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Atmospheric Carbon Dioxide and Carbon Reservoir Changes

Reduction in terrestrial carbon reservoirs since 1850 has resulted in atmospheric carbon dioxide increases.

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The well-documented increase in the carbon dioxide content of the atmosphere since 1957 (1) is often attributed solely to the combustion of fossil fuels. An appreciable net transfer of CO_2 to the

mine the size of the net global biospheric CO_2 flux to the atmosphere. Long-term atmospheric changes are accounted for in the calculations by utilizing measurements of the ¹³C content of tree rings.

Summary. The net release of CO_2 from the biosphere to the atmosphere between 1850 and 1950 is estimated to amount to 1.2×10^9 tons of carbon per year. During this interval, changes in land use reduced the total terrestrial biomass by 7 percent. There has been a smaller reduction in biomass over the last few decades. In the middle 19th century the air had a CO_2 content of approximately 268 parts per million, and the total increase in atmospheric CO_2 content since 1850 has been 18 percent. Major sinks for fossil fuel CO_2 are the thermocline regions of large oceanic gyres. About 34 percent of the excess CO_2 generated so far is stored in surface and thermocline gyre waters, and 13 percent has been advected into the deep sea. This leaves an airborne fraction of 53 percent.

atmosphere, however, may also result from terrestrial biospheric changes such as clearing of forests. Both deforestation and the burning of dead organic materials when primary forests are converted to younger stands can result in a net release of CO₂. Estimates of net biospheric CO₂ release vary widely, and it has been suggested that this flux to the atmosphere may approach 5×10^9 tons of carbon per year (2, 3). The terrestrial reservoir (Fig. 1) is sufficiently large to allow fluxes of this magnitude for at least a couple of decades.

The purpose of this article is to deter-

Calculated in this way, the net global biospheric CO_2 flux is 1.2×10^9 tons of carbon per year for the interval 1850 to 1950.

Carbon Isotopic Ratios

The cumulative amount of CO_2 produced from fossil fuels is a well-known quantity. Between 1850 and 1950, about 60×10^9 tons of carbon were injected into the atmosphere from this source alone (1). The size of the atmospheric carbon reservoir is less well determined. Late 19th-century measurements of the CO_2 content of the atmosphere have considerable scatter, and the size of the atmospheric CO_2 reservoir in 1850 could have been anywhere from 540 to 625×10^9 tons of carbon, representing a CO₂ concentration of 260 to 300 parts per million (ppm).

Before 1950, the ¹⁴C in atmospheric CO₂ was produced by the interaction of atmospheric nitrogen and cosmic rayproduced neutrons and was not yet affected by ¹⁴C from nuclear bombs. The addition of fossil fuel CO₂ reduces atmospheric ¹⁴C levels because the carbon in fossil fuels lacks 14C, since these materials are much older than the 5700year half-life of 14C. Therefore, one would expect that the 60×10^9 tons of carbon in fossil fuel CO₂ released between 1850 and 1950 lowered the atmospheric ¹⁴C level by nearly 10 percent (60 \times 10⁹ tons in about 620 \times 10⁹ tons). However, because of ¹⁴C isotopic exchange with other reservoirs, the atmospheric ¹⁴C reduction is smaller. The actual reduction by 1950, known as the Suess effect (4), amounts to 1.7 to 2.3 percent (5). The magnitude of the Suess effect is not precisely known because changes in atmospheric ¹⁴C are also caused by changes in cosmic ray-produced neutron fluxes related to solar activity and geomagnetic field changes (6).

The ¹⁴C activity of dead organic materials decreases through radioactive decay by 1 percent every 80 years. Materials a few hundred years old have ¹⁴C levels a few percent below "baseline." These changes in ¹⁴C activity are relatively small, and consequently the release of CO_2 from the terrestrial biosphere does not measurably change atmospheric ¹⁴C levels.

