convective boundary downward we use the adiabatic temperature gradient (He/H_2 = 0.2) to obtain a CH_4 abundance of 150 m-amagat for the mean level of CH_3D spectroscopic line formation. The pertinent parameters for our analysis are listed in Table 1. According to the Curtis-Godson approximation (*19*) for a simple transmitting or reflecting atmosphere, the base level of spectroscopic probing is twice this value and a mixing ratio of 9×10^{-5} for CH_3D/CH_4 is obtained. This mixing ratio assumes a solar C/H ratio on Saturn (*20*), instead of an enrichment by a factor of 3 over the solar abundance as has been suggested by some (*16, 17, 21*). Assuming no chemical deuterium fractionation exchange between CH_4 and H_2, we obtain a D/H ratio of 2×10^{-5} for the Saturn atmosphere.

Because a number of observational facts and model parameters were required to derive this ratio, we now discuss our error estimate and give a brief description of the major factors contributing to the uncertainties in the derived D/H ratio. Beer and Taylor (*5*) have estimated the chemical fractionation of CH_3D on Jupiter and calculated a range of possible fractionation factors between 1.22 and 1.36. Subsequent detection in Jupiter's atmosphere of CO (*22*), PH_3 (*13*), and GeH_4 (*23*) has shown, upon comparison with the thermodynamic equilibrium calculations (*24*), that Jupiter's atmosphere must be strongly convective to the 1000 K level (*25*). At that temperature the fractionation factor is only 1.13. The detection of PH_3 in Saturn's atmosphere from the data of Bregman *et al.* (*26*) and Larson and Fink (*11*) indicates that Saturn's atmosphere is also convective to rather deep levels. Thus fractionation of the deuterium should not result in a significant correction to the D/H ratio. The accuracy of this number is largely determined by approximations and assumptions underlying the atmospheric models used (*16*), experimental errors in our temperature determination, uncertainties in the values for the H_2 line strengths (*8*), assumptions concerning the C/H mixing ratio, and the exact mechanism of CH_3D spectral line formation. We believe that the D/H ratio determined in this report is good to within a factor of 2, unless several of the above uncertainties fortuitously add together, in which case a somewhat higher systematic error might result (*27*).

Our observation of CH_3D on Saturn has shown that the 5-μ radiation originates from relatively deep levels within its atmosphere, and has provided a determination of the abundance of CH_3D on this planet. We have also obtained a value for the D/H ratio for Saturn. Our derived value is similar in magnitude to that of Jupiter. Both the Jupiter and Saturn D/H ratios are considerably lower than the terrestrial and meteoritic value of 15×10^{-5} (*28*). We have thus provided additional support for the earlier suggestion (*28, 29*), deduced from the $^3He/^4He$ ratio in the solar wind, that the earth and meteorites are considerably enriched in deuterium. If stellar processing of the deuterium that formed the solar system is minimal (*2*), the D/H ratio obtained for Jupiter and Saturn may be the most representative value determined so far for the primordial D/H ratio at the time of formation of the universe.

UWE FINK
HAROLD P. LARSON
Lunar and Planetary Laboratory, University of Arizona, Tucson 85721

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Radiocarbon Dating with Electrostatic Accelerators: Dating of Milligram Samples

Abstract. *The recently developed direct counting technique for radiocarbon atoms has been used to measure the relative numbers of such atoms in various geological samples which had earlier been dated by the beta-ray counting method. Sample weights ranged from 3.5 to 15 milligrams. The dates determined by the two methods are consistent with each other. Further experience with the new method is also reported.*

The recent reports of successful detection of ^{14}C ions from recent carbon (*1–3*) have been followed by preliminary reports of the first attempts to date milligram samples (*4*) which had earlier been measured by the conventional beta-ray counting method. This report gives an account of the most recent comparative dating results and some of the experience gained from the work.

