Energy Spectrum of Fast Cosmic-Ray Neutrons near Sea Level*

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(Received 27 April 1967)

Measurement of the sea-level spectrum of cosmic-ray neutrons in the energy region 0.05–2.0 MeV has been carried out with 4π hydrogen-recoil proportional counters. Electronic pulse-shape discrimination has been utilized to reject meson- and photon-induced events. In general, the flux per unit energy decreases monotonically with increasing energy. The experimental data reveal some structure which may be due to scattering resonances in elements that dominate the environment. An integral flux of 2.3×10^{-3} neutrons/cm^2 sec in the interval 0.05–2.0 MeV has been obtained for the air-land interface at the Argonne National Laboratory site (53° N geomagnetic latitude).

INTRODUCTION

It was only a few years after the discovery of the neutron by Chadwick[1] that the first investigations of cosmic-ray neutrons commenced.[2–4] Since that time a significant experimental effort has been expended in obtaining the cosmic-ray neutron intensity, in particular, as a function of altitude and latitude. Summaries of this important and extensive work can be found in the reviews of Lingenfelter[5] and Haymes.[6] Subsequently, some attention has been given to the nature of the energy distribution of these neutrons.[7–16]

This experimental effort has been complemented by theoretical descriptions which date back to the work of Bethe, Korff, and Placzek.[17] Later treatments have employed more sophisticated techniques (such as the elaborate numerical procedures developed in nuclear reactor computer calculations) in an effort to obtain (approximate) solutions of the neutron transport equation in the earth's atmosphere.[18–20]

In the lower-energy region, measurements of the cosmic-ray neutron-energy distribution have lacked quantitative detail. To this end, a sequence of experiments has been conducted in an effort to determine the energy spectrum below 2.50 MeV near sea level. A preliminary result of these measurements has already been reported.[21]

The present observations extended from June through October, 1966, at Argonne National Laboratory. This site possesses the following approximate characteristics:

(1) Altitude: 200 m above sea level
(2) Latitude: 42° N
Longitude: 88° W
(3) Geomagnetic latitude: 53° N

In the next section, the experimental method employed in these measurements is described. The following section contains the experimental results so obtained. A comparison of the present results with both theory and the results of other measurements is presented in the concluding section.

EXPERIMENTAL METHOD

Hydrogen-recoil proportional counters were employed in the present experiments. Two cylindrical proportional counters, 5.72 cm in diameter, were constructed of 0.194-cm-thick stainless steel. The active length of each counter was approximately 1 m. A 0.00254-cm-diam stainless-steel wire served as the anode. The anode wire was centered in the cathode by means of hypodermic needles which were, in turn, positioned in ceramic seals at each end of the counter body. The geometric characteristics of these 4π-proportional counters are shown in Fig. 1.

Pulse-shape discrimination was utilized to reject photon- and meson-induced events.[22] In addition, the two proportional counters were centered in an anticoincidence annulus of Geiger counters. The rejection

* Work performed under the auspices of the U. S. Atomic Energy Commission.

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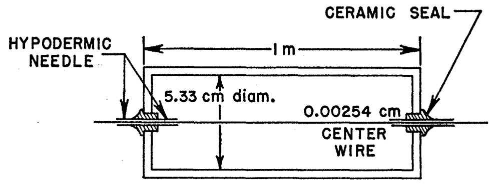

FIG. 1. Geometric characteristics of the 4π hydrogen-recoil proportional counter.

efficiency of this anticoincidence shield was roughly 20% for all of the neutron measurements.

The instrumentation employed is outlined in the logical block diagram of Fig. 2. The outputs of the fast (A) channel (risetime or specific ionization) and the slow (B) channel (energy or total ionization) are analyzed by two independent analog-to-digital converters (ADC). The results obtained from this (64×64) two-dimensional analyzer are punched onto paper tape. The punched paper tape serves as an inexpensive memory storage for the duration of a particular experiment. Upon completion of a given measurement, the paper tape is read directly into a computer for subsequent analysis and data reduction.[23]

A series of experiments were carried out from June–October, 1966. The pertinent characteristics of each experiment are summarized in Table I, which contains

TABLE I. General characteristics for experiments 1–3.

