An Analysis of Long-term Observations of ^{90}Sr Deposition*[1]

Takeshi SOTOBAYASHI, Toshio SUZUKI and Yukio KANDA

Department of Chemistry, Faculty of Science, Niigata University, Niigata

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Measurements of the monthly deposition rate of ^{90}Sr and its time variation[1] have been made on a long-term scale of several years in our laboratory in order to estimate the cumulative deposition of ^{90}Sr per unit area and obtain more detailed information on its geochemical behavior in the atmosphere. We have recently discussed the ^{90}Sr fallout from surface and underground nuclear tests,[2] and, even more recently, the geochemical behavior of fresh nuclear debris of a known origin,[3] on the basis of radiochemical data for ^{89}Sr and ^{90}Sr. In the present paper we will make a brief analysis of our long-term observations of ^{90}Sr, a representative long-lived and hazardous fission product; further, we will estimate the cumulative deposition of ^{90}Sr per unit of area per unit of the activity produced by a series of atmospheric nuclear fission events.

Results and Discussion

More than 4 years after the termination of the atmospheric nuclear tests of 1961—62, measurable amounts of ^{90}Sr activity have been still detected in rain and ground-air samples at Niigata, although the yearly fall-rate had by 1966 lowered to one-eighth that of 1963. Table 1 shows the monthly deposition of ^{90}Sr per unit of area (mCi/km^2) and the monthly mean specific activity in precipitation (pCi/mm) during 1965—66. As can be seen in Table 1, a peak in the monthly deposition rate was observed in March for both years, while the peaks of the specific activity were in March for 1965 and in April for 1966; in 1966 the maximum values of the deposition rate and the specific activity decreased by 40 and 45 per cent respectively compared to those in 1965. The minimum deposition rates for both 1965 and 1966 were obviously found in August, but the 1966 rate dropped to 50 percent of that of 1965; the minimum specific activity in 1965 was clearly found in September, but in 1966 the specific activity ranged from 0.5 to 0.7 pCi/mm, with no definite trend observable during the period from July to December; it was difficult to determine which month of the year had a minimum value. Such a unique pattern of the specific activity remaining rather constant during the 4th quarter of a year was here observed for the first time since the beginning of our measurement of fallout activities.*[2]

TABLE 1. MONTHLY DEPOSITION OF ^{90}Sr AND ITS SPECIFIC ACTIVITY AT NIIGATA DURING 1965—1966

Month of collection	1965		1966	
	Monthly deposition mCi/km^2	Specific activity pCi/mm	Monthly deposition mCi/km^2	Specific activity pCi/mm
Jan.	0.81	3.9	0.21	2.4
Feb.	0.60	5.9	0.27	2.5
Mar.	0.86	11.6	0.35	3.3
Apr.	0.21	3.9	0.33	5.2
May	0.74	6.0	0.29	2.5
June	0.38	2.4	0.26	2.4
July	0.43	1.2	0.19	0.5
Aug.	0.06	1.5	0.03	0.6
Sept.	0.11	0.8	0.11	0.5
Oct.	0.19	1.4	0.14	0.5
Nov.	0.24	1.2	0.15	0.7
Dec.	0.37	1.2	0.12	0.5

Table 2 shows the yearly total of ^{90}Sr deposition per unit of area and the ratio between the two yearly totals of two consecutive years, namely, the yearly decrease rate, during the periods of 1960—61 and 1964—66, together with the yearly amount of rainfall. The decrease rate for 1960—61, 2 to 3 years after the termination of the 1957—58 test series, showed almost the same value as those for 1964—1965 and 1965—1966, 2 to 4 years after the 1961—62 test series. This finding would suggest that the decrease rate seems to have shown a nearly constant value of 0.5 more than one year after the end of each test series in spite of a number of differences in the actual test conditions, *e. g.*, the total energy yields and the altitude at which the nuclear explosions took place. Meanwhile, during the 1964—66 period there were 6 instances of radioactive air contamination at Niigata, resulting from 5 Chinese nuclear tests and an

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1) T. Sotobayashi and S. Koyama, *J. Fac. Sci. Niigata Univ., I*, **3**, 111 (1962).

