 1 / \tau = k$ . Thus, equation (2) can be written in terms of carbon turnover rate as

$$A_t = A_{(t-1)} e^{-k} + (1 - e^{-k}) A_i - A_{(t-1)} \lambda \quad (3)$$

Using ' $A_i$ ' values from the documented changes in atmospheric  $^{14}\text{C}$  concentration at UK latitudes since AD 1900 (Baxter & Walton, 1970;

Ergin, Harkness & Walton, 1970; Nydal, Lovseth & Skogseth, 1980) time-series curves of 'A<sub>t</sub>' were generated for selected values of 'τ' (Fig 1).

*The 'Post-Bomb' Distribution of <sup>14</sup>C*

In Figure 1 the distribution of <sup>14</sup>C enrichment values measured for the past 25 years is compared with the theoretical curves calculated for specific rates of carbon turnover. It is apparent that the superficial leaf litter contained in the O<sub>l</sub> and O<sub>f</sub> layers is readily characterized by turnover times of ca 2 and ca 8 years, respectively. In contrast, the measured temporal variations of <sup>14</sup>C enrichment in the soil humus show marked deviations from the model predictions. Significant features of these deviations are, 1) the onset of a sharp increase in <sup>14</sup>C concentration several years after the peak atmo-

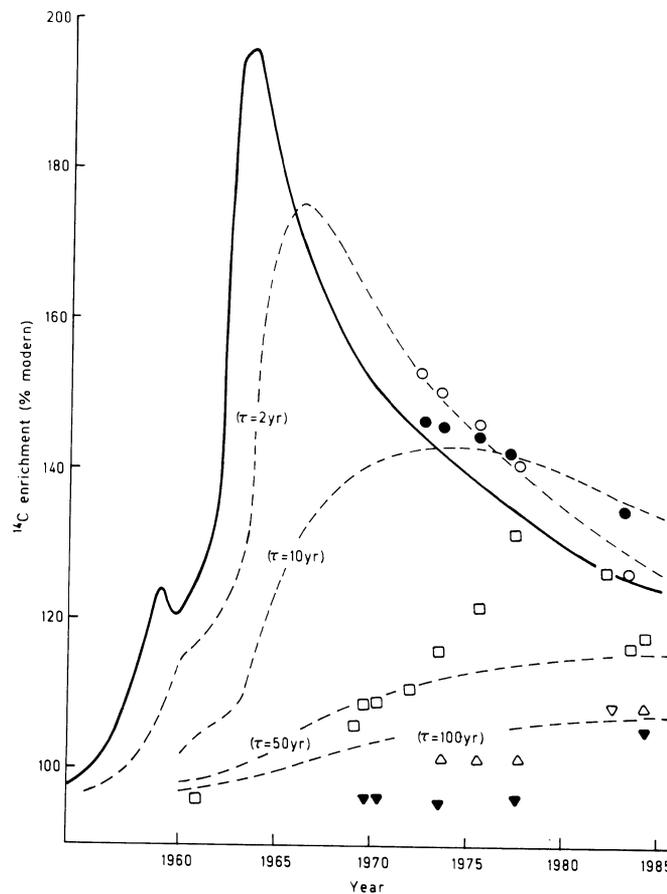


Fig 1. Comparison of measured and modeled <sup>14</sup>C enrichment values. — = free atmosphere; --- = model evaluation for specific turnover times (τ); ○ = leaf litter (O<sub>l</sub>); ● = fermentation layer (O<sub>f</sub>); □ = 0 to 5cm soil humus; Δ = 5 to 10cm soil humus; ▼ = 10 to 15cm soil humus.

spheric values of 1963/64, 2) in the 0 to 5cm horizon, an equally sharp decline from a maximum value attained in ca 1980. A similar, but less pronounced and delayed, maximum turning value may be indicated for the deeper (5 to 10cm and 10 to 15cm) soil horizons, but this possibility must remain speculative pending further  $^{14}\text{C}$  measurements over the next few years. The  $^{14}\text{C}$  concentration in the 0 to 5cm soil has now fallen well below the levels recorded for the post-1960 atmosphere and this can only be due to the retention of a significant amount of 'pre-bomb' carbon. Therefore, contrary to the basic model construction the organic humus in this and, by implication, the underlying soil horizons cannot be regarded as a uniformly mixed carbon reservoir.