Experiment No.	Partial pressure (psi) CH_4	H_2	N_2	Energy region (MeV)	Live-time duration (sec)
1	45.0	···	0.20	0.500–3.10	2.8968×10^6
2	45.0	···	0.20	0.120–0.690	3.5307×10^6
3	4.0	41.0	0.75	0.040–0.144	2.8680×10^6

the counter gas filling employed, the corresponding live-time duration of the measurement, and the energy region explored. Experiments 1 and 2 employed a methane filling to examine the higher-energy regions during June and July–August, respectively. Hydrogen, with a suitable admixture of methane for quenching, was used in experiment 3 for the lower-energy region investigation during September–October.

A small amount of nitrogen, usually a partial pressure of less than 1 psi, was added to each counter filling. Introduction of this small amount of nitrogen serves as a convenient means of evaluating proportional counter operation. The counters were placed next to a small moderating assembly which contained an americium-beryllium neutron source ($\sim10^7$ n/sec). In this environment, monoenergetic protons from the slow neutron-induced $N^{14}(n,p)C^{14}$ reaction arise throughout the entire counter body.

Measurement of this monoenergetic event serves two important purposes. First, it permits an examination of the resolution of the counter as a test for proper quality of operation. Typical detector responses in this environment are displayed in Figs. 3(a) and 3(b) for the hydrogen and methane fillings, respectively. After subtraction of the proton-recoil continuum due to the flux of fast neutrons, one can obtain an estimate of the proportional counter resolution. The resolution of each counter was approximately 12% (full width at half-maximum, FWHM) for all of the experiments described in Table I.

The second important function provided by the $N^{14}(n,p)C^{14}$ reaction is the energy calibration of the detection system. This calibration can be derived from a knowledge of the effective Q of this reaction[24] and the gas gain of the proportional counter. In this treatment, the Diethorn formula for gas gain as a function of anode voltage has been employed. Experimental measurements and theoretical arguments have demonstrated that this formula provides a reliable description over limited regions of gas gain.[25]

In the energy region above approximately 0.5 MeV, distortion of the proton-recoil spectrum arises in these 4π-proportional counters due to wall and end effects. Since the radius-to-length ratio for these proportional counters is approximately 0.02, wall effect will clearly dominate over end effect. A wall- and end-effect response matrix has been obtained from measurements with a specially constructed aluminum proportional counter of the same radius, but possessing a radius-to-length ratio

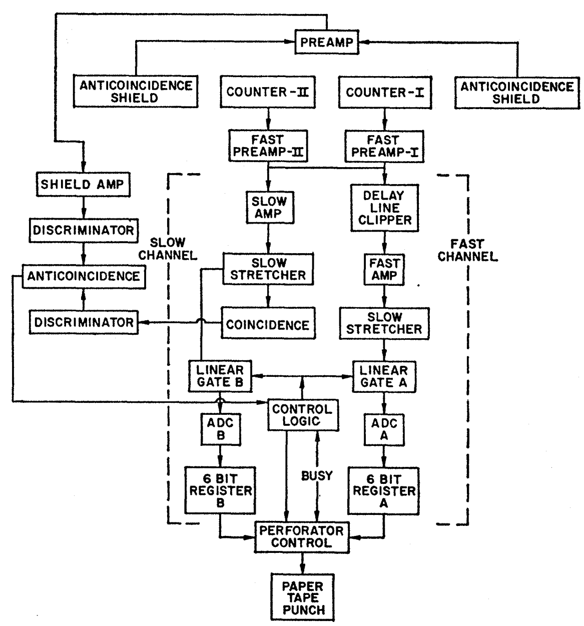

FIG. 2. Logical block diagram of the cosmic-ray neutron two-parameter analyzer.