Fossil fuel CO_2 not only completely lacks ¹⁴C, it also contains reduced amounts of the stable ¹³C isotope. During photosynthesis, plants discriminate against ¹³C in favor of the lighter ¹²C, and as a result the ¹³C/¹²C ratios of organic materials are lower than those of atmospheric CO_2 . The ¹³C/¹²C isotopic ratio (*R*) is generally discussed in terms of the relative difference between the isotopic ratio of the sample and that of the PDB (Pee Dee belemnite) standard

 δ^{13} C per mil =

$$\left(\frac{R_{\rm sample} - R_{\rm PDB}}{R_{\rm PDB}}\right) \times 1000$$

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SCIENCE, VOL. 199, 20 JANUARY 1978

For coal and petroleum the δ^{13} C values range from -22 to -28 per mil on the PDB scale (7), whereas natural gas is lighter, with δ^{13} C values in the range -35 to -48 per mil. Fossil fuels of major importance for the period 1850 to 1950 are coal and petroleum, which contain about 2½ percent less ¹³C than the PDB standard. Atmospheric CO₂ is only 0.7 percent deficient (δ^{13} C = -7 per mil).

Biospheric O_2 has a $\delta^{13}C$ spread similar to that of coal and petroleum. Keeling (8) studied the O_2 in rural air and found $\delta^{13}C$ values ranging from -21 to -26 per mil.

Anthropogenic release of CO_2 from both fossil fuels and the biosphere reduces ¹³C levels in the atmosphere, but ¹⁴C is reduced only by the fossil fuel component. In this article I evaluate both ¹³C and ¹⁴C changes in order to estimate the biospheric flux.

The Carbon-13 Record in Trees

Anthropogenic reduction of ¹³C in atmospheric CO₂ should be reflected in the ¹³C content of tree cellulose. Both cellulose and lignin are basic building materials of the cell wall, but only cellulose is formed entirely during the growing season, whereas a portion of the lignin is formed later with the heartwood. Some of the published ¹³C measurements (9-11) are for whole wood, containing cellulose, lignin, sugars, and so on. A change in the relative proportions of these constituents will change ¹³C ratios because their ¹³C contents differ by up to 4 per mil. Therefore, whole wood ¹³C ratios may give less reliable ¹³C records, especially when heartwood-sapwood boundaries are crossed.

My own ¹³C measurements are for a Douglas fir, covering the years 1820 to 1972. Each sample represented 3 years of growth and weighed about 100 grams. Acid washing (2 percent HCl) and treatment with 2 percent hot alkali were used to obtain soda pulp (soda "cellulose"), and about 35 percent of the wood was lost during this treatment. The end product still contained lignin, and the data plotted in Fig. 2 represent only the heartwood portion (1820 to 1932), which has a fixed lignin content. The tree records atmospheric δ^{13} C changes at 47°46'N, 124°06'W, about 25 kilometers from the Pacific Ocean at an altitude of about 120 meters. It was part of a mature forest logged in late 1972, and the age distribution of the surrounding trees is not known. For most of its life, this tree was probably one of those forming the canopy, and it should have utilized free air (tropospheric) CO₂ during this period. Although it was most likely a good recorder of atmospheric ¹³C variations, in future research it might be better to concentrate on freestanding trees, for which local environmental factors are less important.

The δ^{13} C record (Fig. 2) of the Douglas fir shows a gradual reduction in δ^{13} C values for the interval 1850 to 1932. Similar downward trends are also recorded up to 1932 by the other trees, with the exception of the Russian spruce. This anomalous behavior of the spruce may have



Fig. 1. Major reservoirs of importance for the carbon cycle. Reservoir sizes are drawn to scale; the basic data are from (I).

been due to (i) a reduced supply of recycled biospheric CO_2 if the tree increased in height until it reached above the surrounding canopy or (ii) variations in the chemical composition of the tree rings, which would make this species less reliable as an isotopic indicator.

The δ^{13} C records of the trees represented in Fig. 2 differ between 1932 and the present. The ¹³C content of the Tasmanian tree increased between 1950 and 1970. There was also a ¹³C increase for one of the English trees, but here the upward trend started near 1930. For the Atlantic Coast trees an upward trend is suggested between 1935 and 1960, but the last two measured points are distinctly low. The Brazilian tree shows a continuation of the downward trend in δ^{13} C over its entire record. The Tasmanian tree shows a significant correlation between the ¹³C record and regional temperature. However, the mechanism by which a full 1 per mil change in ¹³C can be linked to temperature, either directly or indirectly, is not clear (11).

The Douglas fir changes from heartwood to sapwood near the 1932 tree ring. During heartwood formation lignin is added to the cells. Thus, as heartwood rings were formed between 1914 and 1932, lignin was added in equilibrium with 1954 to 1972 atmospheric ¹⁴C levels (assuming the sapwood remained at a width of approximately 40 years). These ¹⁴C levels were appreciably higher than normal because of the addition of 14C nuclear bombs, and the lignin added in these years reflects the higher levels. Measurements of the ¹⁴C levels of the heartwood rings, after treatment with alkali as described above, show a maximum ¹⁴C increase of 1 percent. The maximum increase in the atmospheric ¹⁴C content was 80 percent in 1963. Thus, the amount of additional lignin deposited during heartwood formation that was not removed by our treatment was at most 1 percent of the treated sample.

