

Quantifying Ecosystem-Atmosphere Carbon Exchange with a ^{14}C Label

The role of terrestrial ecosystems as sources or sinks for carbon to the atmosphere and their contribution to inter-annual variations in atmospheric CO_2 remain hotly-debated topics. Carbon enters terrestrial ecosystems through a single process, photosynthesis, but it is returned to the atmosphere by the combined metabolic activity of plants, animals, and microbes (Figure 1). The largest uncertainties in our understanding of terrestrial carbon cycling are in these return processes, especially how CO_2 losses from ecosystems are divided among respiration by living plants—termed autotrophic respiration—versus microbial and faunal decomposition of plant residues—termed heterotrophic respiration (Figure 1); and how seasonal and climatic factors that change plant physiological status and soil conditions influence that partitioning.

In order to study ecosystem-atmosphere carbon exchange in a temperate forest, we studied a large release of radiocarbon (^{14}C) that occurred near the Oak Ridge Reservation (ORR), Oak Ridge National Laboratory, Tennessee, in July and August of 1999. This regional ^{14}C release was incorporated into plants as photosynthetic products, and the fate of these ^{14}C -labeled materials is being traced over time. Initial results demonstrate the utility of the ^{14}C label for increasing our understanding of how plants allocate carbon among metabolic respiration, growth, and storage, and what fraction of CO_2 respired from soils comes from autotrophic and heterotrophic sources.

Radiocarbon as a Tracer for the Carbon Cycle

Radiocarbon (^{14}C) is a useful tool for studying the dynamics of carbon exchange between ecosystems and the atmosphere on several time scales. Radiocarbon is naturally produced by the interaction of high-energy cosmic particles with the upper atmosphere. The ^{14}C that is formed quickly oxidizes to CO_2 and enters the Earth's carbon cycle. The residence time of carbon in reservoirs that exchange with the atmosphere on century-to-millennial time scales is determined from the degree to which its ^{14}C has been decreased below atmospheric $^{14}\text{CO}_2$ values by radioactive decay (half-life = 5730 years).

Radiocarbon can also be used to estimate carbon exchange rates on decadal time scales. Atmospheric thermonuclear weapons testing

in the 1950s and 1960s roughly doubled the amount of ^{14}C in atmospheric CO_2 in the Northern Hemisphere prior to the implementation of the Limited Test Ban Treaty in 1963. The rate of incorporation of this "bomb" ^{14}C provides a measure of the rate of carbon exchange among atmosphere, ocean, and terrestrial carbon reservoirs on time scales of years to centuries.

While bomb ^{14}C has been used successfully to study carbon cycling on decadal and longer time scales in ecosystems [Trumbore, 2000], it is of limited use on shorter time scales critical to understanding plant allocation and respiration processes. Radiocarbon in atmospheric CO_2 peaked at about +900‰ in the Northern Hemisphere in 1963, and it has

decreased since then due to exchange with ocean and terrestrial carbon reservoirs (Figure 2). During the 1960s, when rates of atmospheric change were most rapid, only a few laboratories were measuring radiocarbon routinely, and observations relevant to short-term carbon cycling in ecosystems were sparse. By the year 2000, atmospheric $\Delta^{14}\text{CO}_2$ values had fallen to about +80‰, and they continue to decline at rates of 4–8‰/year [Levin and Hessheimer, 2000]. Measurement precision for radiocarbon in our laboratory is $\pm 5\%$ for samples with bomb ^{14}C . Hence, present investigations of short-term carbon cycling are limited to studying carbon exchange on time scales greater than ~2 years.

Much of our understanding of short-term carbon dynamics in plants and soils comes from deliberate additions of ^{14}C tracers [e.g., Coleman and Fry, 1991]. However, environmental regulations on the release of radioactivity and logistical considerations have, for the most part, limited these studies to small-stature vegetation in plots or enclosures. The release