[23] C. E. Cohn, Reactor Physics Division Annual Report, Argonne National Laboratory Report No. ANL-7310 (unpublished).

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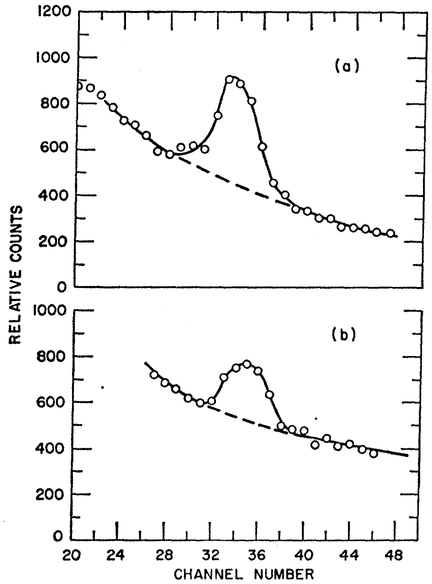

FIG. 3. Calibration response of the 4π proportional counters to the $N^{14}(n,p)C^{14}$ reaction for (a) counter I, experiment 3, 3200 V; and (b) counter II, experiment 1, 2800 V.

of 0.10. This special aluminum counter resulted from a compromise between conflicting requirements. Namely, it was used to approximate the geometry of the longer cosmic-ray neutron counters and at the same time it was short enough for practical use in measurements carried out in the thermal reflector of the Argonne Thermal Source Reactor (ATSR). In addition, employing this special aluminum counter avoided activation of one of the cosmic-ray neutron counters in a strong thermal flux. The background contribution due to such an activation could have otherwise been troublesome, since any given spectral measurement must be conducted over an extended live-time interval (cf., Table I). Using the response matrix obtained with this special aluminum counter, iterative unfolding was employed in an attempt to remove wall- and end-effect distortion from the measured proton-recoil data.[26] A complete description of this procedure will be published elsewhere.[27]

In contrast to the high-energy limitations, the low-energy limit for fast-neutron spectroscopy with 4π recoil proportional counters is normally about 1 keV.[22] Below this energy, the behavior of W (the average energy per ion pair) for protons is not well known. In addition, the measured pulse-height distributions become seriously broadened by statistics, since only a small number of ion pairs are formed per recoil event. While the same lower bound is probably applicable for cosmic-ray neutron measurements, the low-energy limitation in the present experiments was set by the limited capabilities of the punch-paper-tape memory system. For this mechanical system to provide reliable data over extended operating periods, count rates of no more than the order of 30/min could be handled confidently. In the region below 50 keV, the total (anticoincident) event rate in the two counters became too high to be properly handled by the punch-paper-tape system.

Employing alternative means of memory storage should avoid the limited capabilities of a punch-paper-tape system and permit extension of the present cosmic-ray neutron spectral measurements. Whether or not the normal low-energy limit can actually be attained in cosmic-ray measurements depends chiefly on the behavior of the background-event rate which increases rapidly with decreasing energy. For example, preliminary measurements (near sea level) in the energy regions 60–240, 120–480, and 300–1200 keV gave integral values for the background-to-signal ratio of roughly 500, 100, and 4, respectively. In view of this rapid increase in background, the electronic pulse-shape rejection method alone may not provide sufficient discrimination for reliable measurements down to 1 keV. There exists, however, the possibility of improving the over-all rejection efficiency of the system by employing an internal anticoincidence shield. Sikkema[28] has reported a rejection efficiency of over 95% for a large proportional counter with an internal anticoincidence annulus. Consequently, if the pulse-shape rejection method alone is not sufficient, a combination of discrimination methods should afford measurements down to the neighborhood of the 1-keV lower limit.

