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SOLAR MODULATION EFFECTS IN TERRESTRIAL PRODUCTION OF CARBON-14

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ABSTRACT. This paper is concerned with the expected deviations in the production rate of natural ^{14}C on the earth due to changes in solar activity. We review the published estimates of the global production rates of ^{14}C due to galactic and solar cosmic ray particles, and present new estimates of the expected secular variations in ^{14}C production, taking into account the latest information available on galactic cosmic ray modulation and long-term variations in solar activity.

Estimated secular variations are related to data on atmospheric $^{14}C/^{12}C$ ratios based on tree rings. It is concluded that the observed higher frequency wiggles in atmospheric $^{14}C/^{12}C$ ratios occurring with time scales of about two hundred years and of 1 to 2 percent in magnitude (de Vries, 1958; Suess, 1970a,b; 1979; Damon, Lerman, and Long, 1978), are largely due to solar activity dependent modulation of the galactic cosmic ray flux by solar plasma. These variations override a slowly varying sinusoidal change of about 10 percent in magnitude during the last approximately 8000 years, which is believed to be primarily due to changes in the geomagnetic field. The high frequency modulation effect in ^{14}C production is substantial, about 20 percent, which, considering the response function of the atmosphere (*cf* Houtermans, Suess, and Oeschger, 1973), is adequate to explain the observed ^{14}C wiggles (also named Suess wiggles or de Vries oscillations) if in the past, periods of large modulation effects and also periods of weak modulation persisted, *ie,* the sun remained both active and inactive over long periods of time, of the order of several decades to centuries. The pioneering investigations of Eddy (1976; 1977) of the ancient records of solar activity make it plausible that the ^{14}C wiggles in the ancient $^{14}C/^{12}C$ ratios are primarily due to modulation of galactic cosmic ray flux by a varying sun. Thus, the ^{14}C wiggles are good indicators of solar activity in the past.

We also present revised estimates of the production rates of ^{14}C on the earth due to solar flare accelerated cosmic rays and limits of direct accretion of ^{14}C on the earth.

INTRODUCTION

The production of ^{14}C on the earth is nearly constant and its concentration in the different dynamic carbon reservoirs is even more so. These facts form the basis of the radiocarbon dating method (Libby, 1955). However, since the specific activity of ^{14}C can be measured with a high precision, better than 0.2 percent, monitoring "fossil dated" samples of carbon isolated from the dynamic carbon reservoir samples enables the study of the nature of secular variations. These are caused by several factors, such as variations in the cosmic ray intensity and geophysical and geochemical cycles. Studies of the cosmic ray intensity variations based on ^{14}C studies on the earth are linked to the modulation of cosmic rays streaming into the solar system. Cosmic rays depend on the state of the interplanetary medium which is governed by solar plasma

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emission, as well as changes in the dipole moment of the earth. In order to use ^{14}C as a tool for understanding the nature of secular changes in 1) cosmic ray intensity in the past, 2) the earth's magnetic field, or 3) climatic and other changes on the earth that induce perturbations in the dynamic carbon reservoir, (involving "fossil dated" samples from different epochs) must be studied. Supplementing these studies with meteorite data that are free from influences of changes in the earth's magnetic field and geophysical/geochemical parameters would be useful. However, the meteorite record differs appreciably in character from the tree-ring record (meteorites provide time-integrated records) and the record is also affected by other causal parameters, eg, erosion and fragmentation during irradiation.

In this paper we review our present understanding of expected variations in the production of ^{14}C on the earth and examine the question of the secular variations in ^{14}C production during periods of extreme solar activity, eg, very low activity periods such as the Maunder Minimum (1645-1715), when there were virtually no spots on the sun. Fortunately, enough data are available on the production of neutrons in the atmosphere during different periods of solar activity as well as on the nature of modulation of cosmic rays in the interplanetary space with varying solar activity. Reasonable estimates of ^{14}C production in the earth's atmosphere can be made for any postulated modulation although uncertainties remain in estimating corresponding changes in ^{14}C specific activities in the atmosphere. These estimates depend on a number of parameters, the duration and spectral form of solar modulation induced intensity changes and the concomitant changes, if any, in the behavior of the atmosphere-ocean mixing characteristics.

We summarize our present knowledge of these parameters and estimate the magnitude of secular variations in ^{14}C production on the earth due to modulation of galactic cosmic rays as well as cosmic ray particles accelerated by the sun in periods of high activity. The expected deviations in ^{14}C source function seem to be large enough to explain the observed high frequency variations in atmospheric $^{14}C/^{12}C$ ratios, recently termed as Suess wiggles (de Jong and Mook, 1979), provided the sun remained both active and inactive over long periods of time, several decades at a time, as observed during the Maunder Minimum. The results of the present work provide physical basis for the bold postulate made by Stuiver (1961) and Eddy (1976, 1977) that studies of $^{14}C/^{12}C$ ratios in tree rings are relevant to variation of solar activity in the past.

The global production of ^{14}C and the dynamic reservoir of ^{14}C

Radiocarbon is produced continually on the earth in a latitude-dependent manner as a result of nuclear interactions of cosmic radiation in the earth's atmosphere. By far the largest contributions come from thermal neutron capture reaction in nitrogen, ^{14}N (n, p) ^{14}C. The dynamic carbon cycle controls the distribution of radioactivity of long-

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lived ^{14}C on the earth. Per cm^2 column, the largest amount of ^{14}C resides in the oceans. The specific activity of radiocarbon is highest in the atmosphere, as would be expected, since its production on the earth occurs primarily in the atmosphere. The amounts of ^{14}C produced *in situ* in the lithosphere and in the hydrosphere are negligible. For a discussion of production rate of ^{14}C, the reader is referred to the seminal paper by Libby (1955) and subsequent papers by Lingenfelter (1963), Lal and Peters (1967) and Light and others (1973). The global average production of ^{14}C is of the order of 2 atoms/cm^2 sec. For a discussion of the dynamic carbon reservoir and the $^{14}C/^{12}C$, $^{13}C/^{12}C$ ratios therein, reference is made to Craig (1957) and Lal and Suess (1968).

Since global atmospheric mixing occurs rapidly compared to time scales of exchange processes in the carbon cycle, latitude dependence in the source function of ^{14}C is not seen. In fact, the careful measurements of Lerman, Mook, and Vogel (1970) established that geographic variation in atmospheric $^{14}C/^{12}C$ ratios measured in tree rings is small compared to observed secular variations in tree rings.

The response function of the atmosphere, as indicated by the $^{14}C/^{12}C$ ratios, with respect to perturbations in atmospheric source function of ^{14}C and changes in atmosphere-ocean mixing rates has been considered by several authors. Early papers on the mixing of ^{14}C in the atmosphere-biosphere-ocean system (Arnold and Anderson, 1957; Craig, 1957; Revelle and Suess, 1957) are still very interesting. More recent complete studies were made by Houtermans, Suess, and Oeschger (1973) and Oeschger and others (1975). Response function of the atmosphere to changes in the source function, Q (the global average ^{14}C production rate: atoms/cm^2 sec), is conveniently discussed in terms of phase lag and amplitude in variation in atmospheric $^{14}C/^{12}C$ ratio, R for a periodic variation in the source function, Q. Houtermans, Suess, and Oeschger (1973) assumed that variations in Q could be expressed as a sum of sine and cosine of various frequencies. For 2 and 3 reservoir carbon exchange models, they then evaluated the Q-R relationship. Their results (their fig 3) show that the factors by which the variation in R was attenuated relative to variations in Q are about 200, 25, and 10, respectively, for periodicities in Q of 10, 100, and 1000 years, respectively.