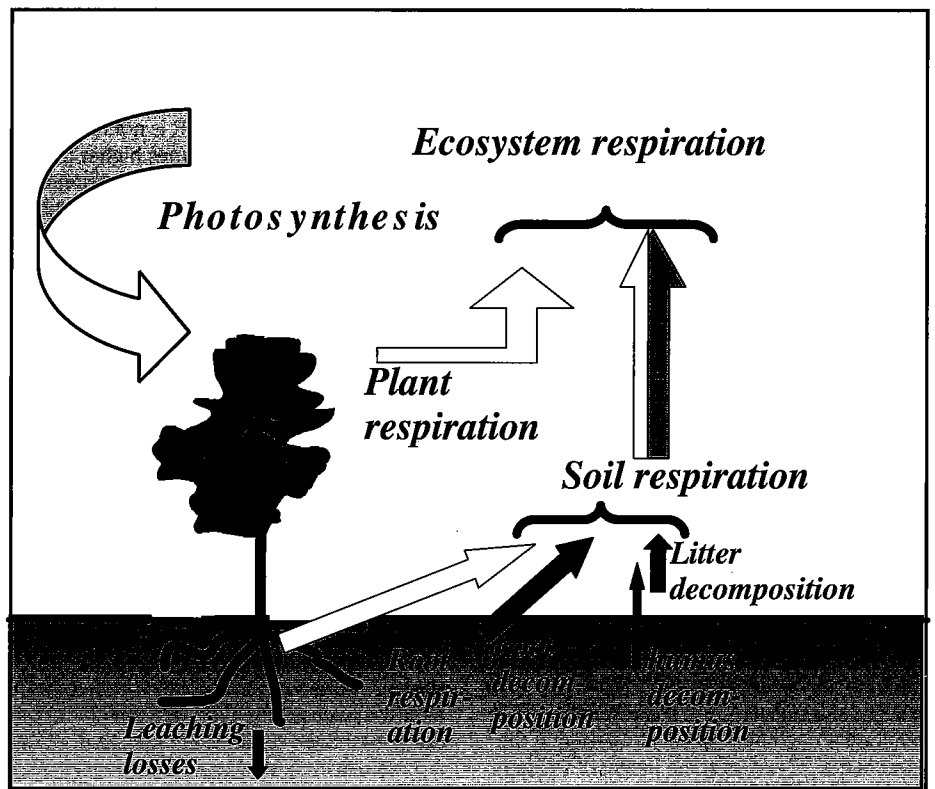


Fig. 1. This diagram shows the flow of carbon through terrestrial ecosystems. The length of time carbon resides in an ecosystem varies depending on how the products of photosynthesis are allocated; for example, some carbon is used to support basal plant metabolism (leaf, stem, and root respiration) and returns to the atmosphere within days to months, some is allocated to growing wood that may last for centuries, and some may be altered by microbes and stabilized in soil minerals for thousands of years.

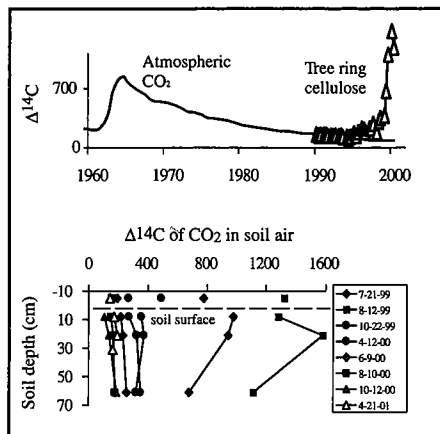


Fig. 2. (top) Radiocarbon in the background atmosphere and cellulose is shown isolated from tree rings in the western Oak Ridge Reservation (black star in Figure 4). (bottom) Radiocarbon in soil air (depth is the y-axis) and surface soil respiration is shown from one site in the central Oak Ridge Reservation (white star in Figure 4). Radiocarbon data are expressed as $\Delta^{14}\text{C}$, the deviation in parts per thousand of $^{14}\text{C}/^{12}\text{C}$ ratio in the sample from that of the primary oxalic acid (I) standard, with the standard corrected for radioactive decay of ^{14}C since 1950. All sample $^{14}\text{C}/^{12}\text{C}$ ratios are corrected to a common $\delta^{13}\text{C}$ of -25‰ to eliminate effects of mass-dependent isotope fractionation on the $\Delta^{14}\text{C}$ value. In $\Delta^{14}\text{C}$ notation, positive numbers indicate the influence of elevated radiocarbon (from either weapons testing or a local ^{14}C release), while negative numbers indicate that the sample has a lower $^{14}\text{C}/^{12}\text{C}$ ratio than 1890 wood due to radioactive decay.

of radiocarbon at the ORR provides a unique opportunity to study shorter-term carbon cycling at the scale of a whole ecosystem by monitoring the ^{14}C label as it moves through various carbon pools.