SPECTRUM MEASUREMENTS

The proton-recoil data obtained in experiments 1, 2, and 3 are presented in Figs. 4, 5, and 6, respectively. The error given with each datum point is simply that due to counting statistics. Each such datum point is the result of a two-dimensional analysis. To demonstrate how these spectra have been obtained, typical two-parameter analyzer data for each of these experiments are presented in Figs. 7(a), 7(b), and 7(c) for proton energies, $E_p = 800$, 306, and 84 keV, respectively. Here the measured cosmic-ray count-rate distribution is dis-

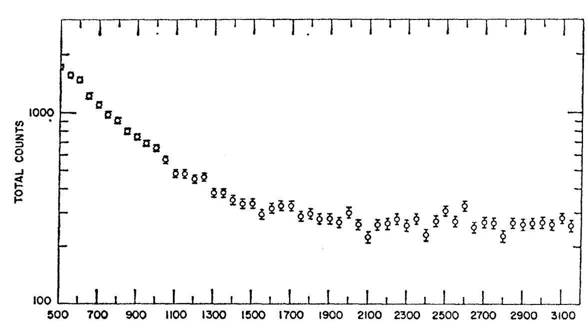

FIG. 4. The proton-recoil data of experiment 1 (0.500–3.10 MeV).

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played as a function of (A/B), i.e., the A (rise-time) channel divided by the fixed B (energy) channel of each respective measurement.

One finds, in general, two overlapping distributions in the experimental data. The more intense distribution, which rises sharply at low values of (A/B), is the background distribution. The less intense and broader distribution occurring at higher values of (A/B) is due to proton-recoil events. In order to determine a proper background subtraction, one must extrapolate the background distribution into the region occupied by the proton-recoil distribution. For the present cosmic-ray data, this procedure has been carried out graphically. Using a semilogarithmic display, the count-rate data are plotted as a function of (A/B). Then, as depicted in Fig. 7(a), 7(b), and 7(c), the background distribution is extended by a linear extrapolation of the data obtained at the lowest values of (A/B). To test the validity of this process, background distributions have been obtained with the aid of external γ-ray sources. If necessary these background distributions can also serve as a guide in the graphical extrapolation. Examples of these γ-ray-induced background distributions are also presented in Figs. 7(a), 7(b), and 7(c).

The neutron spectrum (neutrons per cm^2 per sec per unit lethargy) obtained from these proton-recoil data is presented in Fig. 8 as a function of neutron energy in keV. The approximate resolution for this result is about 30%, since a slope taking interval of 0.30 was utilized in the numerical differentiation process. The magnitude of the slope taking interval depends, in turn, upon the actual resolution of the detection system as well as the statistical quality of the experimental data. For the present case, the actual resolution is about 18 to 20%, corresponding to the simultaneous operation of counters I and II. Since the statistical quality of the present data can only be classified as fair, a slope taking interval of 0.30 is realistic.

The experimental error bars displayed in Fig. 8 include contributions from both counting statistics and the numerical differentiation process. It can be seen that the error bars, which range from about 20% at low energy to 10% in the mid-energy region, become very significant at the high-energy end of the spectrum. This result can be traced directly to the nature of the proton-recoil data above 2 MeV. Figure 4 reveals that the proton-recoil data above 2 MeV are beset with statistical fluctuations due to the low value of accumulated counts. This behavior can be attributed to a combination of decreasing proton-recoil cross section and decreasing neutron spectrum with increasing neutron energy in this region. A number of data points above 2.15 MeV actually correspond to small negative values of flux, but have been arbitrarily plotted at zero flux. Consequently, the data above 2.0 MeV, in Fig. 8, cannot be regarded as reliable.

