Radioactive Contamination of Milk and Milk Products in Japan

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Received May 19, 1959

Radioactive contamination of milk and milk products in Japan was studied extensively. Samples were collected monthly or every season from several districts of Japan.

In this report, total radioactivity of raw milk and skim milk powder and strontium-90 (Sr-90) concentration of skim milk powder and sweetened condensed milk were measured. These experiments show that there is neither seasonal variation nor regional variation in total radioactivity, and that in the measurements of Sr-90 concentration the values in northern districts are higher than those in other parts of Japan.

INTRODUCTION

On account of nuclear fission products born out of nuclear weapon tests, there is an increasing tendency in the radioactive contamination upon air, rain water, soil, cereals, vegetables, farm animals and their products from year to year.

Among nuclear fission products, Sr-90 and cesium-137 (Cs-137) which have long half-lives are said to be the most dangerous elements for human body. Sr-90, as well as calcium, belongs to the alkaline-earth metals, and, therefore, its chemical properties resemble to those of calcium. In this respect, it is reasonable to expect that strontium behaves similarly as calcium in biological processes when it enters into the human body.

Europeans and Americans usually take their calcium mainly from milk and milk products, while Japanese take chiefly from vegetable foods. But recently the consumption of milk and milk products in Japan has gradually been accelerated year by year. Especially, artificial-feeding infants take their nutrition mainly from milk and milk products. Therefore it is of significance to investigate the radioactive contamination in

milk and milk products in Japan.

For this purpose, the measurements of total radioactivity in raw milk and skim milk powder and concentration of Sr-90 in skim milk powder and sweetened condensed milk were carried out accordingly.

EXPERIMENTAL

I. Total radioactivity in raw milk and skim milk powder

(1) Samples

Raw milk and skim milk powder which were collected monthly from June 1956 to May 1957 from 41 and 17 localities respectively were used for radioactivity assay. But occasionally when skim milk powder was not manufactured in some localities, whole milk powder was used for assay instead of skim milk powder.

(2) Measurement of radioactivity

(a) Ashing of samples

Raw milk: 180 ml of raw milk were measured into a beaker, evaporated in boiling water, transferred to a crucible, heated two hours in a crucible type electric furnace until a carbonaceous mass was obtained, and then transferred to a muffle furnace and heated below 550°C until a white ash was obtained.

Skim milk powder: 25 g of skim milk powder (in case of whole milk powder 20 g was used) were weighed out into a crucible, and then treated in the same way as mentioned in the ashing of raw milk.

(b) Measurement of radioactivity: About 0.5 g of ash was weighed out into a counting tray, 25 mm in diameter and 6 mm in depth, manufactured by the Physical and Chemical Research Institute.

After leveling the surface of ash with a glass rod

having a flat end, it was weighed accurately, and then the radioactivity was measured with Model 32-C Geiger Müller counter tube manufacture by the Physical and Chemical Research Institute. The radioactivity was expressed as cpm per 1 g of ash. The time required for

TABLE I. TOTAL RADIOACTIVITY OF RAW MILK (cpm/g of Ash)