This conclusion was also reached by O'Brien & Stout (1978) in the investigation of organic matter turnover in a New Zealand pasture soil. In addition to a relatively fresh organic input labeled with 'bomb  $^{14}\text{C}$ ', the

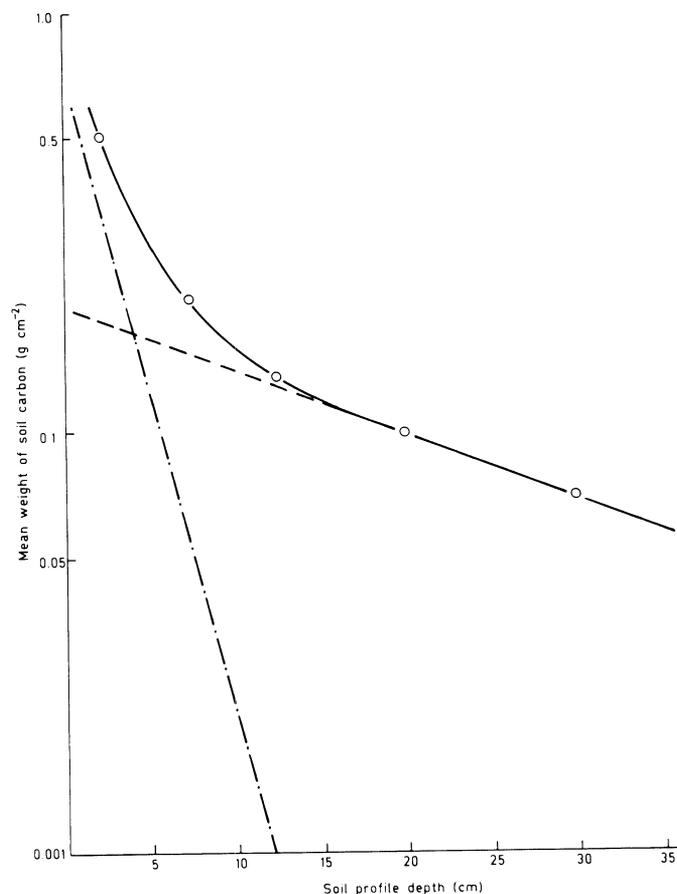


Fig 2. Two-component differentiation of soil humus. —○— = total carbon; —·— = 'fast' component; --- = 'slow' component.

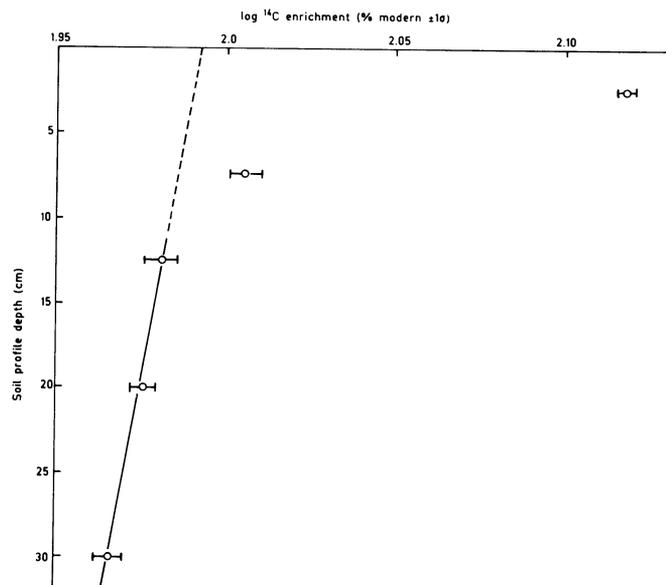


Fig 3. Vertical distribution of <sup>14</sup>C in soil humus at Fall 1977. ◯—◯ = measured data; --- = 'pre-bomb' extrapolation.

presence of 'modern' (<100 yr) and 'old' (≥5700 yr) organic fractions was identified for the uppermost 94cm of the soil profile. The relative abundance and ages of these organic components were resolved by ignoring the evident contribution from 'bomb <sup>14</sup>C'.

We have employed a similar approach in an attempt to improve the model description of the Meathop soil humus in terms of a natural two-component system.

#### The 'Pre-Bomb' Distribution of <sup>14</sup>C

It was assumed that, due to microbial decomposition and eventual mineralization to CO<sub>2</sub>, the concentration of each organic component decreases exponentially with increasing depth in the soil; but at different rates. A graphical differentiation of the concentration of the total humus carbon to 35cm depth (Table 1) was used to assess the relative distribution of 'old' and 'young' carbon in the profile (Fig 2). The corresponding distribution of 'pre-bomb' <sup>14</sup>C (Fig 3) was determined by extrapolation of the 10cm to 35cm age/depth gradient as measured for Fall 1977. At that time, there was no evidence of 'bomb <sup>14</sup>C' having penetrated below 10cm depth (see Fig 1, Table 1).

