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MODELING OF ATMOSPHERIC RADIOCARBON FLUCTUATIONS FOR THE PAST THREE CENTURIES

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INTRODUCTION

Relatively precise quantitative observations of geophysical parameters are available to evaluate the fluctuations of atmospheric ^{14}C activity during the past three centuries. As reviewed by Damon, Lerman, and Long (1978), these fluctuations seem to result from three factors: 1) changes in the earth's dipole magnetic field intensity, which has been decreasing since the first measurements by Gauss (McDonald and Gunst, 1968); 2) solar modulation of the cosmic-ray production, which has been correlated with the sunspot record of Waldmeier (1961), and more recently, to the Aa geomagnetic index by Stuiver and Quay (1980); and 3) the combustion of fossil fuels (Suess, 1955). A relationship between the climatic time series and the ^{14}C-derived record of solar change has not yet been demonstrated (Stuiver, 1980).

To relate ^{14}C fluctuations to geophysical parameters, we must use reservoir models as analogues to the ^{14}C cycle. We have investigated four models of the ^{14}C exchange system (Lazear, Damon, and Sternberg, 1980), a 1-box model (Grey and Damon, 1970), 3-box first-order exchange model (eg, Houtermans, Suess, and Oeschger, 1973), 5- and 6-box first-order exchange models (eg, Bacastow and Keeling, 1973; Ekdahl and Keeling, 1973), and a box-diffusion model (Oeschger et al, 1975). We will concentrate here primarily on the multibox and box-diffusion models that are more or less adequate analogues to nature.

None of the originally parameterized models included a sedimentary sink, which we added to all of the models because it significantly decreases the DC gain and improves their performance as natural system analogues. We suggested that the DC gain (Lazear, Damon, and Sternberg, 1980) and ^{14}C inventory (Sternberg and Damon, 1979) are useful boundary conditions that models and production functions should satisfy to be adequate natural analogues. We make use of these boundary conditions to evaluate both the production function and the amount of carbon entering into the sedimentary reservoir. Walsh et al (1981) indicate that the flux of carbon into the sedimentary sink may

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provide the missing carbon in the global CO_2 cycle required to account for fossil-fuel emission and the pioneer effect (ie, clearance of forests for agriculture).

INVENTORY (I) AND DC GAIN ($\bar{g}$)

One method of determining the total amount of ^{14}C in the geochemical cycle is to inventory the carbon reservoirs. Table 1 is an inventory modified from Damon, Lerman, and Long (1978). The modifications were to reduce the carbon content of the atmosphere to the pre-industrial level and to increase the activity of carbon entering the sedimentary sink to the level of the mixed layer rather than the deep sea because most of the carbon in sediments was derived from the mixed layer rather than the deep sea. The total carbon in the inventory sums to 9.65 g cm_e^{-2} and the decay rate is ca 120 dpm cm_e^{-2}.

TABLE 1. Radiocarbon inventory

Reservoir	C in reservoir (N) g cm_e^{-2}	^{14}C activity* in reservoir (A) dpm g^{-1} C	Decay rate in reservoir dpm cm_e^{-2}
Atmosphere	0.121	14.1	1.85
Terrestrial biosphere	0.108	13.6	1.47
Humus (dead terrestrial organic matter)	0.206	13.5	2.78
Hydrosphere (fresh water)	0.088	8.2	0.72
Hydrosphere (mixed layer of oceans)	0.180	13.6	2.44
Hydrosphere (deep sea)	7.386	12.3	90.85
Biosphere (marine)	0.001	14.2	0.01
Oceans (dead organic matter)	0.588	12.3	7.23
Sediments	0.972	13.6	13.22
	$\Sigma = 9.650$ g cm_e^{-2}	$\bar{A} = 12.3$ (dpm g^{-1})	$\Sigma = 120.5$ dpm cm_e^{-2}
			$= 2.01$ dps cm_e^{-2}

*Modified from Damon *et al* (1978).