Locality	Number	1956 June	July	August	September	October	November	December	1957 January	February	May
Hokkaido	A 1	45	48	46	47	48	45	48	36	39	47
"	B 2		56	40	49	45	44	46	46	54	52
"	C 3	42	47	45	46	51	45	47	39	50	47
"	D 4		50	44	51	40	46	48	30	40	49
"	E 5	54	47	46	43	46	41	45	40	40	39
"	F 6	41	53	41	53	52	46	44	43	42	45
"	G 7	44	51	44	36	49	48	50	44	46	48
Tôhoku	Iwate	8	52		51	45	53	49	40	38	37
"	Miyagi	9	51	47	45	46	45	47	40	52	52
"	Yamagata	A 10	45	51	46	46	49	47	49	33	45
"	"	B 11		49	42	46	44	46	41	35	39
"	Fukushima	12	41	54	45	23	44	48	45	46	42
Kanto	Tochigi	13	42	57	42	49	44	49	48	44	37
"	Gunma	14	48	47	46	45		43	45	49	35
"	Ibaragi	15	45	49	41	45	50	48	49	36	39
"	Saitama	16	49	50	42	43	48	45	48	50	30
"	Tôkyo	17	49	43	49	48	43	50	40	43	46
"	Chiba	18	51	57	47	46	49	51	48	60	53
"	Kanagawa	19	45	40	49	43	52	48	50	39	46
Chûbu	Niigata	20	51	48	48	46		47	52	42	36
"	Nagano	A 21	45	46	33	48	40	39	40	44	34
"	"	B 22	48	47	45	43	47	47	46	35	41
"	Ishikawa	23	57	48	45	49	48	45	47	35	46
"	Yamanashi	A 24	45	53	43	44	45		46	41	47
"	"	B 25	48	47	42	41	37		43	39	50
"	Shizuoka	26	52	54	38	44	46	45	46	40	46
"	Aichi	27	49	45	45	45	48	43	45	42	48
Kinki	Kyôto	28		47	48	47	49	47	48	44	40
"	Ôsaka	A 29	48		40	42	44	46	45	41	45
"	"	B 30	50	51	43	44	44	45	47	43	45
"	Hyôgo	A 31		47	42	46	45	40	45	44	42
—	"	B 32		51	39	43	46	55	50	51	54
Chûgoku	Tottori	33		61	45	44	47	48	46	40	54
"	Okayama	A 34	46	53	42	44	43	49	47	35	53
"	"	B 35		50	38	43	53		50	25	55
"	Yamaguchi	36	48	47	38	43	46	44	46	48	46
Shikoku	Tokushima	37	52		45	47	45	46	46	55	50
"	Ehime	38		49	36	49	55	47	48	40	46
Kyûshû	Saga	39	51	46	43	46	47	48	50	35	47
"	Kumamoto	40	54	56	53	46	49	51	50	55	58
"	Kagoshima	41		46	40	41	47		46	44	38

TABLE II. TOTAL RADIOACTIVITY OF SKIM MILK POWDER (cpm/g of Ash)

Locality	Number	1956 June	July	August	September	October	November	December	1957 January	February	April	May
Hokkaido	A 1		46	39	53	45	51	46			50	36
"	B 2	44	45	38	41	47	40		26	33		
"	C 3	46				45	47					
"	D 4	49	49	49	50	45	40	31	41	42		46
Tôhoku Iwate	5	47	41	41	41	41	46				48	
" Yamagata	A 6	40	46	44	43	44		50			42	38
"	B 7	53	58	44	54	55	40			46	43	45
Kantô Kanagawa	8	50	50	43	47	50	42	43		40		49
Chûbu Nagano	A 9	41		38	51	38	49	45	40	40		
" "	B 10	43		48	49	41	50	44	42		46	48
" "	C 11	49	59	38	54	49	47	48	39			40
" Yamanashi	12		47	48	47	50	45	44				
" Shizuoka	A 13	48	45						41	39	38	37
" "	B 14					41	40				42	
Kinki Hyôgo	A 15	49	50	45				40				
" "	B 16	46			43	43	50	48	44			44
Chûgoku Okayama	17	45	46			43	50	47	43			43

counting was one hour both in background and in samples. The results are shown in Tables I and II.

II. Measurement of Sr-90 concentration in skim milk powder and sweetened condensed milk

(1) Samples

Skim milk powder (or in certain cases whole milk powder) collected from six districts every season (August, October in 1957, February, May in 1958) was used for the measurement of Sr-90 concentration. Besides these, sweetened condensed milk and skim milk powder manufactured before 1957 and 1956 respectively were also used for assay. But in the case of the latter, four kinds of skim milk powder collected from four districts were mixed in proportion to the output of each district, and then this mixture was submitted to assay.

(2) Separation procedure of Sr-90 and the measurement of radioactivity

(a) Outline of separation procedure: In the same way as described in Experimental I, the samples were burnt to ashes below 650°C.

100g of ash were generally used for this assay, but in the case of the samples which contained Sr-90 in comparative large quantity, 20~50g were used.

As to separation procedure, the nitric acid method, having been originated in Noll's method¹⁾ and improved by Asari²⁾, was adopted.

Outline of this method is as follows. Carrier-free sample was dissolved in dilute fuming nitric acid. This solution was boiled and filtered. The filtrate was evaporated in order to remove chlorine gas. After cooling, fuming nitric acid ($d=1.52$) was added, and this solution was allowed to stand overnight, and filtered through sintered glass funnel. The precipitate obtained was dissolved in hot water and filtered. After the filtrate was evaporated, fuming nitric acid ($d=1.52$) was added. The solution was filtered as before. This procedure was repeated two or three times.