Measured secular variations in R comprise at least two components (Suess, 1970a,b; Damon, Long, and Grey, 1970; and also Suess, 1980; de Jong and Mook, 1979). The two components are 1) a slow change which could be approximated by a sine wave with an amplitude of about 10 percent and period of about 10,000 years, and 2) wiggles throughout the available record with a prominent 200-year periodic component; the amplitude of wiggles is of the order of 1 to 2 percent. A detailed discussion of the amplitude of the wiggles back to the seventh millennium is given by Suess (1980).

Long-term slow change is well understood to be primarily due to changes in the geomagnetic field (Bucha, 1970; Cox, 1968; Suess, 1970b). Part of this change could be due to climatically induced change in the

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ocean-atmosphere exchange over the southern ocean, as suggested by Lal and Venkatavaradan (1970). Such slow changes for which the attenuation factor is small, < 5, might also be caused partly by slow time variations (of unknown causes) in the cosmic ray flux. Our present knowledge of long-term secular variations in the cosmic ray flux (Arnold, Honda, and Lal, 1961; Lal, 1972, 1974; Schaeffer, 1975; Forman, Schaeffer, and Schaeffer, 1978) is not precise enough to exclude this possibility. We can only conclude the 10,000 year variation seems principally attributable to changes in the geomagnetic field. Twenty percent or so of the slow variation in R may have other causes but unless more accurate global archaeomagnetic information is available, no comment may be made on the magnitude of slow variation of other causes.

Variation of considerable geo-astrophysical significance in R is the higher frequency component recognized as a variation well above experimental noise as well as random red noise. Fourier analysis of R values by Suess yields a prominent 203 ± 3 year period with an amplitude of up to ± 1.5 percent during the last 8000 years. Even such small variations must be produced by ± 20 to 30 percent variations occurring in a period of 200 years, considering the much larger attenuation for higher frequency variation. Thus, the origin of the wiggles is difficult to understand because it involves rather large short-term variations in Q, not consistent with our present picture of modulation of the galactic cosmic ray flux. Are the wiggles caused by some currently misunderstood variations in the dynamic carbon reservoirs? In this paper we will review our knowledge of variations in solar activity, the nature of cosmic ray modulation, and the expected magnitude in variations in Q directly or indirectly due to solar effects, *eg,* solar plasma caused modulations and solar flare cosmic ray produced ^{14}C in the earth's atmosphere. It seems to us that the tree-ring data may be understood in terms of known physical processes; it is unnecessary to invoke unknown phenomena.

For a recent fairly comprehensive review of the causes of temporal fluctuations of atmospheric ^{14}C, reference is made to Damon, Lerman, and Long (1978). In addition, reference is made to papers presented at the 12th Nobel Symposium held at Uppsala in 1969. Expected secular variation in $^{14}C/^{12}C$ ratios caused by cosmic ray modulations has also recently been treated by Forman, Schaeffer, and Schaeffer (1978).

Cosmic ray flux and solar modulation

Our most reliable and detailed knowledge about galactic and solar cosmic ray fluxes is based on direct observations of cosmic rays during the last few decades. Additional data comes from meteorites and moon samples that have preserved records of galactic and cosmic irradiation (Arnold, Honda, and Lal, 1961; Lal, 1974; Schaeffer, 1975; Reedy, 1977). These data, although accurately recorded by advanced prehistoric methods, generally lack details on, eg, isotopic composition, fluxes of nuclei

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heavier than helium and lighter than iron, and time variations in the past, during 10^7-10^8 years BP.

Thus, we have information on both the contemporary flux and long-term averaged data for cosmic ray protons, alpha particles and iron group and heavier nuclei, averaged in time for millions to billions of years. Cosmic ray fluxes based on studies of meteorites/lunar samples are averaged over intervals corresponding to mean lives of radionuclides studied. The problem is to determine the changes in the cosmic ray fluxes during the last 10,000 years, a period for which accurate tree-ring data are available. To understand variations in $^{14}C/^{12}C$ ratios in tree rings, we must know time variations in the energy spectra of at least the galactic cosmic ray protons averaged over periods of the order of several decades or centuries. Such data cannot be extracted from studies of meteorites that provide integrated flux information. Differential flux information can be had from terrestrial sediments or polar ice caps but such data is not available until radical improvements are made in our present technology. Raisbeck and Yiou (1980) might provide this information.

Modulation of galactic cosmic ray flux

It is well recognized that low energy cosmic ray particles are scattered on the irregularities in interplanetary magnetic fields convected outward by highly conductive solar wind plasma (*cf* Simpson, 1978). Consequently, the energy spectrum of cosmic ray particles near the earth is considerably modified as a result of interactions of incoming cosmic rays with the moving magnetic fields, which lead to scattering, diffusion, and energy losses. This process is termed modulation of the galactic cosmic ray flux by the solar plasma. Modulation is effective within a zone called the heliosphere, extending out 30-50 AU (Axford, 1972; Pyle and others, 1979). Figure 1 shows qualitatively the integral energy spectrum of solar plasma and the galactic cosmic ray fluxes during 1954 (a period of sunspot minimum) and 1959 (a period of sunspot maximum). Also shown in figure 1 are fluxes due to solar cosmic rays, solar black body radiation, and the energy spectra of electrons and protons in the Van Allen belts within the earth's magnetosphere. ^{14}C dominates all other production mechanisms. Solar flare production is generally not significant but may be important for some flares. Thus, solar flare produced ^{14}C may be significant during periods of high solar activity.

Prior to 1972, observations of solar modulation of galactic cosmic ray flux were considered simply as an anti-correlation of flux with solar activity. In recent years these estimates of modulation are no longer considered meaningful because long-term variations in cosmic ray fluxes are involved. Detailed scientific observations and analysis in two areas have caused this radical change: 1) cosmic ray energy spectrum and composition in interplanetary space in recent years particularly during the last decade, and 2) careful analysis of available records of solar activity during the last four centuries. Both investigations provided sub-

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stantial surprises and warrant re-evaluation of expected production rates of ^{14}C and secular variations in the atmosphere.

The new aspects of cosmic ray modulation have been realized through studies of quiet time cosmic ray H and He isotopes using balloons and IMP series satellites. These data for protons and alpha particles (*cf* Garcia-Munoz, Mason, and Simpson, 1977; Webber and Yushak, 1979). These data, in conjunction with the Climax neutron monitor data, clearly show that pre-1972 models for modulation do not work. In particular, the modulation features cannot be correlated in any simple fashion with sunspot numbers. Prior to 1972, clear indications of this existed, as seen from the plot of measured ionization rate at an atmospheric depth of 20gm cm^{-2} versus sunspot number (Pomerantz and Duggal, 1974) in figure 2.

Both magnitude and phase lag in cosmic ray modulation depend on the shape of the solar activity curve. Modulation dynamics depend also on a number of relatively recently recognized solar features, such as coronal holes and high speed streams. Solar activity dependent changes occur within the heliosphere to long distances of ~ 30 to 50 AU (O'Gallagher, 1975; Garcia-Munoz, Mason, and Simpson, 1977; Simpson, 1978; Pyle and others, 1979). Clearly one has to re-examine the expected changes in the cosmic ray flux with the solar cycle in the past, guided by observations during solar cycles 17 through 20.

