Journal of the Korean Nuclear Society
Vol. 3, No. 1, March, 1971

《Original》

Strontium-90 Levels in Milk

Chan Kirl Park, Kyung Rin Yang

Health Physics Division, Atomic Energy Research Institute

(Received October 20, 1970)

Abstract

The levels of strontium-90 in milk produced in Korea were determined during the past six years. Milk samples were collected from dairies and market shops in seoul area.

Strontium-90 in milk was separated from calcium using fuming nitric acid and purified radiochemically. After seculear equilibrium was completed, the radioactivity of yttrium-90 was counted in a low background beta counting system. The determination of stable calcium in milk was also made by volumetric method using 0.1 N potassium permanganate solution.

The highest value of 34.9 pCi ^{90}Sr/g-Ca was determined in August, 1966 and the lowest value was 7.5 pCi ^{90}Sr/g-Ca in August, 1967. From the result we can say that levels of strontium-90 are decreasing year after year and are far bellow the maximum permissible level recommended by International Committe on Radiation Protection.

요 약

방사성 낙진에 의한 우리나라 식품의 오염도를 파악할 목적으로 우리나라의 우유중의 스트론튬-90의 양을 측정하였다.

스트론튬의 분석은 발연질산법에 의하였으며, 스트론튬 단위로 결과를 정리하기 위하여 칼슘의 함량도 동시에 정량하였다.

방사능의 측정은 $^{90}Sr+^{90}Y$의 방사평형이 이루어진 후에 ^{90}Y의 방사능을 측정하였다. 액체 우유의 스트론튬-90의 함량이 가장 많았던 것은 1966년의 34.9 pCi^{90}Sr/g·Ca이었으며, 1966년을 고비로 현저한 감소를 보여주고 있다.

분유의 스트론튬-90의 함량은 액체 우유와 같은 값을 보여 주고 있으며, 방사선 장해면에서 고려할때 안전한 함량임을 알 수 있다.

Introduction

Radiation around us is called the environmental radiation, which comes from both natural and man-made sources. The development and the testing of nuclear weapons brought a substantial amount of man-made radiation into the environment, and to a much lesser degree, reactor operation, and the medical and technological use of radioisotope contaminated the environment.

The continuous survey of environmental radiation reveals that the concentration of

natural radioactivity, its distribution within a specific environment, and its occurrence within the human population living in that environment are fairly constant, but the concentration and distribution of various fission product vary both with time and with given location. The principal causes of change in the levels of fission debris are the frequent nuclear weapons tests of various magnitude on the surface of earth.

Predominantly through the ingestion of food and water contaminated with radionuclides, men are exposed to radiation from nuclear debris. As a study of surveillance on foodstuffs radiocontamination in Korea, strontium-90 radioactivity in milk was measured during the past six years.

As is well known, strontium-90 is one of the most hazardous nuclides among fission debris because of its long physical, biological half-life and a bone seeking beta emitting radionuclide.

In this paper, strontium-90 levels in liquid and powdered milk produced in Korea is presented.

Experimental Procedures [1~2]

(1) Sample preparation

a. Liquid milk-ashing

Pour 3 liters of milk into a stainless steel kettle. Unless the milk is sour already, acidify by addition of acetic acid. Evaporate and pre-ash over a bunsen burner, until the milk fat is burned off. Put the kettle into an incinerator and keep it at 450°C for 8~16 hrs until an almost white ash is obtained. Weigh the ash.

b. Dried milk-ashing

Put 400 g of dried milk into a stainless steel kettle. Put the kettle into an incinerator and ash it at 450°C for 8~16 hrs until an almost white ash is obtained. Weigh the ash.

(2) Radiochemical separation

a. Strontium separation

Weigh 10 g of ash into a 600 ml beaker. Add 50 mg of strontium carrier, 200 ml of distilled water and 40 ml of 16 M nitric acid. Heat until a clear solution is obtained. Allow to cool and filter through a blackband filter. Add 1 ml of syrupy phosphoric acid to the filtrate and neutralize with 17 M ammonium hydroxide until ammonia is in excess. Transfer to two 200 ml centrifuge bottles and centrifuge. Remove supernate and add 100 ml of distilled water to each bottle. Stir up the precipitate to obtain a slurry and centrifuge. Reject the washings.