Since the natural (pre-bomb) <sup>14</sup>C enrichment in the total soil humus ( $\Delta_T\%$ ) compounds the conventional age values of the two-component system, then for a defined depth increment

$$\Delta_T = R_f \Delta_f + R_s \Delta_s \quad (4)$$

where 1)  $R_f$  and  $R_s$  are, respectively, the relative proportions of 'fast' and 'slow' cycling carbon, 2)  $\Delta_f$  and  $\Delta_s$  the corresponding natural  $^{14}\text{C}$  enrichment values as determined by radioactive decay, 3)  $\Delta_{\text{‰}} = [(\% \text{ Modern}/10^2) - 1]10^3$ . In the Meathop soil the 'fast' cycling carbon is seen to have a turnover time of <20 years. Therefore, in the natural situation  $\Delta_f \sim 0\text{‰}$  and

$$\Delta_s = \Delta_f/R_s. \quad (5)$$

This analysis (Table 4) points to a relatively constant turnover rate of ca 350 years for the 'older' organic fraction in the top 15cm of the soil profile and with a gradual increase in age for the underlying horizons. The ca 470-year age obtained for the 0 to 5cm depth increment is considered anomalous; it probably reflects a slight distortion in the graphical analysis due to the influence of fossil-fuel-derived carbon on the  $^{14}\text{C}$  enrichment recorded in the 10 to 15cm horizon in 1977.

#### *Implications for Model Refinement*

Partitioning of the total soil humus contained in the upper 15cm of the soil profile into discrete 'fast' and 'slow' cycling fractions results in much closer model approximation of the distribution of  $^{14}\text{C}$  activities measured since 1961. However, relative to the trend in atmospheric  $^{14}\text{C}$  concentration, the time-delay and peak amplitude as recorded by the 0 to 5cm soil horizon (Fig 1) highlight the need for a quantitative recognition of the biological processes leading to the formation of soil humus.

The 'fast' cycling component, which comprises ca 63% of the soil humus in the top 5cm of the soil, appears to require a transfer mechanism which allows a step-wise delay of several years in the availability of photosynthetically fixed carbon followed by its rapid decomposition and assimilation into the humus carbon pool. In the case of the underlying 10cm of soil a significantly longer delay period is required for the peak transfer of 'bomb  $^{14}\text{C}$ '. Contrary to expectation, a predominant input of carbon via leaf litter and/or root extrudates seems unlikely since this pathway would result in a more immediate and over-damped response to the 'bomb  $^{14}\text{C}$ ' tracer. Roots and other partly-decomposed plant fragments retained in the soil at 1984 obviously comprise a mixture of 'pre' and 'post-bomb' carbon

TABLE 4  
Distribution and age as calculated for the 'slow cycling' carbon  
in Meathop soil humus

Soil horizon depth (cm)	Relative abundance ( $R_s$ )	Natural $^{14}\text{C}$ enrichment		Conventional age (yr BP)
		( $\Delta_f\text{‰}$ )	( $\Delta_s\text{‰}$ )	
0 to 5	0.37	-21	-57	470
5 to 10	0.75	-30	-40	330
10 to 15	0.95	-40	-42	350
15 to 25	0.98	-53	-54	450
25 to 35	1.00	-71	-71	590

(Table 2). Likewise, the decomposing branch litter (Table 3), fragments of which may be incorporated into the soil debris by faunal mixing and/or eluviation. Either input source could account for the initial delay period indicated in the transfer of 'bomb  $^{14}\text{C}$ ' to the soil humus. It is also significant that, even after 20 years or so of sharply falling  $^{14}\text{C}$  concentrations in the living biomass, the plant debris retained in the soil exhibits a marked decrease in  $^{14}\text{C}$  concentration with depth (Table 2). This isotopic pattern is incompatible with a progressive input to the soil of plant material that is uniformly susceptible to decomposition. It can only result from an effective differentiation between 'decomposable' and 'resistant' organic fractions in the overall degradation process.

Consequently, it would seem that for the Meathop soil profile the selective microbial decomposition of root and branch debris must be considered as a major factor in determining the inventory and composition of humus carbon.

#### CONCLUSIONS

The temporal distribution of 'bomb'  $^{14}\text{C}$  in the Meathop soil profile highlights several features that must be recognized in a realistic assessment of soil carbon dynamics. In general, these stem from and relate to the physical and biological complexity of the organic matter contained within the ecosystem.

Foremost in any model evaluation is the need to resolve the 'pre' and 'post-bomb' input of  $^{14}\text{C}$  in defining appropriate carbon reservoirs. Ideally, the  $^{14}\text{C}$  enrichment in these compartments should be quantified and compared relative to the total inventory of organic carbon (humus plus plant fragments) contained within the soil profile.

The direct input of atmospheric carbon to the soil humus via photosynthesis cannot be assumed to be a common characteristic; an initial delay of several years may be appropriate for temperate woodlands. This factor is particularly important where estimates of the transfer and turnover rates for organic carbon is restricted to  $^{14}\text{C}$  data measured over a short period or in a single year as proposed by O'Brien (1984).

The predominant pathways in the overall mass transfer of carbon must also be recognized. For example, the available data for Meathop indicate clearly that much of the organic debris deposited on and within this woodland soil is decomposed and respired without ever becoming incorporated into the soil humus.

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