Lignin is about 4 per mil more deficient in ¹³C than cellulose (9). One expects the sapwood samples, without the additional lignin, to be about 0.04 per mil higher in ¹³C content than the heartwood samples. The actual measurements showed an increase of 0.5 per mil at the heartwood-sapwood boundary, only one-tenth of which can be explained by differences in lignin content. The increased ¹³C content in the sapwood portion of the Douglas fir, and its possible relationship with changes in chemical composition, remain to be investigated. In this article only the heartwood results are used.

Implications of the Carbon-13 Trends in Trees

A composite set of data points for trees for which the $\delta^{13}C$ of either alpha cellulose or soda "cellulose" was measured (12) is given at the bottom of Fig. 2. The differences between the absolute $\delta^{13}C$ values of the various curves are mainly caused by differential fractionation of the various species; only the trends are of importance for this study. The composite data were obtained by matching the average δ^{13} C values for (i) the Brazilian araucaria in the interval 1920 to 1930 and (ii) the Coastal Atlantic trees in the interval 1910 to 1930 with the average $\delta^{13}C$ values for the Douglas fir over the same intervals. The overall reduction in ¹³C content is close to 1.1 per mil between 1850 and 1950.

A complication arises from the temperature sensitivities of the trees. For Monterey pine, for instance, differences in δ^{13} C are found between early and late wood in the same year [Fig. 2 and (9)]. The change in $\delta^{13}C$ amounts to 0.2 per mil/°C. Although the average global temperature variation during the last 100 years was on the order of only 0.5°C, the amplitude of the temperature variation might have amounted to at least 1°C at higher latitudes. The effect of such average long-term temperature changes on the ${}^{13}C/{}^{12}C$ ratio in trees should be nearly an order of magnitude less than that of anthropogenic reservoir changes.

Annual variations in temperature during the growing season would be expected to affect $\delta^{13}C$ values for trees to a greater extent than century-long temperature trends. When averaged over decades, however, this effect should be negligible for the Douglas fir. Between 1890 and 1930, the range of mean annual decadal temperatures on the Olympic Peninsula was only 10.6° to 10.9°C. Mean decadal temperatures for the growing season were only slightly more variable Decadal temperatures for the month of May, for instance, ranged from 12.2° to 12.9°C. Furthermore, the tree's photosynthesis may have been accelerated or delayed depending on temperature, thus attenuating the influence of seasonal changes in temperature on its ¹³C record.

Another complication arises from the bimodal distribution of δ^{13} C in plants. Plants with the C₄ pathway of photosynthesis have δ^{13} C values near -14 per mil, whereas those with the C₃ pathway average about -27 per mil (*13*). The C₃ variety is more common, and as a result δ^{13} C values for coal and petroleum are generally in the range -22 to -28 per mil (7). 20 JANUARY 1978

Agricultural practices, however, may have somewhat shifted the ratio of C_3 to C_4 plants. For instance, a shift may occur when an area having vegetation of the C_3 variety is converted to the cultivation of plants with a C_4 photosynthetic pathway, such as corn. Even if the total biomass remains the same in such an area, there is a net uptake of ¹³C from the atmosphere because the new material is less deficient in ¹³C. Detailed surveys of the distribution of C_4 plants are not available, but their contribution to the total biomass is probably only a few percent. Internal shifts between C_3 and C_4 plants should have a negligible effect on our calculations (but perhaps not in the future, if sugarcane alcohol is burned on a large scale as a fuel).

Similarly, the possibility that there are more C_4 plants in southern forests than in forests in temperate regions should be considered. Little is known of the distribution of C_4 plants in tropical forests. Their contribution again may be negligible, but to evaluate the influence of the more recent clearing of portions of the Amazon forest on the ¹³C record, a more



Fig. 2. Record of δ^{13} C in trees from various regions. The data are obtained from (9-11) except for the Douglas fir. The set of points at the bottom was derived from a combination of three data sets. A description of cellulose is given in the text.



