Biogeochemistry **24**: 115–127, 1994.
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Carbon dioxide consumption during soil development

OLIVER A. CHADWICK[1], EUGENE F. KELLY[2], DOROTHY M. MERRITTS[3] & RONALD G. AMUNDSON[4]
[1] *Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91109;* [2] *Dept. of Agronomy, Colorado State University, Fort Collins, CO 80523;* [3] *Geology Dept., Franklin and Marshall College, Lancaster, PA 17604;* [4] *Soil Science, University of California, Berkeley, CA 95720, USA*

Received 28 September 1993; accepted 20 January 1994

Key words: carbon sequestration, landscape geochemistry, mineral weathering

Abstract. Carbon is sequestered in soils by accumulation of recalcitrant organic matter and by bicarbonate weathering of silicate minerals. Carbon fixation by ecosystems helps drive weathering processes in soils and that in turn diverts carbon from annual photosynthesis-soil respiration cycling into the long-term geological carbon cycle. To quantify rates of carbon transfer during soil development in moist temperate grassland and desert scrubland ecosystems, we measured organic and inorganic residues derived from the interaction of soil biota and silicate mineral weathering for twenty-two soil profiles in arkosic sediments of differing ages. In moist temperate grasslands, net annual removal of carbon from the atmosphere by organic carbon accumulation and silicate weathering ranges from about 8.5 g m^{-2} yr^{-1} for young soils to 0.7 g m^{-2} yr^{-1} for old soils. In desert scrublands, net annual carbon removal is about 0.2 g m^{-2} yr^{-1} for young soils and 0.01 g m^{-2} yr^{-1} for old soils. In soils of both ecosystems, organic carbon accumulation exceeds CO_2 removal by weathering, however, as soils age, rates of CO_2 consumption by weathering accounts for greater amounts of carbon sequestration, increasing from 2% to 8% in the grassland soils and from 2% to 40% in the scrubland soils. In soils of desert scrublands, carbonate accumulation far outstrips organic carbon accumulation, but about 90% of this mass is derived from aerosolic sources that do not contribute to long-term sequestration of atmospheric carbon dioxide.

Introduction

Understanding sources, sinks, and fluxes of carbon in soils is critical to interpreting ongoing anthropogenic perturbations to the natural carbon cycle. More then four times as much carbon is stored in earth's soils as in the atmosphere, about 1550 gT as organic carbon and 1700 gT as carbonate carbon (Eswaran 1994). Although, short-term carbon cycling (photosynthesis and soil respiration) is documented for many ecosystems (Raich & Schlesinger 1992), there are few reliable estimates of long-term net carbon fluxes from atmosphere to soils (Schlesinger 1990; Harden et al. 1992). Our understanding of long-term carbon cycling in soils is limited because of the complexity of vegetation-soil-landscape relationships. The paucity of data is evident to ecologists and earth scientists who are concerned with carbon transfer rates among the atmos-

phere, organisms, surface and ground water, and the organic and inorganic phases in soils (Van Breeman & Feijtal 1990; Schlesinger 1991). It also limits geochemists and paleoclimatologists who model the implications of changes in carbon flux through time (Berner et al. 1983; Olson et al. 1985; Raymo & Ruddiman 1992).

Here, we quantify net long-term carbon transfers from the atmosphere to soils as they develop in moist temperate grassland and desert scrubland ecosystems. Soils developing in terrace sediments of known and increasing age provide a time history of their interaction with the atmosphere. We analysed chemical properties of soil chronosequences sampled from two such localities: marine terraces near the mouth of the Mattole River in northern California and river terraces in the Wind River basin in Wyoming. Carbon-cycle interpretations derived from these measurements are presented in the context of soil evolution and ecosystem functioning.