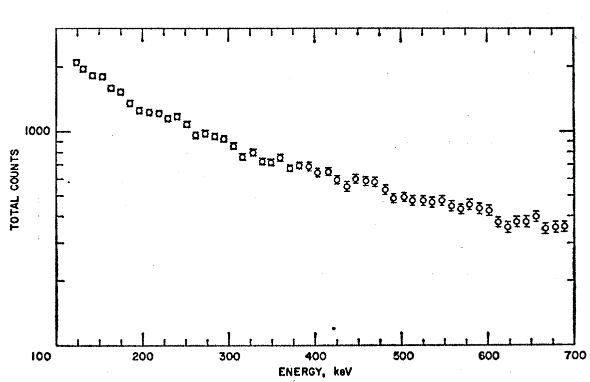

FIG. 5. The proton-recoil data of experiment 2 (0.120–0.690 MeV).

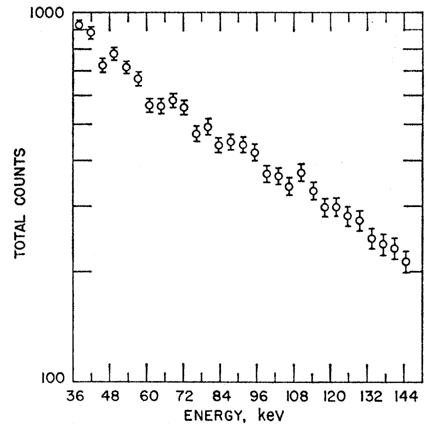

FIG. 6. The proton-recoil data of experiment 3 (0.040–0.144 MeV).

Attempts to unfold wall-effect distortion from the measured proton-recoil distribution (viz., Fig. 4) were unsuccessful. Here again, the reason could be traced to the poor statistical quality of the data. Indeed, in the very region that wall effect becomes more significant, the present data grow more unreliable. Since experimental error due to counting statistics is larger than this effect throughout the region of interest, the process of unfolding this effect cannot be considered meaningful for the present data.

In spite of this shortcoming, the present neutron-energy spectrum is considered reliable (within quoted experimental error) out to 2.0 MeV. This inference is based upon experience in unfolding wall and end effects in much smaller proportional counters, where these effects are more dominant than for the present case.[27] It has been generally observed that significant corrections can occur between the measured proton spectrum and the corrected (infinite-medium) proton spectrum. However, this correction is not reflected in the resulting neutron spectra. Indeed, aside from spectral detail, the uncorrected and corrected neutron spectra are close in shape, trend, and magnitude. Consequently, the neutron spectrum presented in Fig. 8 can be used with confidence below 2.0 MeV.

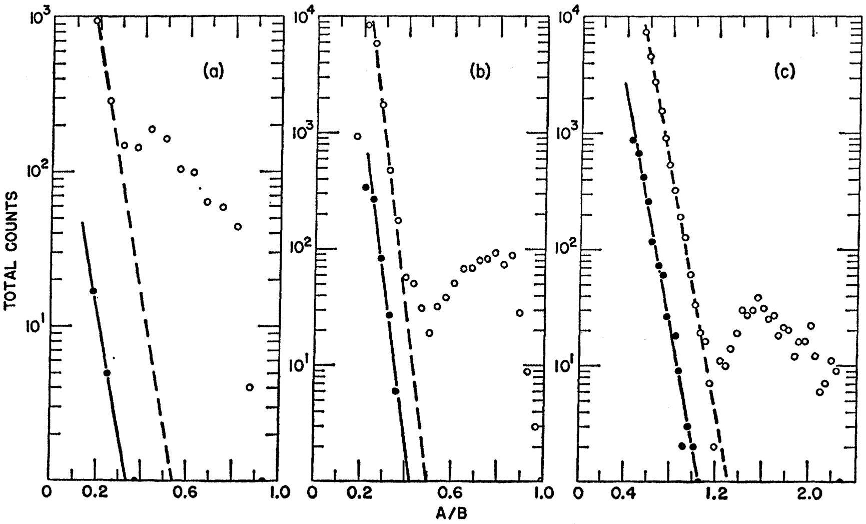

FIG. 7. Cosmic-ray count-rate distribution (open data points) and induced (γ-ray) background distribution (solid data points) as a function of (A/B) for fixed B: (a) experiment 1, $E_p=800$ keV; (b) experiment 2, $E_p=306$ keV; and (c) experiment 3, $E_p=84$ keV.