The apparatus used was essentially the same as that employed in our earlier reports (*1, 3, 4*) with the addition of an improved high-voltage stabilization system (*5*) which is essential for the work we report here. This is necessary because, in contrast to the conventional use of tandem accelerators for nuclear physics, current stabilization is not possible on the very weak ^{14}C beams. The tandem terminal voltage was stabilized to better than 0.1 percent to maximize the transmission of ^{14}C ions (*6*) through the high-resolution analysis system and to minimize the influence of nitrogen ions accelerated to the terminal as $^{14}NH^-$

Charcoal samples of known ages weighing several grams were obtained from the U.S. Geological Survey (*7*); 3.5 to 15 mg of these samples were compressed with an equal volume of KBr on aluminum cones. These cones, when inserted into a sputter ion source (*8*), gave $^{12}C^-$ currents in the range of 1.0 to 7.0 μA which were quite sufficient for these experiments. The surfaces of the carbon

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Table 1. Experimental results.

Carbon samples*	Total run time (min)	Average count/min per microampere of ^{12}C	Age† (years)	
			This work	USGS value
W-3629, Mount Hood, Ore.	90	23.2 ± 0.9	220‡ ± 300	220 ± 150
W-3703, Mount Shasta, Calif.	180	11.7 ± 0.2	5,700 ± 400	4,590 ± 250
W-3663, Lake Agassiz, N.D.	90	7.9 ± 0.5	8,800 ± 600	9,150 ± 300
W-3823, Hillsdale, Mich.	400	0.14 ± 0.02	41,000 ± 1,100	39,500 ± 1,000
Graphite	65	0.06 ± 0.01	48,000 ± 1,300	

*All samples used in this work were about 15 mg except for the Mount Hood sample, which was 3.5 mg. The U.S. Geological Survey (USGS) dates were determined by beta-counting acetylene gas in 1-liter proportional counters for 1 day in each of two USGS measurements. In view of the fact that we used only a small fraction of the material provided by the USGS, inhomogeneities may lead to real differences in age between the two sets of data. †The half-life of ^{14}C was taken to be 5568 years. ‡Assumed.

samples were cleaned and conditioned by the sputtering process in the ion source for a period of up to an hour before measurements were made. After that time, the $^{14}C^+$ counts per minute per microampere of $^{12}C^-$ at the ion source reached a plateau and the numbers quoted are from this plateau region. One-milligram samples of cracked acetylene were also used successfully as sources of C^-, and ^{14}C ions were readily detected. However, the exponential decay of the negative ion current with a half-life of about 12 minutes made measurements difficult. Only measurements of the charcoal plus KBr samples will be presented here. The C^- currents, and hence ^{14}C counts, were found to be very sensitive to the pressure at the ion source, and a liquid nitrogen trap was added to the ion source to minimize the pressure.

The procedure adopted for these measurements was to alternate, at approximately 10-minute intervals, the measurement of the ^{14}C counting rate in the detector and the measurement of the $^{12}C^-$ ion current at the ion source. While the ^{14}C was being counted, the $^{12}C^-$ ion current at the source was monitored with a different Faraday cup that intercepted the $^{12}C^-$ beam. Ideally, one would like to have ^{12}C, ^{13}C, and ^{14}C transmitted through the accelerator and measured simultaneously.

Table 1 shows some of the results we have recently obtained. Earlier dating results have been discussed briefly at conferences (*1*, *3*, *4*). The results of the present direct detection dating are clearly consistent with the dates obtained by beta-counting, and the errors are comparable. The errors quoted, for this work and for the U.S. Geological Survey, are each 1 standard deviation in the mean for several measurements (*9*). The striking fact, however, is that the sample sizes used in the present work are at least a thousand times smaller, and the measuring times are less, than for the conventional methods. This opens the door to entire new classes of dating possibilities.

