Krypton-85 in the Atmosphere

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Measurements of the krypton-85 in atmospheric krypton have been reported by deVries[1] and by Delibrias et al.[2] The atmospheric krypton-85 produced by the fission of uranium or plutonium may possibly serve as a tracer for geophysical circulation problems. This article presents a recent variation in the concentration of atmospheric krypton-85, together with some details on the experimental method.

Experimental

Collection of the Atmospheric Krypton.—The atmospheric krypton of 10~50 ml. was collected in Tokyo from the liquid oxygen obtained from a small-scale nitrogen-liquefier machine after it had run continuously for a day or two. About 5 l. of this liquid oxygen—which is supposed to have more krypton than usual liquid oxygen because the liquid oxygen had been used as a coolant in the nitrogen liquefier—was evaporated in a Dewar vessel until the volume was reduced to about 100 ml. The gas evaporated from the final 100 ml. of liquid oxygen was passed through a trap refrigenated with liquid oxygen. Nearly 50 ml. of the atmospheric krypton gas was obtained from the liquid condensate (20~30 ml.) in the trap by heating the evaporated gas with calcium. Further purifications of the krypton were made by condensation in a trap at 61~65°K and by evaporation at the temperature of liquid oxygen. The purity of the krypton was examined by gas density measurements and by static vapor pressure measurements at the liquid oxygen point.

Radioactivity Measurements.—The radioactivities of the purified krypton samples were measured by using the small-volume Geiger counter shown in Fig. 1. As shown in Fig. 2, the counter worked as an external quenching Geiger counter and had a good characteristic curve for the mixture of the purified krypton of 150~350 mmHg and propane of 2~5 mmHg. The effective volume of the counter was estimated from the geometrical dimensions, and the end effect was measured by the use of the movable teflon rod in the counter, as shown in Fig. 1.

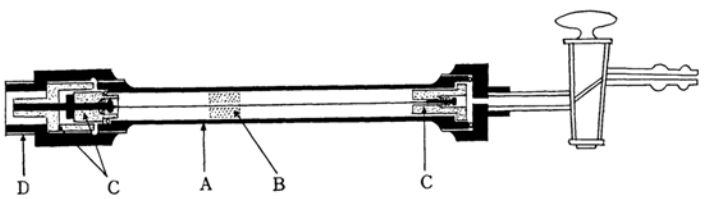

Fig. 1. Small volume counter tube.
A: Copper tubing; D=10.4 mm., L=97.0 mm.
B: Movable teflon rod used for the measurement of effective volume
C: Teflon insulator
D: High voltage supply connector

1) H. deVries, *Appl. Sci. Research*, **B5**, 387 (1956).

2) G. Delibrias and C. Jehanno, *Bulletin d'informations scientifiques et technologiques*, **No. 30**, June, (1959).

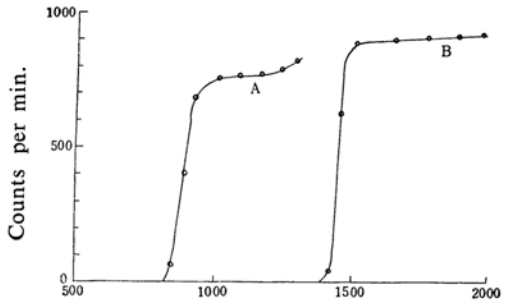

Fig. 2. Characteristic curve for Kr—Propane filled counter.
A: Kr 175 mmHg, C_3H_8 5.5 mmHg
B: Kr 386 mmHg, C_3H_8 5 mmHg

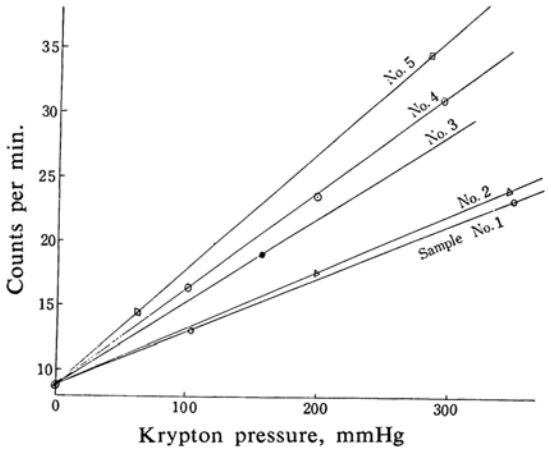

Fig. 3. Measured activities of the atmospheric krypton.

The measured counting rates showed a linear dependence on the amount of krypton in the counter. As shown in Fig. 3, the extrapolated counting rate of every sample at zero pressure met at a background counting rate, which was also checked with the counting with argon and propane. The number of disintegrations per minute was calculated by assuming a counting efficiency of 100% in the measured effective counter volume of 8.25 ml.