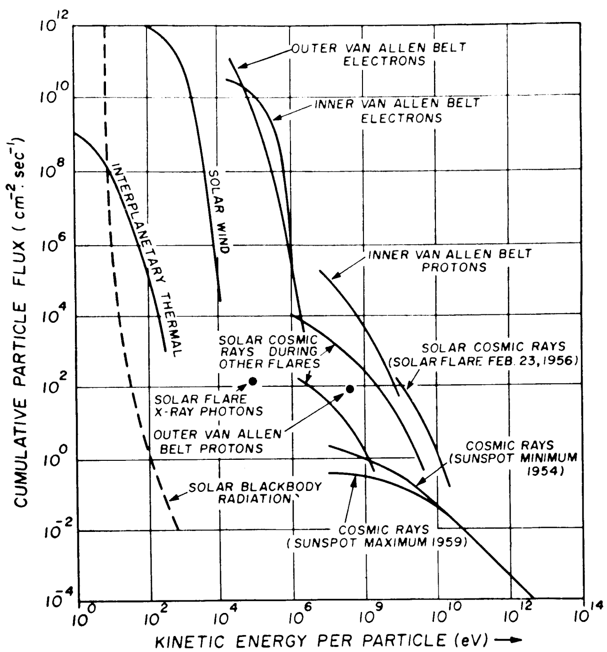

Fig 1. Characteristic integral energy spectra of electromagnetic and particle radiation in space in the near earth environment. Sources of data: published literature. Theme taken from Fillipowsky and Muehldorf (1970).

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In figure 3, we reproduce published data on cosmic ray spectra based on IMP satellites and balloon borne detectors for protons. The proton fluxes were highest during July 1977 (Webber and Yushak, 1979). We have also shown the inferred spectrum outside the heliosphere for 1975 and 1979 (Simpson, pers commun, 1979), based on data for protons, alpha particles, electrons, and some assumptions about source spectra and modulation theory. The problem is in what manner the cosmic ray spectra at 1 AU may vary for sustained periods of low or high solar activity.

Another realization has been that solar activity variations during the past 400 years were quite dramatically different from those of the last 200 years. Good quality data on sunspot number are available for Solar Cycles 1-20. Data prior to Solar Cycle 1 are inferior but sufficient information is available to deduce the character of variation back to AD 1650 as well as an approximate curve for AD 1600 to 1650. Eddy (1976; 1977) described this and concluded that there was a long period of prolonged solar minimum during AD 1645 to 1715. He termed this epoch the Maunder Minimum, after Maunder who first suggested the phenomenon. Eddy also suggested another minimum, during AD 1460 to 1550, and several others based on ^{14}C tree-ring data. Eddy's analysis suggests that there were several periods of Solar Minimum and Maximum during the past 7400 years, 8 periods of high and 10 of low activity, lasting for 100 to 500 years. There is strong evidence for a 70-year Maunder Minimum with essentially no sunspots. It seems quite certain that solar activity has not behaved regularly as during 1730 to the present, and that there probably have been sustained periods of very low and high sunspot numbers, lasting for periods of several decades or centuries. We have to ascertain how galacitic cosmic ray fluxes would vary under conditions corresponding to sustained periods of high or low solar activity.

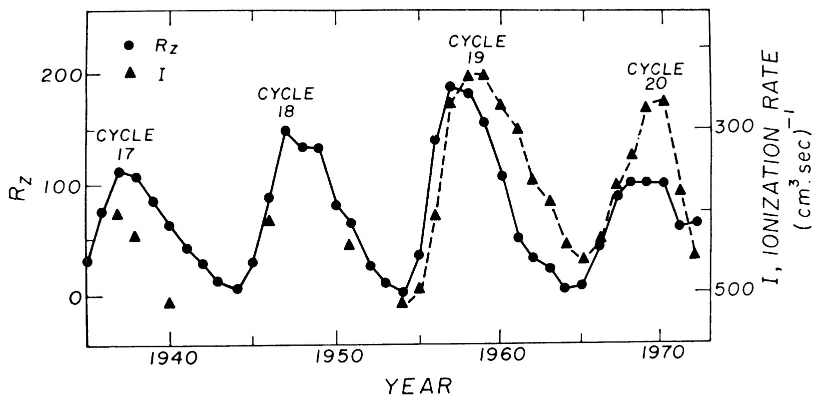

Fig 2. Mean annual ionization at 20gm • cm^{-2} atmospheric depth and Zurich Sunspot numbers for the period 1935-1972. Taken from Pomerantz and Duggal (1974).

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Solar generated interplanetary disturbances inhibit the impingement of the galactic cosmic ray particles on the earth, leading to the well-known Forbush decreases. Transient solar phenomena visually identified in the solar corona have proved to extend far into the interplanetary medium (Hildner, 1977; Pyle and others, 1979) and the phenomena have also been simulated by numerical MHD modeling techniques (Wu, Nakagawa, and Dryer, 1977). Recent observations to distances of ~ 20 AU indicate that the transport of magnetic fields by solar plasma within the entire heliosphere which extends to 30 to 50 AU is effective in producing a strong modulation of galactic cosmic ray flux, not only around the equatorial zone but at least to moderately high latitudes (Bastian and others, 1979; Pyle and others, 1979).

During a long period of low solar activity, eg, during Maunder Minimum, the interplanetary disturbances would have been essentially absent and the heliosphere would almost have been cleared out, whereby the galactic particles would not be hindered from penetrating into the inner solar system. It is difficult to anticipate empirically from the available cosmic ray data during solar cycles 18-20 what fluxes may be anticipated during Maunder Minimum because the conditions for clearing the heliosphere were not attained during these cycles. We, therefore, use modulation theory to estimate spectral shape and fluxes of protons during periods of 'low solar' modulation.

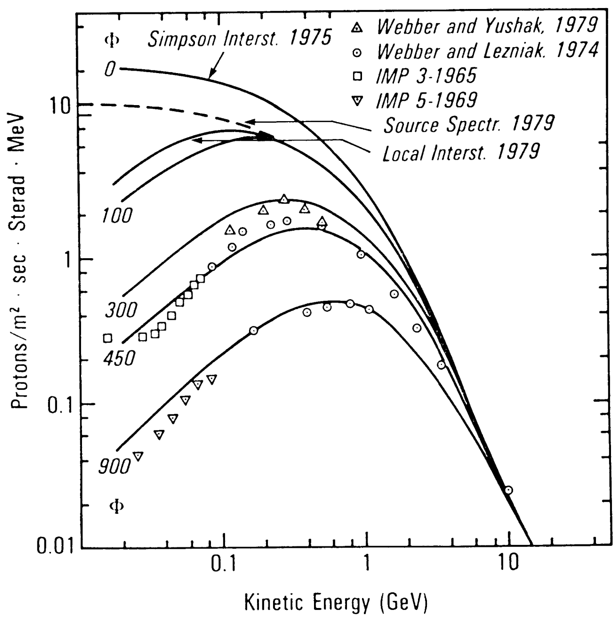

Fig 3. Differential cosmic ray proton energy spectra measured at different periods. Sources of experimental data are given. The smooth curves labelled 0, 100, 300, 450, and 900 are obtained using equation (1) for these values of ϕ. Other curves are analytically based on data and modulation theory (Simpson, pers commun, 1979).

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In the following, our approach is primarily mathematical although based on a physical model. We consider an analytic form for the cosmic ray spectra at different modulation levels using 1) the force field approximate solution of the transport equation as given by Urch and Gleeson (1972) and 2) the galactic differential intensity as given in analytic form by Garcia-Munoz, Mason, and Simpson (1975). We obtain for the proton differential spectra g (T) (particles $sec^{-1}m^{-2}sr^{-1}MeV^{-1}$):

$$g\,(T, \phi) = A\,\frac{T(T + 2E_o)\,(T + m)^{-\gamma}}{(T + \phi)\,(T + 2E_o + \phi)} \qquad (1)$$

where T(MeV) is the proton kinetic energy, E_o its rest energy, ϕ is the modulation parameter (MeV) or the energy lost by particles in traversing the heliosphere while reaching the earth, $A = 9.9 \times 10^8$, $m = 780 \exp(-2.5 \cdot 10^{-4}T)$ and $\gamma = 2.65$.

