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GLOBAL PRODUCTION AND DECAY OF RADIOCARBON

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ABSTRACT. The production rate of ^{14}C during the Holocene averaged 2.4 ± 0.2 atoms $^{14}C/cm_e^2$ sec. Neutrons produced by galactic cosmic rays account for 90% of the ^{14}C production with the remaining 10% resulting from neutrons produced by protons from solar flares. Production and decay of ^{14}C can be reconciled by including ^{14}C permanently or temporarily stored in sediments. Sedimentary reservoirs contain ca 30% of all terrestrial ^{14}C. The lagoons, bays, marshes and deltas of the coastal wetlands alone account for 12% of the ^{14}C inventory. The capacity of the coastal wetlands to store carbon has become the subject of renewed interest.

PRODUCTION OF RADIOCARBON

^{14}C is produced by the secondary neutron flux resulting from spallation of atmospheric isotopes. These neutrons are almost quantitatively involved in interaction with ^{14}N to produce ^{14}C plus tritium in the reactions $^{14}N(n,p)^{14}C$ and $^{14}N(n,T)^{12}C$. There were seven different calculations of the production rate of ^{14}C during the decade 1970–1980. The estimated production rates for the 1965 solar minimum and 1969 solar maximum are shown in Table 1. Six of the calculations are in good agreement and average 2.47±0.19 ^{14}C atoms/s cm_e^2 for solar minimum and 2.02±0.13 ^{14}C atoms/s cm_e^2 for solar maximum. Surprisingly, the lowest estimate by O'Brien (1979) is commonly used in modeling ^{14}C in the carbon cycle (eg, Stuiver & Quay, 1980) because it agrees with the decay rate as estimated from ^{14}C inventories (eg, Siegenthaler, 1985). Siegenthaler calculated a decay rate of 1.73 dps cm_e^{-2}. We obtained a higher value, 2.01 dps cm_e^{-2}, and suggested that the global decay rate was compatible with the higher estimates of the production rate (Damon, Sternberg & Radnell, 1982). The difference lies in the amount of ^{14}C estimated to be in the sedimentary reservoir. According to Siegenthaler, Heimann and Oeschger (1980), "sedimentation is neglected as well as other minor fluxes, because it removes only ca 0.5×10^{-5} parts of the total oceanic amount of carbon (or ^{14}C) per year." However, ^{14}C accumulates in sediments during its mean life of 8270 years and it can be shown that sediments contain a significant part of the total global inventory.

Past changes in atmospheric ^{14}C concentrations ($^{14}C/C$) are evaluated by measuring the carbon isotopic content of dendrochronologically dated tree rings. The ^{14}C concentration is corrected for radioactive decay and isotopic fractionation and compared with mid-19th century wood. The per mil (‰) difference is referred to as $\Delta^{14}C$. The trend of $\Delta^{14}C$ excluding short-term fluctuations can be approximated by a sinusoidal curve with a period of ca 11,000 years back to 6500 BC (Bruns *et al*, 1983; Damon & Linick, 1986) but may deviate from a sinusoidal curve prior to that time (Kromer *et al*, 1986). Since the Twelfth Nobel Symposium held in Uppsala, Sweden in August 1969, this

long-term trend of $\Delta^{14}C$ has been attributed to secular variation of the strength of the geomagnetic dipole moment resulting in changes in the rate of production of ^{14}C (Olsson, 1970). ^{14}C integrates changes in production rate resulting from changes in the dipole moment on a global basis whereas individual archaeomagnetic measurements are strongly affected by the non-dipole components. Generally, it appears that the dipole moment field strength was lower than at present prior to the 5th century BC and higher afterwards (McElhinny & Senanyake, 1982). Thus, the ^{14}C production rate, Q(t), would have been higher prior to the 5th century BC and lower more recently. The global ^{14}C inventory can be calculated from the following relationship (Sternberg & Damon, 1979; Damon, Sternberg & Radnell, 1982):

$$I = -\lambda \int_{\infty}^{0} Q(t)\, e^{-\lambda t}\, dt \qquad (1)$$

where I (inventory) is the global decay rate, λ is the ^{14}C decay constant, and Q(t) is the production rate as a function of time, t. Q(t) is proportional to $M(t)^{-\alpha}$, where $\alpha=0.52$ and M(t) is the time-varying dipole field strength. The function $M(t)^{-\alpha}$ is asymmetric. A decrease in the dipole moment has a much greater effect than a corresponding increase (Sternberg & Damon, 1979, see Fig 12). Thus, fortuitously, the higher earlier production rates offset the more recent lower production rates and the calculated inventory I is not significantly different than the average production rate by galactic cosmic rays during recent solar cycles which is ca 2.2 ^{14}C aps cm_e^{-2} or 132 apm cm_e^{-2} (Table 1).