Fig. 3. Change in atmospheric δ^{13} C after correction for fossil fuel addition (see text). The data reflect atmospheric Δ^{14} C changes introduced solely by net biospheric CO₂ release. The curve was drawn as a visual aid.

detailed study of the distribution of C_3 and C_4 trees is needed.

As discussed above, the carbon exchange between atmosphere and the oceans plus the biosphere causes an attenuation of the maximum possible ¹⁴C reduction expected from fossil fuel CO₂ release. The actual lowering of the atmospheric ¹⁴C level (= S percent) for the interval 1850 to 1950 is obtained from measurements of ¹⁴C in tree rings. Reductions in ¹⁴C can also be calculated from specific models, as done, for instance, by Oeschger *et al.* (14). Their calculated S values are used in this article.

Because fossil fuel CO₂ contains about 18 per mil less ¹³C than atmospheric CO₂, the release of fossil fuel CO₂ lowers atmospheric δ^{13} C values. The ¹³C changes are smaller than the ¹⁴C changes because the difference in ¹⁴C content between fossil fuel and atmospheric CO₂ is 100 percent. The resulting δ^{13} C change, caused by addition of fossil fuel CO₂, is therefore 18/100 of the ¹⁴C change, or

$$\delta^{13}C = 0.18 S$$
 (1)

Substitution of the appropriate *S* values (*14*) into Eq. 1 gives a δ^{13} C lowering of 0.14 per mil in 1910, 0.20 per mil in 1920, and 0.36 per mil in 1950 (for an *S* value of 2 percent). Because fossil fuel CO₂ release has increased at a constant rate over much of this century, the δ^{13} C trend can be extrapolated; this gives a δ^{13} C lowering of 0.7 per mil by 1970.

By adding to the measured δ^{13} C values for trees the calculated fossil fuel contribution, a record reflecting δ^{13} C changes caused solely by biospheric fluxes can be obtained. The resulting curve, given in Fig. 3, shows that the major biospheric injection was between 1860 and 1930.

Up to 1970, the fossil fuel-induced δ^{13} C lowering was 0.7 per mil for a total injection of 120×10^9 tons of carbon. The δ^{13} C lowering due to biospheric CO₂ was also about 0.7 per mil (the interval 1860 to 1930 in Fig. 3). The fossil fuelinduced lowering resulted from an exponential increase in the use of fossil fuels, but the biospheric input function is of a different nature. A wide variety of input functions can yield the 0.7 per mil biospheric δ^{13} C change in Fig. 3, and these functions can be derived from model calculations. However, the scatter in the set of $\delta^{13}C$ data makes such calculations premature, and I prefer to delay precise calculations until several long-term $\delta^{13}C$ records from selected trees are available. I estimate that the integrated biospheric flux between 1850 and 1950 is equal to the fossil fuel flux between 1850 and 1970—that is, 120×10^9 tons of carbon. Because of the uncertainty in the biospheric input function, this estimate may have a fairly large error.

A possible cessation of net biospheric CO₂ release after 1930 would bring the δ^{13} C values in Fig. 3 back to the 1850 level. The response to a single biospheric input is fairly fast. For the box-diffusion model, the transient atmospheric excess CO₂ is reduced to half its original value in 21 years (15). A cessation of net biospheric CO₂ release over the last few decades would more than halve the $\delta^{\scriptscriptstyle 13}C$ anomaly in Fig. 3. Such a trend may be indicated by the 1 per mil increase in δ^{13} C of the Tasmanian tree, and also by the 0.5 per mil increase of the sapwood portion of the Douglas fir. The set of data in Fig. 3, however, is more compatible with a reduced rate of net biospheric CO₂ release over the last few decades. With the present set of tree ${}^{13}C$ data it is not possible to distinguish between (i) the cessation of net biospheric CO₂ release and (ii) a further reduction of total biomass at a reduced rate for the last few decades.

The above discussion suggests an addition to the atmosphere of 120×10^9 tons of carbon through biospheric CO₂ release between 1850 and 1950, together with 60×10^9 tons of carbon from fossil fuel CO₂. The next step is to account for the fate of the 180×10^9 tons of carbon added as CO₂ to the atmosphere between 1850 and 1950.