DISCUSSION

An examination of the neutron spectrum given in Fig. 8 reveals an apparent structure which may not be completely statistical in origin. Some of this structure is probably due to elastic scattering resonances in dominant elements in the environment of the air-land interface. For example, the pronounced spectral dip near 400 keV can be attributed to large scattering resonances in both the oxygen and nitrogen in the neighborhood of 430 keV. Similar structure, which occurs at lower energy, may correspond to scattering resonances in silicon.

The display of this spectrum in terms of the flux per unit lethargy ϕ_μ is quite convenient, since such a presentation immediately reveals the general behavior of the neutron-energy distribution. This follows from the fact that a constant flux per unit lethargy, i.e., $\phi_\mu=\text{const}$, implies a classical $1/E$ behavior for the flux per unit energy. Hence at lower energies (0.05–0.5 MeV), it can be seen that the spectrum decreases more slowly than $1/E$. In the mid-energy region (0.5–1.0 MeV) the qualitative behavior is more nearly that of the classical $1/E$ spectrum, whereas in the high-energy region (1.0–2.0 MeV) the neutron spectrum decreases much more rapidly.

The customary power-law representation of the neutron flux per unit energy ϕ_E, in the form

$$\phi_E=CE^{-\alpha}, \quad (1)$$

where C and α are constants, has been obtained for these regions. The results of a least-squares fit of the experimental data, in each of these regions, are summarized in Table II. These least-squares coefficients provide quantitative support for the general description based on Fig. 8, which was given above. It is evident that the spectrum is monotone decreasing throughout. Moreover, since α increases with increasing energy, the rate of decline in this spectrum also increases with increasing energy.

Comparisons with theory[18–20] are possible within the framework of one serious limitation, namely, that these calculations have been carried out to determine equilibrium spectra in the atmosphere, and therefore do not include boundary effects that may arise from the air-land interface. Below 1.0 MeV there is better agreement

TABLE II. Least-squares power-law coefficients.

Energy region (MeV)	C^a	α
0.05–0.5	$1.14\cdot10^{-3}$	0.492 ± 0.035
0.50–1.0	$1.12\cdot10^{-3}$	0.696 ± 0.134
1.0 –2.0	$1.27\cdot10^{-3}$	$2.81\ \pm0.191$

[a] Units of neutrons/[sec cm² (MeV)$^{1-\alpha}$].

with the S_n calculation of Newkirk[20] than the diffusion-theory calculations of Hess *et al.*[18] or Lingenfelter.[19] Above 1.0 MeV the converse is true. In either case, however, the agreement cannot be considered good. More specifically, while the "plateau" in the spectrum determined by the S_n approximation in the region 0.10–1.0 MeV is smaller than that predicted by diffusion theory, it is not reproduced in the present experimental data. The experimental flux per unit energy possesses a continuously monotonic decreasing behavior. Some

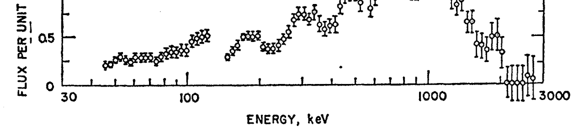

FIG. 8. Composite neutron spectrum in the energy region 0.050–2.50 MeV determined from experiments 1, 2, and 3. The customary definition of lethargy has been used, $\mu=\ln(E_0/E)$, where E_0 is an arbitrary reference energy.

source contribution from evaporation neutrons in this region is still possible, however, since there exists a clear deviation from the classical $1/E$ spectrum. Above 1 MeV, the spectrum obtained from diffusion theory decreases more rapidly than the S_n prediction. Nevertheless, the diffusion-theory result[19] does not decrease nearly as rapidly as the present experimental data.