Temporal aspects of the soil carbon cycle

When photosynthetic organisms colonize freshly exposed geologic substrate, carbon derived from the atmosphere moves to terrestrial ecosystems and their soils. The processes that govern atmosphere-ecosystem-soil carbon cycling are complex; the importance of competing pathways varies as a function of time (Olson et al. 1985). Over yearly time-scales, biological processes strongly control carbon cycling because atmospheric CO_2 consumed during photosynthesis is nearly balanced by CO_2 released during soil respiration (Raich & Schlesinger 1992). However, during each photosynthesis – respiration cycle, a small amount of carbon is not returned to the atmosphere, but is held in living biomass or soil organic matter, leached as dissolved organic matter, or diverted into inorganic carbon phases during chemical weathering. In the absence of anthropogenic perturbations, gains and losses of carbon from biomass and soil organic matter often reach quasi-steady state within 10^2 to 10^4 yr (Jenny 1980), but leaching of inorganic carbon continues to contribute to a longer term carbon cycle (Olson et al. 1985).

The long time-scale ($>10^3$ yr) terrestrial sink for atmospheric carbon is controlled by interactions between soil biota and weathering of silicate minerals (Berner 1992). Biological activity in soil results in local CO_2 concentrations that are commonly 10 to 100 times greater than in the atmosphere (Amundson & Davidson 1990). During soil leaching, HCO_3^- in solution acts to charge-balance cations that are released during mineral weathering (Olson et al. 1985; Van Breeman & Feijtal 1990; Berner 1992; Puckett & Bricker 1992). In essence, some bicarbonate carbon is diverted from the short-term biological cycle into a long-term hydrogeological cycle. Leached bicarbonate carbon is stored in liquid or solid phases within the groundwater/vadose zone or in ocean water and sediments. Vadose zone carbonate deposits may be recylced in the terrestrial environment by erosion and subsequent redistribution by wind (Gillette et al. 1992), but most carbon carried to the oceans is

removed from terrestrial cycling for more than 10^8 yr (Berner et al. 1983; Berner 1991).

Soils play an integral role in both the biological and hydrogeological carbon cycles; they are a site for microbiological breakdown of organic matter (soil respiration) and store the organic and inorganic byproducts of these reactions (soil development). Until recently, little attention was paid to the long-term carbon-cycle record stored in soils (Schlesinger 1990; Berner 1992, Harden et al. 1992).

Materials and methods

Approach and assumptions

Quantification of carbon fluxes during soil development requires integration of geomorphological and pedological techniques. To collect the required data, we sampled soil profiles from two soil chronosequences on stable surfaces of water-laid terraces where sediments containing the soils could be assigned ages with reasonable confidence. Deposit ages were combined with measurements of the organic and inorganic residues of carbon-cycle reactions to calculate long-term rates of carbon transfer for each soil profile. A systematic error inherent in flux determinations from chronosequences is that each successively older soil contains the cumulative record of earlier, often larger fluxes that tend to overestimate the long-term flux (Schlesinger 1990).

The chronosequence concept requires that field-site selection minimize the effects of dominant nonchronological controls on soil development (climate and climate change, organisms, topography, parent material, and anthropogenic activity). For soil-properties determined by long-term weathering processes, chronosequence measurements usually indicate that the soil profile in the oldest sediment is at maximum development for the sequence and soil profiles in progressively younger sediments represent earlier stages of an evolutionary pathway that through time will lead to the maximal stage (Birkeland 1984). Organic carbon accumulation in chronosequences is more difficult to interpret because it is determined both by ecosystem properties that are a function of recent climate and soil properties such as clay content and mineralogy that are a function of soil evolution. Where macro-climate is held constant, changes in organic carbon along a time-sequence of soils represents differences in soil retention of organic carbon due to age-related inorganic soil properties.

Sampling and analytical procedures

Sampling locations were chosen based on many reconnaisance observations. They were selected to minimize post-depositional erosion/deposition influ-

ences on the profiles, so their properties could be related to the age of the terrace deposit. Soils were sampled by horizon to between 2 and 3 m depth from pits that had been excavated by hand in California and by backhoe in Wyoming.

