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On the Deposition of Cosmic-Ray-Produced Beryllium 7

J. F. BLEICHRODT AND E. R. VAN ABKOUDE

Medical Biological Laboratory
National Defence Research Council T.N.O.
Rijswijk, Z.H., Netherlands

Abstract. The observed annual deposition of cosmic-ray-produced Be^7 in the Netherlands exceeds the amount expected from tropospheric production. The difference agrees with an estimated contribution of natural Be^7 originating from the stratosphere.

Introduction. Deposition of cosmic-ray-produced Be^7 has been studied extensively in tropical regions, but until recently only few data have been published for higher latitudes [*Lal and Peters,* 1962 (review); *Rama,* 1960]. During the suspension of testing of nuclear weapons (November 1958 to August 1961) several investigators measured Be^7 deposition at temperate latitudes. The results of this work are becoming available now [*Walton and Fried,* 1962; *Peirson,* 1963; *Schumann and Stoeppler,* 1963]. In this paper some data obtained at Rijswijk, Z.H. (52°3′N, 4°20′E), are presented and discussed in connection with results of others.

Experimental procedure. Rain water and dry fallout were collected in duplicate during monthly periods on two stainless steel trays having a surface area of 1.5 m^2 and a height of 0.1 m, mounted 1.5 m above ground level. During periods of high rainfall, sampling extended over shorter time intervals. Inactive Be carrier was added to the trays before exposure. Samples were evaporated to a small volume after addition of nitric acid. Dust was removed by filtration and Be was precipitated as $Be(OH)_2$ in the presence of ethylenediaminetetra-acetate. The Be was purified radiochemically using the procedure of *Buchanan* [1958]. Purity of several Be preparations was established by repurification procedures and measurement of radioactive half-life.

Radioactivity was measured with a thallium-activated NaI well-type crystal (Harshaw, type F) shielded with 10 cm of lead and a single-channel pulse analyzer under admittance of the full photopeak (0.478 Mev). The counter was calibrated with a standard solution of Be^7 (estimated accuracy 2 per cent) obtained from Nuclear Science and Engineering Corporation, Pittsburgh, Pennsylvania. The resulting efficiency was in good agreement with calibration measurements of *Nablo and Martin* [1961] for this type of crystal.

Deposition of Sr^{90} was determined using the procedure of *Osmond et al.* [1961]. The

TABLE 1. Deposition of Cosmic-Ray-Produced Be^7 at Rijswijk, Z.H.

Sampling Period			Be^7, 10^8 atoms/m^2	Rainfall, mm
	1960			
Nov.	2–Dec.	1	7.8	98
Dec.	1–Dec.	15	6.0	74
Dec.	15–Jan.	12	6.0	78
	1961			
Jan.	12–Feb.	1	3.8	56
Feb.	1–March	1	7.7	64
March	1–March	30	3.0	18.8
March	30–May	2	8.5	58
May	2–June	1	9.2	22
June	1–July	1	7.9	57
July	1–Aug.	1	11.1	104
Aug.	1–Sept.	1	10.6	80
Sept.	1–Oct.	2	13.5 (0.4)*	92
Oct.	2–Nov.	1	11.3 (2.0)	129
Nov.	1–Dec.	1	10.6 (3.2)	97
Dec.	1–Jan.	2	7.2 (3.4)	103
	1962			
Jan.	2–Feb.	5	7.5 (2.8)	92
Feb.	5–March	2	3.8 (1.2)	28
March	2–April	2	4.1 (0.9)	41
April	2–April	27	4.2 (0.9)	54
April	27–June	1	5.3 (0.8)	49
June	1–July	2	4.2 (0.3)	14
July	2–July	31	21.5 (1.0)	113
July	31–Aug.	31	9.7 (0.5)	67

* Values in parentheses represent the amount of artificial Be^7 deposited.

