that accompany the phase transition. Before the phase transition (–1 ps), we calculated N_1 to be 12.2 ± 0.3, which, within error, is the expected number for an fcc lattice (12). The loss of lattice structure and the subsequent atomic rearrangements reduce N_1 to 10.0 ± 0.3 at 6 ps. There is no observable change on longer time scales, as N_1 = 10.0 ± 0.3 at 50 ps as well.

The new technique of femtosecond electron diffraction has provided an unprecedented atomic-level view of ultrafast solid-liquid phase transition dynamics. In this instance, the picture it provides can be expressed in terms of an average pair correlation function, $H(r)$, which contains detailed information on nearest-neighbor distances, coordination numbers, and mean square vibrational amplitudes of the nonequilibrium state at each instant during the transition. This is a general technique, however, and can be applied to myriad systems in which photoinduced structural dynamics occur on the femtosecond time scale. Excellent candidates for investigation include condensed-phase processes, surface chemistry, and even time-resolved protein crystallography in cases where electron diffraction is viable. The single most important achievement is that of subpicosecond time resolution with sufficient structural sensitivity to reveal the atomic details of even nonreversible transition state processes that are central to concepts in chemistry and biology.

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Supporting Online Material

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Impacts of Fine Root Turnover on Forest NPP and Soil C Sequestration Potential

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Estimates of forest net primary production (NPP) demand accurate estimates of root production and turnover. We assessed root turnover with the use of an isotope tracer in two forest free-air carbon dioxide enrichment experiments. Growth at elevated carbon dioxide did not accelerate root turnover in either the pine or the hardwood forest. Turnover of fine root carbon varied from 1.2 to 9 years, depending on root diameter and dominant tree species. These long turnover times suggest that root production and turnover in forests have been overestimated and that sequestration of anthropogenic atmospheric carbon in forest soils may be lower than currently estimated.

Roots provide a path for movement of carbon and energy from plant canopies to soils; thus, root production and turnover directly impact the biogeochemical cycle of carbon in terrestrial ecosystems. However, accurate estimates of root life-spans and turnover rates have been elusive, and the impacts of root turnover on belowground processes in terrestrial ecosystems are not well known (*1*). Uncertainties in estimates of root longevity prevent proper quantification of net primary productivity (NPP) and belowground C allocation in forests. The contribution of root C to the formation of soil organic matter depends on root productivity, turnover rates, exudation, mycorrhizal colonization, and soil characteristics, all of which vary with forest type. Current estimates have indicated that fine root production contributes from 33 to 67% (*2*–*4*) of the annual NPP in forest ecosystems. These estimates assumed root turnover rates of about 1 year, despite reported turnover rates ranging from days to several years (*1*). Estimates of root turnover and longevity have been obtained through conventional biomass assessments (*4*, *5*), nitrogen budgeting (*6*), direct observation in minirhizotrons (*7*), pulse-labeling experiments using ^{14}C- (*8*) or ^{15}N-enriched fertilizers (*9*), and measurements of bomb-derived radiocarbon in root

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tissues (*10*). The disparity among estimates of root turnover has been debated for the past several decades, but global-change models have yet to incorporate the variability in root turnover rates associated with different forest types and/or tree species. Here, we used the isotopic composition of CO_2 added to air in elevated CO_2 experiments (*11*) to quantify turnover of C in fine root populations in two different forested ecosystems. Our findings demonstrate major differences in root C turnover times for contrasting tree species.