The Radiocarbon Release in 1999

During routine sampling at two field sites close to the center of the Oak Ridge Reserve (ORR; Figure 3) in July and August 1999, elevated levels of radiocarbon were observed in air, soil CO_2 , and CO_2 emitted from the soil surface (Figure 2, bottom). The ^{14}C concentration measured in CO_2 in boundary layer air on 22 July 1999, was $+435\text{‰}$, nearly half that of the peak bomb ^{14}C values in 1963. By August 1999, $\Delta^{14}\text{CO}_2$ values observed in soil respiration had reached a maximum of $+2000\text{‰}$ —twice the level of the ^{14}C excess at the height of weapons testing in 1963. These observations can only be explained by a local release of radiocarbon to the atmosphere that entered plant carbon pools through photosynthesis, was translocated to plant roots, and subsequently respired again through root metabolism, or decomposition of roots or root exudates.

Subsequent measurement of ^{14}C in the cellulose from tree rings formed between 1950 and 1999 (Figure 2, top) confirmed that a growing season release of ^{14}C in 1999 occurred and was

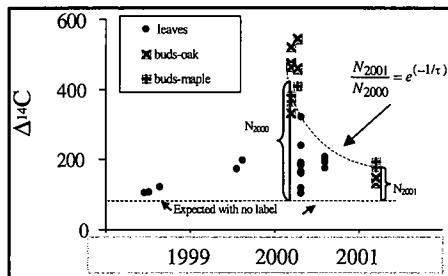


Fig. 3. Samples of radiocarbon in leaves, leaf buds, and parasitic plants taken at the central ORR site (white star on Figure 4) from 1998–2001 are shown. The rapid drop in $\Delta^{14}\text{C}$ values in the spring of 2000 occurs when buds grow into full leaves. The carbon in the buds comes from over-winter stores, while the leaves start to produce their own carbon through photosynthesis.

unique in its magnitude, although the tree ring record began to deviate from background troposphere $^{14}\text{CO}_2$ values as early as 1996. Highest amounts of ^{14}C label were observed in trees on the western portion of the Reserve, near two hazardous waste incinerators that are presumed to be the source of the emitted radiocarbon. Monitoring of ^{14}C levels in CO_2 in air, initiated in the fall of 2000, and sampling of annual plants grown in subsequent years, demonstrate that no subsequent growing season ^{14}C release of similar magnitude has occurred.

The amount of excess radioactivity present in the vegetation on the ORR is not hazardous; at most, it was only a few times higher than what is normally present in the atmosphere. However, the size of the release is sufficient to provide us with an unprecedented opportunity to trace carbon cycling within the ecosystem on brief time scales of less than 3 years.

Allocation of the Label

Previous studies with deliberate ^{14}C labeling [Hanson et al., 2000] have shown that $>90\%$ of the added ^{14}C may be respired within a few days, with $<10\%$ allocated to longer-lived carbon pools such as leaves, roots, and wood. The incorporation of the 1999 ^{14}C release provides a measure of carbon allocation and storage times in these plant components. High values of ^{14}C measured in 1999 tree ring cellulose (Figure 2, top) and in roots known to have grown between April and August 1999 (not shown) mean that carbon fixed immediately after the release was allocated directly to production of these tissues. In contrast, whole-leaf $\Delta^{14}\text{C}$ values increased to a lesser degree over the same period (Figure 3). Leaf buds that grew in early spring 2000 had higher levels of ^{14}C (Figure 3), indicating that the label was incorporated into non-structural carbohydrate pools of carbon that were stored over the winter of 1999, and then used to grow leaves in 2000. As buds grew into full leaves, the $\Delta^{14}\text{C}$ values dropped as fresh photosynthetic product without the ^{14}C label was allocated to growth.

