Geochemical Behavior of Fresh Nuclear Debris of Known Origin in the Atmosphere

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Recently we have made measurements[1,2] of two unique fallout activities which resulted from a Chinese atomic bomb explosion at ground-level on October 16, 1964 and the venting of a large-scale Russian underground nuclear test on January 15, 1965. In this paper we have briefly discussed the geochemical behavior of fresh nuclear debris. A relatively short-lived isotope ^{89}Sr (half-life, 50.4 days) could serve as a tracer which followed the behavior of fresh debris. Further, it was a favorable weather condition to estimate the air concentration of nuclear debris by measuring the activity concentration in precipitation that showery rain or snow occurs quite frequently during the period from October to March of a year in the Niigata district on the northern Japan Sea side of Honshu.

Results and Discussion

The mean specific activities in rain of ^{89}Sr and old ^{90}Sr (in pc/mm), and the ^{89}Sr/old ^{90}Sr activity ratio are shown in Fig. 1. An old ^{90}Sr fraction resulting mainly from the earlier nuclear test series (1961—1962) was evaluated in the same way as in the previous paper.[2]

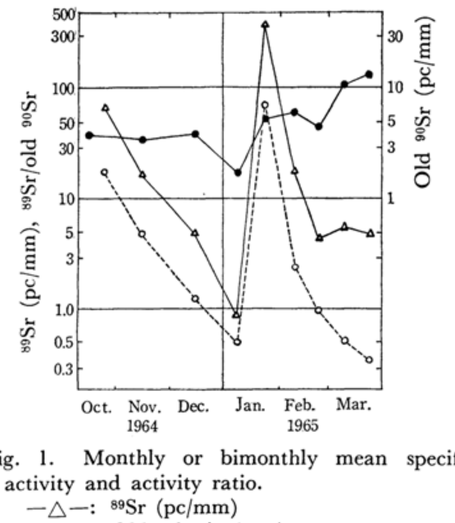

Fig. 1. Monthly or bimonthly mean specific activity and activity ratio.
—△—: ^{89}Sr (pc/mm)
--●—: Old ^{90}Sr (pc/mm)
--○--: ^{89}Sr/old ^{90}Sr ratio

First, it should be noted that the mean specific activity of ^{89}Sr in rain decreased at a more rapid rate than the radioactive decay rate of ^{89}Sr during the period from late October to December 1964. By contrast, the specific activity of old ^{90}Sr was kept almost unchanged. This finding suggested that old ^{90}Sr remained stationary in its air concentration for the whole period concerned, but that ^{89}Sr had been rapidly removed from the atmosphere three months after the Chinese explosion. Such independent behaviors of the two isotopes would imply that the Chinese ^{89}Sr was almost pure tropospheric in contrast to the old ^{90}Sr of stratospheric origin.

It is useful for studying the transitional behavior of fresh debris to evaluate an apparent decrease rate of the ^{89}Sr concentration in air from its measured specific activity in precipitation, in relation to the atmospheric mean residence time of nuclear debris. In this connection, since nuclear clouds were relatively new in this case, the concentration of nuclear debris in air over Niigata could vary with time for a number of reasons, *i. e.*, by lateral and vertical diffusion or merely by the passage of the cloud. However, when the specific activity of nuclear debris in precipitation is assumed to be proportional to its average air concentration, it is reasonable on the basis of a rough exponential decrease of the specific activity in rain of ^{89}Sr that the average air concentration decreases roughly following an exponential function of time as a result of the combined effects resulting from such various factors as mentioned above. Thus, an apparent decrease rate constant ($\lambda_R+\lambda$) of the air concentration of ^{89}Sr for a given period was calculated from the ratio between the two consecutive mean specific activities for each period; λ_R is a resultant rate constant consisting of vertical, lateral and other components mentioned above, λ being the radioactive decay constant of ^{89}Sr. On the other hand, ^{89}Sr/old ^{90}Sr ratio was regarded as a mean specific activity corrected for the reasonal factor associated with spring peak of stratospheric ^{90}Sr deposition—a specific activity for average stratospheric-debris conditions. The apparent decrease rate constants calculated are presented in Table 1, together with an apparent mean time for the decreasing process of ^{89}Sr concentration in air,

1) S. Koyama, T. Sotobayashi and T. Suzuki, *Nature*, **209**, 239 (1966).
2) T. Sotobayashi and S. Koyama, *Science*, **152**, 1059 (1966).