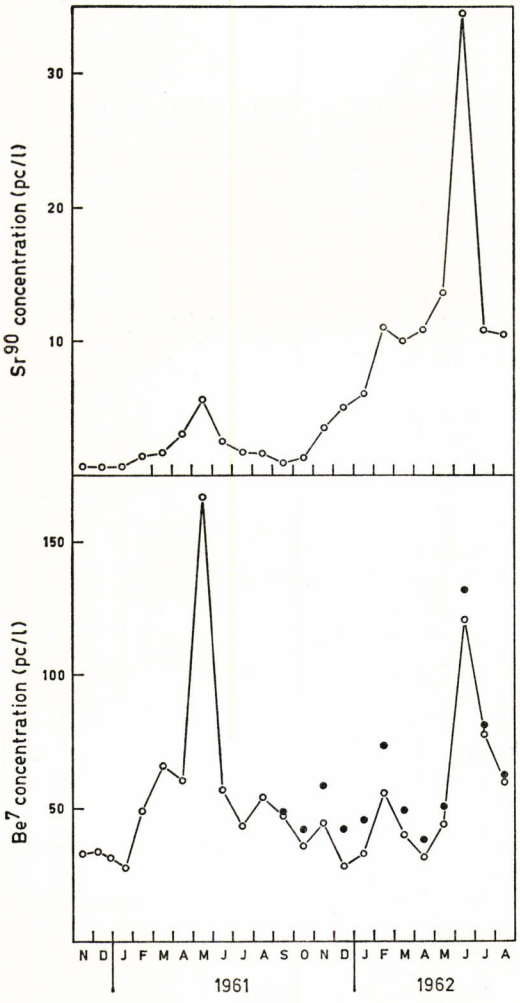

Fig. 1. Concentration of Be^7 and Sr^{90} in rain water at Rijswijk. Total Be^7 is represented by solid circles, natural Be^7 by open circles.

amount of rainfall was measured with a rain gage. Samples of stratospheric radioactivity were taken by aircraft using glass fiber filters. Be^7 and Sr^{90} were extracted from the filters with nitric acid in the presence of inactive Be and Sr carriers. Radiochemical purification of the nuclides was analogous to the above-mentioned procedures.

Results. In Table 1 average values of duplicate measurements are given for the period November 1960 to September 1962. Duplicates usually differed less than 15 per cent. In calculating the amount of Be^7 at the middle of the sampling period a half-life of 53.4 days was used

TABLE 2. Annual Deposition of Cosmic-Ray-Produced Be^7

Measured	
Nov. 1960 to Nov. 1961	10.6×10^9 atoms/m²
Sept. 1961 to Sept. 1962	10.3
Calculated	
Using average monthly mean tropopause heights (1953 to 1959)	
Residence time = 30 days	5.7×10^9 atoms/m²
Residence time = 40 days	5.2
Using monthly mean tropopause heights (Nov. 1960 to Nov. 1961)	
Residence time = 30 days	5.4
Using average global tropospheric production	
Residence time = 30 days	6.1

[*Ajzenberg-Selove and Lauritsen,* 1959], and the number of γ's per disintegration was assumed to be 0.103 [*Taylor and Merritt,* 1962].

The results for the period September 1961 to September 1962 have been corrected for the contribution of artificial Be^7 from the Soviet nuclear test explosions during September and October 1961, assuming a ratio of 0.038 for disintegration rates of artificial Be^7 and Sr^{89} on November 1, 1961 [*Bleichrodt and van Abkoude,* 1963]. The corrections have been included in Table 1. During May and June 1962 the contribution of Sr^{89} from atmospheric tests by the United States, started on April 25, was negligible, as shown from the relative concentration of short-lived nuclides in fission products. In July and August the contribution of these tests and those of the Soviet Union in August was appreciable. However, unless the ratio of Be^7 to Sr^{89} is considerably higher in debris from nuclear explosions in 1962 (this ratio does not change much with time because the half-lives of Be^7 and Sr^{89} are approximately equal), the error in the concentrations of natural Be^7 made by assuming all the Sr^{89} to originate from the Soviet 1961 detonations is small, because corrections for artificial Be^7 do not exceed 5 per cent during these months. Both the concentrations of total Be^7 (pc/l = deposition of Be^7 in pc/m² per mm of rain) and of natural Be^7 in rain are plotted in Figure 1, together with the concentration of Sr^{90} from nuclear explosions. The sharp increase of Sr^{90} after September 1961 was due to the resumption of nuclear testing.