TABLE 1

Comparison of ^{14}C production calculations (atoms/sec cm_e^2).

Year	Solar max+min	Merker (1970)	L&R* (1970)	Light *et al* (1973)	Povinec (1977)	O'Brien (1979)	K&M* (1980)	C&L* (1980)
1965	15.1	2.15	2.40	2.42	2.6	1.91	2.58	2.66
1969	105.6	1.86	2.18	1.93	2.1	1.60	1.93	2.12

*L&R: Lingenfelter and Ramaty
K&M: Korff and Mendell
C&L: Castagnoli and Lal

DECAY OF RADIOCARBON

There is general agreement concerning the ^{14}C inventory of preanthropogenic ambient reservoirs, *ie*, reservoirs with transfer times that are much less than the half-life of ^{14}C (5730 years). The inventory for ambient reservoirs is given in Table 2. Note that the summed inventory is ca 20% less than the current average production rate. If the model from which I in equation 1 was estimated is correct, the difference must lie either in the calculation of production rates or in a missing component of the inventory. The missing component is the controversial sedimentary reservoir.

Much attention has been given in recent years to storage of carbon in sediments because of the need to account for the "missing" anthropogenic carbon resulting from the combustion of fossil fuels and agricultural practices such as the clearing of forests. I have taken advantage of this flourishing lit-

TABLE 2

^{14}C inventory of pre-anthropogenic ambient reservoirs

Ref*	Reservoir	C in reservoir (g/cm_e^2)	^{14}C activity of reservoir (dpm/gC)	Reservoir decay rate (dpm/cm_e^2)
1	Atmosphere	0.116±0.002	14.1±0.5	1.64±0.06
2	Terrestrial biosphere (rapid, vascular, heterotroph and litter)	0.172±0.020	13.6±0.6	2.34±0.29
2	Soil humus	0.393±0.029	13.2±0.6	5.19±0.45
2	Hydrosphere, mixed layer of ocean (0-75m)	0.122±0.005	13.4±0.5	1.63±0.09
2	Hydrosphere, intermediate and deep ocean	7.420±0.030	11.8±0.5	87.56±3.73
2	Saprosphere (total organic matter in oceans)	0.274±0.078	13.3±0.5	3.64±1.05
3	Fresh surface water	0.0002±0.001	13.0±0.6	0.03±0.02
		Σ = 8.50±0.14		Σ = 102±4

*1. Oeschger *et al* (1975), 280±5 ppm; 2. Olson *et al* (1985); 3. Kemp (1979)

erature to estimate the ^{14}C inventory of sediments (Table 3). This estimate differs from our earlier estimates in the addition of organic matter in the coastal wetlands which are now considered to be as important as the marine carbonate reservoir. Much of the carbon storage in this reservoir is in the form of macrophyte (large marine plants such as eel grass, kelp and intertidal marsh grasses). A higher fraction of macrophyte than phytoplankton escapes oxidation and is buried in marsh and other coastal sediments (Baes, Björkstrom & Mulholland, 1985) or transported to the shelf area (Walsh, 1984). Carbon that is rapidly cycled is not included in the inventory. Table 3 only includes carbon that is stored for a sufficiently long time to allow most of the ^{14}C to decay.

TABLE 3

^{14}C inventory in buried sediment (long-term storage)

Ref*	Reservoir	C accumulated during mean life of ^{14}C (g/cm_e^2)	^{14}C activity of C flux (dpm/gC)	Reservoir decay rate (dpm/cm_e^2)
1	Continental shelves (carbonate)	0.033±0.016	13.4±0.6	0.44±0.21
1	Continental slopes and deep ocean (carbonate)	0.853±0.178	13.4±0.6	11.43±2.86
2	Continental shelves (organic)	0.178±0.089	13.4±0.6	2.38±1.20
2	Continental slopes and deep ocean (organic)	0.026±0.013	13.4±0.6	0.35±0.18
3	Coastal wetlands (lagoons, marshes, bays and deltas)	1.135±0.378	13.2±0.6	14.98±5.95
4	Freshwater lakes and wetlands	0.24±0.03	13.4±0.6	3.22±0.42
		Σ = 2.46±0.42		Σ = 32.86±6.7

