PATTERNS OF ATMOSPHERIC ¹⁴C CHANGES

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ABSTRACT. Natural atmospheric ¹⁴C changes are caused by fluctuations in upper atmospheric ¹⁴C production rates (Q) that are related to earth geomagnetic field variations and changes in solar wind magnetic shielding properties. Climate variability may also be responsible for some of the changes because it influences exchange rates of ¹⁴C between the various terrestrial carbon reservoirs.

Upper atmospheric ¹⁴C production rates Q_{M} , in at/sec cm² (earth), were calculated for the past 1200 years from the atmospheric ¹⁴C record and a carbon reservoir model. The changes in Q_{M} are compared in detail with the predicted Q variability derived from an Aa solar modulation mechanism and 20th century neutron flux observations. The influence of earth geomagnetic field changes on the magnitude of the solar wind modulation is discussed, and it is shown that the variations in this magnitude agree with the known differences in earth magnetic field intensity during the past 1200 years. The larger calculated Q_{M} oscillations during the sixth millennium BP also agree with this concept.

Solar wind magnetic as well as geomagnetic forces modulate the incoming cosmic ray flux and explain the main features of the atmospheric ¹⁴C record. It is argued that climatic fluctuation is not a dominant cause.

The oscillations between 3200 and 3700 RC, as measured by de Jong, Mook, and Becker, differ in rise time from those found for the current millennium.

INTRODUCTION

Atmospheric ¹⁴C levels are subject to change because both the ¹⁴C production rate in the atmosphere and the exchange rate between atmosphere and other carbon reservoirs (such as the ocean and the biosphere) are not constant with time. In a recent paper (Stuiver and Quay, 1980) we discussed the atmospheric ¹⁴C record of the present millennium and used a carbon reservoir model (Oeschger and others, 1975) to calculate the past ¹⁴C production rate changes Q_M that were needed to produce such an atmospheric record in a carbon reservoir model. This model describes the terrestrial carbon exchanges between the atmosphere, ocean, and biosphere and incorporates an eddy diffusive ocean. The calculated production rate changes Q_M were compared with ¹⁴C production rate changes Q derived from 20th century cosmic ray measurements as well as with basic time patterns of the historical record of sunspot numbers.

In the current paper, the solar modulation is discussed in more detail by taking into account earth geomagnetic field intensity changes, and by comparing the Q_M values calculated for the 6th millennium BP with Q_M values calculated for the last millennium. A further addition is the extension of the atmospheric ¹⁴C and Q_M record from AD 1000-1860 to AD 730-1860.

Solar modulation

Solar modulation of the cosmic ray flux arriving in the upper atmosphere is caused by changes in magnetic properties of the solar wind. The changes of the solar wind properties are also reflected in other parameters that relate to solar variability, such as sunspot numbers and Aa geomagnetic indices (Mayaud, 1972). The 20th century measurements of

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neutron and cosmic ray fluxes, and the observations of sunspot numbers and Aa geomagnetic indices, can be used to derive relationships between global atmospheric ¹⁴C production rates Q (in atoms/sec cm² earth surface) and either Aa index or the average sunspot number \bar{S} of each 11year cycle. These relationships were derived previously (Stuiver and Quay, 1980) and are the following:

$$Q = 2.310 (\pm 0.114) - 0.024 (\pm 0.005) Aa$$
 (1)

and

$$Q = 2.091 (\pm 0.126) - 0.0041 (\pm 0.0010) \bar{s}$$
(2)

The relationship between observed average sunspot number \bar{S} and Aa index given in figure 1 was used to derive equation 2 from equation 1 and is

$$Aa = 9.13 (\pm 1.29) + 0.17 (\pm 0.02) \bar{s}$$
(3)

Equation 3 is clearly valid when average sunspot numbers exceed 30 (see fig 1). A straightforward extrapolation to zero average sunspot number results in Aa = 9.13. Substitution of $\bar{s} = 0$ in equation 2, or Aa = 9.13 in equation 1, yields a production rate Q of 2.091 at/sec cm² (earth). A further increase in the ¹⁴C production rate to 2.310 at/sec cm² (earth) is still possible by reducing Aa to zero in equation 1. This formalism agrees with the idea of a residual solar wind originating at the sun's polar regions that is still further reduced in intensity when sunspots are absent (Stuiver and Quay, 1980).