The most important backgrounds that can interfere with the detection of ^{14}C ions are the breakup products of the $^{13}CH^-$ and $^{12}CH_2^-$ molecules injected simultaneously with the $^{14}C^-$. These molecules are over 10^9 times as intense as the $^{14}C^-$ ions at the ion source, but they are destroyed during the conversion from negative to positive ions at the terminal of the tandem. Some of the resulting carbon ions undergo a second charge exchange in the residual gas of the accelerating tube and give ^{12}C and ^{13}C ions of the same magnetic rigidity as the ^{14}C ions. This process produces the ^{12}C and ^{13}C peaks shown in our previous reports. They are not useful for normalization in the ^{14}C dating because the $^{12}CH_2$ and ^{13}CH concentration and production rates vary from sample to sample. They could be removed by further electrostatic analysis.

Since the first observation of ^{14}C ions from a contemporary sample in May 1977 (*1*), a variable background of ^{14}C ions from graphite has been observed. The origin of this background is unclear at present, and no detailed study has been undertaken. The background can be interpreted as an age for the graphite and this has varied from 40,000 to 70,000 years. It has often been observed that the counting rate of ^{14}C ions from graphite varied with time and usually decreased rapidly at the beginning of a measurement. This decrease is presumably due to a superficial layer of contemporary carbon on the surface of the graphite cone being sputtered away in the ion source. We have also noticed that medium-aged samples (about 5000 years) often show a lower ^{14}C counting rate for up to an hour before reaching a plateau, and in this case a superficial sputtered graphite layer could be the explanation. This problem of cone "cross-talk" can be eliminated by modification of the ion source. The following possible source of contamination should also be noted. Accelerators, over the years, generate ^{14}C atoms from nuclear reactions, and so the hydrocarbons in the vacuum system could be slightly enriched in ^{14}C. We have measurements which perhaps indicate a slight enrichment in ^{14}C for the Rochester MP tandem Van de Graaff accelerator. Contamination of the Berkeley cyclotron has also recently been reported (*10*).

C. L. BENNETT
Nuclear Structure Research Laboratory, University of Rochester, Rochester, New York 14627
R. P. BEUKENS
Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7
M. R. CLOVER, D. ELMORE
H. E. GOVE
Nuclear Structure Research Laboratory, University of Rochester
L. KILIUS, A. E. LITHERLAND
Department of Physics, University of Toronto
K. H. PURSER
General Ionex Corporation, Ipswich, Massachusetts 01938

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Radioisotope Dating with an Accelerator: A Blind Measurement

Abstract. *The age of a sample of carbon dioxide has been determined by accelerating the carbon with a cyclotron and detecting the carbon-14 ions in the beam. Nitrogen-14 was eliminated as a background through the use of a range-separation technique. To avoid all possibility of experimenter bias, the measurement was conducted in a blind fashion.*

Muller recently proposed that the use of high-energy particle accelerators for the direct detection of radioactive atoms could greatly extend the age of samples that could be dated while simultaneously reducing the sample size required (*1*). He also reported in (*1*) that the 88-inch (224-cm) cyclotron at Berkeley had been used in the first successful test of the technique: the $^3H/^2H$ ratio had been measured to determine the age of a sample approximately one mean-life old. In that experiment a range-separation technique had been used to eliminate potential backgrounds, such as ^3He, that are produced in the ion source with a charge-to-mass ratio (e/m) very similar to that of ^3H. Soon after the publication of that result, several other groups demonstrated (*2*) that the range-separation stage could be avoided in the case of ^{14}C if a tandem Van de Graaff accelerator were used in place of a cyclotron, since the main source of background, ^{14}N, does not form negative ions. Dates have now been obtained for samples several milligrams in mass with previously known ages up to about 40,000 years (*3*). The sensitivity appears to be limited by ^{14}C associated with the accelerator. It may be possible to eliminate the background if the specific source can be found and removed, or if an accelerator with low radiation background is constructed specifically for radioisotope dating.