2) T. Sotobayashi and S. Koyama, *Science*, **152**, 1059 (1966).

3) T. Sotobayashi, S. Koyama and T. Suzuki, This Bulletin, **40**, 1269 (1967).

*[2] According to our long-term ivnestigation of detailed seasonal variation in the monthly ^{90}Sr deposition at Niigata, the monthly value usually showed an increasing trend from October to December of any given year. *Nippon Kagaku Zassi (J. Chem. Soc. Japan, Pure Chem. Sect.)*, **89**, 206 (1968).

accidental release of nuclear debris due to a Russian underground test,[4] but the contribution to the total by the ^{90}Sr originating from these tests was small; the percentages of contributions by the ^{90}Sr fraction newly arising from each nuclear detonation to the yearly total were estimated to be 0.7, 3.0, and 2.4 per cent for 1964, 1965, and 1966 respectively. (In this estimation the two respective ^{90}Sr fractions resulting from the May 14, 1965, and the December 28, 1966, tests have not been taken into account, because data for the former were unavailable and the latter made no significant contribution to the yearly total of ^{90}Sr in 1966.)

TABLE 2. THE YEARLY TOTAL OF ^{90}Sr DEPOSITION AND THE YEARLY DECREASE RATE

Year of collection	Yearly total of ^{90}Sr mCi/km^2	Rain fall mm	Decrease rate %
1960	1.67	1780	53
1961	0.88*	2030	
1964	9.35	1880	54
1965	5.00	1880	50
1966	2.48	1950	

* This figure represents a value corrected for the contribution from the ^{90}Sr fraction originating from the 1961—1962 test series.

Table 3 gives two 10 year total depositions of ^{90}Sr per unit of area after the end of each test series; they were evaluated on the assumption that the yearly deposition of ^{90}Sr originating separately from the 1957—58 and the 1961—62 tests would independently continue to decrease at a decrease rate of 0.5 for 1962—68 and 1967—72 respectively.

4) S. Koyama, T. Sotobayashi and T. Suzuki, *Nature*, **209**, 239 (1966).

TABLE 3. ESTIMATED TEN-YEAR TOTAL OF ^{90}Sr DEPOSITION AND ITS RATIO TO TOTAL ^{90}Sr ACTIVITY PRODUCED

Test series	Estimated 10-year total of ^{90}Sr per unit area, mCi/km^2	Total ^{90}Sr activity produced* MCi	Total ^{90}Sr deposition/ ^{90}Sr produced mCi/km^2/MCi
1957—1958	26	4.0	6.5
1961—1962	55	9.6	5.7

* U.S. Federal Radiation Council Data (Ref. 5)

The values obtained are approximately equal to the expected total deposition at Niigata of ^{90}Sr which would be derived separately from each of the two test series. Thus, from these figures, the ratio of a 10 year total of ^{90}Sr deposition per unit of area to an estimated total of ^{90}Sr activity[5] produced by each of the two test series (Table 3) was estimated to be 6.5 and 5.7 mCi/km^2 per megacurie for the test series of 1957—58 and 1961—62 respectively. The fact that these ratios nearly equalled each other leads to the possible conclusion that a total deposition of ^{90}Sr per unit of area obtained at an observation site as a result of considerably long-term measurements would be proportional to the total ^{90}Sr activity produced, so long as nuclear tests have been performed in the atmosphere, in spite of the actual different test conditions. However, it should be noticed that this is not true for the case of a land-surface burst, such as the October 16, 1964, test by the Chinese; the corresponding ratio for the Chinese test[2] was estimated to be 32 mCi/km^2 per megacurie, about 5 times greater than those obtained above.

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5) Value taken from data in the Fourth Report of the U. S. Federal Radiation Council (July, 1963).