The inventory (I) of radiocarbon decay rates in all reservoirs should have been balanced by past production:

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

where Q(t) is the production as a function of time (t), where t is measured from the present to the past, and λ is the decay constant of ^{14}C. This was evaluated by Sternberg and Damon (1979) for a sinusoidal dipole moment using the production function of Lingenfelter and Ramaty (1970) and the assumption that the average long term heliomagnetic activity has remained constant. The result was that I, calculated from equation 1, was ≥ 122 dpm cm_e^{-2} with a most probable value of 124 dpm cm_e^{-2}. This is not significantly different that the value of 120 dpm cm_e^{-2} obtained from the reservoir inventory.

O'Brien (1979) obtained a much lower production function than we calculated using a linear approximation relating Lingenfelter and Ramaty's production data to the Zurich sunspot numbers. The two production functions are:

$$Q = 2.434 - 0.00264\ S\ \text{dps}\ cm_e^{-2} \quad \text{(Lingenfelter \& Ramaty, 1970)} \tag{2}$$

$$Q = 1.937 - 0.00242\ S\ \text{dps}\ cm_e^{-2} \quad \text{(O'Brien, 1979)} \tag{3}$$

These two production functions were evaluated for different years and compared with other ^{14}C production calculations. Light et al (1973) agree with Lingenfelter and Ramaty (1970) and O'Brien's values are ca 30% lower. According to Lingenfelter (pers commun, 1982), O'Brien's calculations are theoretically correct but not adequately related to measurements of the neutron flux. Also, the inventory for O'Brien's production function calcuated from equation 1 yields a value that is ca 20% lower.

Lazear, Damon, and Sternberg (1980) state that the observed DC gain (g_{obs}) is equal to the ratio of the steady state ^{14}C content of the atmosphere ($\bar{N}_a^*$) to the steady-state production rate ($\bar{Q}$) which they approximated by mean values for the observed record and evaluated at 111 ± 22 years.

Using the standard parameters of the authors, we obtained the DC gain of the 3-box, 5-box, and box-diffusion model (table 2).

TABLE 2. Standard model DC gain ($\bar{g}$), steady-state atmospheric ^{14}C activities ($\bar{A}_a^*$), transfer constants (K_{sed}), ratios of mixed layer to atmospheric carbon contents (N_m/N_a), and required flux to the sedimentary sink (ϕ)

Model	$\bar{g}$ yr	$\bar{A}_a^*$ dpm g^{-1}	$\bar{A}_a^{**}$ dpm g^{-1}	K_{sed} yr^{-1}	N_m/N_a	$\phi^\dagger$ $\times 10^{15}$ g yr^{-1}
3-box	151	19.9	14.6	2.07×10^{-1}	1.3	1.66
5-box	150	19.8	14.5	1.28×10^{-3}	2.0	1.58
box-diffusion	142	18.7	13.7	1.73×10^{-3}	1.3	1.39

*Standard model with Lingenfelter and Ramaty (1970) production function and no sedimentary sink

**Standard model with O'Brien (1979) production function and no sedimentary sink

†Flux to the sedimentary sink required to reduce $\bar{g}$ to 111 yr

The model DC gains are high by ca 30%. Consequently, if the Lingenfelter and Ramaty (1970) production function is used, the models generate a high steady-state atmospheric ^{14}C content and high steady-state ^{14}C activities ($\bar{A}_a$):

$$\bar{N}_a^* = \bar{g}\,\bar{Q} \qquad (4) \qquad\qquad \bar{A}_a = \frac{\bar{N}_a^*}{N_a \bar{T}} \qquad (5)$$

where N_a is the pre-industrial atmospheric carbon content and $\bar{T}$ is the mean life of ^{14}C. Table 2 shows that the model-generated ^{14}C activities are unacceptably high (18.7 to 19.9 dpm $g^{-1}C$).