The approach to quantifying carbon sequestered during soil development is two fold: (1) direct measurement of (a) organic carbon in all soil profiles, and (b) carbonate carbon stored in arid soil profiles, and (2) calculation of carbon that was removed from the atmosphere during silicate weathering and subsequently leached from humid soil profiles. Carbon stored in soil organic matter and carbonates was measured using oxidation and acid-reacted manometric procedures, respectively (Nelson & Sommers 1982). In arid soils, the sampled mass of carbonates stored in the top 2.5 m was measured as an estimate of carbon consumption due to inorganic processes because bicarbonate-rich soil solution seldom moves far below the surface before evapotranspiration effects carbonate precipitation by drying (McFadden et al. 1991). Mass accumulation of carbon is calculated as follows.:

$$\delta_{j,w} = (\rho_w C_w F_w - \rho_p C_p F_p)/100. \quad (1)$$

Where 'j' refers to mobile elements, 'p' refers to parent material, 'w' refers to weathered soil, ρ is bulk density, F is the < 2mm fraction of the soil, C is elemental concentration in wt. %.

Calculation of carbon consumed during weathering of humid-environment soil profiles is based on functional, mass-balance relationships among soil chemical composition, bulk density, and volume change in relation to the soil parent material (Chadwick et al. 1990; Merritts et al. 1992). Because cation (Na^+, K^+, Ca^{2+}, Mg^{2+}) leaching controls the quantity of bicarbonate removed from each profile (Puckett & Bricker 1992), we calculate mass loss of cations as follows (Brimhall et al. 1992):

$$\delta_{j,w} = (\rho_w C_{j,w} F_w(\varepsilon_{i,w} + 1) - \rho_p C_{j,p} F_p)/100. \quad (2)$$

Where 'p', 'w', 'ρ', 'F' and 'C' are the same as in (1) and 'j' refers to mobile elements, 'i' refers to an immobile element (in this case, Zr), and $\varepsilon_{i,w} = ((\rho_p C_{i,p})/(\rho_w, C_{i,w})) - 1$. Cation mass-loss values were integrated over the 2 to 3 m sampling depth for each soil, and are presented on a whole-soil basis with error estimates based on uncertainties in parent material assignments, bulk density, and rock fragment quantity. The mass of cations in soil samples and parent material was determined by plama emission spectroscopy on a borate fusion. The mass of Zr was measured using X-ray fluorescence.

The mass loss of each cation is converted to moles of positive charge which is summed for each profile. For the moist temperate grassland soils, we assume that all the positive charge is balanced by negative charge from bicarbonate and calculate the mass of carbon removed by leaching waters. This is justified because with the exception of small amounts of NO_3^-, HCO_3^- provides the charge balance to cations in the leaching waters because there are no

significant mineral sources for Cl^- or SO_4^{2-} in the beach sand (Merritts et al. 1992) and local atmospheric deposition is charge-balanced (Kennedy & Malcolm 1978). Our measurements and calculations quantify net long-term changes as opposed to yearly infiltration of elements, thus we do not subtract yearly inputs as part of the budget.

Physiography of study areas

To quantify rates of long-term atmospheric carbon sequestration in a grassland ecosystem, we sampled nine soil profiles on seven marine terraces at the mouth of the Mattole River in northern California. Because of dynamic interplay between glacio-eustatic sea-level fluctuation and tectonic uplift in the lee of the Mendocino triple junction, a sequence of emergent Quaternary shore platforms are notched and preserved in the easily abraded graywackes of the Franciscan coastal terrane. Ages of the seven terraces span the past 240 ky and were obtained from two methods, radiometric dating and altitudinal spacing analysis. The youngest terrace is assigned an age of 3.9 ka (+0.1/–3.9 ka) on the basis of C-14 dating of shells found in original growth position. The remaining terraces are assigned inferred ages of 29, 40, 118, 124, 214, and 240 ka on the basis of correlation with dated worldwide glacio-eustatic sea-level highstands (Merritts & Bull 1989, Merritts et al. 1992). Error estimates are +5/–15 ka for the 29- to 124-ka terraces and +15/–20 ka for the 214- and 240-ka terraces (Merritts et al. 1992).