Carbon Dioxide Transfer to the Oceans

With a net flux of CO_2 from the terrestrial biosphere to the atmosphere, the only major sink for excess atmospheric CO_2 appears to be the world ocean. The regions where anthropogenic CO₂ is taken up can be identified by investigating the transfer of ¹⁴C and ³H from nuclear explosions. These tracers were measured at the University of Washington (¹⁴C) and the University of Miami (¹⁴C) and ³H) as part of the GEOSECS (Geochemical Ocean Sections) oceanographic program. Some of the ¹⁴C data for West Atlantic waters collected in 1972 at northern Atlantic GEOSECS stations have been published (16, 17). Carbon-14 activity is often expressed as Δ^{14} C, which is the per mil deviation, after correction for isotope fractionation, of the measured ¹⁴C activity from a ¹⁴C standard of the National Bureau of Standards (16, 17).

The distribution of ¹⁴C in the upper 1200 m of the West Atlantic in 1972 is



Fig. 4. Isograms of Δ^{14} C in the West Atlantic, constructed from Δ^{14} C profiles at the indicated GEOSECS stations (see Fig. 6 for geographic locations). The δ^{13} C data for stations 3, 5, 11, 29, 31, 37, and 40 are from Ostlund *et al.* (17).

given in Fig. 4. The Δ^{14} C isograms show two well-defined regions of maximum 14C penetration near 30°N and 30°S. Carbon-14 concentrations in surface waters increased from a value of -57 per mil before nuclear testing (18) to an average value in 1972 of +127 per mil between 35°S and 45°N. Similar patterns are observed for ³H (19), and measurable ³H has penetrated to approximately the -80per mil 14C isogram between 0°S and 45°N. Negligible amounts of ¹⁴C are found in the thermocline in tropical regions. The deepest penetration of ¹⁴C is near the center of large surface gyres. The various mechanisms and models explaining the Δ^{14} C distribution will be discussed in detail elsewhere (20). Nuclear bomb 14C, as calculated from these models, was distributed in 1972 between 35°S and 45°N according to the pattern in Fig. 5. Using the standard exponential fossil fuel CO₂ increase, about 4 percent per year, the detailed calculations (20) show that excess CO_2 in the oceanic mixed layer plus gyre reservoirs amounts to 0.64 times the atmospheric CO₂ excess.

Transfer of excess atmospheric CO₂ directly to the deep sea (>1000 m) takes place in the Arctic and Antarctic deepsea outcrop areas. Most of the deep-sea ¹⁴C activities have not vet been influenced by nuclear bomb carbon, and the changes in natural ¹⁴C specific activities give the progressive aging of the deep waters. The aging of North Atlantic deep water, from its source near Greenland to a latitude about 40°S, is shown in Fig. 6 for both the East and West Atlantic. The oldest water, which is about 300 years old, is encountered in the East Atlantic. This age is an upper limit because there may have been mixing with some Antarctic bottom water, which has a lower ¹⁴C content. The rate of formation of Atlantic deep water is determined by the rate of aging given above. Each year the sinking waters remove about 0.5 percent of the excess atmospheric CO₂ to the deep ocean (5, 20).

The integrated amount of excess CO_2 advected directly into the deep ocean between 1850 and 1950 depends on the actual change in atmospheric CO_2 content. For an exponential increase in atmo-

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spheric CO_2 of 4 percent per year, an amount equal to about 14 percent of the 1950 atmospheric excess would have been removed to the deep sea. However, the total biospheric and fossil fuel CO_2 flux (Fig. 7) gives a more linear increase. The amount of excess CO_2 advected to the deep sea between 1850 and 1950 equals 25 percent of the atmospheric CO_2 increase for 1950.

Summarizing, by 1950 an amount of CO_2 equal to 64 percent of the atmospheric CO_2 excess is found in the surface and thermocline gyre areas of the oceans, and 25 percent has been advected directly into the deep oceans. Al-



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Fig. 5 (top). Calculated nuclear bomb ¹⁴C concentrations in the West Atlantic in 1972 between 35°S and 45°N. The main penetration is in the center of large gyres. Fig. 6 (bottom). Values of Δ^{14} C for North Atlantic deep water originating in the western and eastern Atlantic. Samples were collected at the stations and depths indicated. Station 31 data are from Ostlund *et al.* (17). The scale at the right gives the aging derived from the change in Δ^{14} C.

Fig. 7. Cumulative biospheric and fossil fuel CO₂ release to the atmosphere. The biospheric contribution was estimated from the $\delta^{\rm 13}C$ data in Fig. 3. The slope of the biospheric curve is approximate.

together, there has been a nearly equal (53:47) division between the atmosphere and the oceans of the excess CO_2 released up to 1950. This agrees with the estimate of 49 ± 12 percent (21) for the airborne fraction of fossil fuel CO₂ released between 1959 and 1969, when net biospheric fluxes were small.