We monitored root C turnover in two forest plantations growing under free-air CO_2 enrichment (FACE): loblolly pine, *Pinus taeda*, near Durham, North Carolina, planted in 1980 (*12*), and sweetgum, *Liquidambar styraciflua*, in Oak Ridge, Tennessee, planted in 1988 (*12*). The two stands were similar in many respects: Trees were about 12 m tall when CO_2 treatments commenced and were in a linear growth phase, having fully occupied the site. The continuous CO_2 fumigation with FACE technology uses a depleted $^{13}CO_2$ source, which alters the $\delta^{13}C$ of plant tissues. In the FACE treatment, air was enriched in CO_2 by 192 parts per million (ppm) in pine and 150 ppm in sweetgum with the use of depleted $^{13}CO_2$ [$\delta^{13}C$ equaled −44 per mil (‰) and –55‰ relative to the Pee Dee Belemnite standard, respectively], which changed the $\delta^{13}C$ of CO_2 in the FACE treatment from –8‰ (ambient) to –20 and –21‰ in the pine and sweetgum forests, respectively. This change affected the ^{13}C composition of the vegetation, which is also determined by discrimination against atmospheric $^{13}CO_2$ during photosynthesis. Trees in FACE plots incorporated depleted $^{13}CO_2$ into new biomass, including root tissues. As the experiment continued, the standing crop of fine roots was progressively dominated by roots that reflected the new signature of ambient CO_2, which allowed us to estimate root C turnover during a 5-year period while the trees were exposed to added CO_2.

Fine roots were obtained from intact soil cores and from root ingrowth cores (*12*). Roots sampled from intact cores exhibit changes in the ^{13}C of the root population over time, which reflect the mixture of new and old roots (Fig. 1). Roots sampled from ingrowth cores exhibit only the ^{13}C signature of new roots produced after fumigation started and provide the end members for mass-balance calculations (Table 1). Live roots were sorted into three diameter (*Rt*) size classes: $Rt \leq 1$ mm, 1 mm $< Rt \leq 2$ mm, and 2 mm $< Rt \leq 5$ mm. Roots were considered living if they showed tensile strength and had white vascular tissue. At each sampling, roots in each size class were pooled within each FACE plot, and the $\delta^{13}C$ was determined by stable isotope ratio mass spectrometry (*12*).

The difference in $\delta^{13}C$ of the newly grown FACE roots from ingrowth cores as compared to pretreatment- or ambient-CO_2 grown roots was 11.4‰ for pine and 11.2‰ for sweetgum, averaged across all diameter size classes. The $\delta^{13}C$ of roots from ingrowth cores was similar to the measured $\delta^{13}C$ of new leaves (–39.2 ± 0.83 SE ‰) produced after CO_2 enrichment began (*13*). Thus, new root tissues with a diameter <5 mm were produced entirely from C fixed after the start of fumigation, and for pine this C was newly fixed and not mobilized from storage (*14*). New leaf and new root $\delta^{13}C$ signatures were similar, so incorporation of soil CO_2

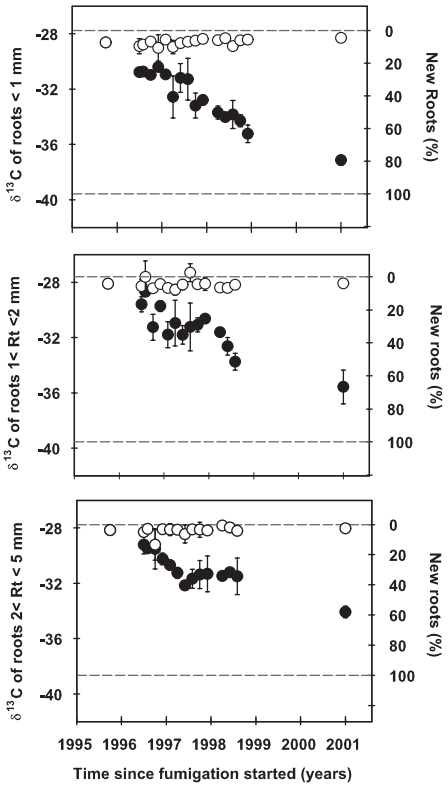

Fig. 1. The $\delta^{13}C$ of live pine roots harvested by sequential soil coring from June 1997 to December 2001 in the elevated CO_2 FACE treatment (filled symbols) and control (open symbols) experimental sites. Dashed lines are the average $\delta^{13}C$ of roots harvested from ingrowth cores and represent the end members for the mass balance calculation. The right axes show the percentage of roots initiated after CO_2 fumigation started.