The curves for different ϕ values are shown in figure 3 together with the experimental data. The curves for $\phi = 450$ MeV and $\phi = 900$ MeV are good approximations for the quiet sun year, 1965, and the active sun year, 1969, spectra, respectively. We note that during 1977, a value of ϕ as low as 300 MeV can be used to approximate the spectrum at maximum intensity, in agreement with the experimental value obtained by Badhwar and Golden (1979). It should also be noted that the new source spectrum proposed by Garcia-Munoz and Simpson (Simpson, pers commun) gives a local interstellar spectrum (in the vicinity of the solar system) not much different from that obtained by formula (1) with $\phi = 100$ MeV.

In the limiting case, the theoretically expected unmodulated spectrum would be given by that labeled as Local Interstellar, 1979 (fig 3). As stated above, confidence in this rests on the fact that the cosmic ray spectra during different epochs are well represented by the modulation treatment in terms of the Fokker-Planck equation for diffusive particle transport within the heliosphere (Urch and Gleeson, 1972; O'Gallagher, 1975; Morfill, Volk, and Lee, 1976). Based on these calculations, (results of calculations of Simpson given in figure 3, and in Morfill, Volk, and Lee, 1976, fig 8) we feel that $\phi = 100$ would be a conservative case for the expected proton flux during solar minimum periods such as the Maunder Minimum. We cannot however rule out $\phi = 0$ for this period. In periods of Solar Grand Maximum, $\phi > 900$ would be expected; however, it is difficult to realistically estimate the highest value attainable in periods of intense modulation. The modulation effect is asymmetric with change in ϕ and a substantial difference is not expected between the lowest ^{14}C production and that of $\phi = 900$. Thus, we assume that the Solar Minimum and Maximum periods correspond to $\phi = 100$ and 900, respectively.

Global ^{14}C production rates in periods of high and low solar activities

The production of ^{14}C at any latitude at a given time can be calculated from knowledge of the cosmic ray spectrum at that time and the

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universal yield function Y(T), the total number of ^{14}C atoms produced per incident cosmic ray particle at the top of the atmosphere. The ^{14}C production rate, Q_λ (cm^2 sec^{-1}) at geomagnetic latitude, λ is then given by:

$$Q_\lambda = \pi \int_{T_\lambda}^{\infty} g(T) \cdot Y(T)\, dT \qquad (2)$$

In equation (2) T_λ is the vertical cut-off energy at the latitude λ, and g(T) • dT is the flux of cosmic ray particles between kinetic energies, T and T + dT. Relation (2) is too over-simplified to describe the actual case in which must be considered 1) the yields separately for all components of cosmic radiation, 2) the cut-off energy as a function of zenith and azimuth angles. However, no appreciable error is made in deducing Q_λ values using relation (2) if the values of Y(T) themselves are derived in an identical manner; the values of Y(T) are then merely operational values and not true ^{14}C yields. Appendix 1 presents the method of obtaining "operational" values of Y(T) based on estimated ^{14}C production rates as a function of latitude during (1953-1954)/1965 and (1957-1958)/1969 corresponding to ϕ = 450 and 900, respectively; *cf* equation 1 and figure 3. Latitudinal Q values for these periods are based on data of Lingenfelter (1963), Light and others (1973) and Korff and Mendell (1980).

Based on the estimated operational values of Y(T) given in figure 4 and g(T) values with ϕ = 0, 100, 300, 450, and 900 as given in figure

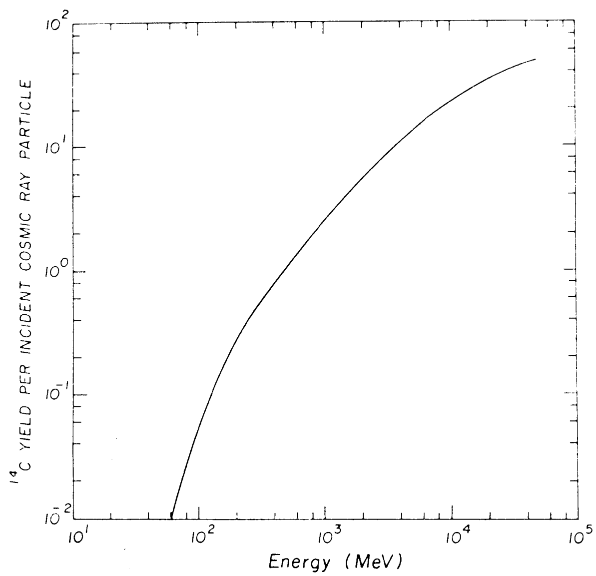

Fig 4. Yield of ^{14}C per incident cosmic ray particle as a function of energy. See text for definition of yield which includes leakage of neutrons in the upper atmosphere.

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3, we have estimated Q_λ, *ie,* Q_λ ($>T_\lambda$), for present geomagnetic dipole field (M_o). These values are given in figure 5 alongside vertical cut-off values at six latitudes for M/M_o = 0.1, 0.25, 0.5, 1.0, 2.0, and 4.0. The same information is plotted in figure 6. In figure 6A and B, latitudinal Q_λ values for $M/M_o = 1$, and $\lambda = 20°, 30°, 40°, 50°, 70°$, and the global average Q values for several M/M_o values are respectively given as a function of ϕ. The ϕ dependence of the global Q value for $M/M_o = 1$ is well represented by the relation:

$$Q\ (M/M_o = 1) = 3.2 - 1.2 \times 10^{-3}\ \phi\ (\mathrm{cm^2\ sec^{-1}}) \qquad (3)$$

As stated earlier, we may assume that the value of ϕ during Maunder Minimum was close to $\phi = 100$. Consequently, the extreme global ^{14}C production rates corresponding to $\phi = 100$ and 900 are 3.08 and 2.0. The amplitude around the mean value (2.54 ± 0.54) is ± 20 percent. This amplitude is quite adequate to explain the ^{14}C wiggles if the modulation occurs with a period of ~ 100 to 300 years. It also becomes clear from figure 6b that the amplitude in Q for an identical variation in ϕ is larger for lower values of M/M_o. Since the geomagnetic field decreased steadily between about 1000 to 8000 years BP, the amplitude of the wiggles should progressively increase in the past. The effect is, however,

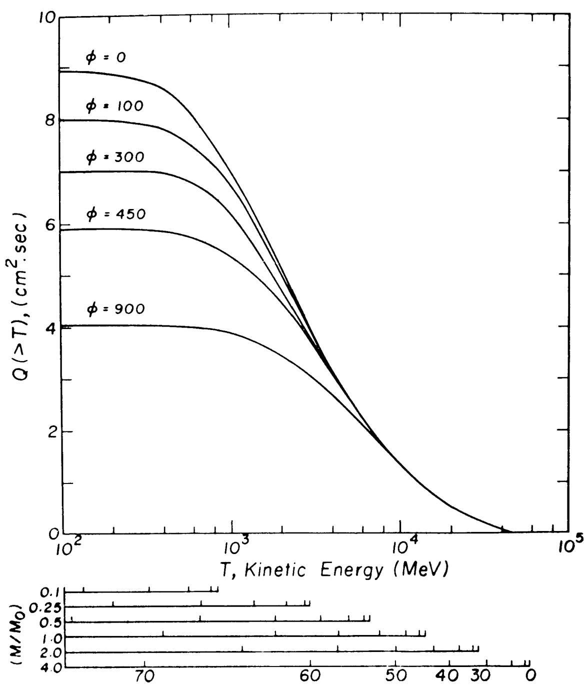

Fig 5. Calculated production rates of ^{14}C due to all cosmic ray particles above a certain energy for different values of the parameter ϕ (eq 1). The vertical cut-off energies for protons for geomagnetic latitudes 0°, 30°, 40°, 50°, 60°, and 70° can be seen from the scale given for different geomagnetic field strengths.

