JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 68, NO. 15 AUGUST 1, 1963

Artificial Beryllium 7 in the Lower Stratosphere

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

Medical Biological Laboratory
National Defence Research Organization TNO
Rijswijk (Z.H.), the Netherlands

In a previous communication [*Bleichrodt,* 1962] evidence was presented that Be^7 had been produced by the Soviet nuclear test explosions in September and October 1961. For samples of stratospheric radioactivity taken at an altitude of 12.5 to 13 km over the Netherlands during October–November 1961 the following relation was found to hold:

$$[Be^7] = C + x[Sr^{89}]$$

where $[Be^7]$ and $[Sr^{89}]$ represent the concentrations (pc kg^{-1}) of Be^7 and Sr^{89}, respectively, C is the concentration of natural, cosmic-ray-produced Be^7 which was assumed to be approximately constant, and x is the ratio of disintegration rates of artificially produced Be^7 and Sr^{89}. As the half-lives of Be^7 and Sr^{89} are approximately equal [*Ajzenberg-Selove and Lauritsen,* 1959; *Osmond and Owers,* 1959], x changes only slowly with time and may be assumed to be constant for relatively short time intervals. Assuming the number of γ rays per disintegration of Be^7 to be 0.115 we obtained a value of 0.034 for x. From the more recent value of 0.103 of *Taylor and Merritt* [1962] x is found to be 0.038.

As cosmic-ray-produced Be^7 is used as a tracer in atmospheric circulation studies, it seemed worth while to extend measurements on artificial Be^7 in order to know its contribution to the concentration of Be^7 in the atmosphere. During 1962 a number of stratospheric samples taken at 12.5 to 13 km when the tropopause was at least 1 km below this altitude have been analyzed for Be^7 and Sr^{89}. The method of sampling and radiochemical analysis was the same as described previously [*Bleichrodt,* 1962] except for the samples taken after August 1, 1962. For these the radiochemical purification of Be^7 has been extended by two successive evaporations with $HClO_4$ and Ru as an extra precaution against Ru activity remaining in the final Be preparations (Ru^{103} has a half-life of 39.8 days and γ energy of 0.498 Mev; Be^7 has a half-life of 53.4 days and γ energy of 0.478 Mev). The counting setups have been calibrated with standardized solutions of Be^7 and Sr^{89} obtained from Nuclear Science and Engineering Corporation, Pittsburgh, Pennsylvania, and Isotope Division, Atomic Energy Research Establishment, England, respectively (estimated accuracy 2 per cent).

When comparing the results obtained in 1962 for debris of the Soviet explosions of 1961 with those obtained during October and November 1961 it is necessary to apply a correction for the slow increase of x with time which is due to the small difference in half-life of Be^7 and Sr^{89}. In the above-mentioned formula, x should be replaced by $xe^{0.00078t}$, where t is the time in days since November 1, 1961, the average date of the measurements of 1961. Moreover, any seasonal variation of the concentration of natural Be^7 (C) should be taken into account.

Some data concerning the concentration of cosmic-ray-produced Be^7 during various periods of the year are presented in Table 1. The predicted value for a geomagnetic latitude of 55°N in a stagnant atmosphere [*Lal and Peters,* 1962], when production and radioactive decay are in equilibrium, is also given for comparison. No significant seasonal variation of C is observed.

Be^7 concentrations for samples of the period January–July 1962 are plotted in Figure 1 versus $[Sr^{89}]e^{0.00078t}$. In these samples the contribution of debris from nuclear explosions of the United States during 1962 was negligible as followed from the relative concentration of short-lived nuclides. The curve in Figure 1 represents the expected relation between Be^7 and Sr^{89}. It intersects the ordinate at a concentration equal to the average of the data of Table 1 and has a slope x of 0.038. The points do not

TABLE 1. Concentration of Cosmic-Ray-Produced Be^7 at 12.5 to 13 Kilometers over the Netherlands

Period	Number of Samples	Be^7 pc/kg*
1960		
November–December	3	3.8 ± 2.2†
1961		
January–March	8	5.1 ± 1.2
April–June	12	5.2 ± 2.1
July–August	10	4.0 ± 1.2
Equilibrium value		7

* Only samples taken at least 1 km over the tropopause have been included. 1 pc/kg = 10^{-12} curie per kilogram of air = 2.46×10^5 atoms/kg.
† Standard deviation.

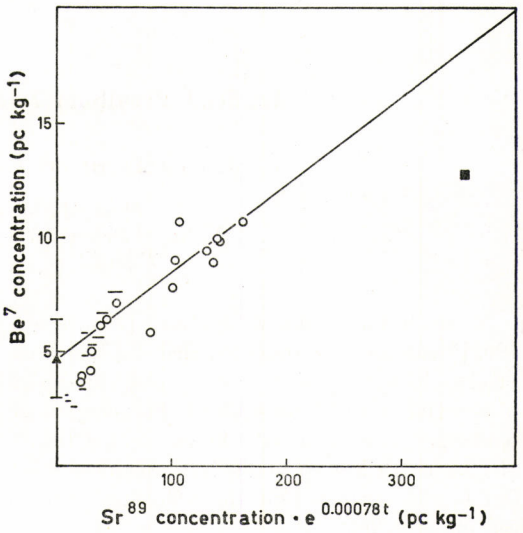

Fig. 1. Be^7 concentration versus $[Sr^{89}]e^{0.00078t}$ in air at 12.5 to 13 km over the Netherlands during 1962. Circles represent samples taken during the period January–July. Samples taken after August 5 (the very radioactive one not included) are represented by a bar, the left-hand side of which coincides with the Sr^{89} concentration observed (for Sr^{89} from recent tests t is small) and the right-hand side with $[Sr^{89}]e^{0.00078t}$ (it is assumed then that all Sr^{89} originated from the 1961 tests). The true value to be considered is somewhere in between these end points. The square represents only 'hot' cloud from the Soviet 1962 tests sampled (time correction assumed to be negligible, $t = 0$). The triangle represents average concentration of Be^7 due to cosmic rays.

deviate significantly from the curve. The best-fitting straight line through these points and those of October–November 1961 [*Bleichrodt*, 1962] has a slope x of 0.038 ± 0.002.