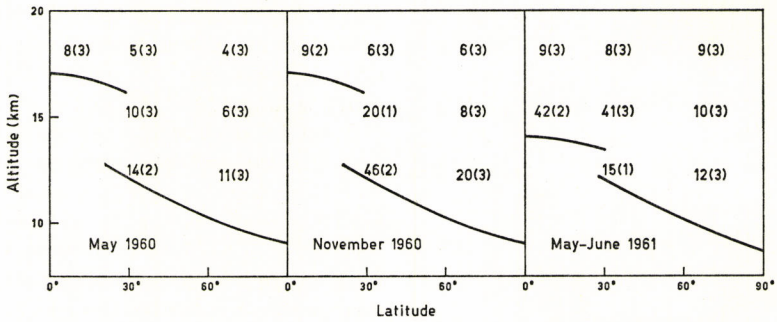

Fig. 2. Radioactivity ratio (curie/curie) of Be^7 and Sr^{90} in the stratosphere at various altitudes and latitudes in the northern hemisphere. Numbers in parentheses represent the number of measurements. Solid lines indicate approximate tropopause levels.

Discussion. The concentration of natural Be^7 in rain shows a seasonal variation that is in phase with that of the Sr^{90} concentration (Figure 1). This was also observed by *Peirson* [1963] and *Schumann and Stoeppler* [1963]. Since the amount of Be^7 per kilogram of air is higher in the stratosphere than in the troposphere a spring maximum in the concentration of Be^7 in rain suggests an increased exchange of air between these two parts of the atmosphere in spring. However, the influence of differences in rainfall characteristics between various seasons cannot be excluded [*Cruikshank et al.*, 1956; *Hvinden et al.*, 1961].

Table 2 shows the deposition of natural Be^7 during the years November 1960 through October 1961 and September 1961 through August 1962 as computed from Table 1. During the second period 15 per cent of the total Be^7 deposition was due to nuclear tests. Table 2 also contains values for the annual deposition of Be^7 which is produced in the troposphere by cosmic rays, calculated in the following ways:

1. From *Lal and Peters'* [1962] production rates of Be^7 in the troposphere at a geomagnetic latitude of 50° (the geomagnetic latitude of Rijswijk, Z.H., is about 53°) and the average values of the monthly mean tropopause heights for the years 1953 to 1959 at De Bilt (52°6′N, 5°11′E), depositions were calculated for a mean residence time (of Be^7 atoms in the troposphere) of 30 days, a value observed by *Stewart et al.* [1955] at temperate latitudes for nuclear weapons debris, and for a residence time of 40 days, which was found for cosmic-ray-produced nuclides in tropical regions [*Lal and Peters*, 1962]. Mean residence time τ, mean life of a Be^7 atom λ^{-1} (λ = disintegration constant), tropospheric production P (atoms/m² sec), and deposition D (atoms/m² sec) of Be^7 are correlated by $P = D\ (1 + \lambda\ \tau)$.

2. From Lal and Peters' production rates in the troposphere at a geomagnetic latitude of 50° and the monthly mean tropopause heights at De Bilt during the period November 1960 through October 1961, the deposition was calculated for a mean tropospheric residence time of 30 days.

3. From Lal and Peters' average global production rates in the troposphere, the deposition was calculated for a mean tropospheric residence time of 30 days.

The measured deposition of Be^7 is found to exceed by a factor of about 2 the expected fallout of Be^7 that is produced in the troposphere. It cannot be concluded, however, that half of the Be^7 reaching the earth's surface is of stratospheric origin. Besides uncertainties in the production rate of Be^7 in the atmosphere, the estimated deposition depends on the mean tropospheric residence time used for the calculation. Moreover, the yearly deposition at a given location will be approximately proportional to the amount of annual precipitation. *Möller* [1951] estimated the total amount of precipitation per year in the latitude belt 50° to 55°N to be 715 mm. During the years November 1960 to November 1961 and September 1961 to September 1962 rainfall at Rijswijk exceeded this value by 30 and 23 per cent, respectively. Deposition of tropospheric Be^7 might, therefore, be approximately 25 per cent

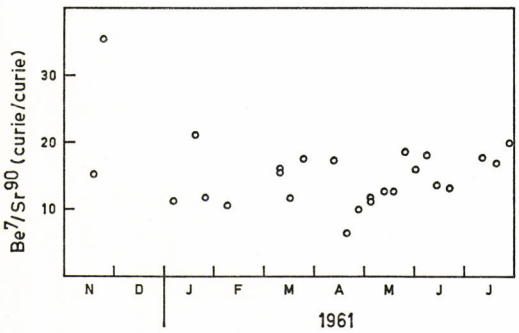

Fig. 3. Radioactivity ratio of Be^7 and Sr^{90} at 13 km over the Netherlands (52° to 53°N) during November 1960 to August 1961. Only samples taken at least 1 km above the tropopause have been included.

