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.

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Radiocarbon Production Rate near Sea Level*

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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 $\text{N}^{14}(n,p)\text{C}^{14}$ reaction. Electronic pulse-shape discrimination is utilized to discriminate against meson- and photon-induced events. A C^{14} production rate of $(1.89\pm0.08)\times10^{-7}/\text{sec/cm}^3$ 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.

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

[1] W. F. Libby, Phys. Rev. **69**, 671 (1946).

[2] R. E. Lingenfelter, Rev. Geophys. **1**, 35 (1963).

[3] J. G. Greenhill, J. Phillips, K. B. Fenton, A. G. Fenton, and M. Bowthorpe, in *Proceedings of the Ninth International Conference on Cosmic Rays* (The Institute of Physics and The Physical Society, London, 1965), Vol. I, p. 532.

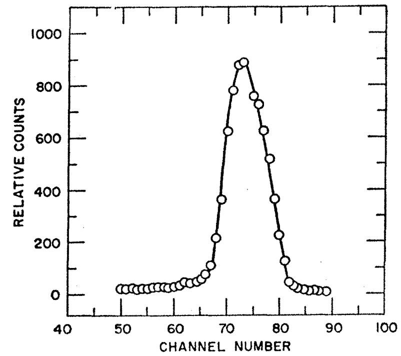

FIG. 1. Calibration response of counter I due to the $N^{14}(n,p)C^{14}$ reaction with a 30-psi nitrogen filling and an anode voltage of 3600 V.

The present effort is a direct experimental measurement of the C^{14} production rate at the air-land interface. The measurement described herein has been carried out at the Argonne National Laboratory site, which possesses the following approximate characteristics:

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

A preliminary account of this work has already been given.[4]

It will become clear, in the sequel, that the establishment of an experimental method can serve not only to complement calculation of the world-wide C^{14} production rate, but can actually go farther. In principle, the higher precision available with the present experimental method affords a determination of the world-wide C^{14} production rate with greater accuracy than has heretofore been possible.

The experimental method used to determine the C^{14} production rate is described in the section immediately below. The experimental results are then presented. The concluding sections contain an analysis and discussion of these experimental results.

EXPERIMENTAL METHOD

Measurements were carried out with the very same experimental apparatus used for the cosmic-ray neutron experiments described in the previous paper.[5] Figures 1 and 2 of that paper display the proportional counter detectors and instrumentation employed. In the neutron measurements, the utility of the $N^{14}(n,p)C^{14}$ reaction for evaluation of the detection system has already been demonstrated. In fact, the only change that actually need be made for observation of the C^{14} production rate is the gas filling of the proportional counters. Use of a pure nitrogen gas filling will permit observation of the $N^{14}(n,p)C^{14}$ reaction rate in a given environment and, consequently, a straightforward determination of the C^{14} production rate for that environment. For the present measurement a 30-psi nitrogen filling, in both counter I and counter II, was used.

Calibration of the proportional counters for this experiment followed the same procedure described in the previous paper.[5] Response distributions were obtained with the detection system placed next to the small, neutron-moderating assembly. Under these conditions, a typical pulse-height distribution obtained from the $N^{14}(n,p)C^{14}$ reaction is displayed in Fig. 1. Here the response of counter I is displayed for an anode voltage of 3600 V with the 30-psi nitrogen filling. The resolution of both counters was determined over a range of anode voltages and was found to vary from about 12 to 15% (full width at half-maximum, FWHM).

As in the neutron measurements, both the anticoincidence annulus and electronic pulse-shape discrimination were utilized to eliminate photon- and meson-induced events.[5] The capabilities of the pulse-shape rejection system are qualitatively depicted in Figs. 2(a) and 2(b). This figure contains two cathode-ray oscilloscope photographs obtained with counter II at an anode voltage of 3500 V. Photograph 2(a) has been obtained from a 1-min time exposure with the neutron source in the moderating assembly and exhibits the separation of the γ-ray-induced events from the proton events. Photograph 2(b) has been obtained from a 3-min exposure to an external Co^{60} γ-ray source and therefore represents the detection-system response to background events only. In this case, the γ-ray-induced events lie close to the origin of the two-dimensional $(A\times B)$ pulse-height space. (Here the A channel represents risetime or specific ionization and the B channel represents energy or total ionization.)

EXPERIMENTAL MEASUREMENT

The anode voltages of counters I and II were adjusted to yield a response peak at the midpoint of the B axis (pulse-height channel number $B=32$). Data were collected from mid-November until mid-December, 1966 at the Argonne site. The actual total live-time of the measurement was $T=2.2791\times10^6$ sec. The accumulated pulse-height distribution is given in Fig. 3, where each experimental point is the result of a two-dimensional $(A\times B)$ analysis similar to that used in the

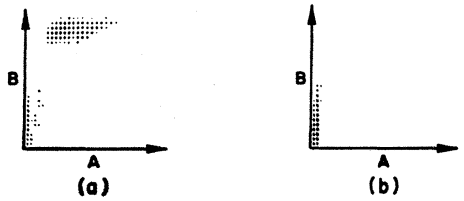

FIG. 2. Cathode-ray oscilloscope photographs of the two-dimensional (AXB) pulse-height response for: (a) $N^{14}(n,p)C^{14}$ reaction; and (b) Co^{60} γ source.