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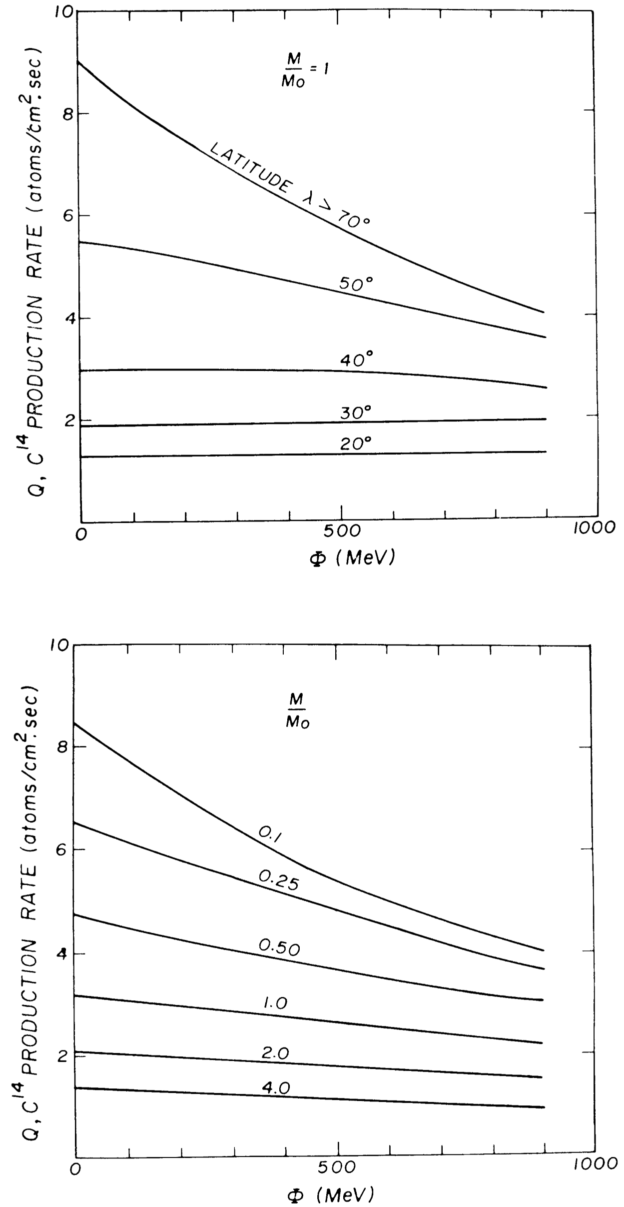

A.

B.

Fig 6. Latitudinal ^{14}C production rates for present geomagnetic field strength ($M/M_o = 1$) are given as a function of ϕ for geomagnetic latitudes 20°, 30°, 40°, 50°, and $\geqslant$ 70° (A) based on data in figure 5. In (B) the global average ^{14}C production rates are given for different M/M_o values as a function of ϕ.

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not very large and needs to be ascertained carefully in the tree-ring record. Theoretically, the amplitude is expected to increase by 25 percent only as the magnetic field reduces to 50 percent of its present value.

Variations in global ^{14}C production arise mainly from changes in the flux of medium energy particles of kinetic energy $\sim$ 1 BeV. Modulation is largest for the lowest energy particles (fig 3) but low energy particles are not effective for two reasons. First, the ^{14}C yield drops monotonically with energy (fig 4) and second, the low energy particles are incident only on a smaller area of the earth (at high latitudes where they are not excluded by magnetic screening). The appreciably higher ^{14}C rate expected during periods of very low solar activity, particularly the Maunder Minimum, was suggested earlier by Simpson (Lal, 1965, p 90).

Production of ^{14}C by solar flare particles

As discussed earlier, the primary nuclear source of ^{14}C in the earth's atmosphere is the reaction ^{14}N (n, p) ^{14}C. To a first approximation, solar flare production of ^{14}C is given by the total number of neutrons produced by interactions of the solar flare particles. Since the solar flare spectrum is soft (see fig 8) and proton is the abundant constituent, neutron production is given approximately by considering the individual neutron producing reaction, (p, n), (p; xp, yn) in N and O; *ie,* production of neutrons due to fast secondary neutrons can be neglected since we are primarily dealing with a beam composed of tens of MeV protons. The present problem, then, is reduced to calculating the yield of neutrons per proton of kinetic energy, T in nuclear interactions in the atmosphere before it is brought to rest by ionization losses. We made these estimates guided by the measured cross-sections for the reactions (p, n), (p, pn) and (p; xp, yn) reactions in light nuclei (Wong and others, 1961; Audouze, Epherre, and Reeves, 1967; Tobailem, de Lassus, and Levesque, 1971; Lorenzen and Brune, 1974; Ramaty, Kozlovsky, and Lingenfelter, 1975; de Lassus, St-Genies, and Tobailem, 1972; 1977).

Based on the neutron producing excitation functions mentioned above, we have estimated the neutron yield per proton, $Y_n(p)$ as a function of energy. For convenience, we have translated available data on neutron modulation cross sections as a function of residual range of proton in air (fig 7). In a narrow energy interval, the neutron yield is well approximated by the following relations in kinetic energy/rigidity:

$$Y_n(p) = 2.6 \times 10^{-7} T^{2.53} \quad (4)$$

or

$$Y_n(p) = 1.4 \times 10^{-14} R^{4.63} \quad (4)'$$

where T and R are in MeV and MV, respectively. These relations are valid in the kinetic energy interval (10-200 MeV; at higher energies, the yield drops below that predicted by the power law. The estimated neutron yields differ somewhat from those estimated earlier by Lingenfelter and Ramaty (1970).

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The production of ^{14}C by solar flare protons is estimated by the equation 4 in T, or the corresponding expression in R. The time averaged solar flare cosmic ray spectra are well represented over a wide range of energies by a single exponentional rigidity spectrum of the type:

$$dN = \text{const } e^{-R/R_o} dR \tag{5}$$

where R_o is the characteristic slope (Lal and Venkatavaradan, 1967; Lal, 1972; Reedy, 1977). Pomerantz and Duggal (1974), Lanzerotti (1977), and Duggal (1979) have recently reviewed observations of solar cosmic rays during solar cycles 18 through 20. The long-term average fluxes based on lunar studies correspond to $R_o = (100\text{-}150)$ MV and $N > 137$ MV = 100 protons/cm^2 sec; this is the 4 π omni-directional flux in space averaged over the past several million years (Wahlen and others, 1972; Bhandari, Bhattacharya, and Padia, 1976). In figure 8, we have plotted the range of proton energy spectra based on the lunar data cited above, along with the galactic cosmic ray proton spectra for $\phi = 0$, 100, and 900, to allow an intercomparison of the fluxes. It is seen that on a time-averaged base, solar protons are significant only at kinetic energies below a few hundred MeV.

It is clear that the neutron yield will depend very sensitively on the characteristic slope, R_o. Since the spectral shape varies significantly from event to event (Reedy, 1977), we have estimated the neutron yields as a function of R_o keeping the total flux above 10 MeV kinetic energy (R = 137 MV) constant at 100 protons/cm^2 sec (omnidirectional flux).