higher than the calculated amount during these periods, i.e., about 7×10^9 atoms/m² year. At first sight this kind of argument seems to be of little value since it presupposes absence of latitudinal transport of air masses with different Be^7 content. However, as a first approximation, latitudinal exchange may be assumed not to influence the result for two reasons. First, the production of Be^7 in a column extending from ground level to tropopause is approximately independent of latitude [*Lal and Peters,* 1962] and, second, annual rainfall does not vary widely between latitude belts of 5 degrees from 30°N to 60°N [*Möller,* 1951]. The average yearly precipitation in the belt 30° to 60°N computed from Möller's data differs less than 1 per cent from that in the belt 50° to 55°N; the average for the northern hemisphere amounts to 820 mm.

The question remaining is whether Be^7 originating from the stratosphere may account for 3×10^9 atoms/m² year, i.e., the difference between the measured deposition and the above-mentioned tropospheric fallout. A rough estimate of the contribution of Be^7 produced in the stratosphere by cosmic rays to the deposition of this nuclide on the surface of the earth may be obtained on the basis of the following considerations.

Radioactive fission products from a nuclear bomb that reach the ground several months after the explosion originate from the stratosphere because debris injected into the troposphere is removed relatively rapidly by rain. If, therefore, the ratio of concentrations of Be^7 and a fission product, e.g. Sr^{90}, in the stratosphere in the period of the suspension of nuclear weapons tests (November 1958 to August 1961) is known, the deposition of stratospheric Be^7 can be calculated from the fallout of Sr^{90} and the tropospheric residence time for Sr^{90} and Be^7 atoms. A comprehensive program of sampling and analysis of stratospheric radioactivity as a function of latitude, altitude, and time during the moratorium of nuclear testing was undertaken in the United States [*HASL-115 and 117,* 1961]. The radioactivity ratio (curie/curie) of Be^7 and Sr^{90} in samples collected in the northern hemisphere during th[illegible] program (only data with a standard error smaller than 20 per cent have been used) is shown in Figure 2 for the periods May 1960, November 1960, and May to June 1961. Some values for the lower stratosphere over the Netherlands obtained in collaboration with the Royal Netherlands Air Force and the Royal Netherlands Meteorological Institute are plotted in Figure 3 and are found to agree well with those of Figure 2.

As can be seen from Figures 2 and 3, the average Be^7/Sr^{90} ratio in air that entered the troposphere during the period June 1960 through May 1961 can be estimated at 15 with an error of less than 50 per cent. A few much higher values are found just over the tropopause at lower latitudes, one in November 1960 (the average of 12.8 and 80, respectively), two during May to June 1961 (at 15-km altitude and 12°N the average of 9.6 and 74; at 15 km and 32°[illegible] the average of 13.7, 52, and 56). The hig[illegible] ratios were probably measured in air that recently left the troposphere, where Be^7/Sr^{90} ratios were usually found to be higher. Measurements made during project Star Dust [*Friend and Feely,* 1961] in July and August 1961 agree with a ratio of about 15 at lower altitudes and latitudes in the stratosphere (22 at 15 km and about 40°N, 17 at 17 km, 30°N). The high values have been ignored, an important reason being that it is generally supposed that influx of stratospheric radioactive debris into the troposphere occurs north of 30°.

Using a value of 15 for the ratio in the lower stratosphere and assuming a mean residence time in the troposphere of 30 days for both nuclides, we can calculate the concentration of stratospheric Be^7 in rain from the concentration

of Sr^{90}. Sr^{90} data for stations of the worldwide network of the United Kingdom Atomic Energy Authority [*Crooks et al.*, 1961; *Cambray et al.*, 1962] and of the Health and Safety Laboratory of the United States Atomic Energy Commission [*HASL-132*, 1963] have been used to obtain Figure 4, which shows the average concentration of stratospheric Be^7 in rain as a function of latitude in the northern hemisphere during the period July 1960 through June 1961.