The northern California coastal region has a cool, temperate Mediterranean climate; mean annual temperature is about 13 °C; mean annual precipitation is about 1 m. Climatic variations between interglacial and glacial periods are minimized by maritime influences on temperature (CLIMAP 1981; Merritts et al. 1992). Full glacial precipitation may have been greater than at present because of steepened north-south temperature gradients. Present vegetation is prairie bunch grass; the invasion of coniferous trees is prevented by wind (Major 1977). Full glacial vegetation may have been coniferous trees (Johnson 1977), but the soil profiles provide little evidence to support this possibility.

The beach sand parent material is dominated by equal amounts of quartz and plagioclase derived from Franciscan complex argillaceous sandstone. Over 240 ky, weathering reduced plagioclase form 450 kg m^{-3} to about 100 kg m^{-3}; Fe- and Mg-chlorite and mica were weathered also. These reactions released 70–80% of the Na^+ and Ca^{2+}, and 20–30% of the K^+ and Mg^{2+} to leaching waters (Chadwick et al. 1990).

To quantify long-term rates of atmospheric carbon sequestration in a desert scrubland ecosystem, we sampled twelve soil profiles on nine glaciofluvial terraces along the Wind River in west central Wyoming. Because of the dynamic interplay among glaciofluvial sediment flux, erodable lithologies, and epiorogenic uplift of the Rocky Mountain region, extensive sequences of river terrace remnants are preserved in the basins of Wyoming (Reheis et al. 1991). In the Wind River basin, fifteen terrace levels are mapped whose ages span the past 1,740 ky and were obtained from three methods, radiometric

dating, tephrochronology, and incision rate modelling. Soil profiles were sampled from the following terraces (Chadwick et al. 1993): WR 1 (10 ± 2 ka), WR 3 (100 ± 20 ka), WR 4 (130 ± 26 ka), WR 8 (870 ± 170 ka), WR 9 (950 ± 190 ka), WR 11 (1090 ± 220 ka), WR 12 (1110 ± 220 ka), WR 13 (1320 ± 260 ka), WR 15 (1740 ± 250 ka). Error estimates are placed conservatively at ± 20% based on worst case scenarios about incorrect field relationships.

The Wind River basin has a cold, temperate, semiarid climate; mean annual temperature is about 6 °C and the mean annual precipitation is about 22 cm. Climatic variation between interglacial and glacial periods has been severe. Mean annual temperature during the last glaciation was at least 10 °C colder than present (Mears 1981), annual precipitation may have been less than present (Whitlock & Bartlein 1993,), and effective soil moisture was similar to present (Nettleton & Chadwick 1991). Soil moisture seldom infiltrates below 2 m in the soil profiles sampled for this study. Present vegetation types are wheatgrass-needlegrass shrubsteppe and sagebrush steppe (Kuchler 1970). Glacial plant assemblages may have included sagebrush and tundra herbs (Whitlock & Bartlein 1993).

Soil parent material is composed of mixed volcanic and crystalline lithologies overlain by about 50 cm of fine-textured eolian material derived from diverse lithologies. In contrast to the grassland chronosequence, atmospheric deposition is a significant contributor of soil cations (dominated by Ca) (Reheis 1990; Gillette et al. 1993) that precipitate to form carbonate in the subsoil. These carbonates provide a well preserved record of inorganic carbon accumulation during pedogenesis.

Results and discussion

Carbon sequestration during soil evolution

For the moist grassland soil profiles, cumulative conversion of atmospheric carbon dioxide to bicarbonate increases in a curvilinear fashon and begins to level off after carbon transfer of 10 to 15 kg m^{-2} (Fig. 1A). The long-term weathering-induced carbon flux (Fig. 1B) ranges from 0.11 g m^{-2} yr^{-1} for relatively recent soil profiles to about 0.06 g m^{-2} yr^{-1} for the oldest soil profiles. The flux decrease is due to exhaustion of easily weatherable minerals (Chadwick et al. 1990). Conversion of atmospheric carbon dioxide to organic carbon in the grassland soil profiles levels off after cumulative carbon transfer of about 150 kg m^{-2} at about 120 ky (Fig. 1C). Initial net carbon flux into organic matter is about 7.4 g m^{-2} yr^{-1} (Fig. 1D), but fluxes decrease rapidly and in the oldest soil profiles are about 0.5 g m^{-2} yr^{-1}. Although these soils store more than average amounts of carbon, the organic carbon fluxes are within the range of published values for Holocene soils (Schlesinger 1990).