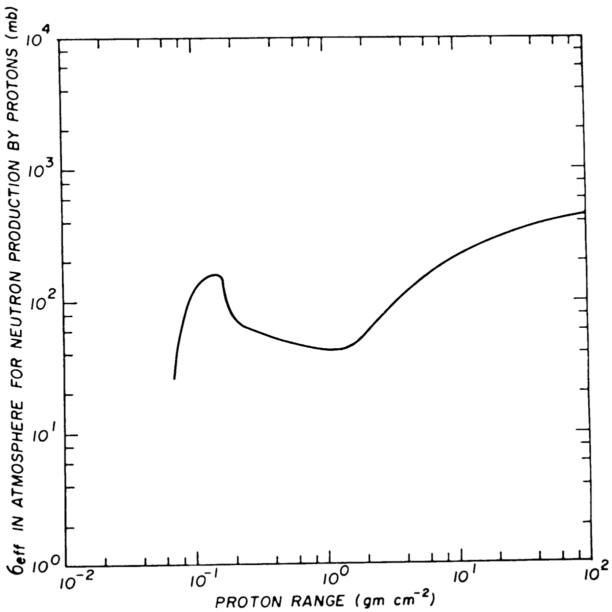

Fig 7. The effective neutron production cross-section as a function of range for protons in air. See text for cross-section data.

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The calculations are given in figure 9 where the total number of neutrons produced/cm^2 sec due to solar protons of rigidity exceeding R is given, for four R_o values in the range 50-400 MV. Based on the vertical cut-off rigidity as a function of geomagnetic latitude ($R_\lambda = 1.49 \times 10^3 \cos^4\lambda$MV), the estimated surface area averaged production rate on the earth between (0°-λ°) is given in figure 10. Only the neutron production rate is given in figure 10. The production rate of ^{14}C will be less than this because of loss of neutrons through leakage; also an appropriate factor for the capture of neutrons by ^{14}N must be considered. However, the calculations in figures 9 and 10 are approximate and should merely serve as a comparison between the relative contribution of solar particles to ^{14}C production and that due to galactic particles. Earlier calculations of solar flare ^{14}C production were made by Lingenfelter and Flamm (1964).

In the above, we have presented estimated ^{14}C production rates on the earth for certain assumed spectra and flux of flare particles. In

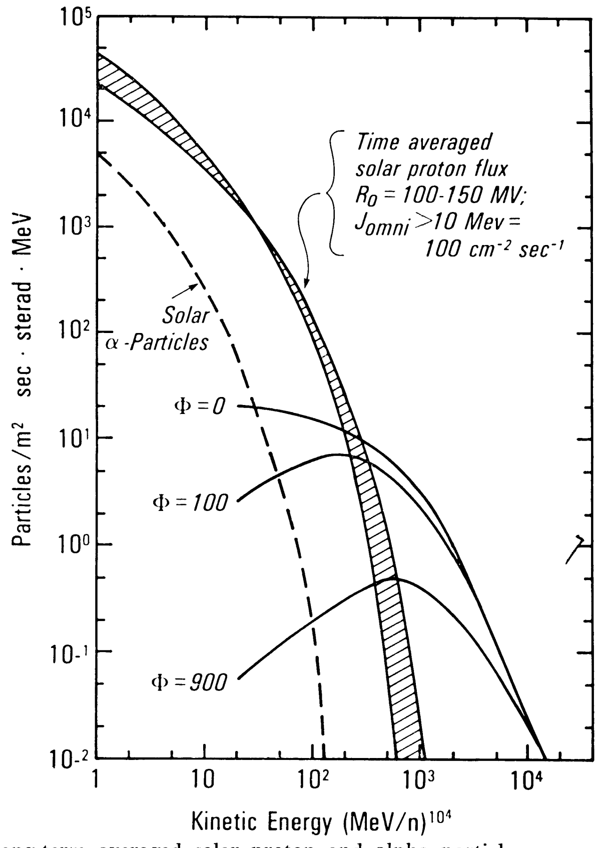

Fig 8. Long-term averaged solar proton and alpha particle energy spectra based on lunar data and galactic cosmic ray proton spectra for different modulation levels ($\phi = 0$ refers to no modulation).

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order to make meaningful predictions of the secular variations due to flare production of ^{14}C in the past, one must understand the basic processes involved. The detailed analysis of 185 solar events with respect to fluxes and spectra and heliolongitude of the flare by Van Hollebeke, Ma Sung, and McDonald (1975) is a case in point. The authors clearly indicate that a preferred connection region exists with the sun for events seen at the earth, 20°-80°W heliolongitude. Cosmic ray production by flares cannot be adequately understood from our small statistical sample and furthermore, it seems that solar fluxes at 1 AU may, in fact, be quite different from those at 3-4 AU, the primary place of irradiation of meteorites.

Meteorite or lunar data (fig 8) provide time-integrated averages of solar flare production. Qualitatively, for example, if during the past

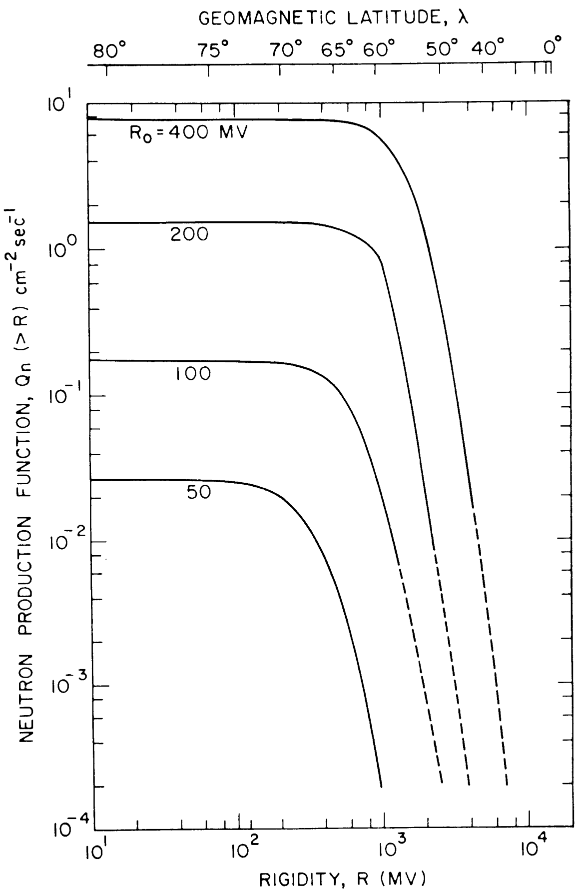

Fig 9. Neutron production rate per cm^2 column due to protons above a certain rigidity for an exponential proton rigidity spectra is plotted as a function of R. Irrespective of spectral shape, characterized by R_0 parameter, the number of protons greater than 10 MeV has been kept fixed at 100 protons/$cm^2 \cdot$sec (omnidirectional value).

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1 million years (corresponding to information based on ^{26}Al in lunar samples) there were 10 grand maxima of 100 years' duration, each with average cosmic fluxes of $10^2 \times$ that during the normal period, this enhanced flux would reflect only a ten percent contribution to the grand average. Enhanced production would, however, clearly be seen in a differential record as that of tree rings.

Variation in flux from one solar cycle to another is quite appreciable. During solar cycle 19 (1954-64), the total proton fluence above 10 MeV was five times that during solar cycle 20 (1965-75). The cycle 20 flux corresponds closely to the million year average flux. Moreover, it must be noted that most of the solar cycle 20 protons were accelerated in August 1972 events (Reedy, 1977). Dicke (1978) recently discussed the character of the sunspot cycle based on solar-terrestrial weather studies.

It is premature to anticipate temporal changes in the solar cosmic ray particle production rate particularly when solar activity was high.