Results

The results of the measurements are shown in Table I. Sample 1 was collected from a large-volume liquid oxygen reservoir in an air liquefaction plant in Tokyo. Therefore the exact date of the collection of this sample is unknown. The data shown here and the data by Delibrias et al.[2] (1954; 1.160 d. p. m./ml. Kr; 1958: 4.200 d. p. m./ml. Kr) and by deVries[1] (March 1955: 26000 c.p.m./mol. Kr) show a steady increase in the concentration of atmospheric krypton-85 between 1954 and 1961. The increase in the atmospheric krypton-85 after the end of 1958 gives a maximum possible rate of supply of stratospheric krypton-85 to the troposphere, because the increase arose from the transfer of krypton-85 from the stratosphere to the troposphere and the direct supply to the troposphere by the atomic piles.

TABLE I. VARIATION OF KRYPTON-85 ACTIVITY IN THE ATMOSPHERIC KRYPTON

Sample No.	Collection date	Measured activity d.p.m./ml. Krypton (NTP)
1	Sept.—Nov. 1959	3.90±0.05
2	18—19 May 1960	4.14±0.04
3	9—11 July 1960	5.97±0.05
4	1— 2 Oct. 1960	6.86±0.09
5	10 Jan. 1961	8.32±0.09

The amount of man-made carbon-14 transported into the stratosphere from the troposphere after the biginning of 1959 can be estimated from the annual variation in mean atmospheric carbon-14 concentration. Using the measured variation of carbon-14 in tree rings[3,4] as the variation range of the atmospheric carbon-14, and assuming a mean residence time of 6 years for the carbon atoms in the troposphere before transfer to surface ocean water[5], the amount of injection of carbon-14 into the troposphere is estimated as follows:

$$1959: \quad 8\times10^{26} \text{ atoms/year}$$

$$1960: \quad 4\times10^{26} \text{ atoms/year}$$

The estimated total amount of man-made carbon-14 in the troposphere and ocean water is about 1.3×10^{28} atoms at the end of 1958[6]. Almost the same ratio of the amount injected into the troposphere from the stratosphere to the total amount in the troposphere and in ocean water may hold in the case of man-made carbon-14 and in the case of krypton-85 produced by nuclear tests.

The solubility of krypton in the sea water is so small that we can disregard sea water as a reservoir of krypton-85. When we assume that all the krypton-85 in the atmosphere in 1959 was produced by nuclear tests, the total atmospheric krypton-85 of 2.5×10^{26} atoms at that time corresponds to the carbon-14 of 1.3×10^{28} atoms in the troposphere and in sea water. Using these figures, the amount of krypton-85 injected from the stratosphere to the troposphere is estimated as 6×10^{24} atoms per year in 1959 and 3×10^{24} atoms per year

3) K. Kigoshi and Y. Tomikura, This Bulletin, **33**, 1576 (1960).
4) K. Kigoshi, *Journal of Radiation Research*, **1**—2, 111 (1960).
5) H. Craig, *Tellus*, **9**, 1 (1957).
6) F. Hagemann, J. Gray, L. Machta, Jr. and A. Turkevich, *Science*, **130**, 542 (1959).

in 1960. These figures correspond to the increase in atmospheric krypton activities of 0.24 d. p. m./ml. Kr/year in 1959 and 0.12 d. p. m./ml. Kr/year in 1960. The observed slow increase up to May, 1960, does not contradict the above estimate based on the parallelism between carbon-14 and krypton-85.

The increase after July, 1960, can not be attributed to the transfer of stratospheric krypton-85 to the troposphere. Even if the total fission energy released by nuclear tests between 1953 and 1958 were equivalent to the energy produced by 200 megatons of TNT, the total calculated amount of produced krypton-85 is about 7×10^{25} atoms. This amount is too small to explain the atmospheric krypton-85 after July, 1961, because the krypton activity of 8 d. p. m./ml. Kr corresponds to about 5×10^{26} atoms of krypton-85 in the whole atmosphere. The escape of krypton-85 from the atomic piles and from chemical processing plants for used fuel might be the main sources of the supply of atmospheric krypton-85 after July, 1960.

In the present situation, information on the mixing and movement of the air masses in the troposphere can be obtained when detailed measurements are made on the local distribution and variation in atmospheric krypton.

Summary

The activities of atmospheric krypton-85 were measured on the samples of krypton collected in Tokyo. The observed activities show a remarkable increase after May, 1960, which can not be explained by the production of krypton-85 by nuclear tests. The slow increase before May, 1960, does not contradict an assumed parallelism between the behavior of the man-made carbon-14 in the atmosphere and that of the krypton-85 produced by nuclear tests.

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