A Whole-Ecosystem Carbon-14 Label in a Temperate Forest

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The role of terrestrial ecosystems as present sources or sinks of carbon to the atmosphere, and their contribution to interannual variations in atmospheric CO_2, remain hotly debated topics. Carbon enters terrestrial ecosystems through a single process, photosynthesis, but is returned to the atmosphere by many different pathways (Figure 1). The largest uncertainties in our understanding of terrestrial C cycling are in the return processes – respiration and decomposition.

A large release of radiocarbon (^{14}C) 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 we are tracing these ^{14}C-labeled materials to their ultimate fate over time. Our initial results

demonstrate the utility of the ^{14}C label for increasing our understanding of how plants allocate carbon among respiration, growth and storage, and what fraction of CO_2 respired from soils comes from respiration versus decomposition.

Radiocarbon as a tracer for the carbon cycle

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

Radiocarbon can also be used to estimate C exchange rates on decadal timescales. Atmospheric thermonuclear weapons testing in the 1950's to 1960's 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 between atmosphere, ocean, and terrestrial carbon reservoirs on timescales of years to centuries.

While bomb ^{14}C has been used successfully to study carbon cycling on decadal and longer timescales in ecosystems, it is of limited use on shorter timescales critical to understanding plant allocation and respiration processes. Radiocarbon in atmospheric CO_2 peaked at about +900 ‰ in the northern hemisphere in 1963, and 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 $\Delta^{14}CO_2$ values had fallen to about +80‰, and they continue to decline at rates of 4-8‰/year (Levin, 2000). Measurement precision for radiocarbon in our laboratory is ±5‰ for samples with bomb ^{14}C. Hence, present investigations of short-term C cycling are limited to studying carbon exchange on timescales greater than ~2 years.

Much of our understanding of short-term (<1 year) C 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 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, we observed elevated levels of radiocarbon in air, soil CO_2, and CO_2 emitted from the soil surface (Figure 2b). The ^{14}C concentration measured in CO_2 in boundary layer air on July 22, 1999, was +435‰, nearly half that of the peak bomb ^{14}C values in 1963. The maximum $\Delta^{14}CO_2$ values we observed in soil respiration reached +2000‰ (twice the level of the ^{14}C excess at the height of weapons testing in 1963) by August 1999. These observations can only be explained by a local

release of radiocarbon to the atmosphere that entered plant C pools through photosynthesis, was translocated to plant roots, and subsequently respired again through root metabolism, and/or decomposition of roots or root exudates.

Subsequent measurement of ^{14}C in the cellulose from tree rings formed between 1950 and 1999 (Fig. 2a) confirmed that a growing season release of ^{14}C in 1999 occurred and was unique in its magnitude, although the tree ring record began to deviate from background troposphere $^{14}CO_2$ 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 we presume are the source of the emitted radiocarbon. Monitoring of ^{14}C levels in carbon dioxide 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.

It should be stressed that the amount of excess radioactivity present in the vegetation on the ORR is not hazardous (at the maximum 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 C cycling within the ecosystem on short (< 3 year) time scales.

Allocation of the label

Previous studies with deliberate ^{14}C labeling (summarized in 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 C allocation and storage times in these

plant components. High values of ^{14}C measured in 1999 tree ring cellulose (Figure 2a) and in roots known to have grown between April and August, 1999 (not shown) mean that C fixed immediately after the release was allocated directly to production of these tissues. In contrast, whole leaf $\Delta^{14}C$ 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 label was incorporated into nonstructural carbohydrate pools of C that were stored over the winter of 1999, then used to grow leaves in 2000. As buds grew into full leaves, the $\Delta^{14}C$ 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 $\Delta^{14}C$ values than maple.

The rate of dilution of the ^{14}C label can be used to estimate the residence time of C 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 $\Delta^{14}C$ from 2000 to 2001 in squaw root (*Conopholis Americana*), a parasitic plant thought to derive its carbon directly from oak roots, we similarly obtain a residence time for nonstructural

carbohydrate pools in oak of ~5 years (Figure 3). Few measurements of the residence time of C in nonstructural carbon pools exist for comparison with our estimates.

