

on purely scientific matters, but also on questions more susceptible to distortion by human passions, always carried great weight.

Fermi was also an incomparable teacher, from the time, in his college years, that he introduced me and a few other fellow-students to the first mysteries of theoretical physics. He was gifted with great clarity and orderliness of exposition; many times a question asked by one of his students or colleagues was answered by a masterful, exhaustive, improvised lecture on the subject which, if taken down word for word, could have been sent almost unchanged to the editor of a scientific journal. Physicists familiar with his work well know the outstanding clarity and thoroughness of all his publications; for example, the treatment given in his historical papers on Fermi statistics and the theory of beta-decay is so perfect that hardly a word need be changed or added even today, more than a score of years after those articles were written. Also well known for easily readable treatments of difficult subjects are his 1932 article on the quantum

theory of radiation and his book, *Elementary Particles*, which records the contents of the 1950 Silliman lectures.

Fermi was probably the most sought-after lecturer in physics of the last two decades. Universities, academies, and other scientific institutions all over the world vied in securing his participation in lectures, meetings, and symposiums, certain that his presence would stimulate creative discussion and often lead to important advances. During the summer of 1954 Fermi lectured, for the last time, at the schools of advanced physics at Les Houches in France and Varenna in Italy. His mind was as brilliant as ever, but his body was beginning to suffer from the attacks of a fatal disease. Most of his colleagues hardly suspected the gravity of his illness, and they were greatly shocked, when the man, who, more than any other single individual, fathered the advent of the atomic age, passed away in Chicago on 28 November 1954.

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## Beryllium-7 Produced by Cosmic Rays

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IN his classic paper of 1946, W. F. Libby (1) predicted the existence of cosmic-ray-induced radioactivities in the atmosphere, in particular  $C^{14}$ , with a half-life of 5600 yr. and tritium, with a half-life of 12.4 yr. Both nuclides have now been discovered and used to study a wide variety of processes having time-scales comparable to their respective half-lives (2). Carbon-14 is made by low-energy neutron capture in nitrogen, whereas tritium results chiefly from high-energy interactions, or "stars."

Two other nuclides may be expected to result from these high-energy interactions in nitrogen and oxygen. These are  $Be^7$ , with a half-life of 53 days, and  $Be^{10}$ , with a half-life of  $2.5 \times 10^6$  yr. Because of their well-spaced half-lives these species should be of geochemical interest. This paper (3) reports the discovery of cosmic-ray produced  $Be^7$ .

Our picture of the history of this nuclide is as follows. The peak of the star production occurs about 15 km above the surface (4). Beryllium formed at this height will form  $BeO$ , or just possibly  $Be(OH)_2$ . The nonvolatile molecule will diffuse in the atmosphere until it encounters a dust particle, to which it will adhere. Its further history is that of the upper atmosphere dust.

It appeared probable that the great majority of upper atmosphere dust particles ultimately form cloud nuclei, which are carried down in rain, or are otherwise washed out by rain. Rain water then

seemed a likely location for  $Be^7$ . A series of rain-water samples taken at Chicago and at Lafayette, Indiana, were analyzed for this nuclide.

In each case a sample of rain water of 5 to 50 gal was collected from a roof (it therefore included some "between-rains" dust), 1 to 2 ml of  $Be^{++}$  carrier per gallon (5 mg  $BeO/ml$ ) was added, with sufficient nitric acid to bring the pH to about 3. After thorough agitation, the solution was brought to pH 9 with ammonia, precipitating  $Be(OH)_2$  along with substantial quantities of  $Fe(OH)_3$  and other species present in rain water in this industrial area or originating in the cans used for collection in some cases. This flocculent precipitate served as a general carrier. After the sample had been allowed to stand for 30 min or more, it was filtered, and the clear filtrate was discarded.

An early experiment showed that all the activity remained in the precipitate at this point. The precipitate was ashed and fused with  $KHSO_4$ . After the supernate was leached and centrifuged, it was scavenged with  $CuS$  and then was made strongly basic to precipitate  $Fe(OH)_3$  and other group-III hydroxides. The supernate was acidified and then made basic with  $NH_4OH$ , yielding a precipitate containing the  $Be(OH)_2$ . This was then put through an acetate-chloroform cycle following McMillan (5) and finally was precipitated as  $Be(OH)_2$  and ashed to  $BeO$ .