Atmospheric Carbon Dioxide and Climatic Change

Of the 180×10^9 tons of carbon added as CO_2 to the atmosphere between 1850 and 1950, about half has been transferred to the oceans, according to the calculations above. Unless we can identify other large sinks for fossil fuel CO₂ uptake, the same amount remained in the atmosphere. The 1950 atmospheric reservoir contained about 650 \times 10⁹ tons of C, and consequently an atmospheric reservoir containing about 560×10^9 tons is derived for the year 1850. The corresponding atmospheric CO₂ content of 268 ppm in 1850 appears low when compared with most measured values for the 19th century (22). This value is more compatible with Brown and Escombe's measurement (23) of 274 ± 5 ppm for the turn of the century. Callendar (24) used their values in the first description of the influence of atmospheric CO₂ increase on climate.

The relatively low atmospheric CO_2 value for the middle of the last century implies an 18 percent increase in atmospheric CO₂ content between 1850 and the present. This increase was approximately linear up to 1960 (Fig. 7). Its effect on climate is not clear because there have been other influences on the global climate during this interval. Global temperature increases of 2° to 3°C have been predicted for early in the next century, when the atmospheric CO₂ content is expected to have doubled. The equivalent temperature increase for 18 percent excess atmospheric CO_2 is 0.5° to $0.6^{\circ}C$. Average global temperatures increased by nearly 0.5°C between 1880 and 1940, but since 1940 about half of the increase has been canceled by a cooling trend. Although it is, of course, tempting to relate the warming trend between 1880 and 1940 to the release of 170×10^9 tons of anthropogenic CO_2 , the subsequent cooling shows that the CO₂ increase is not



yet sufficiently large to produce a clear signal distinguishable from natural climatic variability. Our future anthropogenic "hypsithermal" has not yet been proved.

Changes in Terrestrial Carbon Reservoirs

Between 1850 and 1950 about 120 \times 109 tons of carbon was moved from the biospheric reservoir to the atmosphere. The total amount of carbon in the terrestrial biosphere is estimated as 1760×10^9 tons [$\sim 680 \times 10^9$ tons live and $\sim 1080 \times$ 10^9 tons dead (1, 25)]. Thus, changes in land use reduced the total terrestrial biomass by 7 percent in 100 years. With the method used here it is not possible to distinguish between live and dead organic materials or to give the relative contributions of these components to the overall 7 percent reduction.

Two-thirds of the CO₂ added to the atmosphere between 1850 and 1950 was derived from biospheric sources, and only one-third came from fossil fuel combustion. The deforestation of the Great Lakes region starting in 1870 and the changes in forest composition resulting from large-scale logging operations in the Pacific Northwest early in this century are examples of a worldwide pattern of deforestation contributing to the biospheric CO₂ release.

At present, fossil fuel combustion is the dominant factor because net biospheric fluxes appear to have been negligible over recent decades (Fig. 3). A negligible net biospheric flux, however, does not mean a status quo in land use. Increased levels of atmospheric CO₂ cause higher rates of photosynthesis in many plants. The dead organic carbon reservoir is also being enlarged by increased sedimentation related to increased nutrient (phosphate) availability. These fluxes back into the biosphere may have cancelled the effects of net reductions in forested areas.

The rates of annual biospheric CO₂ release derived here depend strongly on the δ^{13} C change, and a difference in the trend of a few parts in 10,000 may change the estimate by about 0.3×10^9 tons of carbon per year. More (and longer) precise records of δ^{13} C in trees, combined with model calculations, are needed for a better understanding of the net biospheric CO_2 release.

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 Supported by NSF Climate Dynamics grant ATM75-22650 and by NSF International Decade of Ocean Exploration grant OCE76-03930. The ¹⁴C oceanographic data given here will be utilized for the calculation of CO₂ atmosphere-ocean gas exchange coefficients and for modeling of natural ¹⁴C variations (20). The sample collection at sea was carried out by personnel of the GEOSECS Operations Group, with A. Bainbridge of the Scripps Institution of Oceanography directing the development of equipment and the fieldwork. Comments by W. S. Broecker, H. Oeschger, and R. L. Burk resulted in many improvements of this article. provements of this article.