Determining Sources of Soil CO_2 emissions

Emission of CO_2 from soils is the largest terrestrial flux of carbon, and variability in soil CO_2 emission rates dominates year-to-year variation in net ecosystem carbon exchange. However, fundamental questions remain about the factors that influence soil CO_2 fluxes. A key factor is determining what fraction of soil respiration comes from autotrophic (below-ground plant metabolism) versus heterotrophic (decomposition of litter residues and humus) sources (Figure 1), and how seasonal and climatic factors that change plant physiological status and soil conditions influence that partitioning (reviewed in Hanson et al., 2000).

We took advantage of the large differences in $\Delta^{14}C$ between C 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 to soil CO_2 emissions. Using a chamber to isolate air in contact with the soil surface, we measured the radiocarbon content of CO_2 emitted from soils ($\Delta^{14}C_{Total}$; Gaudinski et al., 2000). We determined the radiocarbon signature of root metabolized C (root respiration; $\Delta^{14}C_{Root}$) 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 ($\Delta^{14}C_{Decomp}$) was determined by incubating soil and litter samples in jars for a period of one week.

The relative contributions of heterotrophic and autotrophic sources to total soil respiration are derived from the isotope mass balance: $\Delta^{14}C_{Total} = F*(\Delta^{14}C_{Root}) + (1-F)*(\Delta^{14}C_{Decomp})$, 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‰ ($\Delta^{14}C_{Total}$), +84‰ ($\Delta^{14}C_{Root}$), and +185‰ ($\Delta^{14}C_{Decomp}$), indicating that 35% of the C 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‰ ($\Delta^{14}C_{Total}$), +84‰ ($\Delta^{14}C_{Root}$), and +580‰ ($\Delta^{14}C_{Decomp}$), 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.

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 DOE, we plan to manipulate the inputs of ^{14}C to further study the dynamics of C cycling in the ORR.

We collected large amounts (over 2.5 hectares) of leaf litter that fell in autumn of 2000 at sites in the western and central portion of the ORR (Figure 4, inset). At four sites on the Oak Ridge Reservation, including two soil types and two levels of ^{14}C exposure in 1999, we have established replicated permanent plots (8 per site) for the manipulation of forest litter through reciprocal transplants of highly enriched ($\Delta^{14}C$ = +1000 ‰) and less enriched ($\Delta^{14}C$ = +250‰) 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 belowground 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, we will trace the radiocarbon label through fine roots and leaf litter to study the dynamics and fate of C – how fast they decompose and what fraction of their C is respired as CO_2 versus incorporated in microbial biomass or soil organic matter. Measurements of 14C 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.

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

Acknowledgements

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Figure Captions

Figure 1. Flow of carbon through terrestrial ecosystems. The 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.

Figure 2. (a). Radiocarbon in the background atmosphere and cellulose isolated from tree rings in the western ORR (black star on Figure 4a). (b) Radiocarbon in soil air and surface soil respiration from one site in the central ORR (white star on Figure 4a). Radiocarbon data are expressed as $\Delta^{14}C$, the deviation in parts per thousand of $^{14}C/^{12}C$ 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 $^{14}C/^{12}C$ ratios are corrected to a common $\delta^{13}C$ of –25‰ to eliminate effects of mass dependent isotope fractionation on the $\Delta^{14}C$ value. In $\Delta^{14}C$ 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 $^{14}C/^{12}C$ than 1890 wood, due to radioactive decay.

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

Figure 4. Map of the ORR, with values of $\Delta^{14}C$ measured in leaves during the summer of 2000. Q are *Quercus* spp (Oak leaves); A are *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 ORR; black: western ORR). Insets: The state of Tennessee, with location of the Oak Ridge Reserve marked, and a photograph showing tarps for litterfall collection in the Fall of 2000.

Figure 1.