Sampling of leaves in the summer of 2000 showed that the amount of label incorporated

was highest in the western portion of the ORR, closer to the presumed source (Figure 4). Differences were seen between leaves of oak (Q: *Quercus* spp.) and maple (A: *Acer* spp.), with oak leaves having consistently higher $\Delta^{14}\text{C}$ values than maple.

The rate of dilution of the ^{14}C label can be used to estimate the residence time of carbon in tree storage pools used to fuel new leaf growth. Assuming the drop in the amount of ^{14}C label from 2000 to 2001 was due to dilution with unlabeled photosynthetic products, and assuming storage pools are homogeneous, we estimate the residence time (τ) of carbon by solving the relation $N_{2001} = N_{2000} e^{(-1/\tau)}$, where N_{2000} and N_{2001} are the amounts of label in leaf buds (Figure 3) in the buds in years 2000 and 2001. We estimate τ to be ~ 6 yr (5.4–6.8 yr) for maple, and ~ 4 yr (3.5–5.2 yr) for oak. Using the drop in $\Delta^{14}\text{C}$ from 2000 to 2001 in squaw root (*Conopholis Americana*), a parasitic plant thought to derive its carbon directly from oak roots, a residence time is similarly obtained for non-structural carbohydrate pools in oak of ~ 5 years (Figure 3). Few measurements of the residence time of carbon in non-structural carbon pools exist for comparison with our estimates.

Determining Sources of Soil CO_2 Emissions

We took advantage of the large differences in $\Delta^{14}\text{C}$ between carbon fixed in late summer 1999 and in subsequent years to test an isotope mass balance approach for quantifying the contribution of autotrophic versus heterotrophic components of CO_2 emitted from the soil surface. Using a chamber to isolate air in contact with the soil surface, we measured the radiocarbon content of CO_2 emitted from soils ($\Delta^{14}\text{C}_{\text{Total}}$; Gaudinski et al. [2000]). We determined the radiocarbon signature of root-metabolized carbon (root respiration; $\Delta^{14}\text{C}_{\text{Root}}$) by isolating freshly sampled roots in a container and sampling the CO_2 evolved in the hour after their removal from the soil. The ^{14}C content of CO_2 derived from decomposition ($\Delta^{14}\text{C}_{\text{Decomp}}$) was determined by incubating soil and litter samples in jars for 1 week.

The relative contributions of heterotrophic and autotrophic sources to total soil respiration are derived from the isotope mass balance: $\Delta^{14}\text{C}_{\text{Total}} = F * (\Delta^{14}\text{C}_{\text{Root}}) + (1-F) * (\Delta^{14}\text{C}_{\text{Decomp}})$, where F is the fraction of total respiration derived from root metabolism. For example, in August 2000, samples taken in the central ORR had values of $+150\text{‰}$ ($\Delta^{14}\text{C}_{\text{Total}}$), $+84\text{‰}$ ($\Delta^{14}\text{C}_{\text{Root}}$), and $+185\text{‰}$ ($\Delta^{14}\text{C}_{\text{Decomp}}$), indicating that 35% of the carbon respired by the soil is derived from root respiration ($F = 0.35$). In the western ORR, which received higher amounts of label, the equivalent values were $+370\text{‰}$ ($\Delta^{14}\text{C}_{\text{Total}}$), $+84\text{‰}$ ($\Delta^{14}\text{C}_{\text{Root}}$), and $+580\text{‰}$ ($\Delta^{14}\text{C}_{\text{Decomp}}$), indicating a similar value of 42% of total soil respiration derived from root metabolism. Sites exposed to very different amounts of the label thus yield very similar results.