*1. Hay (1985); 2. Berner (1982); 3. Baes *et al* (1985); 4. Kemp (1979)

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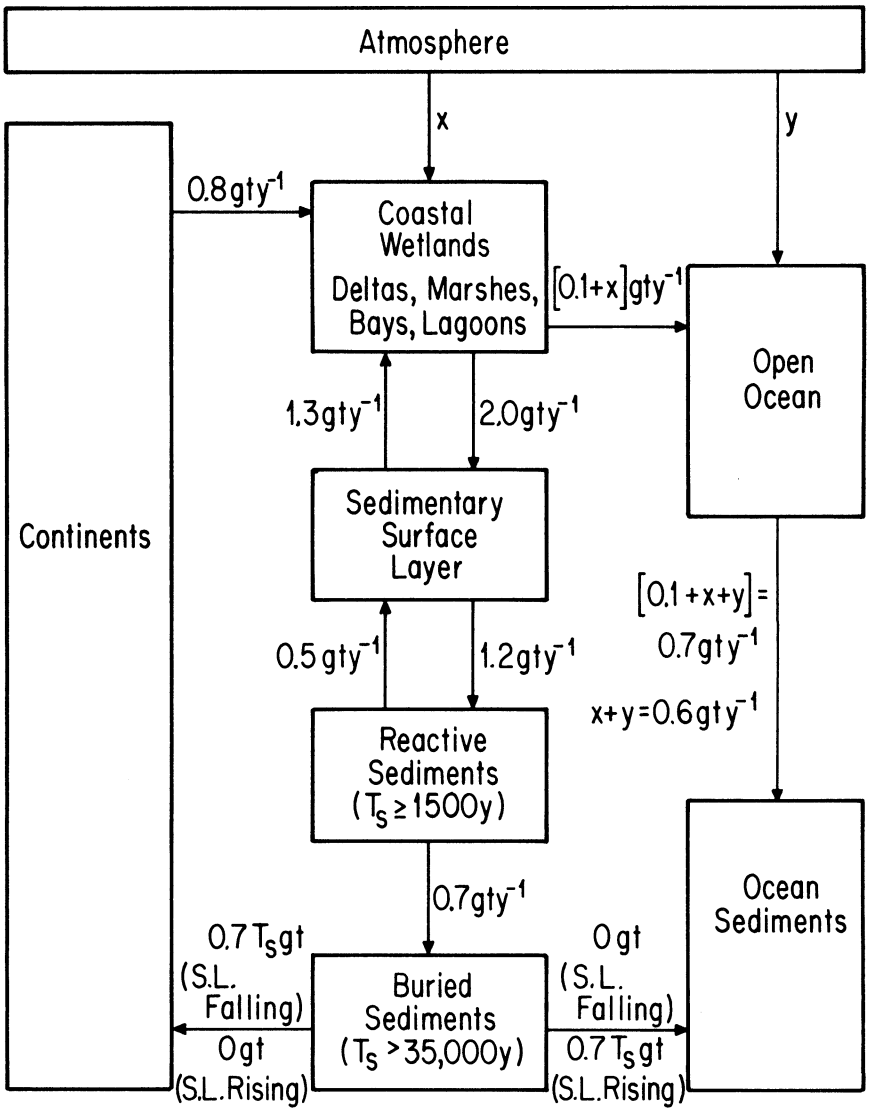

Fig 1. Marine carbon fluxes. The fluvial input of carbon is augmented by photosynthesis. Ca 40% of the flux to the surface sediments is rapidly recycled with negligible decay of ^{14}C. Ca 25% is recycled from the reactive sediments after appreciable decay of ^{14}C and ca 35% is transferred to the buried sediments where all ^{14}C decays. Falling sea level may return coastal sediments to land whereas rising sea level shifts the coastal sediments to the open ocean. Part of the coastal wetland carbon flux is transferred to the open oceans where further augmentation by photosynthesis takes place. Carbon in the open ocean goes through a similar sequence of stages, not shown here, before a similar amount is permanently stored long enough for all ^{14}C to decay.