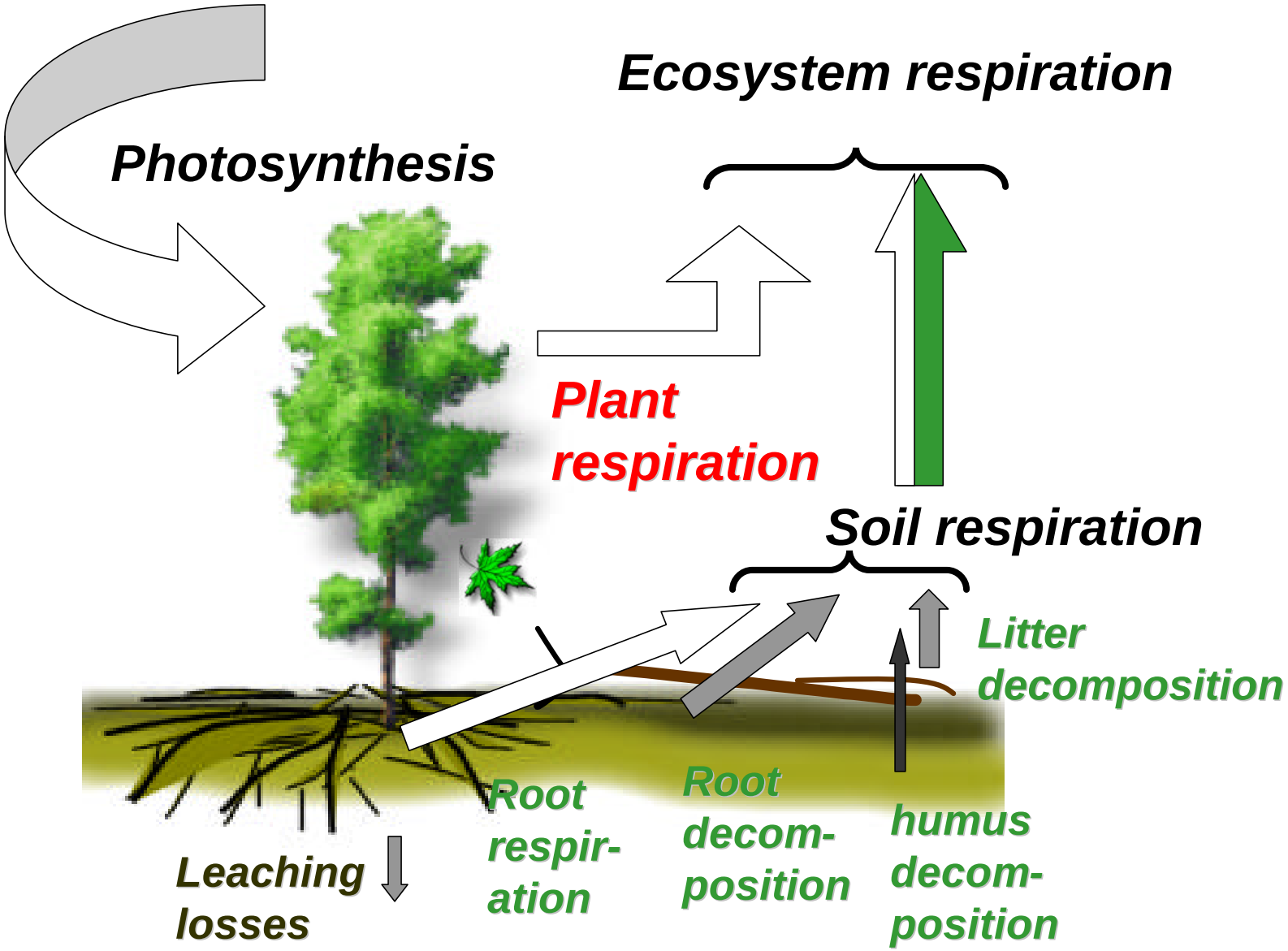

Ecosystem respiration
Photosynthesis
Plant respiration
Soil respiration
Litter decomposition
Root respir-ation
Root decom-position
humus decom-position
Leaching losses