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[4] R. Gold, Bull. Am. Phys. Soc. **12**, 591 (1967).

[5] R. Gold, preceding paper, Phys. Rev. **165**, 1406 (1967).

neutron measurements. Here the error bars represent counting statistics only.

As an example of this treatment, Fig. 4 presents the count-rate distribution obtained for $B=31$, as a function of (A/B). This figure also contains a γ-ray background distribution induced with an external Co^{60} source. With the aid of this background distribution, an extrapolation (as depicted in Fig. 4) can be carried out to determine the proper background subtraction. Each experimental B-channel data point displayed in Fig. 3 has been determined in this manner.

ANALYSIS OF EXPERIMENTAL DATA

The nature of the experimental results in Fig. 3 reveals, in addition to the $N^{14}(n,p)C^{14}$ response peak, the existence of a background continuum. This continuum can be attributed to a number of effects. For the purpose of the present analysis, it is convenient to introduce the following classification of these effects:

Class 1: Fast-neutron-induced $N^{14}(n,p)^{14}$ reactions.
Class 2: Cosmic-ray events not rejected by pulse-shape discrimination.

An example of events that fall in *Class 2* would be N^{14} recoils from elastic scattering with high-energy cosmic-ray neutrons.

The dashed curve displayed in Fig. 3 is a linear representation of the background continuum. It has been obtained from a linear extrapolation of the data in the pulse-height region above the response peak. The smooth curve given in Fig. 3 is the result of a Gaussian fit of the experimental data above pulse-height channel No. $B=26$, after subtraction of the background continuum. The deviation of the experimental data from Gaussian behavior below a pulse-height value of $B=26$ can be attributed to wall effect. Experiments have been performed to determine the wall- and end-effect response matrix for these cosmic-ray proportional counters.[6] These measurements reveal that for this pressure filling and at this energy, the cosmic-ray counters possess a Gaussian response function of relative area 0.87. Consequently, division of an experimentally determined Gaussian area by this constant will account for losses due to wall effect.

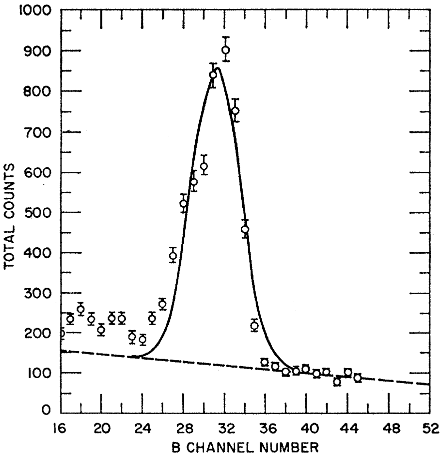

FIG. 3. Experimental pulse-height distribution obtained from mid-November to mid-December, 1966 at the Argonne site. The smooth curve is a Gaussian fit to the experimental data above the background continuum.

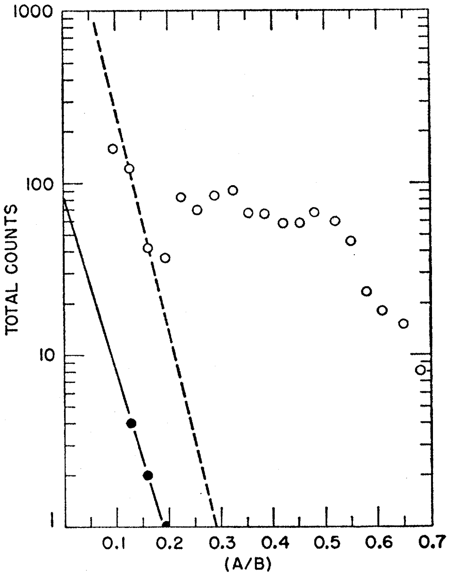

FIG. 4. The observed count-rate distribution (open circles) and the induced (γ-ray) background distribution (solid circles) as a function of (A/B) for $B=31$.

Employing this correction alone, one can determine the slow-neutron-induced C^{14} production rate. Here the distinction between slow and fast neutrons can be defined in terms of the proportional counter response function. The present detection system (i.e., simultaneous operation of counters I and II) possesses a total resolution of approximately 18% for the thermal neutron induced $N^{14}(n,p)C^{14}$ reaction. Since the Q of this reaction corresponds to an effective energy of about 0.6 MeV, it is clear that $N^{14}(n,p)C^{14}$ events induced by neutrons of up to roughly 0.1 MeV will not be distinguishable from thermal neutron-induced events. Hence, neutrons that possess an energy below 0.1 MeV will be considered slow. It also follows that the wall-effect correction discussed above will provide the slow-neutron-induced C^{14} production rate, i.e., the production rate due to neutrons below 0.1 MeV.