The precipitate was dissolved in hot water and evaporated below 100 ml. 1.5M sodium chromate and ammonia were added to this solution. Then the solution was adjusted pH 5.5~6.0 with acetic acid, heated at 90°C, and filtered. After the filtrate was evaporated below 100 ml, ammonium acetate and barium carrier (barium chloride) were added. The solution was heated at 90°C, and filtered as before. The procedure was repeated once again. By this procedure barium and radium were separated from calcium and strontium.

Ammonia and excess of ammonium carbonate were added to the filtrate. The solution was boiled and filtered. The precipitate obtained was dissolved in dilute nitric acid and evaporated to dry up. The residue was dissolved in small amount of water and to this solution fuming nitric acid was added until the specific gravity became just 1.45. By this procedure only strontium nitrate was precipitated. The precipitate was dried and

1) W. Noll, *Z. Anorg. Chem.*, **199**, 193 (1931).

2) T. Asari and H. Watanabe, *Reports of the Physical and Chemical Research Institute*, **23**, 402 (1944).

TABLE III. Sr-90 CONCENTRATION OF SKIM MILK POWDER OR WHOLE MILK POWDER

Kind of Sample	Locality		Date		Ca% in ash	Sr/100 Ca	Sr ⁹⁰ μμc/g of Ca (S.U.)	Sr ⁹⁰ μμc/7 mg Sr (M.U.)	Sr ⁹⁰ μμc/kg of Sample
Skim	Hokkaido		August	1957	14.4	0.038	4.3±0.4	79.4	50.1
"	"		October	"	14.6	0.033	5.2±0.5	110.0	63.8
"	"		January	1958	14.7	0.062	6.7±1.5	76.5	82.2
"	"		May	"	14.7	0.032	4.2±0.97	86.9	48.1
Whole	Tōhoku	Miyagi	August	1957	15.0	0.030	1.2±0.1	28.0	10.5
Skim	"	Yamagata	October	"	13.5	0.036	2.0±0.2	39.4	21.7
Whole	"	"	January	1958	15.4	0.023	0.9±0.06	28.5	8.8
"	"	"	May	"	13.2	0.027	1.7±0.85	44.2	13.8
"	Kanto	Gunma	October	1957	15.1	0.021	1.1±0.2	35.5	10.1
Skim	"	"	January	1958	14.5	0.041	1.4±0.3	24.0	17.1
"	"	"	May	"	14.8	0.024	1.0±0.2	29.8	12.4
Whole	Chūbu	Yamanashi	August	1957	15.7	0.031	1.5±0.2	34.8	13.9
"	"	Nagano	October	"	14.9	0.034	1.2±0.1	25.4	11.1
"	"	"	January	1958	14.4	0.046	2.2±0.3	33.0	19.6
"	"	"	May	"	14.6	0.033	0.8±0.3	15.8	6.7
"	Kinki	Hyōgo	August	1957	14.0	0.026	1.3±0.3	35.9	11.6
Skim	"	"	October	"	14.5	0.025	0.61±0.12	17.3	7.6
"	"	"	January	1958	14.5	0.041	1.0±0.25	17.4	12.5
"	"	"	May	"	15.1	0.019	0.5±0.1	16.4	5.8
"	Chūgoku and Tokushima	Shikoku,	August	1957	13.6	0.022	1.2±0.2	36.2	13.9
"	"	"	October	"	14.3	0.041	1.4±0.3	23.2	16.6
"	"	"	January	1958	14.9	0.024	1.6±0.25	45.4	19.6
"	"	"	May	"	13.5	0.031	1.0±0.5	22.3	11.2

TABLE IV. Sr-90 CONCENTRATION OF SWEETENED CONDENSED MILK

Whole of Sample	Locality		Date		Ca% in ash	Sr/100 ca	Sr ⁹⁰ μμc/g of Ca (S.U.)	Sr ⁹⁰ μμc/7 mg Sr (M.U.)	Sr ⁹⁰ μμc/kg of Sample
Whole	Kanto	Chiba		1950	12.5	0.017	0.1±0.06	4.0	0.24
"	Hokkaido		July	1956	14.4	0.05	2	38.0	6.7
Skim	"		September	1957	14.8	0.043	3.4±1.0	54.4	12.2
"	Kanto	Kanagawa	"	"	14.5	0.035	2.6±0.05	51.4	9.0

TABLE V. Sr-90 CONCENTRATION OF SKIM MILK POWDER

Kind of Sample	Locality		Date	Ca% in ash	Sr/100 Ca	Sr ⁹⁰ μμc/g of Ca (S.U.)	Sr ⁹⁰ μμc/7 mg Sr (M.U.)	Sr ⁹⁰ μμc/kg of Sample
Skim	{ Hokkaido Yamagata Ishikawa Hyōgo }		{ 1951 1952 1953 }	14.0	0.018	0.054±0.03	2.1	0.65
Skim	{ Hokkaido Yamagata Ishikawa Hyōgo }		1956	14.4	0.07	2.1	21.0	2.2

weighed. After weighing, the strontium nitrate was converted to carbonate, and then submitted to counting.