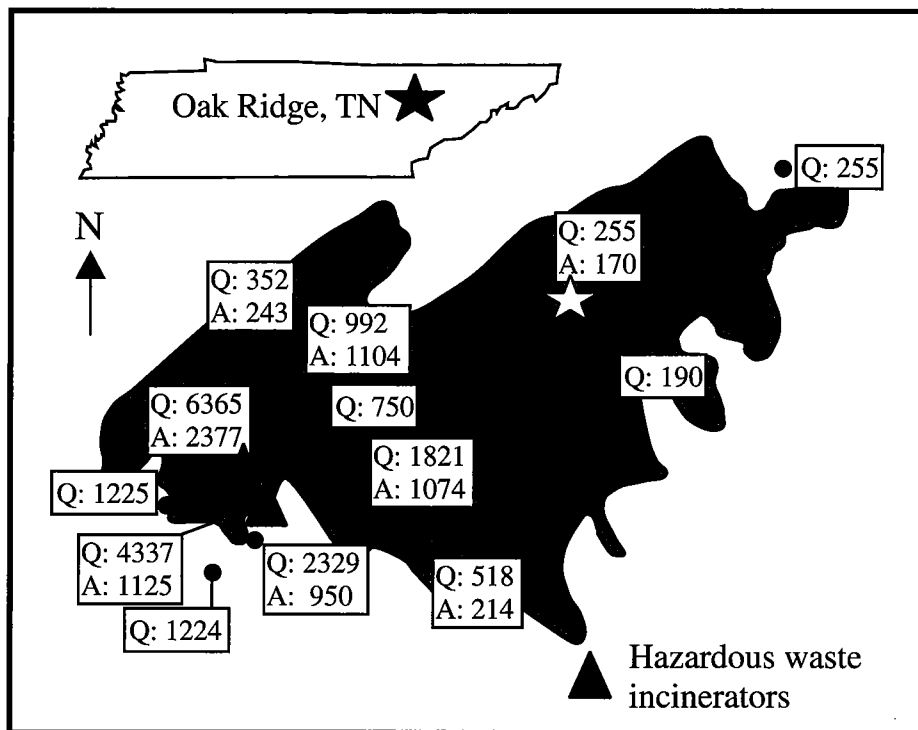


Fig. 4. This map of the Oak Ridge Reservation (an ~11,000-ha area) provides values of $\Delta^{14}\text{C}$ measured in leaves during the summer of 2000. Q is *Quercus* spp. (oak leaves); A is *Acer* spp. (maple leaves). The two triangles mark the position of hazardous waste incinerators thought to be potential sources of the ^{14}C label. The stars show sites where data reported here were collected (white: central Oak Ridge Reservation; black: western Oak Ridge Reservation). Inset: Tennessee is shown, with location of the Oak Ridge Reserve marked.

Future Studies

Our results to date demonstrate the utility of the radiocarbon label for tracing short-term carbon cycling in ecosystems. With new funding from the U.S. Department of Energy, we plan to manipulate the inputs of ^{14}C to further study the dynamics of carbon cycling in the ORR.

Over 2.5 hectares of leaf litter that fell in the autumn of 2000 were collected at sites in the western and central portion of the ORR. At four sites on the ORR, including two soil types and two levels of ^{14}C exposure in 1999, permanent plots were replicated for the manipulation of forest litter through reciprocal transplants of highly enriched ($\Delta^{14}\text{C} = +1000\text{‰}$) and less enriched ($\Delta^{14}\text{C} = +250\text{‰}$) leaf litter among sites. The manipulation plots will produce all combinations of high- and low- ^{14}C -labeled roots with high- and low- ^{14}C -labeled leaf litter.

This experimental design will allow us to examine several key processes in below-ground carbon cycling. Using the mass balance method described above, we will track the changes in the sources of soil CO_2 emissions over several seasons and years in the manipulated plots.

The tracking of ^{14}C respired from the different manipulations will further allow us to separate contributions of leaf litter and root litter decomposition to heterotrophic respiration.

As part of annual sampling of these plots, the radiocarbon label through fine roots and leaf litter will be traced to study the dynamics and fate of carbon—how fast they decompose, and what fraction of their carbon is respired as CO_2 versus that incorporated in microbial biomass or soil organic matter. Measurements of ^{14}C in soil solution will elucidate the role of leaf and root litter as sources of dissolved organic carbon and its role in vertical transport of organic matter in soil profiles.

Discovery of the radiocarbon label in the ORR was serendipitous and unlikely to be repeated in other environments. However, its overall utility leads us to reconsider the use of radiocarbon labeling in natural environments. The use of accelerator mass spectrometry (AMS) increases the sensitivity for detection of ^{14}C by a factor of nearly 10,000 over the decay counting methods used in past pulse labeling experiments. Local labeling experiments may now be feasible for studying the response

of plants and microbes in their environment without the addition of a large amount of radioactive tracer to the environment.

Acknowledgments

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