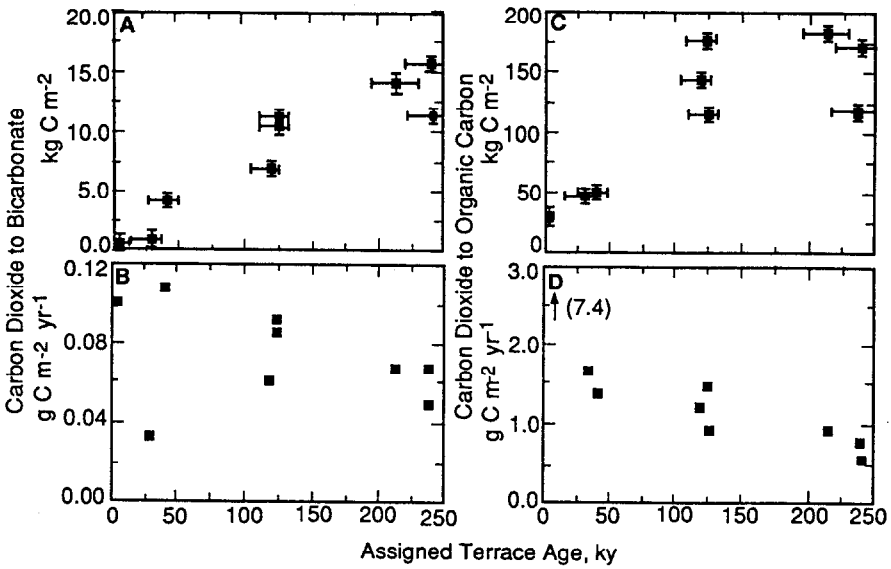

Fig. 1. Amount (A) and flux (B) of bicarbonate carbon leached from, and amount (C) and flux (D) of organic carbon accumulated in the chronosequence of moist temperate grassland soils from the mouth of the Mattole River, California. Y-axis values were integrated over 2 to 3 m sampling depths, and are presented on a whole-soil basis with error estimates based on uncertainties in parent material assignments, bulk density, and rock fragment quantity. Marine terrace ages and error bars are based on radiometric dating and altitudinal spacing analysis. Inherent in flux determinations from chronosequences is that each successively older soil contains the cumulative record of earlier, often larger fluxes that tend to overestimate flux for older soils.

For the desert scrubland soil profiles, cumulative mass of carbonate carbon reaches about 25 kg m^{-2} after 1,700 ky (Fig. 2A). Minimal weathering of silicate minerals (Nettleton & Chadwick 1991) and nearly linear carbonate accumulation suggests relatively constant long-term atmospheric deposition at the soil surface of calcium carbonate and dissolved Ca (Gillette et al. 1992; Reheis 1990). The gross long-term carbon flux (not corrected for external carbonate input) ranges from 0.02 to 0.035 g m^{-2} yr^{-1} for recent soil profiles to 0.015 g m^{-2} yr^{-1} for the oldest soil profiles (Fig. 2B). For comparison, values range from 0.1 to 0.6 g m^{-2} yr^{-1} for 8 calcic-soil chronosequences in warmer parts of the southwestern United States (Machette 1985) and about 0.1 to 1.0 g m^{-2} yr^{-1} for calcic soils in Arizona (Schlesinger 1985). The amount of carbon actually lost from the atmosphere is more difficult to evaluate because much of the atmospheric deposition of Ca is spatially variable and comes from calcite derived from continental sources (Gillette et al. 1992; Harden et al. 1991). Using $^{87}Sr/^{86}Sr$ of parent silicate minerals and aerosolic input we have shown that only 5 to 10% of the carbonate accumulation in arid and semi-arid soils represents CO_2 consumption due to silicate mineral weathering (Capo & Chadwick 1993). For the rest of the data discussion in this paper,