The sunspot record, as observed historically (Waldmeyer, 1961; Eddy, 1976) can be used to calculate an average ¹⁴C production rate for the AD 1650 to 1860 period. An Aa record is not available for this interval because magnetometer observations were only made after AD 1860. Between AD 1705 and 1860, the average number of sunspots observed per year was 44.8 which corresponds with an Aa value of 16.75 (from equation 3). Between AD 1650 and 1705, during the Maunder minimum when sunspots were absent, the Aa value can only be estimated. For this interval we took



Fig 1. Observed sunspot numbers and Aa indices averaged over the 11-year solar cycle.

an Aa value of 4.6 which is the midpoint between $\bar{s} = 0$ (or Aa = 9.13) and Aa = 0. Combining both intervals gives an average Aa value of 13.6 for the AD 1650 to 1860 period. The substitution of this value in equation 1 yields an average ¹⁴C production rate of 1.984 at/sec cm² (earth) for the AD 1650 to 1860 interval.

The percentage deviation ΔQ from the average production rate is given by

$$\Delta Q = \frac{(Q - 1.984)}{1.984}$$
 100 percent

For S > 0 the relationship between ΔQ and \bar{S} is derived from equation 2:

$$\Delta Q = 5.4 - 0.207 \text{ \bar{s} percent} \tag{4}$$

The production rate changes ΔQ around the mean are given as a function of sunspot number by the solid line in figure 2, where the ΔQ scale is on the right. The ¹⁴C production rate changes ΔQ derived from the relationship between neutron flux measurements, \bar{S} and Aa are compared with the production rate changes ΔQ_M calculated from carbon reservoir modeling which are relative to the average ¹⁴C production rate of 1.57 at/sec cm² (earth) of the Oeschger and others type of carbon reservoir model (Stuiver and Quay, 1980; Oeschger and others, 1975). The post AD 1650 ΔQ_M values average -5 percent relative to the AD 1000 to 1860 average ¹⁴C production rate of 1.57 at/sec cm² (earth). We have, therefore, matched the -5 percent ΔQ_M value (left scale) with $\Delta Q = 0$ percent (right scale in fig 2).



Fig 2. A comparison of ¹⁴C production rate changes ΔQ , based on 20th century observations, with carbon reservoir model calculated production rate changes ΔQ_M . The solid line gives the ΔQ changes predicted from 20th century observations. The ΔQ_M values calculated from the atmospheric ¹⁴C record for each 10-year interval between AD 1660 and 1860 are given as dots. Different interpretations of the observed sunspot number record result in a possible shift of two points, as indicated by the squares.

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For $\bar{s} = 0$, and Aa = 0, ΔQ can reach maximally + 16.2 percent, as indicated by the vertical extension in figure 2 of the line representing equation 4.

The agreement between 19th century ΔQ_M values calculated from the carbon reservoir model (open circles in figure 2) and the 20th century ΔQ values derived from the dependence of observed cosmic ray fluxes on sunspot numbers and Aa indices is excellent. The slope $dQ/d\bar{s} = -0.0041 \pm 0.0010$ (derived from equation 2) is similar to the $dQ_M/d\bar{s}$ slope of -0.0048 ± 0.0013 derived from the least squares linear fit through the 19th century points (see Stuiver and Quay, 1980). The 1660-1790 points also generally agree with the concept of higher ¹⁴C production during lower sunspot numbers, but the overall agreement with the curve derived from the 20th century cosmic ray flux observations is less satisfactory. This could be due to several causes:

1) The sunspot numbers are less accurate for the older parts of the sunspot number record, as discussed previously by Eddy (1976). Some of the outliers could be attributed to this. For instance, the 5 numbers used in figure 2 were derived from the Waldmeyer compilations for the post AD 1700 interval and from Eddy's estimates prior to AD 1700. Eddy's interpretation of the historical record, however, differs from Waldmeyer's for the AD 1700 to 1715 interval. The use of Eddy's numbers would substantially change the anomalous position of the 1695 to 1705 and 1705 to 1715 decade points by moving these values to the squares in figure 2.

2) The standard deviations in the ¹⁴C measurements of the 17th and 18th century is in the 1.5 to 2 per mil range. This corresponds, on average, with a ΔQ_M inaccuracy of about 4 percent. Some of the more negative outliers that are based on only a single ¹⁴C determination may be in error by up to 2 sigma (8 percent) in production rate. Additional single year analysis for these periods is presently in progress in our laboratory in order to reduce this uncertainty.