The yields for this procedure were erratic but improved with experience; they range from 20 to 80 percent in the samples reported. Radiochemical purity

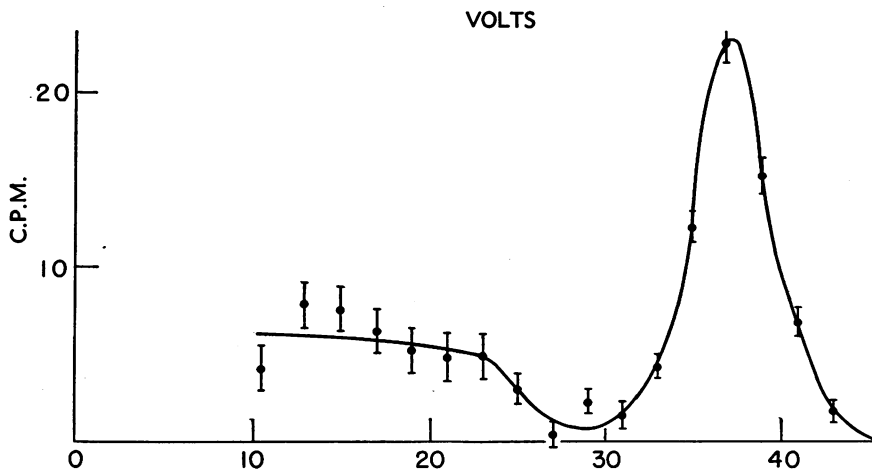


Fig. 1. Spectrum of composite sample, using a 2-in. well crystal.

was tested by counting the samples in an end-window beta counter. No activity was observed. The samples were counted on our low-level scintillation spectrometer, using a 1-in. NaI (Tl) crystal, at a setting admitting the full photopeak of  $\text{Be}^7$ .

The nature of the activity observed was verified in three ways. First, repeated cycles of the highly specific acetate-chloroform procedure showed constant specific activity (which would be expected, since this procedure has almost no interferences). Second, the scintillation spectrum of the activity shows the characteristic photopeak of a pure gamma emitter, with energy  $472 \pm 10$  kev. Figure 1 shows the spectrum of

a composite sample, taken in a 2-in. well crystal. Finally, the half-life checks the literature value; Fig. 2 shows the decay curve of sample RS, and Fig. 3 that of a composite sample.

The results for a series of rain and snow samples are shown in Table 1. The wide variations in successive rains are obvious and parallel Libby's earlier tritium results. In the cases where rain from the same storm was taken at both locations (RZ and PA, SE and PD), the agreement is satisfactory. More important for our purpose is the absence of a systematic drift with time. Seven samples taken before 13 March 1954 average 2.12 count/min gal, while 15 taken on or after that date average 2.00. Taken together with the 53-day half-life, the absence of an increase during and after the American nuclear weapons test of last spring substantially demonstrates the natural origin of this nuclide.

We may attempt to estimate the rate of production  $Q$  of  $\text{Be}^7$  from the foregoing data. First, the average value of 2.04 counts/min gal must be converted to absolute units. Using McGowan's (6) values for a source-to-crystal distance of 3 mm, we get a total counting efficiency of 0.19 and the fraction in the photopeak 0.47. Since only 11 percent of the decays produce the 479-kev gamma ray, the over-all counting efficiency is 0.0099, and the absolute assay is 54 decays/min lit, or  $6 \times 10^6$  atoms/lit. In units equivalent to Libby's T units this is 0.09.

Assuming 1 m of annual rainfall, each square centimeter of earth's surface in the Chicago area receives  $6 \times 10^5$  atoms/yr, or 0.019 atoms/sec. In order to translate this into a production rate, some estimate must be made of the time-scales of a number of atmospheric processes: (i) the vertical mixing in the lower stratosphere where most of the production occurs; (ii) vertical mixing in the troposphere, from the tropopause down to cloud level or below; (iii) north-south mixing (because of the great difference between high- and low-latitude production rates); (iv) east-west circulation, which plays a role because of the differences between geographic and magnetic latitude; and (v) frequency of substantial rainfall.

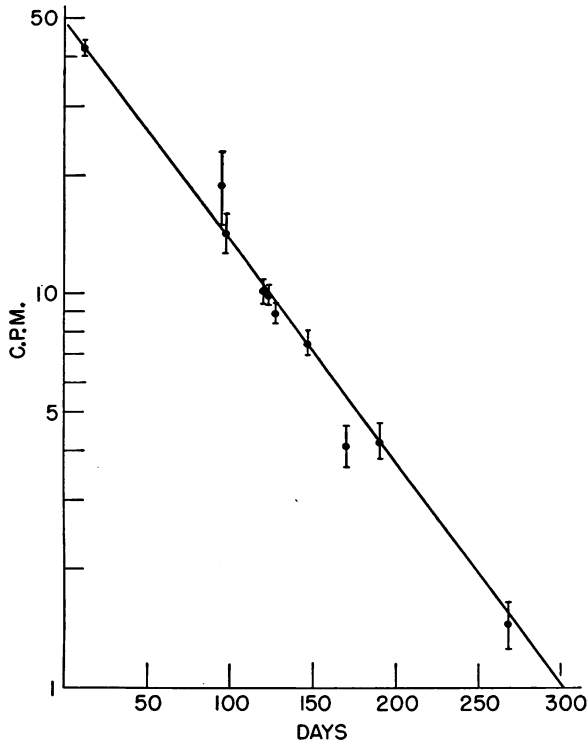


Fig. 2. Decay curve of sample R.S.

Of course none of these processes occur in a systematic pattern, either in time or space, but gross averages may be useful. The fluctuations seem ample to account for the observed variations. This is especially true of the north-south mixing, since Chicago is almost at the southern edge of the high-latitude plateau of the cosmic-ray flux.