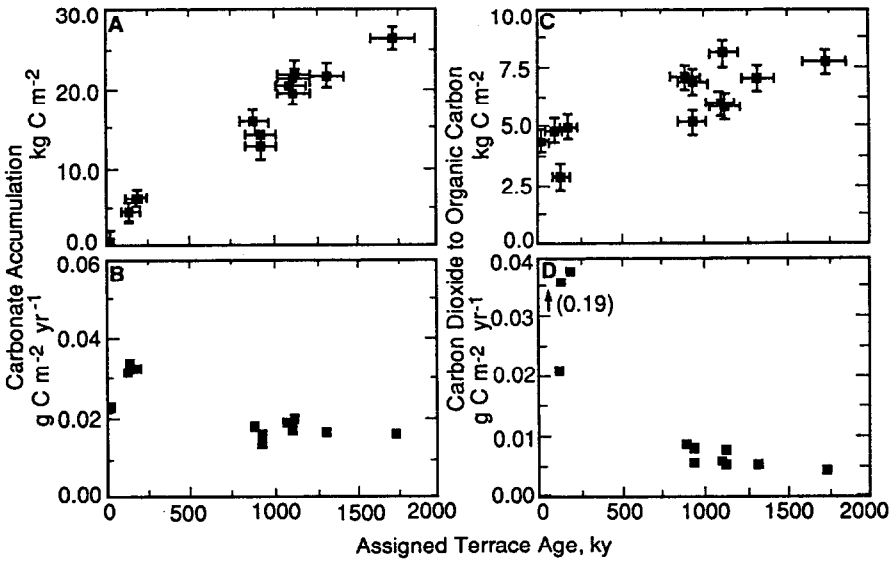

Fig. 2. Amount (A) and flux (B) of carbonate carbon and amount (C) and flux (D) of organic carbon accumulated in the chronosequence of desert scrubland soils from the Wind River Basin, Wyoming. Carbonate was measured directly, integrated over 2 to 3 m sampling depths, and presented on a whole-soil basis with error estimates based on uncertainties in parent material assignments, bulk density, and rock fragment quantity. Fluvial terrace ages and error bars are based on interpolation and extrapolation from tephra-dated terraces and correlation to glacial deposits.

we assume that only 10% of the carbonate-C measured in these profiles is derived from silicate mineral weathering. Cumulative conversion of atmospheric carbon dioxide to organic carbon results in small amounts of carbon transfer with maximum values of about 7.5 kg m^{-2} (Fig. 2C). Initial carbon flux into soil organic matter is 0.19 g m^{-2} yr^{-1} which is similar to published values for Holocene arid-region soils (Schlesinger 1990), but for the oldest soil profiles the flux decreases to about 0.005 g m^{-2} yr^{-1} (Fig. 2D).

In both ecosystems, carbon movement from the atmosphere to soil is partitioned into dominance of organic accumulation early in a soil's history with silicate mineral weathering becoming more important as the soil matures (Fig. 3). In young moist temperate grassland soils, the rate of weathering-induced sequestration is about 2% of that due to organic matter accumulation; for older soils, it is somewhat greater than 8%. In relatively young desert scrubland soils, the rate of inorganic carbon sequestration due to weathering is also about 2% of that due to organic matter accumulation; but for older soils it increases steadily to nearly 40%. In both environments, 240 ky soils have about the same ratio of carbon sequestered by silicate weathering relative to carbon stored in soil organic matter (≈ 8–12%). In desert scrubland ecosystems, there is less weathering and less organic C accumulation.