We report here on the first accelerator dating of a "blind" sample of carbon. The 88-inch cyclotron at Berkeley was used to accelerate $^{14}C^{3+}$ ions to an energy of 60 MeV, and the range-separation technique was used to separate and eliminate the background from $^{14}N^{3+}$. The sample was obtained in the form of CO_2 from R. Berger at the University of California at Los Angeles, who had measured the age using the standard radiocarbon "decay dating" technique invented by W. R. Libby. Our measured age of 5900 ± 800 years was obtained and announced (*4*) without prior knowledge of the age obtained by Berger's group.

The range-separation technique is based on the observation that the range of ^{14}C is 1.3 times that of ^{14}N (*1*). We accomplished the separation by passing the output beam of the cyclotron through a xenon gas cell about 10 cm long. Gas was used in order to assure a uniform material; xenon was chosen because its high Coulomb barrier minimizes nuclear interactions. The gas was separated from the vacuum of the cyclotron with a window of platinum and gold less than 1 μm thick. This window was supported on a tungsten grid with a clear aperture of approximately 65 percent. With the tungsten support, the window was able to hold a pressure difference of greater than 1 atm.

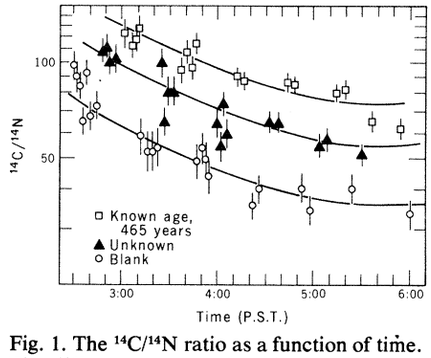

Fig. 1. The $^{14}C/^{14}N$ ratio as a function of time. The ^{14}C was measured with an ionization chamber–silicon detector telescope. We measured the ^{14}N by integrating the current from slits which collimate the beam soon after it emerges from the cyclotron and well before it enters the xenon gas cell. Three samples were alternated: one of known age 465 years, a blank known to contain no ^{14}C, and the unknown. The fit shown corresponds to an age of 5900 years. The time scale is in hours.

Adjacent to the xenon gas cell was an ionization chamber–silicon detector telescope to measure the ionization and total energy of the emerging particles. This telescope provided unambiguous identification of the nuclear charge of the particles emerging from the xenon cell.

The range-straggling curve for the nitrogen ions in the xenon gas cell was very steep. When the xenon pressure was maintained at 1.15 times the pressure corresponding to a 10-cm range of the nitrogen, all of the nitrogen ions stopped in the cell. In over 10 hours of operation with 1 to 10 nA of $^{14}N^{3+}$ incident on the xenon, not a single nitrogen ion emerged into the particle telescope to register a coincidence. Thus the discrimination against nitrogen was better than 10^{-14}.

Although the nitrogen background was effectively eliminated, another background has thus far limited the sensitivity of the measurements. Even when measuring a sample known to contain no ^{14}C, we found that carbon counts register in the detector telescope. The most plausible origin of the background counts is highly radioactive carbon within the cyclotron vacuum. Our level of background ^{14}C appears to be higher than that reported by the Rochester group (*3*).

Because of the high ^{14}C background, it is necessary to make a subtraction in order to obtain an age. Figure 1 shows the results of an experiment in which the cyclotron source gas was switched rapidly between three gas samples, one known to be 465 years old, the unknown, and a blank known to contain no ^{14}C. All three samples had been measured at Berger's laboratory. The data were fitted to a simple model with five constants: the age of the unknown, the background level, the overall normalization (that is, the cyclotron efficiency), and two constants to describe the slow variation of the source output with time.

From the fit we obtained an age (*5*) for the unknown of 5900 ± 800 years. The errors are statistical only and we obtained them by varying the age parameter and refitting the data. The variation corresponding to a chi-square change of one gives an estimated error of 1 standard deviation. Although our statistical error is twice that reported by the Rochester group, it is comparable to the accuracy they obtained for a sample of similar age (*6*).

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