North of 30° the concentration of stratospheric Be^7 is found to be, within a factor of about 2, 4×10^6 atoms per liter of rain water. In tropical regions it is approximately a factor of 5 lower. The order of magnitude of the annual deposition of stratospheric Be^7 at a given location can be estimated using Figure 4 and the yearly amount of precipitation. Because dry fallout is included in the data plotted in Figure 4, the actual average concentration of stratospheric Be^7 in rain is lower. Especially in areas with extremely low rainfall the contribution of dry deposition may be high and therefore Figure 4 may not be applicable for such regions. The value of 26×10^6 atoms/l at 27°N, for instance, concerns Dhahran, Saudi Arabia, where the total amount of precipitation amounted only to 63 mm from July 1960 to July 1961.

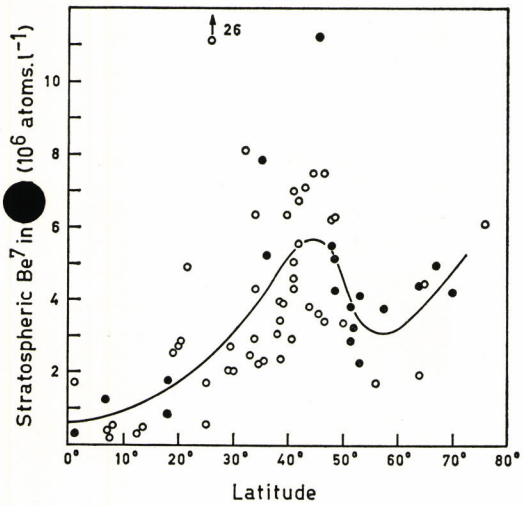

Fig. 4. Annual average of the concentration of Be^7 originating from the stratosphere in rain as a function of latitude in the northern hemisphere, calculated from Sr^{90} data for the year July 1960 through June 1961 of the United States network (open circles) and the United Kingdom network (solid circles). The curve is drawn through the averages of the data of 10° latitude bands (the value of 26 at 27°N not included).

TABLE 3. Estimated Deposition of Stratospheric Be^7 during the Period June 1960 through May 1961

Location	Rainfall, mm	Sr^{90} Deposition, pc/m²	Stratospheric Be^7, atoms/m²
Rijswijk, Z.H. (52°3′N, 4°20′E)	953	1220	$3.2\ 10^9$
Westwood, N. J. (40°59′N, 74°2′W)	1298	2160	$5.7\ 10^9$

In Table 3 is given the estimated deposition of stratospheric Be^7 during the period June 1960 to June 1961 for Rijswijk, Z.H., and Westwood, New Jersey, again calculated assuming a Be^7/Sr^{90} ratio of 15 in stratospheric air and a tropospheric residence time of 30 days. Sr^{90} deposition and precipitation data for Westwood have been taken from a report of *Walton and Fried* [1961].

At Rijswijk the deposition of Be^7 of stratospheric origin is about 3×10^9 atoms/m² year. It can be concluded, therefore, that the measured deposition at Rijswijk (10×10^9 atoms/m² year) agrees with the sum of the expected tropospheric fallout (7×10^9 atoms/m² year) and the estimated stratospheric contribution. At Westwood *Walton and Fried* [1962] investigated fourteen rains for Be^7, two in December 1960 and twelve in the first 8 months of 1961, and found an average concentration of 6.1×10^6 atoms/l. For the year June 1960 to June 1961 a total deposition of 7.9×10^9 atoms/m² is therefore expected, which seems to be somewhat low in view of the estimated stratospheric contribution (Table 3). Walton and Fried pointed out, however, that their data may be biased because only rains greater than 7 mm were analyzed. Moreover, the contribution of dry fallout is not included in the calculated annual deposition.

It should be re-emphasized that the results acquired are only tentative because the calculations are based on various assumptions concerning atmospheric processes that are still poorly known. A residence time of 30 days, for instance, which was used in the present considerations because it is generally accepted as the best average value [*Junge*, 1963, p. 253],

may be unsatisfactory for describing atmospheric phenomena at a given location. Moreover, the mean tropospheric residence time may vary with season.

The influence of the residence time on the result of the calculations, which may be easily appreciated from the above-mentioned relation between deposition and production, is found to be rather small. If the tropospheric residence time were only 10 days, for example, both the deposition of stratospheric Be^7 and of Be^7 produced in the troposphere would increase by 23 per cent. However, in view of the roughness of the calculations, differences of this magnitude do not warrant extensive discussion.

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