On August 5, 1962, the Soviet Union started a new series of nuclear tests. Samples taken after this date are also shown in Figure 1. Only one 'hot' radioactive cloud was sampled on September 25. The filter was cut into two pieces which were analyzed separately. Results differed by less than 8 per cent. The radioactivity ratio of Sr^{89} and Sr^{90} was 114 indicating that at least a large fraction of the debris originated from a recent detonation. As the Be^7 concentration in this sample differed significantly from the value observed for natural Be^7, it is probable that during the Soviet 1962 tests Be^7 was also produced artificially.

If LiD is used for nuclear devices in the megaton range [*Thirring*, 1955], Be^7 is probably produced by nuclear reactions of Li, since no Be^7 has been detected in fission products [*Cook*, 1952]. The most likely reactions are Li^6 (n, t) He^4 followed by Li^6 $(t, 2n)$ Be^7 [*Roy et al.*, 1960; *Thirring*, 1955], Li^6 (d, n) Be^7 [*Roy et al.*, 1960], and Li^7 $(d, 2n)$ Be^7 [*Garisson and Hamilton*, 1951]. Use of Li in the Soviet bombs is suggested by an increased Li twilight emission at Saskatoon during November 1961 [*Sullivan and Hunten*, 1962] and at Uppsala during November 1962 [*Stoffregen et al.*, 1963].

If the above interpretation is correct, the ratio of Be^7 and Sr^{89}, which is a fission product, is dependent on the ratio of fusion and fission energy release of the weapons tested. The fusion to fission ratio probably differs widely for individual detonations. The approximately constant ratio of Be^7 and Sr^{89} observed for the 1961 explosions may, therefore, be the result of a relatively rapid mixing of debris injected into the lower stratosphere. For debris reaching higher levels, e.g. debris from the 60-megaton test on October 30, 1961, which was assumed to have a small fission to fusion ratio (see appendix of *Machta et al.* [1962]), the Be^7/Sr^{89} ratio might have been quite different. The influence of high-altitude debris on the concentration of Be^7 in the lower stratosphere will be small and therefore will remain undetected because of radioactive decay during downward transport.

Note added in proof. Samples of stratospheric air taken on March 19 and 26, and on April 2 and 9, 1963, showed concentrations of Be^7 of 10.6, 13.5,

8.1, and 11.5 pc kg^{-1}, and concentrations of Sr^{89} of 182, 195, 100, and 143 pc kg^{-1}, respectively. These data are consistent with the curve of Figure 1. Assuming all the Sr^{89} to originate from the Soviet 1962 series (August–December), $e^{0.00078t}$ amounts to about 1.14.

Acknowledgments. We are indebted to Dr. Joh. Blok for fruitful discussions. Collaboration of the Royal Netherlands Air Force and the Royal Netherlands Meteorological Institute in stratospheric sampling is gratefully acknowledged.

References

Ajzenberg-Selove, F., and T. Lauritsen, Energy levels of light nuclei, 6, *Nucl. Phys., 11,* 1–340, 1959.

Bleichrodt, J. F., Increased concentration of beryllium-7 in the stratosphere after the nuclear test explosions during September-October 1961, *Nature, 193,* 1065–1066, 1962.

Cook, C. B., Search for beryllium-7 in uranium fission, *Nature, 169,* 622, 1952.

Garrison, W. M., and J. G. Hamilton, Production and isolation of carrier-free radioisotopes, *Chem. Rev., 49,* 237–272, 1951.

Lal, D., and B. Peters, Cosmic ray produced isotopes and their application to problems in geophysics, chapter 1 in *Progress in Elementary Particle and Cosmic Ray Physics,* vol. 6, edited by J. G. Wilson and S. A. Wouthuysen, North-Holland Publishing Company, Amsterdam, 1962.

Machta, L., R. J. List, and K. Telegadas, A survey of radioactive fallout from nuclear tests, *J. Geophys. Res., 67,* 1389–1400, 1962.

Osmond, R. G., and M. J. Owers, The half-life of Sr^{89}, *J. Inorg. Nucl. Chem., 9,* 96–98, 1959.

Roy, J. C., M. Bresesti, and J. J. Hawton, On the production of Be^7 in the NRX reactor by B^{10} (p, α) Be^7, Li^7 (p, n) Be^7, and Li^6 (d, n) Be^7 reactions, *Can. J. Phys., 38,* 1428–1435, 1960.

Stoffregen, W., H. Derblom, and B. Ånger, Lithium emission in twilight at Uppsala during November 1962, *Nature, 197,* 783–785, 1963.

Sullivan, H. M., and D. M. Hunten, Lithium twilight at Saskatoon, 1960–1961, *Nature, 193,* 1064–1065, 1962.

Taylor, J. G. V., and J. S. Merritt, Branching ratio in the decay of Be^7, *Can. J. Phys., 40,* 926–929, 1962.

Thirring, H., Thermonuclear power reactors—Are they feasible?, *Nucleonics, 13*(1), 62–66, 1955.

(Received February 27, 1963;
revised April 25, 1963.)