This problem can be rectified by using the lower production function of O'Brien (1979) or by adding a sedimentary sink to the standard models. Table 2 also shows that the O'Brien production function generates acceptable atmospheric ^{14}C activities. We believe this is fortuitous because the carbon inventory demands a sedimentary sink and O'Brien's production function yields a lower inventory and production rates compared to all other calculations. Inclusion of a sedimentary sink predicts sedimentary fluxes that are acceptably close to values required by inventories of the carbon cycle. The carbon in the sedimentary reservoir in table 1 is based on a flux (ϕ) of 0.6×10^{15} g yr^{-1} (Damon and Wallick, 1972). Hay and Southam (1977) estimated the minimum Holocene flux to the sedimentary reservoir at 0.43×10^{15} g yr^{-1} and the maximum Holocene sedimentary flux at 0.86×10^{15} g yr^{-1}. Walsh et al (1981) require a post-industrial flux of 1.8×10^{15} g yr^{-1}, with up to 0.75×10^{15} g yr^{-1} due to increased eutrophication since the industrial revolution, and 1.05×10^{15} g yr^{-1} entering the sedimentary sink prior to the industrial revolution. Thus, the sedimentary sink flux required by the models when evaluated with the Lingenfelter and Ramaty (1970) production function agrees reasonably well with independent estimates. Both Hay and Southam (1977) and Walsh et al (1981) emphasize the importance of the flux to shelf sediments. The latter authors suggest that this flux may account for the missing carbon in the global cycle that is required when the pioneer-effect emission is added to the fossil fuel emission. Use of the DC gain and inventory as boundary conditions also suggests that the surplus carbon produced by the pioneer effect can be accounted for by the flux to the sedimentary sink.

As suggested by Houtermans, Suess, and Oeschger (1973), the DC gain acts as a scaling factor because of its effect on the apparent ^{14}C activity of the atmosphere. Thus, the high ^{14}C activities produced by neglect of the sedimentary sink in table 2 will reduce ^{14}C fluctuations (Δ ‰) by 36% in all three models. We will demonstrate this in the model of the ^{14}C fluctuations observed during the past three centuries.

COMPARISON OF MODELS

We used a general-purpose electrical-circuit analysis program (SPICE, University of California) for evaluation of the 3-box, 5-box, and box-diffusion models and trapezoidal method of numerical integration. The program is convenient and useful for ac, dc, and transient analysis. We accepted the standard parameters used by the authors, modifying only the production function and adding a sedimentary sink when required by the DC gain. We were concerned with only the past three centuries for which sunspot data are available. We used Waldmeir (1961) sunspot numbers (S) as modified for the Maunder minimum by Eddy (1976). Fine tuning of the models was not necessary because performance of the models is insensitive to small changes in initial conditions after the first 100 years; we limited our comparisons to the 19th and late 18th centuries. We raised the atmosphere and mixed layer by ca 1.5% at AD 1650 to simulate the Maunder minimum while leaving the deep sea unchanged. For the box-diffusion model, we also assumed a 1.5% increase for the uppermost part of the ocean immediately below the mixed layer, exponentially decreasing to standard deep sea values with depth. We approximated the variation [Q(t)] resulting from the decreasing geomagnetic dipole field intensity [M(t)] using the geomagnetic field intensities from McDonald and Gunst (1968) and the relationship between Q(t) and M(t) derived by Elsasser, Ney, and Winckler (1956).

Figure 1 shows that the transfer functions are the same below periods of 1000 years for the standard models with high DC gain (~150) as for the models with DC gain adjusted to 111 by adding a sedimentary sink. Figure 2 shows the resulting response of the models with sedimentary sinks to the sunspot record using the Lingenfelter and Ramaty (1970) production function (eq 2). The response curve compares favorably with measured data after the reservoirs adjust to the initial conditions (see Damon, Lerman, and Long, 1978, fig 2; Stuiver and Quay, 1980, fig 5). There is a minimum, eg, at AD 1790 and a maximum at AD 1825 which also occur in the measured data (table 3), but, with a