A limited comparison is possible with other neutron measurements carried out at the air-land interface. The magnitude of the present measurement is lower than that found by Kastner *et al.*[12] and therefore considerably lower than that determined by Hess *et al.*[8] It should be noted, however, that these measurements focused on a different energy region, 1–10 MeV. The integral flux between 0.10–1.0 MeV can be compared with the value of 1.6×10^{-3} n/cm^2 sec obtained by Yamashita *et al.*[16] at 44° N geomagnetic. This value is consistent with an integral of the least-squares power-law representation [viz., Eq. (1)] of the present experimental data in the region 0.10–1.0 MeV, which yields 1.57×10^{-3} n/cm^2 sec. Over the entire interval of these measurements, 0.05–2.0 MeV, one finds an integral flux of 2.3×10^{-3} n/cm^2-sec.

ACKNOWLEDGMENTS

The author is grateful for the support and encouragement extended by the Radiological Physics Division of Argonne National Laboratory. The author is indebted to Dr. E. F. Bennett, Dr. C. E. Cohn, Dr. Y. Feige, and Dr. J. Kastner for many interesting discussions concerning various aspects of this experiment. Thanks are also due to R. Brenner and M. G. Strauss for assistance with the instrumentation and to R. Grajek and R. Hopf and Miss I. Olson for help with data collection and reduction.

PHYSICAL REVIEW VOLUME 165, NUMBER 5 25 JANUARY 1968

Radiocarbon Production Rate near Sea Level*

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(Received 27 April 1967)

The natural production of radiocarbon by cosmic-ray neutrons has been observed experimentally. Proportional counters, filled with 2 atm of nitrogen, have been employed to detect the exoergic $N^{14}(n,p)C^{14}$ reaction. Electronic pulse-shape discrimination is utilized to discriminate against meson- and photon-induced events. A C^{14} production rate of $(1.89\neq0.08)\times10^{-7}$/sec/cm^3 has been obtained for the air-land interface at the Argonne National Laboratory site. This result represents the average value for a time duration of one month, November–December, 1966. Fluctuations as large as 25% from this average value were observed. The density of slow neutrons can also be determined from this experimental result. An average value of $(1.19\pm0.06)\times10^{-8}$ neutrons/cm^3 was found for the air-land interface at the Argonne National Laboratory site (53° N geomagnetic latitude).

INTRODUCTION

THE world-wide inventory of radiocarbon has been of general interest since the original work of Libby.[1] An extensive list of investigations and calculations related to C^{14} production by cosmic rays can be found in the summary of Lingenfelter.[2] In this review, it is concluded that a nonequilibrium condition exists in the present world-wide inventory of radiocarbon. It is estimated, in fact, that the natural C^{14} production rate exceeds the natural decay rate by as much as 25%. This conclusion must, however, be considered tentative in view of the experimental error assigned to the production and decay rates, i.e., 20 and 10%, respectively. In fact, the more recent experimental work of Greenhill *et al.*[3] indicates a lower world-wide C^{14} production rate. While these measurements are consistent with an equilibrium condition, they also possess large relative error. Consequently, more precise determinations are necessary, especially for the production rate, before definitive conclusions can be reached. Moreover, more precise data could also serve as a quantitative test for proposed models that describe long-term changes in the earth's magnetic field.

Estimates of natural radiocarbon production have been deduced from a knowledge of cosmic-ray neutron intensity as a function of altitude and latitude. In a more complete treatment,[2] calculated cosmic-ray neutron equilibrium energy distributions as a function of altitude and latitude have been employed in an attempt to improve over-all accuracy.

* Work performed under the auspices of the U. S. Atomic Energy Commission.

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