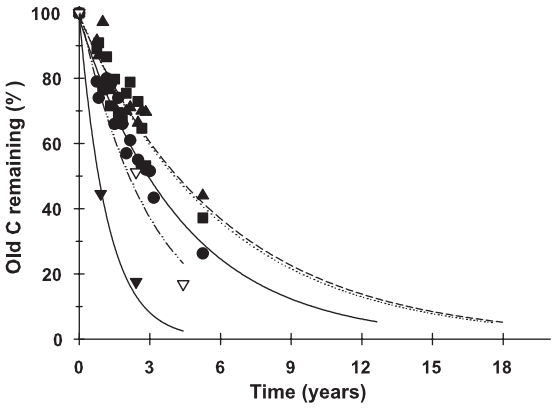

Fig. 2. The percent of old C remaining in roots, for each tree species and for each of the diameter size classes, was fitted to time (with the use of the SAS nonlinear model procedure) with a nonlinear equation [$F(t) = aexp^{-kt}$, where a is initial amount of old C and k is the decay rate of the old C] that best explained the $\delta^{13}C$ changes in the root population] over time. Roots ≤1 mm were fitted to $F(t) = 0.99exp^{-0.2343t}$, $r^2 = 0.99$, and $P(k) < 0.0001$ for pine (●) and $F(t) = 0.99exp^{-0.8356t}$, $r^2 = 0.995$ and $P(k) = 0.016$ for sweetgum (▼). Roots $1 < Rt \leq 2$ mm were fitted to $F(t) = 1.0exp^{-0.175t}$, $r^2 = 0.987$ and $P(k) < 0.0001$ for pine (■) and $F(t) = 1.0exp^{-0.3333t}$, $r^2 = 0.978$ and $P(k) = 0.122$ for sweetgum (▽). Roots $2 > Rt \geq 5$ mm were fitted to $F(t) = 0.99exp^{-0.160t}$, $r^2 = 0.988$, and $P(k) < 0.0001$ for pine (▲). The MRT of the roots was calculated as $-1/k$.

Table 1. Isotopic ^{13}C signatures of roots before (ORNL pretreatment, 1997) and after CO_2 fumigation started (ACO_2 indicates ambient CO_2; ECO_2, elevated CO_2). The $\delta^{13}C$ of roots from the ingrowth cores grew from placement in June 1997 through collection in August 1998 for the pine site and from placement in October 2001 through collection in October 2002 for the sweetgum site. Ingrowth sweetgum roots <2 mm in diameter were pooled together.

Root size class	Pretreatment $\delta^{13}C$ (‰)	Ingrowth cores $\delta^{13}C$ (‰)	
		ACO_2	ECO_2
	Pine roots		
$Rt < 1$ mm		–27.6 ± 0.2	–39.6 ± 0.1
$1 < Rt < 2$ mm		–27.6 ± 0.4	–39.5 ± 0.2
$Rt > 2$ mm		–27.7 ± 0.4	–38.5 ± 0.8
	Sweetgum roots		
$Rt < 1$ mm	–27.0 ± 0.2	–27.3 ± 0.1	–37.5 ± 0.4
$1 < Rt < 2$ mm	–26.5 ± 0.2	–27.3 ± 0.1	–37.5 ± 0.4

into roots via phosphoenolpyruvate carboxylase (PEPC) was negligible in these forests (*15*, *16*).

The disappearance of the old C in the root population followed an exponential decay function (Fig. 2) (*17*). We estimated mean residence time (MRT) of root C as the average number of years that a molecule of C, once it is fixed by photosynthesis and incorporated to new root mass, stays in the root system. This MRT is not sensitive to, but integrates, short-term climatic variability. The MRT of C in fine roots, <1 mm in diameter, was 4.2 years (with an error range of 4.0 to 4.6 years) for the pine forest and 1.25 years (1.1 to 1.4 years) for the sweetgum plantation. Because of the exponential loss of C from any annual cohort of roots, C has a maximum residence (i.e., 95% turnover) in roots <1 mm in diameter of about 12 years for pine and 4 years for sweetgum. The MRT of C in roots with diameters between 1 and 2 mm was 5.7 years (5.1 to 6.6 years), with a calculated maximum residue that can be >18 years for pine (Fig. 2). Sweetgum roots of this diameter had a MRT of 3 years (2.7 to 3.3 years) and a maximum residue of over 9 years. Similarly, the MRT of thicker pine roots, 2 to 5 mm in diameter, was 6.3 years (5.5 to 7.2 years), with a maximum residue greater than 18 years (Fig. 2).