TABLE 3. Comparison of model predictions with measurement

	Lingenfelter and Ramaty (1970) with sedimentary sink			O'Brien (1979) without sedimentary sink			Measured values Stuiver & Quay (1980)
	3B*	5B	BD	3B	5B	BD	
Min (°/oo) (AD 1790)	-6.26 (-4.52)	-3.22	-3.33	-5.83	-3.23	-2.87	-7
Max (°/oo) (AD 1825)	+0.33 (+0.20)	+0.58	+0.97	+0.27	+0.46	0.94	+3
Peak to peak (°/o)	6.59 (4.52)	3.80	4.30	6.10	3.69	3.81	10 (11)**
Phase lag (yr)	15	14	14	14	14	14	15
Sunspot min (°/oo) (AD 1856)	-0.87 (-0.64)	-0.94	-0.69	-0.71	-0.76	-0.72	?
Sunspot max (°/oo) (AD 1848)	-2.60 (-1.94)	-2.30	-2.40	-2.34	-2.02	-2.28	?
Peak to peak (°/o)	1.73 (1.30)	1.36	1.71	1.63	1.26	1.56	?
Phase lag (yr)	3	2	3	3	2	3	?

*Number in parentheses corresponds to Lingenfelter and Ramaty (1970) production function and 3B model without sedimentary sink

**Number in parentheses is from figure 2 of Damon, Lerman, and Long (1978)

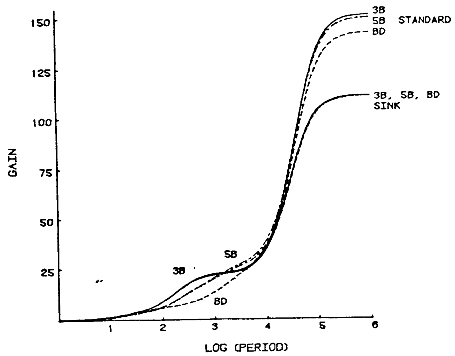

Fig 1. Transfer functions for 3-box (3B), 5-box (5B), and box-diffusion models (BD) with and without sedimentary sink.

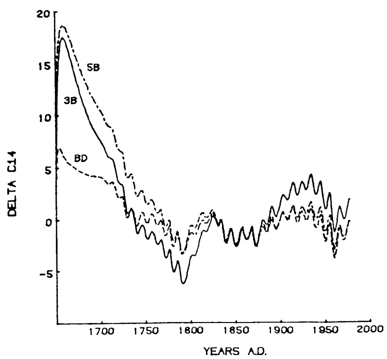

Fig 2. Response of 3B, 5B, and BD models with sedimentary sink to the Lingenfelter and Ramaty (1970) production function.

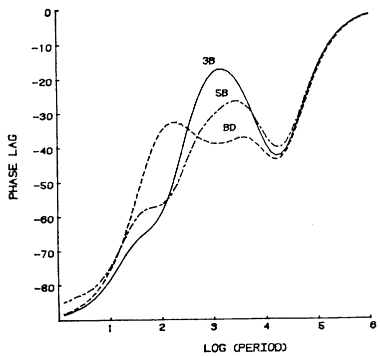

Fig 3. Phase lag of the 3B, 5B, and BD models with sedimentary sink.

greater amplitude. Amplitudes from the models are only 38% to 66% of the measured values but the phase lag corresponds to theory (fig 3). The standard models without sedimentary sink and using O'Brien's (1979) production function yield slightly lower amplitudes. The models also predict a peak-to-peak variation of 1.3‰ to 1.7‰ for the sunspot cycle with a minimum at AD 1856 (S = 4) and a maximum at AD 1848 (S = 125). Annual measurements at ± 2‰ for an entire sunspot cycle should be able to resolve a variation of that magnitude.

If the standard models without sedimentary sink are evaluated with Lingenfelter and Ramaty's (1970) production function, the "wiggles" are further reduced by the expected amount (eg, see the numbers in parentheses for 3B model in table 3. However, unreasonably high steady-state ^{14}C activities are generated compared to natural values (table 2) and initial ^{14}C activities must be increased to match the steady-state values. If the modeler is not aware of the high steady-state ^{14}C activities generated by the model and inputs reasonable ^{14}C activities for the natural environment, the ^{14}C activity and Δ values will start low and steadily climb to the steady-state value. The modeler may then be tempted to avoid the catastrophe of apparent atmospheric ^{14}C buildup by lowering the production rate to values that correct the problem at the expense of unacceptably low production rates relative to the natural environment. The modeler might also ignore the importance of the sedimentary sink in reducing the DC gain and, hence, the steady-state atmospheric ^{14}C activity. Thus, an important source of information would be lost.