Dissolve the precipitate by adding 20 ml of fuming nitric acid to each bottle and stirring using a glass stick. Combine the two solutions in a measuring cylinder and measure the volume. Return a half of the solution (v) to each centrifuge bottle and add to each 2.5 V–70 ml of fuming nitric acid. Cool in water with stirring for 30 minutes. Centrifuge and reject supernate.

Dissolve the residues and transfer to one bottle using 40 ml of distilled water. Add 90 ml of fuming nitric acid and cool with stirring for 30 minutes. Centrifuge and reject supernate. Transfer the residue to a 40 ml centrifuge tube with 20~30 ml of water.

Make the solution in the centrifuge tube alkaline with ammonium hydroxide, add solid ammonium carbonate, and heat in a boiling water bath to coagulate the carbonate precipitate. Centrifuge and reject the supernate.

Add 10 ml of water to the residue in the centrifuge tube and then 22.5 ml of fuming nitric acid cautiously. Cool with stirring for 30 minutes, centrifuge and reject supernate.

Dissolve the residue in 10~15 ml of water, add 2 mg of barium carrier and 1 drop of methyl red indicator. Neutralise the excess acid with 6 M ammonium hydroxide and add 1 ml of 6M acetic acid and 2 ml of 3 M ammonium acetate. Dilute the solution to 30 ml and heat in a boiling water bath. Add 1 ml of 1.5 M sodium chromate and continue heating for 5 minutes. Centrifuge and transfer supernate to a centrifuge

tube.

Make the solution alkaline with 17 M ammonium hydroxide, add solid ammonium carbonate and heat in a boiling water bath to coagulate the carbonate precipitate. Centrifuge and reject the supernate.

Dissolve the residue in a few drops of dilute nitric acid, and 1 drop of 100-vol. hydrogen peroxide, and add 5 mg of iron (Fe^{+++}) carrier. Heat in a water bath and stir to remove carbon dioxide. Dilute to 15~20 ml and make alkaline with carbonate-free ammonium hydroxide, heating for 2~5 minutes to complete precipitation. Centrifuge and transfer supernate to a 40 ml centrifuge tube.

Acidify the solution with 6 M nitric acid and add 10 mg of yttrium carrier and store for at least 20 days.

b. Yttrium separation and counting

After the ^{90}Sr-^{90}Y equilibrium has been attained, make the solution alkaline with carbonate-free ammonium hydroxide and heat in a boiling water bath to coagulate the precipitate. Note date and time of precipitation. Centrifuge and transfer supernate to a tube. Redissolve the precipitate in a few drops of 6 M nitric acid, dilute to 10~15 ml and precipitate with ammonium hydroxide. Heat in a boiling water bath. Centrifuge and add supernate to that from the previous precipitation.

Redissolve the precipitate in the minimum of 6 M nitric acid and add 10 ml of 8% oxalic acid solution. Heat in a water bath for 10~15 minutes. Cool and filter through a filter stick using a weighed filter paper. Wash with methanol taking care not to suck air through the filter. Dry and weigh. Put the filter with the precipitate on the disc of a ring-disc set, add two drops of dilute collodion solution, cover with a Myler film and put the ring on. Start counting immediately in a low background beta counter.

The yield of the strontium and yttrium are determined by the gravimetric method.