The pine forest showed a very slow replacement of old root C, suggesting that the fine root population is composed mainly of semipermanent roots that survive for many years. In contrast, the rapid turnover of C in roots under sweetgum forest suggests that its fine root system is composed mostly of ephemeral roots that do not survive more than one growing season. The smaller average diameter of sweetgum roots (0.5 mm) as compared to pine roots (0.8 mm) is consistent with their more ephemeral nature (*18*, *19*).

With the use of the conventional method, i.e., the division of the maximum standing crop by annual gross production, we estimated that the MRT of fine roots <1 mm in diameter would be 3 years for pine (*20*) and 0.6 years for sweetgum (*21*). The discrepancy between conventional and $\delta^{13}C$-based estimates is substantial. It is possible that our $\delta^{13}C$-based estimates of root C turnover have been influenced by the exposure of the trees to atmospheric CO_2 enrichment. However, the long MRT of fine root C in the pine forest was confirmed by ^{14}C dating of root tissues collected under ambient CO_2 (roots <1 mm in diameter were dated at 4 to 5 years old by ^{14}C dating). Further, no difference in turnover rates between ambient and elevated plots was found in the sweetgum forest (*21*). Estimates of MRT by isotopic turnover of carbon includes the time from photosynthesis until its incorporation into the root system. Although this time is short, it could, in part, explain differences between conventional and isotopic estimates. Our isotopic estimates suggest that pine forests have much slower root C turnover rates than previously reported, which may indicate a conservative use of resources and energy in nutrient-poor soils (*22*). Assuming a 1-year MRT for fine roots, Jackson *et al*. (*2*) estimated that roots account for a third of terrestrial NPP. If the turnover of fine roots is much slower, this will substantially reduce estimates of the contribution of roots to the global annual NPP of terrestrial ecosystems.

The two forests exhibited very different ecosystem dynamics. The NPP of sweetgum increased by 21% under elevated CO_2 during the first 3 years of the study. During this period, there was a shift in C allocation from woody biomass to fine roots, increasing the proportion of forest NPP diverted to roots from 12 to 16% under elevated CO_2 (*19*). The pine forest also showed a CO_2-treatment effect, increasing NPP by 25% under elevated CO_2 (*23*). Although root production increased moderately, only 5 to 7% of total pine forest NPP was directed to roots (*20*). Although there are other pathways that deliver C to soils (for example, root exudation, turnover of mycorrhizal fungi, and surface litter), root turnover is a major input to soil organic matter. Thus, ecosystem differences in C allocation to roots and root turnover rates could be important determinants of changes in soil C storage resulting from atmospheric CO_2 enrichment.

Schlesinger and Lichter (*24*) measured C accumulation in the soil of the pine forest after 3 years of CO_2 fumigation. They found an increased sink for C in the forest floor, but the C in soil mineral horizons did not differ significantly from that under ambient CO_2 conditions. Although root production increased under CO_2 fumigation, the slow root C turnover in the pine forest is consistent with minimal effects on soil C, because after 3 years only 45% of the root C was replaced and available for entry into the soil pool. Overall, root production in the sweetgum forest was 45 to 50% greater than that in pine forest. The greater response of root production in sweetgum grown at elevated CO_2, coupled with the higher annual turnover of these roots, means that annual C inputs to soil in the sweetgum forest could be two times greater than in the pine forest. Thus, we might expect a greater increase in soil C after a shorter time period in the sweetgum forest than in the pine forest. Preliminary results show a significant increase in C concentration for the surface 5 cm of the mineral soil under the sweetgum plantation after only 3 years of CO_2 enrichment (*25*). Models predicting ecosystem C sequestration in soils must attend the possibility that this potential is strongly affected by root production and the MRT of C in the root system.

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