Figure 2

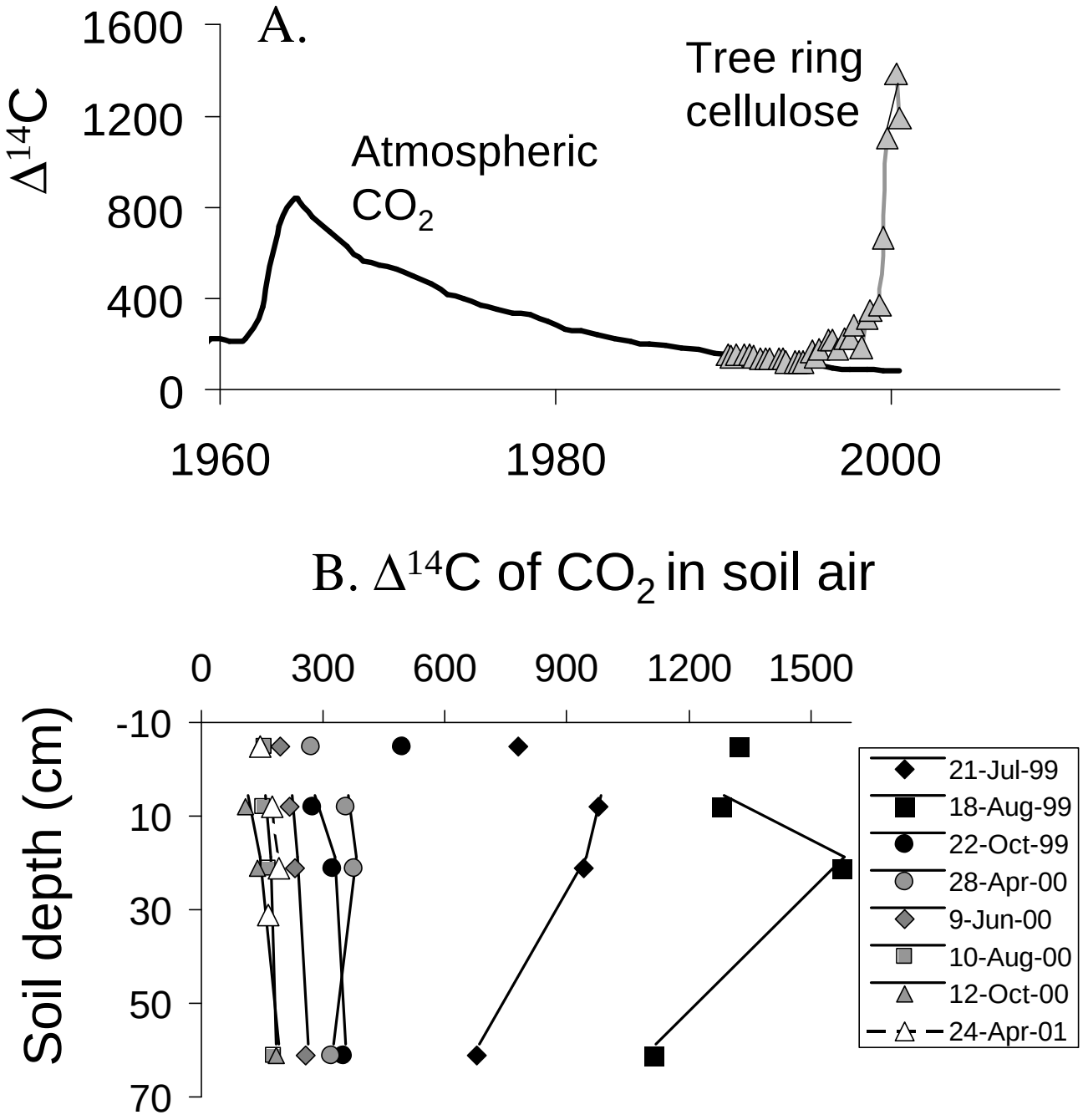