DISCUSSION AND CONCLUSIONS

O'Brien's (1979) production function yields rates of production that are low compared to other independent estimates. It also yields an inventory that is 20% lower than that obtained by summing the disintegration rates in all reservoirs (table 1), whereas Lingenfelter and Ramaty's (1970) production function predicts an inventory (124 dpm cm_e^{-2}) very close to the estimate from table 1 (120 dpm cm_e^{-2}). We did not include ^{14}C dissolved in the water content of sediments in table 1 (Lazear, Damon, and Sternberg, 1980) which would raise the inventory and provide a source of ^{14}C-depleted carbon resulting from diagenesis and consequently, lower the ^{14}C activity of deep water. O'Brien's production function does not require a sedimentary sink to lower the DC gain. This further suggests that his estimates of ^{14}C production are too low because independent estimates demonstrate that the flux of carbon to sediments is very significant and, indeed, may account for the "missing" carbon in the carbon cycle.

In contrast, the production function that we obtain from Lingenfelter and Ramaty (1970) agrees reasonably well with estimates other than O'Brien's (1979) and predicts an acceptable inventory. Coupled with a sedimentary sink, it produces a reasonable response with standard values in qualitative agreement with measurements. Both production functions predict amplitudes for "wiggles" that are lower than indicated by measurements. This may be a difficiency in the models as natural analogues (Lazear, Damon, and Sternberg, 1980) or an inadequacy of a production function based on a sunspot vs neutron flux relationship derived solely from three 20th-century sunspot cycle (Stuiver and Quay, 1980). The one-box model does predict the correct amplitude for the AD 1790 - AD 1825 "wiggle" because its gain is 1.5 times the gain of the 3B model at that frequency. However, this is fortuitous because the 1B model is not an adequate natural analogue and it would predict gains that are too high for periods above 200 years.

Based upon experience with multibox models, we do not expect that reasonable manipulation of the model parameters will yield a sufficiently high gain at periods between 50 and 200 years. Rather, as Stuiver and Quay (1980) suggest, a production function based upon modulation during the 11-year cycle is probably not adequate for longer periods. Thus, the solar modulation process, like the ^{14}C reservoir system, may also be acting as a low pass filter or, perhaps, it is affected by cosmic-ray drift in the interplanetary field (Shea and Smart, 1981; Jokipii, 1981). Shea and Smart (1981) showed that the correlation between the Mt Washington neutron monitor counting rate and the geomagnetic Aa index significantly changes from one solar cycle to the next. Jokipii (1981) demonstrated that cosmic-ray drifts can produce the sense of the observed shift in correlation. Also, Stuiver and Quay (1980) suggest modulation of cosmic-ray intensities probably continues beyond zero sunspot number.

The Lingenfelter and Ramaty (1970) production function does predict an inventory and a sedimentary flux that are in reasonable agreement with independent estimates. Thus, their average production rate for the three 20th-century solar cycles, when corrected for past changes in the geomagnetic dipole field intensity (Sternberg and Damon, 1979), appears also to be about the average for the past eight millennia. If so, the ^{14}C flux to the sedimentary sink may account for all or a large part of the missing anthropogenic carbon.

An 11-year cycle in ^{14}C must exist and should be measurable at the 2‰ (sd) precision level. Failure to observe it may be due either to measurement error, or, most probably, to a combination of geographic effects, suggested by Baxter and Farmer (1973 and Damon (1982), solar flare production of ^{14}C (Lingenfelter and Ramaty, 1970), and the annual injection of stratospheric ^{14}C into the troposphere. We note in this regard that the annual injection of artificial ^{14}C into the troposphere still persists at measurable levels long after cessation of major tests in the early 1960's (Nydal, Lövseth, and Gulliksen, 1979).

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