Rough relaxation times might be guessed as follows:

Process	Time scale
Vertical mixing, stratosphere	
Temperate zone	60 to 180 days
Tropic zone	30 days
Vertical mixing, troposphere	15 days
North-south mixing	
Stratosphere	100 days
Troposphere	10 days
East-west mixing, troposphere	5 days
Frequency of substantial rainfall	
Temperate zone	15 days
Tropic zone	15 days

All these numbers, except the one for the east-west mixing process, are comparable to the 53-day half-life. This makes calculation difficult, but we can still make an attempt. First, if all processes were assumed fast, the world-wide  $Q$  would be the Chicago rate of 0.019 atoms/sec cm<sup>2</sup>. If we assume that the newly formed Be atoms spend 80 days in descending to cloud level, and that the north-south and east-west mixing lower the concentration of Be<sup>7</sup> at Chicago by 30 percent (which is half the effect of complete north-south mixing) before rainfall brings it down 10 days later, we get a value for the world-wide  $Q$  of  $0.019 \times 2^{90/53} \times 0.4/0.7 = 0.035$  atoms/sec cm<sup>2</sup>.

The data on the primary cosmic-ray flux on which the effect of north-south mixing is calculated are those of Winckler and Stix (7) and Simpson (8). These

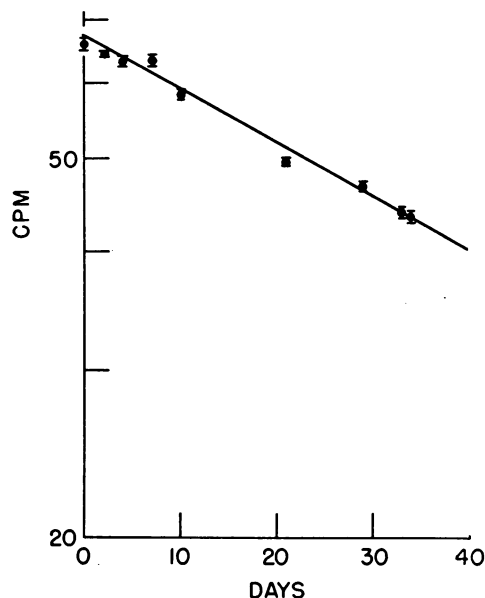


Fig. 3. Decay curve of composite sample.

Table 1. Cosmic-ray-produced Be<sup>7</sup> in some rain and snow samples.

Sample	Place collected	Date	No. of gallons	(counts/min gal)
RS	Chicago	10-26-53	30	2.30 ± 0.10
RX		12- 2-53	15	1.42 ± 0.22
RZ		1-20-54	30	2.81 ± 0.34
SA		1-26-54	10	3.26 ± 0.26
SB		2-28-54	10	0.70 ± 0.16
SC		3- 3-54	9	0.56 ± 0.10
SD		3-13-54	10	1.15 ± 0.30
SE		3-18-54	5	1.27 ± 0.35
SF		3-24-54	10	1.67 ± 0.11
SG		3-29-54	6	1.65 ± 0.18
SK		4-21-54	5	5.00 ± 1.12
SL		4-22-54	5	2.75 ± 0.40
SN		5- 4-54	5	1.37 ± 0.33
SO		5-11-54	5	5.40 ± 0.45
ST		6-17-54	5	3.52 ± 0.14
SU		7- 3-54	17	1.20 ± 0.04
SV	7- 6-54	10	2.44 ± 0.07	
PA	Lafayette	1-20-54	50	3.76 ± 0.24
PB		3-20-54	15	0.94 ± 0.09
PD		3-19-54	15	0.88 ± 0.13
PE		4-17-54	15	0.35 ± 0.06
PF		4-27-54	10	0.48 ± 0.07

give 1 per square centimeter per second for the primary flux at high latitude, from which we compute a value of 0.4 per square centimeter per second for the world-wide average.

The expected value of  $Q$  cannot be computed accurately on the basis of presently known cross sections, the greatest uncertainty being in the number of Be<sup>7</sup> atoms produced by secondaries. The best estimates, using cross sections of Baker and Hudis (9) and estimating production by secondaries from the neutron data (4), give values that range between extremes of 0.015 and 0.06. This lends support to our proposed mechanism of formation and distribution of Be<sup>7</sup> in the atmosphere.

Interestingly enough Be<sup>7</sup> is the easiest of all the cosmic-ray-produced nuclides to detect, despite its comparatively low production rate, because it occurs substantially carrier-free. The possibility of using this nuclide to study the afore-mentioned atmospheric processes is apparent.

#### References and Notes

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- We wish to express our thanks to W. H. Johnston and his students at Purdue University for gathering and partially processing the Lafayette samples. We are indebted to Luis Marquez, Anthony Turkevich, Gerhart Friedlander, and W. F. Libby for helpful discussions, and particularly to Herbert Riehl, who was of great assistance on meteorological questions. Trans World Airlines furnished assistance in some preliminary experiments. The Office of Ordnance Research contributed generous support.
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