The slow-neutron-induced C^{14} production rate found in this experiment was $(4.75\pm0.19)\times10^{-27}$/sec/nitrogen atom or $(1.89\pm0.08)\times10^{-7}$/sec/cc for the ICAO (International Civil Aviation Organization) standard atmosphere.[7] This value, obtained near seal level, should only be attributed to the air-land interface. Both theory[8]

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[6] R. Gold and E. F. Bennett (to be published).

[7] R. C. Weast, S. S. Selby, and C. D. Hodgman, *Handbook of Chemistry and Physics* (The Chemical Rubber Publishing Co., Cleveland, Ohio, 1965), 46th ed.

[8] H. A. Bethe, S. A. Korff, and G. Placzek, Phys. Rev. **57**, 573 (1940).

and experiment[9,10] imply a different result for the air-water interface at sea level.

The experimental error given above is a composite of (Poisson) counting statistics and the error in the experimentally determined wall-correction factor. The relative error for the wall-correction factor is approximately 3%. For this measurement, the relative error due to counting statistics is close to 2%. Hence the total relative error for this result has been taken as approximately 4%.

Since events of *Class 1* have been subtracted from the analyzed data, it is clear that an independent estimate of C^{14} production due to fast neutrons is required. That this contribution cannot be large follows directly from the experimental evidence already displayed in Fig. 3. Here the approximate Gaussian decrease of the response peak for increasing pulse height implies that the relative fast-neutron contribution is not large. Quantitative estimates of this contribution can be obtained from the behavior of both the $N^{14}(n,p)C^{14}$ cross section and the cosmic-ray neutron flux above 0.1 MeV. The spectrum obtained in the previous paper[5] together with recent compilations of this cross section,[11] provide all the information necessary for such an estimate.

Integrating over the energy region, 0.1–2.15 MeV, one finds a fast-neutron production estimate of roughly 5×10^{-29}/sec/nitrogen atom or 2×10^{-9}/sec/cc for the ICAO standard atmosphere.[7] Comparison of this estimate with the slow-neutron production rate reveals that it is indeed negligible, in support of the qualitative conclusion deduced above. Since this fast-neutron contribution is less than experimental error, it will be completely neglected.

An additional interpretation of the present wall-corrected C^{14} production rate is also possible. Namely, one can determine the slow-neutron density at the air-land interface from this result. This conclusion follows directly from the $1/v$ behavior of the $N^{14}(n,p)C^{14}$ cross section for slow neutrons (i.e., below 0.1 MeV). Employing the most recent recommendation for this thermal cross section,[12] $\sigma(n,p)=1.81\pm0.05$ b, one finds a slow-neutron density of $(1.19\pm0.06)\times10^{-8}$/cc for the air-land interface near sea level.

The total experimental error quoted above includes a contribution due to the error in the $N^{14}(n,p)C^{14}$ cross section. Based on the recommended value, a relative cross section error of 3% has been employed. Consequently the total relative error for the slow-neutron density measurement is approximately 5%.[13]

DISCUSSION

One must exercise care in the application or interpretation of the results given above. The experimental error quoted for both the C^{14} production rate and the slow-neutron density correspond to values averaged over a duration of approximately one month (November–December, 1966). During this time period, fluctuations occurred in the production rate that were well outside experimental error. In fact, the production rate was observed to change by as much as 25% from the average value. The origin of such fluctuations can be attributed to variations in many factors which affect the environment. For example, changes in the primary cosmic-ray intensity, or the local weather conditions, or even the local humidity, or a combination of changes in all three of these factors could alter the measured production rate.

The present determination of the C^{14} production rate at the air-land interface may be compared with the sea-level value calculated by Lingenfelter,[2] within the framework of one serious limitation, namely, that the calculation does not account for the presence of the air-land boundary. Although the present experimental measurement is at least a factor of 2 higher than the calculated value, the effect of the boundary is expected to considerably enhance the production rate.[9] Consequently these two results are in qualitative agreement.

A comparison with the neutron-density measurements of Miles is also possible.[14] Unfortunately, the very same limitation again exists, since these measurements did not extend to the earth's surface. Using the absorption mean free path found by Miles (165 g cm^{-2}) to extrapolate his lowest experimental data point, one finds a sea-level neutron density which is a factor of 5 lower than the present experimental measurement. However, the boundary contribution from the air-land interface, the uncertainty of the large extrapolation, and the large experimental error of this lowest data point could account for most of this discrepancy. It is interesting to note that the maximum neutron density obtained by Miles $(4.8\pm1.2)\times10^{-7}$ neutrons/cm^3, is some forty times larger than the air-land interface density obtained in the present experiment.

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[9] G. V. Gorshkov, V. A. Zyabkin, and O. S. Tsvetkov, At. Energ. (USSR) **17**, 492 (1964).

[10] M. Yamashita, L. D. Stephens, and H. W. Patterson, J. Geophys. Res. **71**, 3817 (1966).

[11] J. H. Ray, G. Grochowski, and E. S. Troubetzkoy, UNC-5139 1965 (unpublished).

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