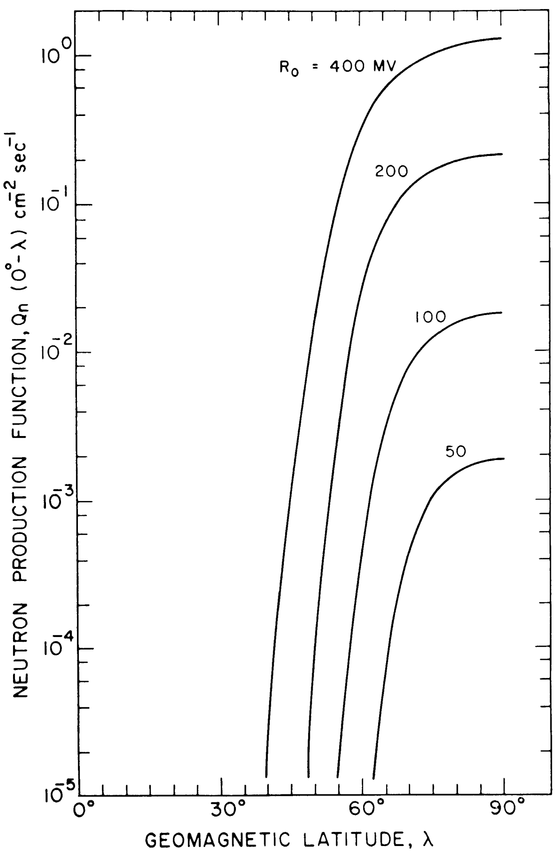

Fig 10. The integrated average neutron production rate per cm^2 column on the earth between (0° — λ°), based on calculations presented in figure 9.

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But there does seem to be a good positive correlation between solar activity and solar cosmic ray production (Lanzerotti, 1972).

In addition to uncertainties discussed above, the calculation of ^{14}C flare production is somewhat uncertain because changes take place in the magnetic environment of the earth following flares in which the cut-off with rigidities are appreciably altered (Yoshida, Akasofu, and Kendall, 1968; Tanskanen, Kananen, and Blomster, 1973; Agrawal and others, 1974; Rao, 1976).

Ring current effects on cosmic rays have been studied by Yoshida, Akasofu, and Kendall (1968). They showed that during geomagnetic storms, the ring current decreases the cut-off rigidity which leads to a local increase in the cosmic ray flux. Of particular interest to us are special situations such as the August 1972 flares when four major proton flares occurred during August 2-9. Agrawal and others (1974) reported that the superimposed intensity increase between 0400 and 0800 UT on 4th August for stations with cut-off rigidity exceeding 1.4 GV, was probably due to the lowering of cut-off rigidity due to a geomagnetic storm leading to decrease of 200 gammas in H at low latitudes. During the Solar Grand Maximum, therefore, successive flares occurring within a few days could possibly be associated with appreciably higher global production of ^{14}C by flare particles due to lowering of cut-off rigidity at low latitudes.

At present, only rough guesses can be made about the solar flare production of ^{14}C in the past. It cannot be ruled out (fig 10) that there have been periods in the past when appreciable solar flare production occurred. If we make the assertion that the most intense flares occurred only during high solar activity periods, then probably the decreases due to solar modulation are more significant than the increases due to flare production.

Accretion of ^{14}C by the earth

^{14}C could be accreted by the earth along with solar wind or high energy galactic solar cosmic ray particles. For the latter, only low energy particles with residual ranges much smaller than the mean free path for nuclear interaction would survive as ^{14}C nuclei. In this section, we present upper limits for the rate of ^{14}C atoms accreted by the earth from these sources.

In the case of solar wind, fairly good upper estimates can be derived from the results of Begemann (1972), and Fireman, DeFelice, and D'Amico (1976; 1977) who found that the lunar rock surfaces had excess ^{14}C compared to what was expected to be produced by galactic and solar cosmic ray particles. The excess, if attributed to implantation by solar wind, corresponds to a ratio of $^{14}C/^{1}H \cong 6 \times 10^{-11}$. Lingenfelter (pers commun) has shown that if solar ^{14}C was continually produced in the solar photosphere due to neutron capture and/or due to spallation of ^{16}O, one would expect a substantially higher quiet time flux of 2.2 MeV and 4.4 MeV gamma rays, respectively, than observed. The theoreti-

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cally expected fluxes are, in fact, even higher than those observed during the August 1972 flares.

However, even taking the $^{14}C/^{1}H$ ratio of 6×10^{-11}, one obtains a value of $< 2 \times 10^{-2}$ ^{14}C atoms/cm^2 • sec for the solar wind transported flux at 1 A U, assuming an average flux of 3×10^8 protons/cm^2 sec in the solar wind. The global average influx of solar wind ^{14}C accreted by the earth would be lower by at least a factor of 10^3, considering the fraction of the earth's area in the polar regions where solar wind penetrates. The global average accretion rate of ^{14}C is, therefore, $< 2 \times 10^{-5}$ ^{14}C atoms/cm^2 sec, which is smaller by a factor of 10^5 compared to its *in situ* production in the atmosphere by the galactic cosmic radiation.

In the case of galactic cosmic radiation, an upper limit of ^{14}C in the cosmic ray beam can be obtained by considering a model that allows a maximal production of ^{14}C in the cosmic ray beam during its traversal in the galaxy. The ratio $^{14}C/p$ is estimated to be $< 10^{-4}$ for energies below 1 BeV, considering production of ^{14}C is spallations of cosmic ray oxygen nuclei with hydrogen in the interstellar medium. Even assuming that all of the ^{14}C in the primary cosmic ray beam is brought to rest without suffering any nuclear reactions, the maximum amount of ^{14}C accreted with the galactic cosmic ray beam is $< 10^{-6}$ ^{14}C/atoms/cm^2 sec, which is lower by six orders of magnitude compared to its *in situ* production in the atmosphere by the galactic cosmic radiation. A similar conclusion is derived for the case of solar cosmic radiation.

Thus, it seems that the accretion of ^{14}C by the earth is not of any consequence compared to its *in situ* production in the atmosphere by galactic and solar cosmic ray nuclei.

CONCLUSION

The main aim of the present paper has been to explicitly consider whether the observed high frequency ^{14}C wiggles of 200 years in the $^{14}C/^{12}C$ ratios, R, in the atmosphere could be understood in terms of secular variations in *in situ* ^{14}C production in the earth's atmosphere. It is amply recognized that the ratio R depends not only in global ^{14}C production rate, Q, but also on a number of geophysical parameters, *eg*, climatic fluctuations, which may change the rate of mixing within and between dynamic carbon reservoirs. This aspect is not considered here. We merely wish to understand the wiggles in terms of secular variations in the production rate of ^{14}C in the earth's atmosphere. This question is in turn related to secular variations in the primary galactic cosmic ray flux outside the heliosphere, secular variations in solar activity which modifies the heliosphere itself and the degree of cosmic ray modulation at 1 AU, and finally, the production of cosmic rays by the sun. We have also considered whether an appreciable amount of ^{14}C could be accreted by the earth as a result of influx of solar wind and cosmic ray particles. We find that this mechanism is not an important one.

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Until only recently we could not make accurate predictions of secular changes in Q (*cf* Lingenfelter, 1963; Lingenfelter and Ramaty, 1970). As a result of work in the last decade, we know more about secular variations in solar activity and modulation of cosmic rays and feel less confident about making accurate predictions. Only a decade ago cosmic ray physicists assumed that the sun was well behaved and sunspots were fairly good indicators of solar plasma modulation effects. But now astronomers, astrophysicists, and cosmic ray physicists have independently learned otherwise. Unusually large fluctuations in sunspot numbers has been eloquently discussed by Eddy (1976; 1977).