TABLE 1. APPARENT DECREASE RATE CONSTANT OF AIR CONCENTRATION AND MEAN RESIDENCE TIME FOR STRONTIUM-89

Period of collection	Apparent decrease rate constant ($\lambda_R+\lambda$)		Apparent mean residence time	
	By specific activity	By isotope ratio	From specific activity	From isotope ratio
	day^{-1}		day	
Oct. 18—Nov. 30, 1964	0.058	0.058	23	23
Nov. 1—Dec. 31	0.040	0.046	36	31
Dec. 1—Jan. 15, 1965	0.078	0.044	16	33
Jan. 16—Feb. 14	0.278	0.258	3.8	3.9
Feb. 1—28	0.100	0.078	12	16
Feb. 15—Mar. 15	—	0.047	—	30
Mar. 1—31	—	0.027	—	77[a]

a) This high value would be derived from the result of a progressive mixing of ^{89}Sr with old ^{90}Sr in upper levels of the atmosphere.

which is here designated as "an apparent mean residence time."

Comparison of the two methods may be readily made from the figures in Table 1. It will be clear that apparent mean residence times obtained by the two methods were comparable with each other during the period from late October 1964 to mid-January 1965. An average value of 27±6 days was estimated for the Chinese ^{89}Sr. This value is nearly equal to an average value of tropospheric mean residence times obtained by measurement of older and well-mixed nuclear debris,[3,4] and is also in good agreement with recent Beck's value[5] derived from the ^{89}Sr/^{90}Sr ratio in nuclear debris arising from the Chinese atomic bomb test (October 16, 1964). The Russian debris showed a shorter apparent residence time of 4 days from late January to mid-February and it agreed well with a shorter mean residence time (3 to 6 days) obtained by measurement of a nuclide ratio between short-lived fission products,[6,7] or between naturally present decay products of radon.[8,9] Consequently such a shorter value of 3 to 6 days would mostly be caused by an atmospheric residence of the material which directly entered lower levels of the troposphere and which remained there not yet well-mixed. From late February to early March, the apparent mean time of residence for the Russian ^{89}Sr attained to a 30-day value which equals a well-established tropospheric mean time of residence for stratospheric debris. The Russian ^{89}Sr gave this 30-day value two months after its production, while the Chinese ^{89}Sr showed this value a month after its formation. The gradual change ranging from 4- to 30-day values would reflect a slower atmospheric mixing of atomic cloud derived from the underground explosion. The intermediate value of two weeks can be considered as a resultant apparent mean time of residence resulting from the combination of a shorter 4-day value and a longer 30-day one.

The ^{89}Sr isotope derived from relatively new debris clouds could be used as a tracer for studying the geochemical behavior of fresh debris. When an atmospheric residence of fresh debris is estimated indirectly by the measurement of its decrease rate constant of specific activity in precipitation, it should be taken care of that an apparent mean time of the atmospheric residence of fresh debris depends largely upon the time interval between explosion and sampling and, further, upon whether the source of debris was present substantially above or under the ground surface.

3) W. Anderson, R. E. Bentley, L. K. Burton, I. O. Crookell and C. Greatorex, *Nature,* **186**, 925 (1960).
4) T. Sotobayashi and S. Koyama, *J. Fac. Sci. Niigata Univ.,* **I**, **3**, 111 (1962).
5) J. N. Beck and P. K. Kuroda, *J. Geophys. Res.,* **71**, 2451 (1966).
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7) G. Hermann, H. Houser and H. J. Riedel, *Nukleonik,* **1**, 305 (1959).
8) L. Lehmann and A. Sittkus, *Naturwissenschaften* **46**, **1**, 9 (1959).
9) L. M. Fry and K. K. Menon, *Science,* **137**, 995 (1962).