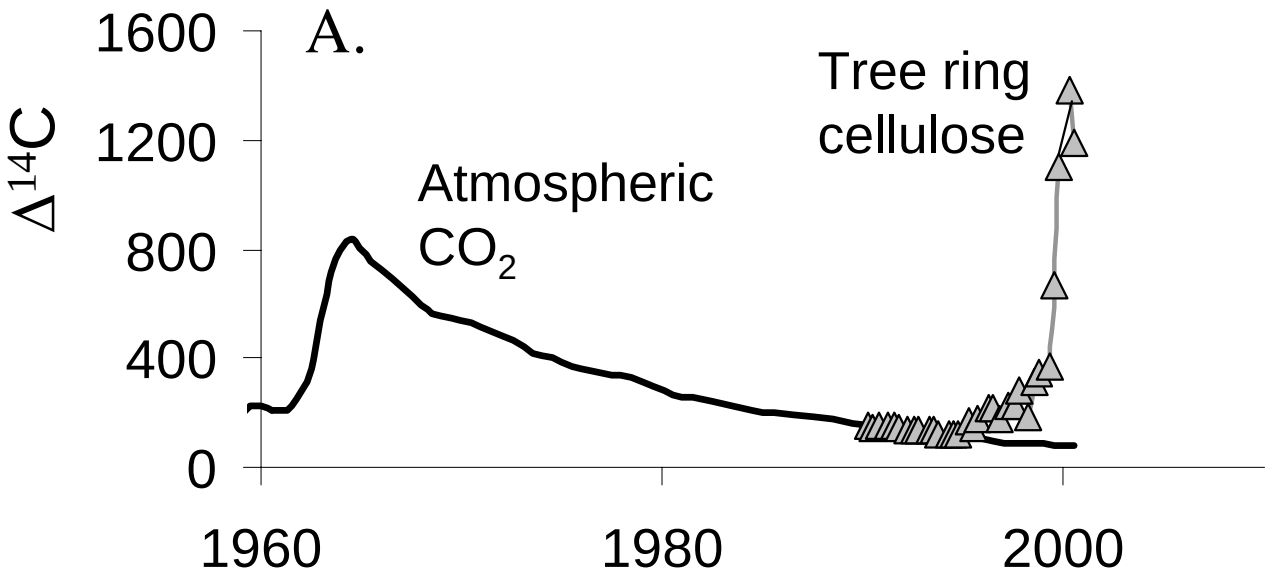
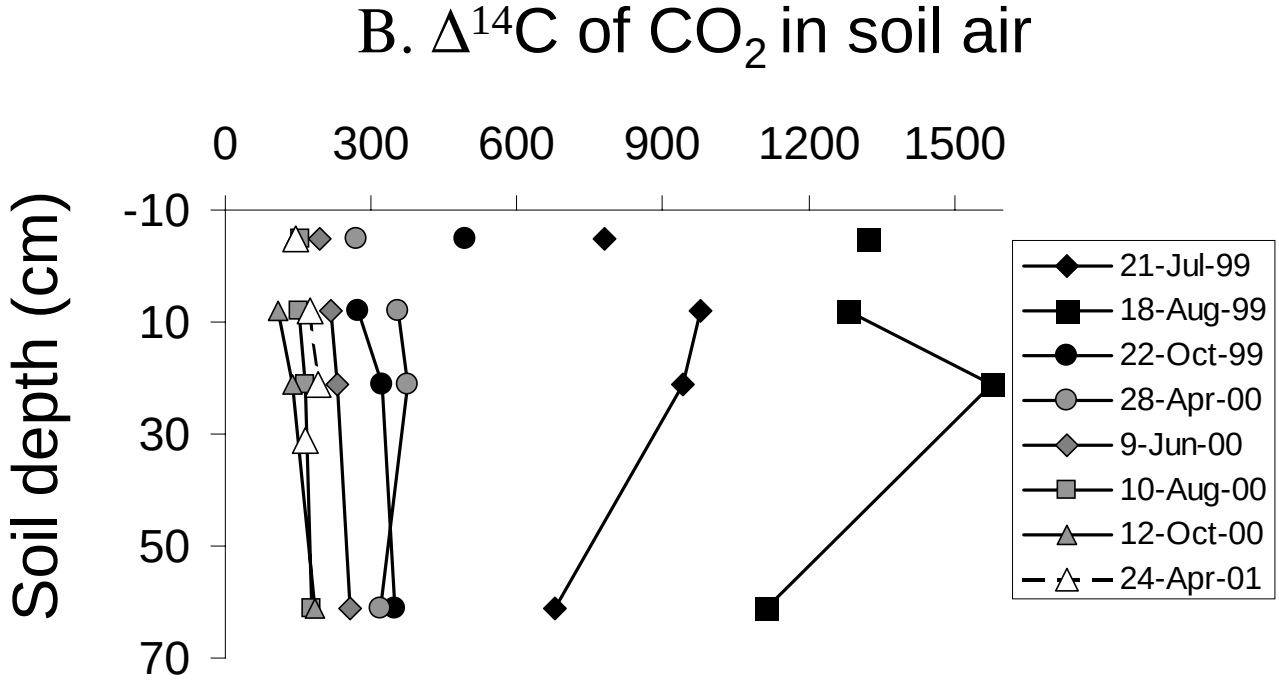