This strontium carbonate was left for two or three weeks until radiation equilibrium was obtained between Sr-90 and Y-90, and then dissolved in hydrochloric acid. To this solution ferric chloride was added as carrier for measurement of yttrium, and carbonic acid free water saturated with ammonia was added. Yttrium was precipitated as yttrium hydroxide and this precipitate was submitted to counting.

(b) Measurement of radioactivity: Radioactivity was measured at the Yamazaki Laboratory of the Physical and Chemical Research Institute. The results are shown in Tables III, IV and V.

Sr-90 concentration in samples is given as Sr-90 $\mu\text{c/g}$ of Ca (S.U., Sunshine Unit). And, in addition, Sr-90 $\mu\text{c/kg}$ of sample was calculated for convenience.

DISCUSSION

I. Total radioactivity

It was found that there was scarcely any difference in the total radioactivity between raw milk and skim milk powder. The values shown in Tables I and II were averaged monthly as shown in Table VI.

TABLE VI. TOTAL RADIOACTIVITY AVERAGED MONTHLY

Kind of Sample	Date 1956 June	July	August	September	October	November	December	1957 January	February	April	May
Raw milk	48	50	44	45	47	46	47	42	45	—	44
Skim milk powder	46	47	43	48	45	46	45	40	40	44	43

Table VI shows that seasonal variation in total radioactivity is hardly found in both raw milk and skim milk powder. And also regional variation is hardly noticeable in Table I and II.

From these results, it is reasonable to expect that most of the radioactivity in milk could be attributed to natural radioactive substances and therefore variation in radioactivity owing to artificial radioactive substances, if any, hardly affects the total radioactivity.

In order to ascertain this, simple experiment was carried out. 0.5 g of potassium chloride, which was commonly used as reagent in chemical analysis, was weighed out into counting tray and its radioactivity was measured in the same way as mentioned in the measurement

of radioactivity.

As a result, the radioactivity of potassium chloride showed 118 cpm per 1 g. If all of the radioactivity of potassium chloride is originated in potassium, the radioactivity of potassium will be 226 cpm per 1 g. As to potassium sulfate, the same result was obtained. Further, it was shown by Nakae³⁾ that the content of potassium in milk ash was averaged 0.21 g per 1 g of ash. From these results, consequently, it is recognizable that the radioactivity of potassium in 1 g of milk ash become about 48 cpm.

Bergh⁴⁾ showed that most biological materials contained appreciable K-40 activity, in case of milk 3000~5000 dpm per liter. In authors' experiment, 40~50 cpm per 1 g of ash is converted to 2400~4000 dpm per 1 liter of milk.

Putting all these results together, it is considered that most of the radioactivity in milk would originate in natural radioactive substances, especially such as K-40.

It is considered that this is the reason why neither seasonal variation nor regional variation

in total activity was found in this experiment.

II. Sr-90 concentration

Table III shows that Sr-90 concentration in skim milk powder is higher in the northern districts of Japan than in the central or southern districts and the highest in Hokkaido.

A report from the Japan Meteorological Agency⁵⁾ showed that the accumulative amount of Sr-90 (presumptive value) was higher in Hokkaido and Tohoku (the north-east district of Honshu) than on the Pacific side of Japan. It

3) T. Nakae, *Bulletin of the National Institute of Agricultural Sciences, Japan*, No. 36 (1938).

4) H. Bergh, A Method for the Determination of Very Small Amounts of I-131 in Biological Materials, Especially in Milk A/CONF. 15/P/586, Norway, 28 May, 1958.

5) The present condition of observation of artificial radioactivity in Japan, March 1958: Reports of Japan Meteorological Agency.

is considered that this is one of the reasons why Sr-90 concentration in skim milk powder is higher in Hokkaido or in the north-east. Besides, the condition of feeding cows is considered as another reason.