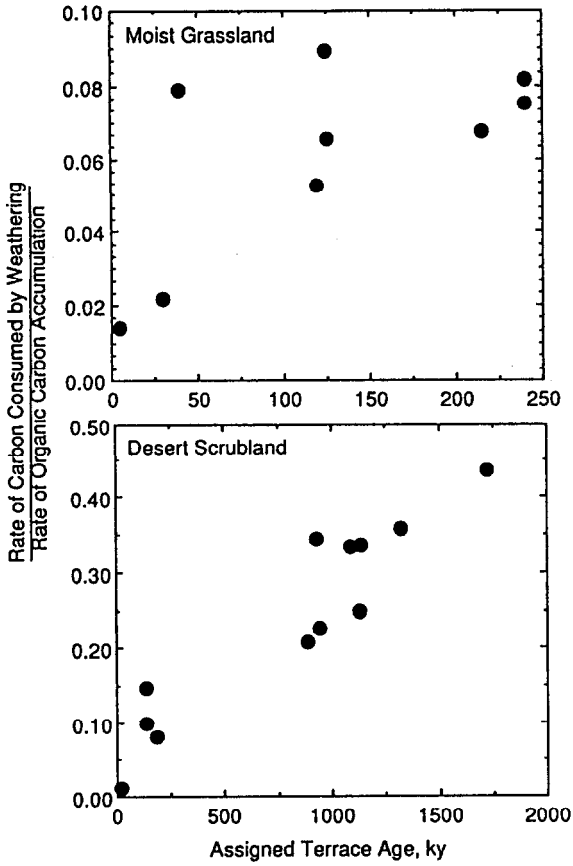

Fig. 3. Ratio of carbon flux due to silicate mineral weathering to carbon flux due to organic accumulation as a function of soil age.

Carbon sequestration in an ecosystem context

Although the carbon cycle is dominated by large, biologically controlled yearly throughput, it is the small soil-mediated fluxes that are key to understanding the ultimate magnitude and direction of carbon movement. Figure 4 summarizes our understanding of both short-term and long-term carbon-cycle fluxes. Moist temperate grasslands cover about 5×10^{12} m^2 or 3% of continental surfaces and on average have an annual net primary productivity (NPP) of about 540 g C m^{-2} yr^{-1} (Ajtay et al. 1979). Desert scrublands cover about 21×10^{12} m^2 or 14% of continental surfaces and on average have no NPP of about 100 g C m^{-2} yr^{-1} (Ajtay et al. 1979). Using these published NPP values as representative of our sites, we may calculate the percent carbon partitioned into soil organic matter and bicarbonate/carbonate for each

ecosystem. Chronosequence end-members are used because intervening soil profiles have intermediate values. It suggests the following interpretations: (1) more than 98% of average yearly NPP is released to the atmosphere by soil respiration, (2) as soils age, less carbon is diverted from the annual photosynthesis/respiration cycle into the long-term cycle – this implies that highly weathered soils such as Ultisols and Oxisols play a relatively insignificant role in carbon sequestration, (3) as soils age, more atmospheric carbon is removed by weathering and bicarbonate leaching and less by organic matter accumulation – the same point as shown by Fig. 3, and 4) as a function of NPP, the amount of atmospheric CO_2 sequestered by desert scrubland ecosystems is an order of magnitude less than for the grassland ecosystem.

For comparison, we constructed Fig. 5 from mass balance data for three post-glacial, moist temperate deciduous forest soils developed in arkosic sediments in northeastern New York and Vermont (using data for the Adams, Becket, and Houghtonville soils in Jersak 1991 and Jersak et al. 1994). Overall, the flux of CO_2 into these soils is similar to the grassland soils, but organic carbon accumulation is less and the weathering controlled carbon flux is slightly greater that for the grassland soils. It is logical that each soil-ecosystem should have specific process-controlled carbon sequestration relationships. However, it should be recognised that the NPP values used in the preceding evaluation are global averages that may vary by up to a factor of 5 (Ajtay et al. 1979). Although our chronosequence selections were meant to minimize climate-change driven changes in ecosystems, we cannot quantify a time-weighted NPP for each of the soil profiles.