A.
1600
1200
800
400
0
Δ14C
Atmospheric
CO2
Tree ring
cellulose
1960
1980
2000
B. Δ14C of CO2 in soil air
0
300
600
900
1200
1500
-10
10
30
50
70
Soil depth (cm)
21-Jul-99
18-Aug-99
22-Oct-99
28-Apr-00
9-Jun-00
10-Aug-00
12-Oct-00
24-Apr-01

Figure 2


A.
1600
1200
800
400
0
Δ14C
Atmospheric
CO2
Tree ring
cellulose
1960
1980
2000


B. Δ14C of CO2 in soil air
0
300
600
900
1200
1500
-10
10
30
50
70
Soil depth (cm)
21-Jul-99
18-Aug-99
22-Oct-99
28-Apr-00
9-Jun-00
10-Aug-00
12-Oct-00
24-Apr-01

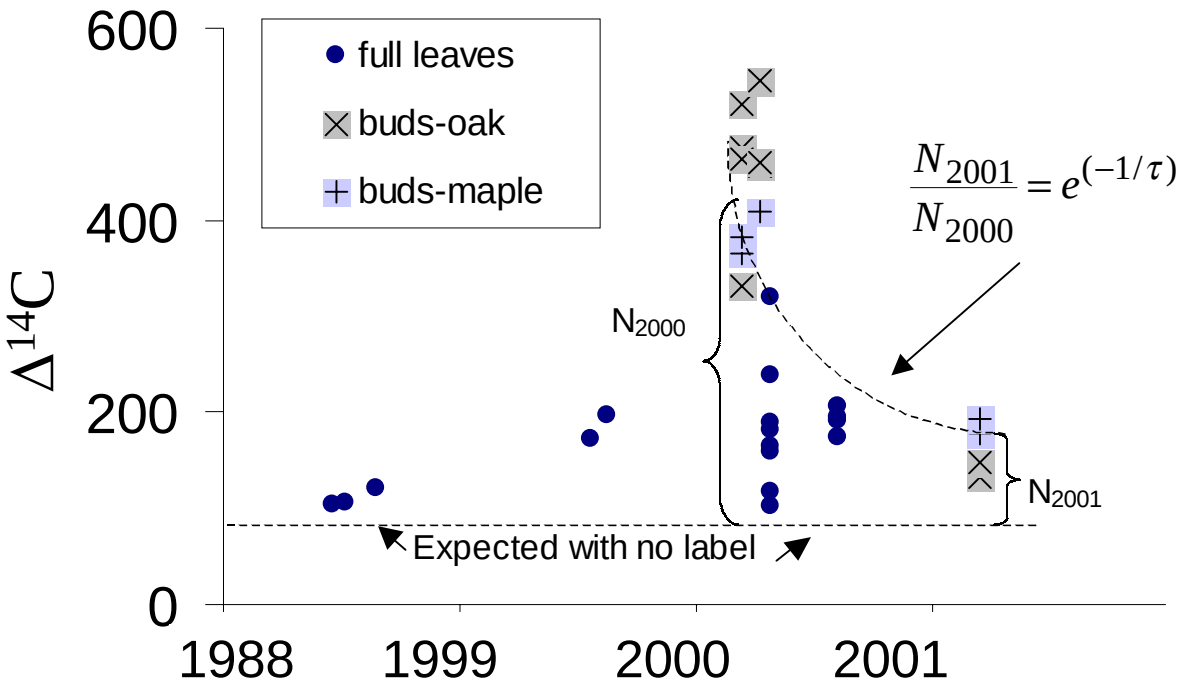

600
400
200
0
Δ14C
full leaves
buds-oak
buds-maple
$\frac{N_{2001}}{N_{2000}} = e^{(-1/\tau)}$
N2000
N2001
Expected with no label
1988
1999
2000
2001