An additional uncertainty is the error of the regression analysis that results in equation (1). A ΔQ value of 19.4 percent can be reached for $\bar{s} = 0$ and Aa = 0, instead of the +16.2 percent given before, when one standard deviation is included.

The actual baseline ${}^{14}C$ production rate is also important. The Aa value of 13.6 over the AD 1650 to 1860 interval, and the associated Q value, is approximate only.

3) Carbon reservoir modeling is still an art that attempts to simulate natural processes. It is possible that the reservoir model over-attenuates production rate changes by inaccurately describing ¹⁴C reservoir sizes and/or exchange rates. As a consequence, our ΔQ_M values could be systematically too large. A plot of ΔQ_M values 1.5 times smaller than given here, would indeed provide for a better fit, but of course, we do not know whether the suggestion of over-attenuation of the Oeschger and others model will be borne out by further research.

Changes in internal ¹⁴C distribution, associated with climatic change, also could contribute to deviations from the predicted ΔQ curve. For in-

stance, it is possible that the higher ¹⁴C level reached during the Maunder minimum was partially caused by climate-induced changes in atmosphereocean CO₂ exchange rates, ocean eddy diffusion coefficients, or biospheric reservoir size. The difference between the calculated production rate change of 21 percent for the Maunder minimum, and the 11 percent ΔQ_M production rate increase predicted by the solid line in figure 2, could, for instance, be explained by a 20 percent reduction in either CO₂ exchange rate or vertical eddy diffusivity (Stuiver and Quay, 1980). Studies of global scalar wind velocities over decade intervals suggest that changes of this order of magnitude in CO₂ exchange rate are possible (J M Fletcher, pers commun).

If the magnitude of the above effects is too small to resolve the discrepancy between the calculated changes in Q and Q_M , one can conclude that 20th century solar modulation is not identical to 17th and 18th century modulation. The 17th and 18th century $dQ_M/d\bar{s}$ increase should then be attributed to a change in solar behavior. A small portion of the increase can also be attributed to earth geomagnetic field changes, as discussed in the following section.

Earth geomagnetic field

So far, we have discussed the influence of the interplanetary solar wind magnetic field properties on ¹⁴C production rates. The interplanetary field acts on the particles traveling from the interstellar medium to the inner solar system. Closer to earth, the earth geomagnetic field interacts with the incoming cosmic ray flux and deflects part of this flux. The global ¹⁴C production Q(t) at time t decreases with increased earth magnetic dipole moment M(t).

Our relationship 2 between Q and S is only valid for the 20th century earth magnetic dipole moment M value of 8×10^{25} Gauss cm³. The ¹⁴C production rate Q(t) for a dipole moment M(t) relates approximately to the 20th century production rate Q(O) (Lingenfelter and Ramaty, 1970) as follows:

$$Q(t)/Q(O) = {M(O)/M(t)}^{0.5}$$

Substitution of relationship 2 gives:

$$Q(t) = \{M(O)/M(t)\}^{0.5} (2.091 - 0.0041 \ \overline{S})$$
(5)

Thus, for the same level of solar modulation of the interplanetary medium, the absolute change ΔQ in ¹⁴C production rate Q(t) is larger when earth geomagnetic intensity is smaller. For instance, the predicted change in the 20th century ¹⁴C production rate Q(O) is 0.205 at/sec cm² (earth) for an Aa change of 0 to 17.6 but the Q change is $\sqrt{2} \times 0.205 = 0.289$ at/sec cm² (earth) when the dipole moment is half the 20th century value.

In our carbon reservoir modeling we have removed the long-term trend in atmospheric ¹⁴C level attributed to earth geomagnetic dipole moment changes by deducting the sinusoidal approximation given by Houtermans (1971). The remaining Δ^{14} C variations are used to calculate

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¹⁴C production rate changes ΔQ_M around the steady state value of 1.57 at/sec cm² (earth). As discussed previously, solar modulation of the cosmic ray flux can account for the main features of the ΔQ_M record obtained for the post AD 1650 interval (Stuiver and Quay, 1980). Thus, for the older ¹⁴C variations, larger ΔQ_M values would be expected for intervals of low earth geomagnetic field intensity, in accordance with equation 5.

The long-term sinusoidal trend that is removed from the atmospheric Δ^{14} C record is not necessarily the best approximation of the changes in earth magnetic dipole moment. Therefore, in the following discussion, we use the direct measurements of magnetic dipole intensities, as compiled by Barton, Merrill, and Barbetti (in press), and as given in figure 3.