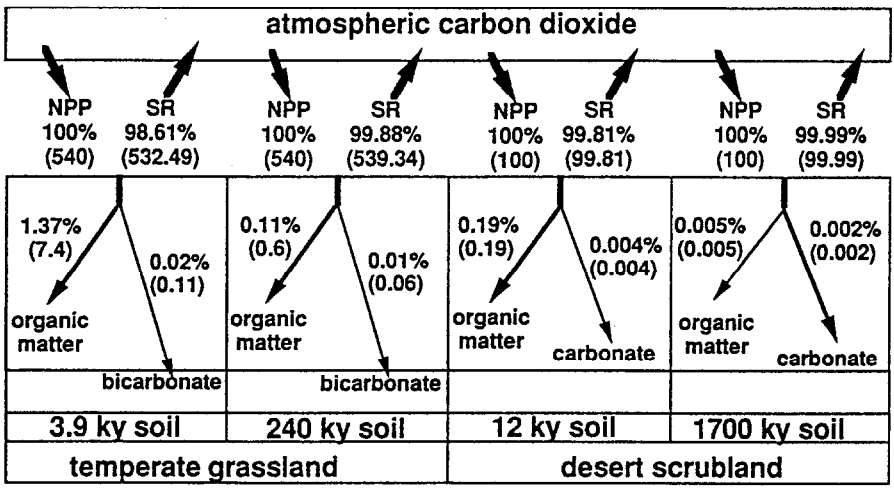

Fig. 4. Flux diagram showing net yearly carbon transfers among atmosphere, soil organic matter, and bicarbonate/carbonate (after correction for eolian input) for chronosequence end-member soils in moist temperate grassland and desert scrubland ecosystems. Values in parentheses are given in g C m^{-2} yr^{-1}. Soil respiration (SR) values are based on the difference between net primary production (NPP) and soil-derived carbon numbers.

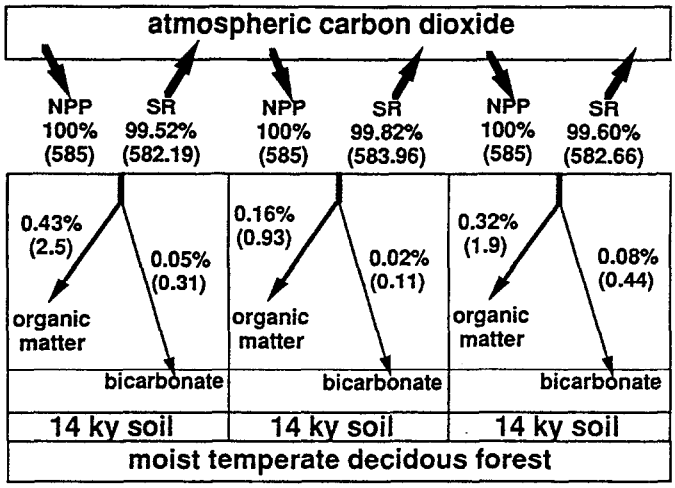

Fig. 5. Flux diagram for three postglacial soils from moist temperature deciduous forests in northeastern New York and Vermont (Adams (left), Becket (center), and Houghtonville (right)). Values in parentheses are given in g C m^{-2} yr^{-1}. Soil respiration (SR) values are based on the difference between net primary production (NPP) and soil-derived carbon numbers.

Conclusions

We have demonstrated clear relationships between net long-term soil carbon sequestration, and soil age and soil-ecosystem properties that are the logical result of the interaction of carbon fixation by ecosystems and the susceptibility of the geologic substrate to weathering. It is possible that regional to global estimates of net soil carbon sequestration can be derived based on these relationships. However, we have not yet addressed the extent of areal variation in long-term carbon sequestration during soil development as a function of vegetation-soil-landscape relationships. For instance, soils developing in mafic and carbonate lithologies or in catenary sequences probably have different relationships between NPP, weathering, and carbon sequestration. These sources of regional variability in long-term carbon sequestration need to be assessed further by sampling along carefully defined lithological, climatological, topographical, and ecological gradients.

Acknowledgements

We thank Susanne Anderson, Robert Berner, Rosemary Capo, Roberto Gwiazda, Jennifer Harden, William Schlesinger, Susan Trumbore, and two anonymous reviewers for comments on this paper. David Hendricks, Chuck Peacock, Elise Pendall, and Bud Burke and the Humboldt State Diggers

provided critical laboratory and field assistance. The research described in this paper was carried out by the Jet Propulsion Laboratory, California Institute of Technology on contract to NASA's Geology Program, Mission to Planet Earth.

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