Figure 3

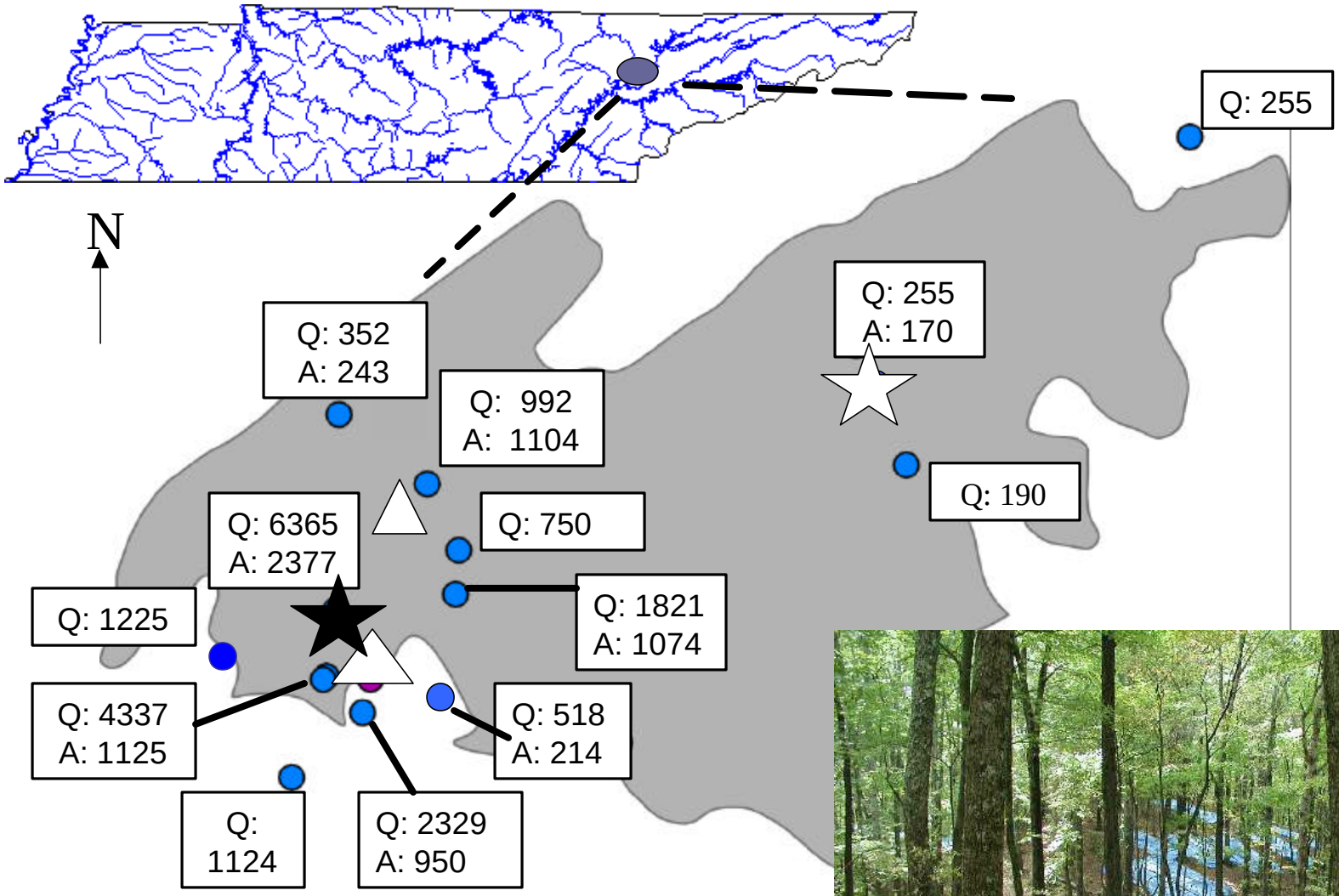

N
Q: 255
Q: 255
A: 170
Q: 352
A: 243
Q: 992
A: 1104
Q: 190
Q: 6365
A: 2377
Q: 750
Q: 1821
A: 1074
Q: 1225
Q: 4337
A: 1125
Q: 518
A: 214
Q:
1124
Q: 2329
A: 950
Hazardous waste
incinerators

Figure 4