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The amount of carbon stored temporarily and returned to the ambient reservoirs before all ^{14}C has decayed is even more difficult to evaluate. From Table 3, (column 3) ca 0.7 g t y^{-1} is stored in the wetlands and approximately the same amount is stored in ocean sediments long enough for ^{14}C to decay. The amount of carbon entering the oceans is ca 0.8 g t y^{-1} (Baes, Björkstrom & Mulholland, 1985). Figure 1 shows some features of the fluvial flux, ocean, atmosphere and sediment cycle. The fluvial flux enters the wetlands carbon cycle, depositing a large amount on the sedimentary surface annually. Part of this is rapidly returned to the aqueous phase with negligible ^{14}C decay. Another part is stored temporarily in reactive sediments but long enough for significant ^{14}C decay and the remainder is deposited permanently, *ie*, long enough for all ^{14}C to decay. CO_2 also enters the ocean through photosynthesis. Part of the carbon passes into the open ocean where photosynthesis fixes more carbon and sediments accumulate on the sea floor. An attempt at balancing the cycle is given in Figure 1. Falling sea level yields sediments to land for recycling and rising sea level transfers buried sediment to the open ocean.

TABLE 4

^{14}C inventory of reactive sediments

Ref*	Reservoir	C in reservoir (g/cm_e^2)	^{14}C activity of reservoir (dpm/gC)	Reservoir decay rate (dpm/cm_e^2)
1	Continental paleosols, peat and sediment	0.100±0.030	10.7±0.8	1.07±0.33
1	Marine organic	0.127±0.040	4.9±0.8	1.32±0.46
2	Marine carbonate	0.490±0.160	5.5±0.8	2.74±0.96
2,3,4	Coastal wetlands	0.196±0.065	11.0±0.8	2.16±0.73
		Σ = 0.913		Σ = 8.38±1.35

*1. Sundquist (1985); 2. Baes *et al* (1985); 3. Wollast and Billen (1981); 4. Mulholland (1981)

TABLE 5

Distribution of ^{14}C in terrestrial nature

Reservoir	Decay rate (dpm/cm_e^2)	Relative amount (%)
Hydrosphere (mixed, intermediate and deep ocean)	89.2	62.4
All other ambient reservoirs	12.8	9.0
Coastal wetlands (lagoons, bays, marshes and deltas)	15.0	10.4
Reactive sediments	8.4	5.8
Carbonate in marine sediments	11.9	8.2
All other sediments	6.0	4.2
		100.0
Σ = 143±8 dpm/cm_e^2		

As mentioned above, the reactive sediments were not included in Table 3. An estimate is given in Table 4. Addition of this component increases the sedimentary ^{14}C storage by 25%.

Table 5 summarizes the distribution of ^{14}C in terrestrial nature. The marine hydrosphere is the largest reservoir, including 62.4% of all ^{14}C. All other

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ambient reservoirs include an additional 9.0%, leaving 28.6% of the total in the sedimentary reservoirs. The sum is not significantly higher than the estimated average production rate by galactic cosmic rays. Nevertheless, solar flares also produce a significant amount of ^{14}C (Lingenfelter & Ramaty, 1970; Damon, Cheng & Linick, 1989) on the order of 10% of the galactic component. Thus, a total ^{14}C production rate of 143 dpm/cm$_e^2$ (Table 5) would not be unreasonable. If this is added to the galactic component, a total ^{14}C production rate of 2.4 aps cm$_e^{-2}$ is obtained.

CONCLUSIONS

The following conclusions seem to be warranted by the available data:

1) The average rate of ^{14}C production during recent sunspot cycles is 2.4 ± 0.2 ^{14}C atoms/cm$_e^2$ sec. Production rates are 90% by galactic cosmic rays and ca 10% by solar flare protons. This is also close to the average production rate during the Holocene calculated from equation 1.

2) This rate of production can be reconciled with the inventory of ^{14}C decay in terrestrial reservoirs if ^{14}C stored permanently or temporarily in buried sediments is taken into consideration. Reconciliation is required because atmospheric ^{14}C has not deviated from steady state by more than ± 5% during the last 14,000 years (Damon & Sonett, 1989). The total inventory is 2.38 ± 0.13 ^{14}C atoms/cm$_e^2$ sec.

3) Recent work has demonstrated that the lagoons, bays, marshes and deltas of the coastal wetlands constitute a ^{14}C reservoir approximately equal in magnitude to the ^{14}C stored in the sediments of the continental shelves, slopes and deep ocean. Ca 28.6% of terrestrial ^{14}C is stored in sedimentary reservoirs, 62.4% is present in the marine hydrosphere and 9% is stored in all other reservoirs. Future work should include a re-evaluation of the last reservoir in Table 3. Alexander Wilson of this laboratory has suggested that the evaluation by Kemp (1979) does not adequately account for the significant storage of carbon tundra wetlands.

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