The behavior of the sun is extremely relevant to the ^{14}C wiggles. The sun accelerates particles to energies sufficient to produce ^{14}C on the earth, and the solar plasma modulates the galactic cosmic ray flux in interplanetary space. The sun, therefore, both attenuates and augments the cosmic ray flux. The range of energies over which the two processes occur are not the same, and the two processes occur in opposition. During periods of higher solar activity, the galactic flux is further depressed, and acceleration of fast particles by the sun occurs preferentially in periods of high solar activity. Numerically, the sun gives back more than it modulates during periods of high solar activity, (see, eg, fig 8), but in effect, the ^{14}C production on the earth decreases during periods of high solar activity because of a substantial depression of the galactic cosmic ray flux of energetic particles of energies below (1-2) BeV. Thus, when the sun becomes active a decrease would be generally expected in ^{14}C production on the earth (see figs 3, 9, and 10).

As discussed above, we feel that during long periods of low solar activity (eg, the Maunder Minimum), the cosmic ray spectrum may be characterized by $\phi = 100$ (eq 1, fig 3). We have shown that such cosmic ray spectra if incident on the earth for periods of the order of decades or centuries can easily explain the observed ^{14}C wiggles. Forman, Schaeffer, and Schaeffer (1978) have also studied galactic ^{14}C production during the Maunder Minimum. They have not given the ^{14}C production rates, but, based on observations of ^{39}Ar activities in meteorites they have concluded that ^{14}C production on the earth was probably sufficiently augmented during the Maunder Minimum to explain the wiggles in ^{14}C data.

In this paper, we have also presented estimates of the production rates of ^{14}C due to solar flares for different spectral shapes of accelerated particles. For earlier calculations of flare production of ^{14}C, see Lal and Peters (1962) and Lingenfelter and Flamm (1964). It is clear from the present work (figs 9 and 10) that sufficient ^{14}C could be produced in single flares to appreciably change the atmospheric $^{14}C/^{12}C$ ratio. We, of course, do not know the probability with which hard flares (high intensity) occur. Meteorite and lunar data, unfortunately do not provide this information. Some theoretical work on the subject has recently been done (King, 1974).

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Our calculations show that if the sun was active or quiet for long periods of time, of the order of several decades, or a few centuries, corresponding secular changes in ^{14}C production rate would be large enough (± 20 percent from the mean) to explain the observed ^{14}C wiggles. The observed excursion in R during the Maunder Minimum period (Stuiver, 1978) can be understood very well in terms of persistent irradiation of the earth by a weakly modulated cosmic ray flux ($\phi = 100$) for about a century.

We have shown that, generally, the flare production of ^{14}C on the earth is not important but that one or a few intense solar flares with high R_0 value could produce an appreciable change in the atmospheric $^{14}C/^{12}C$ ratios. It should be possible to see such solar flare effects in careful ring by ring analyses.

During the past few decades, several indices have been used for solar activity, sunspot numbers, and geomagnetism. Sunspot numbers have been shown to be inadequate. Geomagnetic indices may be more meaningful (Stuiver and Quay, 1979, 1980; Feynman and Crooker, 1978) since modulation occurs through solar magnetic fields. However, in our opinion, use of the index is limited because the value of the index is not known for the past to be useful for a prediction. And predictions are not necessary for the future.

In summary, the results of the present investigations lead us to the conclusion that high frequency temporal changes in ^{14}C activity in the terrestrial atmospheric reservoir (de Vries oscillations, recently termed Suess wiggles) are primarily of solar origin. We have shown that change in solar activity directly produces change in the production rate of ^{14}C on the earth, but this change is due primarily to modulation in the flux of galactic cosmic ray particles streaming into interplanetary space. The solar flare production of ^{14}C is generally negligible. Our work provides a physical basis for the postulate made by Stuiver (1961) and Eddy (1976; 1977) that observed secular variations in $^{14}C/^{12}C$ ratios in tree rings reflect on variations in solar activity in the past. We, therefore, hope that in the near future, closer examination of the tree-ring record of $^{14}C/^{12}C$ ratios will result in a chart of solar activity in the past 10,000 years.

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APPENDIX

In situ production of ^{14}C in the atmosphere and the yield function of ^{14}C

The primary ^{14}C producing nuclear reaction in the atmosphere is the exothermic reaction, ^{14}N (n, p) ^{14}C which has a large cross-section at thermal energies (1/v behavior with resonances in the MeV region). The production of ^{14}C as a neutron slows down by successive interactions occurs primarily at thermal energies. ^{14}C is also produced in interactions of high energy protons and neutrons ^{16}O (p; 3p) ^{14}C, ^{16}O (n, 2pn) ^{14}C and the lesser important nuclear reactions with less abundant target nuclei in the atmosphere, ^{15}N (p, 2p) ^{14}C and ^{15}N (n, pn) ^{14}C, and in complex spallations of Ar, Ne, etc. These are all low-threshold nuclear reactions, important only at low energies. Since neutrons dominate the secondary cosmic ray beam at low energies, the principal ^{14}C producing mechanism is the ^{14}N (n, p) ^{14}C reaction. (High energy neutrons produce further neutrons in nuclear reactions; any neutron produced is slowed down as a result of inelastic collisions and it mostly ends its life producing a ^{14}C nucleus by the (n, p) reaction with ^{14}N). In the upper and lower regions of the atmosphere, some neutrons escape into the atmosphere before being thermalized. These processes are well known and detailed estimates exist for the production, slowing down, and loss of neutrons as a function of latitude (see Lingenfelter, 1963; Merker and others, 1973; Light and others, 1973, and Mendell and others, 1973).

Average yields of ^{14}C as a function of energy

It would be impossible to estimate with any precision the ^{14}C production rate in the atmosphere were it not for the fact that world wide distribution of atmospheric neutrons has been determined (Light and others, 1973). Pioneering work in this field has been done over more than two decades by S A Korff and his collaborators (see Korff and Mendell, 1980). As stated above, the loss of neutrons due to leakage from the upper atmosphere before being captured by ^{14}N must be considered. These estimates have been made very successfully (Lingenfelter, 1963; Light and others, 1973). Although considerable (ca 20%) uncertainties remain in absolute production rates of ^{14}C, relative latitudinal production rates as well as secular variations are better known. Galactic cosmic ray ^{14}C production rates for different latitudes given by Lingenfelter (1963) and Korff and Mendell (1980) for solar active and solar quiet periods are in fair agreement.

The present work seeks to determine relative production rates of ^{14}C during different epochs characterized by different cosmic ray spectra, such as given in figure 3. For this purpose the latitudinal data for ^{14}C production (cm^{-2} sec^{-1}) is used to obtain operational values of the yield parameter Y(T) in eq (1) in the text. The functional behavior of g(T) is known for the epochs considered for which ^{14}C production rates have been estimated by Lingenfelter and Korff. The functional behavior of Y(T) was assumed to be a power law within a narrow energy interval defined by the vertical cut-off energies $T_{\lambda i}$ and $T_{\lambda j}$, between successive latitude intervals considered in the computations. Initially, a value of the slope was assumed for the power law dependence of Y ($Y = const\ T^{s}$) and then the correct value of both the slope and the constant were determined by iteration. This method resulted in satisfactory estimates of the parameter Y(T), for energies above 300 MeV (fig 4). At lower energies, we estimated the ^{14}C yields per cosmic ray particle of energy T on the basis of available cross-section data. ^{14}C production due to alpha particles and heavier cosmic ray nuclei must also be considered. The yield of neutrons for alpha particles has been estimated by Lingenfelter and Ramaty (1970). On the basis of estimated neutron yields for protons (fig 7) and heavier particles, we have determined effective operational values for the yield parameter Y(T) above T = 100 MeV. These values are given in fig 4. Galactic production of ^{14}C for energies below 100 MeV kinetic energies is unimportant.

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