In Hokkaido the grazing of cows is practised extensively, while in the other districts of Japan cows are usually fed in stalls. In case of grazing, there are more frequent occasions that cows directly take grass which is contaminated by radioactive rain water and directly exposed to radioactive rain water or dust than in case of feeding in a stall. Therefore it is considered that the milk produced in Hokkaido contains more Sr-90 than that in the other districts of Japan.

A report from the United National General Assembly⁶⁾ showed that the average Sr-90 concentration in milk produced in six stations of Canada was 5.0 S.U. in 1956 and 6.2 S.U. in 1957. These are similar values in Hokkaido, but are much higher than in the other districts of Japan. And the same report showed the average Sr-90 concentration in milk produced in New York City was 3.9 S.U. in 1956 (from January to May) and 4.5 S.U. in 1957 (from January to August) and, further, average Sr-90 concentration in milk produced in Mandan was 9.2 S.U. in 1956 and 9.3 S.U. in 1957 (from January to July).

From above, it is recognized that Sr-90 concentration in milk is higher in Mandan than Hokkaido.

A report from West Germany⁷⁾ showed that the rate of radioactive contamination on milk produced in West Germany was similar to that in the United States of America and Canada, and that the averages Sr-90 concentration in milk was 4~5 S.U. in 1956.

Putting these data together, it is conclusively evident that the milk produced in Japan is less contaminated by Sr-90 than in Canada, the United States of America and West Germany.

6) United Nations General Assembly. Scientific committee on the effects of atomic radiation, The effects of atomic radiation, A/AC. 82/R. 65/Add. 3 8 May 1958.

7) Physikalisches Institut der Bundesforschungsanstalt für Milchwirtschaft, Kiel, Private communication (1958).

On the other hand, summarizing all results (shown in Tables III, IV and V) it is recognizable that the radioactive contamination on condensed milk and skim milk powder has shown an increasing tendency in its accumulation for the past decade in Japan. For instance, the Sr-90 concentration in condensed milk manufactured in 1956 is twenty times as high as that 1950.

But, in comparison with other foodstuffs, Radiological Data in Japan II⁸⁾ showed that the Sr-90 concentration in milk and milk products was much lower than in vegetables, tea, soybean and rice. Vegetables and cereals are directly contaminated by radioactive substances, while milk is filtered by passing through the cow's body. This is the reason why Sr-90 concentration in milk and milk products is lower than that in vegetables and cereals.

Table III shows that there is some seasonal variation in Sr-90 concentration, but judging merely from the results obtained, it is not possible to say about the relation between seasonal variation and atomic weapon test. As to this point, further investigations are required.

In order to account for the fact that Sr-90 concentration in skim milk powder manufactured in Hokkaido is higher than that in the other districts of Japan, it is necessary to investigate the Sr-90 concentration in food-stuffs, rain water and soil near the milk products factory in detail.

SUMMARY

Raw milk and skim milk powder collected monthly from several districts in Japan (from June 1956 to May 1957) were burnt to ashes and their total radioactivities were measured.

As a result, total radioactivities showed 40~50 cpm per 1 g of ash. There was neither seasonal variation nor regional variation in the total radioactivity.

Skim milk powder (or whole milk powder) collected every season from six districts in Japan

8) Y. Hiyama, Radiological Data in Japan II. Concentrations of Sr-90, Cs-137, Pu-239 and others in various materials on earth's surface. Sept. 1958.

(August, October in 1957, February, May in 1958) was burnt to ashes and its Sr-90 concentration was measured.

Results obtained showed that Sr-90 concentration in skim milk powder manufactured in the northern districts were higher than those in the central or southern districts of Japan.

The Sr-90 concentrations are as indicated in the following districts.

Hokkaido	: 4.2~6.7	Sr-90 $\mu\mu\text{C/g}$ of Ca
Tohoku	: 0.9~2.0	"
Kanto	: 1.0~1.4	"

Chubu	: 0.8~2.2	"
Kinki	: 0.5~1.3	"
Chugoku and Shikoku	: 1.0~1.6	"

The authors wish to thank Dr. Yamazaki and Mr. Hashizume, Physical and Chemical Research Institute, for having taken charge of counting in the measurement of Sr-90 concentration, and to Dr. Asari, Japan Inspection Co. Ltd., for his valuable counsel and good offices given in the measurement of Sr-90 concentration.