In figure 4 we give the post AD 730 calculated ΔQ_M values, in percent deviation from the mean steady state production rate of 1.57 at/sec cm² (earth). The ratio of the 8th and 20th century AD earth geomagnetic dipole moments (figure 3) is 1.38. The 8th century AD calculated ΔQ_M values, therefore, would be 1.17 times smaller than the 20th century AD values (equation 5). Figure 4 shows this basic trend by the dashed lines. The increase in ΔQ_M values towards the present evidently agrees with the trend calculated from the earth geomagnetic field changes given in figure 3.

A more sensitive test of the influence of earth geomagnetic field changes is a comparison of the ΔQ_M oscillations for the current millennium with those of the 6th millennium BP. The midpoints of the intervals to be discussed (0 to 1000 and 5150 to 5650 BP) are 500 and 5350 BP. The earth magnetic dipole moment ratios M(t)/M(O) are, respectively, 1.18 and 0.68 for these mid-points. Thus, for the older interval, a deviation 1.32 times larger in ¹⁴C production rate is expected.

The atmospheric Δ^{14} C oscillations of the 5150 to 5650 BP interval were measured by de Jong, Mook, and Becker (1979). Figure 5 illustrates our calculated production rate changes ΔQ_M after removal of the main



Fig 3. Earth magnetic dipole moments M(t), relative to the 20th century value M(O), for the past 7000 years. The data are from Barton, Merrill, and Barbetti (in press).



Fig 4. Reservoir model calculated changes in ¹⁴C production rate ΔQ_M , relative to the average AD 730 to 1860 production level. The earlier part of the record (AD 730 to 940) was derived from an atmospheric ¹⁴C record obtained by measuring wood samples that cover 20-year intervals. Samples of the remaining part of the record were either from a single decade, or from single years (see also, Stuiver and Quay, 1980, for further discussion).



Fig 5. Reservoir model calculated ΔQ_M changes, derived from an atmospheric ¹⁴C record measured by de Jong, Mook, and Becker (1979).

sinusoidal trend. The production rate changes ΔQ_M are again relative to the 1.57 at/sec cm² steady state value. The magnitude of the increase in Q_M averages nearly 66 percent for the three oscillations of the 6th millennium BP (figures 5 and 6). They average 43 percent for the 4 oscillations of the current millennium (figures 4 and 6). Thus, the ΔQ_M oscillations in the 6th millennium BP are 1.52 times larger than the 4 most recent changes. From the geomagnetic dipole intensity variations an increase of 1.32 times was predicted. The difference between predicted and actual increase may be due to underestimating the reduction in magnetic field intensity around 5300 BP. A M(t)/M(O) ratio of 0.51, instead of 0.68, would agree with the observed ΔQ_M increase. Of course, an alternative explanation could be a change in solar modulation behavior. The 6th millennium BP oscillations all have a faster rise time (30 to 40 years) than the oscillations of the current millennium (about 70 years, see also fig 6). The faster change from active to quiet sun mode in the 6th millennium BP may reflect different solar behavior that could conceivably increase cosmic ray modulation 1.15 times.

Up to this point, we have neglected the influence of climate change on atmospheric ¹⁴C levels. Within the framework of a carbon reservoir model, the atmospheric ¹⁴C levels depend on atmosphere-ocean CO₂ exchange rates, oceanic eddy diffusion, and biospheric reservoir size. These parameters may all undergo changes when global temperature, wind



Fig 6. The increase in ¹⁴C production rate ΔQ_M measured from the lowest ΔQ_M value preceding a maximum. The oscillations of the 6th millennium BP are given by roman numerals (see also fig 5); the even numbers correspond with ΔQ_M increases of the current millennium.

velocity, and precipitation rates change. The influence of climate on the carbon reservoir parameters is difficult to estimate, especially since the three parameters need not cause an atmospheric ¹⁴C change in the same direction. However, we think that the influence of climate change on atmospheric ¹⁴C levels during the Holocene is probably a secondary effect to solar modulation. The reasoning is as follows:

1) Extrapolation of the 20th century knowledge of solar modulation can explain the main portion of the Maunder minimum ¹⁴C increase. The Maunder minimum ¹⁴C production level Q_M (calculated from the observed ¹⁴C record) is 30 percent above the Q_M levels calculated for the 19th century (AD 1800 to 1860 interval, fig 2). The average number of sunspots observed from AD 1800 to AD 1860 was 42. The predicted production rate increase (equations 1 and 2) is from 1.919 to 2.310 at/sec cm², or 20.4 percent. Thus, 2/3 of the increase can be explained by extrapolating 20th century knowledge of cosmic ray modulation. As previously discussed, the difference between model calculated Q_M values and predicted Q values can be attributed to several causes, of which, climatic change is one possibility. If climatic change did indeed influence the Maunder minimum ¹⁴C level, it would have increased atmospheric ¹⁴C levels in phase with solar modulation and account for about 1/3 of the total increase.

2) The increase of 1.52 times in Q_M amplitude found for the 6th millennium BP oscillations is entirely compatible with the difference in magnitude of solar modulation caused by the change in earth dipole moment. The clear dependence of the Q_M oscillations on earth geomagnetic field intensity points to solar modulation, and not climate, as the main causal factor for the secular ¹⁴C variations.

In summarizing our discussions, we can state that the forces acting on the incoming cosmic radiation evidently are responsible for the main features of the atmospheric ¹⁴C record. Long-term changes (10³ years or longer) are ascribed to earth geomagnetic field variations. Atmospheric ¹⁴C changes, lasting one or more centuries, are caused by solar modulation of the cosmic ray flux as described by the Aa solar modulation mechanism. The magnitude of the solar modulation of the cosmic ray flux is also modified by earth geomagnetic field changes.

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DISCUSSION

Lal: I do not yet see how the Aa index provides a better handle on the ¹⁴C source function. There is, of course, a numerical advantage in the case of the Aa index since the Aa variations are well above the zero line and accommodate the possibility of a non-linear behavior at low values. The Aa index relates to the magnetic field in the space near one AU, but we believe that the cosmic ray flux seen at one AU results from conditions in the interplanetary space as far out as 10 AU. I believe that the correct procedure, which would have a physical basis, would be to correlate cosmic ray flux with a two-parameter index, say sunspot numbers and the shape of the sunspot curve. The latter could possibly take into account the time intervals involved in "coasting" of the solar wind to outer reaches of the modulation region.

Stuiver: The Aa index relates directly to the magnetic properties of the solar wind. In our model we assume that the time dependent changes in solar wind interaction at one AU, near the ecliptic plane, also represent the time dependent changes of solar wind interaction further in space. By using the Aa index solar wind properties are taken directly into account. Sunspots are less suitable because they do not deflect cosmic rays.

Damon: The authors have been forced to use an adjustment to prevent the box-diffusion model from over-attenuating and piling up ¹⁴C in the model atmosphere. The box-diffusion model of Oeschger and others (1975) is the best mechanistic ¹⁴C model yet introduced, but, like all multi-box models, it tends to over-attenuate. The adjustment made by the authors is one that we used (unpublished) *ie*, they reduced the "steady state" production rate to 1.57 at/cm_e² sec. They used standard box-diffusion model parameters for which the DC gain (g = H (O)) is 130 years. But, g is also an observable quantity: $g_{obs} = H(O) = \bar{N}_a */\bar{Q}$, *ie*, the DC gain is equal to the radiocarbon content of the atmosphere divided by the "steady state" production rate. Thus, since g is equal to 130 years, a necessary consequence of the model, using $\bar{Q} = 1.57$ at/sec cm_e², is that it will out-put an absurdly low atmospheric ¹⁴C content equivalent to an activity of 11.5 dpm/gC. This activity is used, raising the relative size of the De Vries effect "wriggles" (à la Suess).

Stuiver: The problem of the discrepancy between model ¹⁴C production rates and estimated terrestrial ¹⁴C decay rates was discussed in our Science (1980) paper. By adding a reservoir (residence time = 1000 years) containing additional radiocarbon the "steady state" production rate in the model was raised to 1.89 at/cm_e² sec. Our model calculated ¹⁴C production rates are expressed as the percent deviation around the "steady state" ¹⁴C production rate. When expressed in this manner, the differences between the calculated percent production rate changes derived from the 1.57 at/cm_e^2 sec model and the 1.89 at/cm_e^2 sec model are small (eg, a 21 percent calculated change in production rate for the first model becomes 20 percent for the second).

^{*} Additional discussions of the Oeschger and others model are also given in "Modeling the Carbon System" by Broecker and others in the Proceedings.