Calculate the strontium-90 content of the sample using the formula:

Sr-90-Activity=Y-90-Activity

$$=(A_y-B)\frac{F_t\times F_y\times 10^4}{R_{sr}\times R_y\times 2.22}\text{pCi}$$

Where A_y : Sample count in cpm

B : Background count in cpm

F_y : Efficiency factor of counter

F_t : Correction factor for decay of Y-90 from separation to midpoint of counting period

R_{sy}, R_y : Respective Sr and Y yields in %

(3) Calcium determination

Weigh 180 mg of milk ash into a 250 ml beaker, add 40 ml of distilled water and 5 ml of concentrated hydrochloric acid. Heat for a few minutes and pour the hot solution into a 250 ml centrifuge bottle. Add 20 ml of 8% oxalic acid, a few drops of bromocresol green indicator and enough of 4 M ammonia solution to change the colour of the indicator to blue-green (pH 4). Let the solution stand at room temperature for 4 hours, centrifuge and reject the supernate. Wash the precipitate twice with water, centrifuge and reject the washings. Dissolve by addition of 100 ml water and 5 ml of 2 N sulphuric acid and heating in a water bath with stirring. Titrate the hot solution with 0.1 N potassium permanganate.

Calculate the calcium content in the sample using the relation

1 ml 0.1 N $KMnO_4$=2,0035 mg Ca

and the formula

$$(Ca)_a=\frac{100\times V\times F\times 2,0035}{W_a}$$

Where $(Ca)_a$: Calcium content in ash sample in %

W_a : Weight of ash sample aliquot in mg

V : $KMnO_4$ consumption in ml

F : Factor of $KMnO_4$ solution, determined with oxalate standard

Table 1. The Strontium-90 to Calcium ratio in Liquid Milk Seoul, Korea

pCiSr-90 per gram Ca

	Jan	Feb	Mar	Apr	May	June	July	Aug	Sep	Oct	Nov	Dec	Annual Average
1965	☆	☆	☆	☆	28.9	24.2	32.5	20.9	24.0	24.8	21.9	23.6	25.1
1966	25.3	24.1	20.1	23.7	29.5	27.7	29.3	34.9	☆	☆	☆	☆	26.8
1967	☆	19.6	22.0	17.7	13.7	9.2	15.4	7.5	10.0	12.5	11.2	11.9	13.7
1968	☆	☆	23.0	11.8	13.7	31.9	21.7	16.0	14.8	11.8	19.2	17.9	18.2
1969	☆	14.0	10.4	21.3	☆	14.8	☆	16.2	13.2	12.9	16.7	24.0	15.9
1970	24.5	18.1	14.2	8.9	14.0	17.2	19.3	11.4	16.5	11.8	12.3	11.9	15.0

☆ Samples not collected

Table 2. The Strontium-90 to Calcium ratio in Powdered Milk Seoul, Korea July 1970

No.	pCi/g-Ca
1.	18.4±0.9
2.	16.6±0.8
3.	12.6±0.7
4.	22.8±1.1
5.	15.0±0.8
6.	14.4±0.9
7.	9.2±0.9
8.	4.9±0.8
Mean	14.2

Results and discussion

Nuclear explosions produce two radioisotopes of strontium: strontium-90 with a half-life of 28 years, and strontium-89 with a half-life of 51 days. Of all radionuclides, strontium-90 represents the greatest potential long-term hazard; in the body it moves with calcium and is incorporated into bone, where, because of its long half-life, it remains as an internal source of radiation.

Dairy products and foods of plant origin are the primary routes of strontium-90 to man. They involve both surface contamination of plants and contamination of soil. By the influence of the amount of testing on soil contamination, the relative importance of these two routes varies with time. The behavior of calcium influences the movement of strontium radionuclides from soil to man to some extent. For this reason, levels of strontium-90 are often expressed in terms of strontium unit, picocuries of strontium-90 per gram of calcium. In assessing the strontium-90 hazard, the total intake of both strontium-90 and calcium must be taken into account. For any single food two factors must be considered: How it is contaminated with strontium-90 and how much calcium it contributes to the diet.

Milk has been the most important single item used for analysis and evaluation of food contamination, including contamination with strontium-90, for several reasons: dairy products are the target source of dietary calcium, milk is produced regularly the year round; it is convenient to handle; it can be obtained from various geographical areas; and it contains the most important radiocontaminant. Table 1 and Table 2 show the results of 153 samples of liquid milk during the past six years and 8 samples of powdered milk which collected in 1970. From liquid milk, the highest level of 34.9 pCi ^{90}Sr/g-Ca was determined in August, 1966 and the lowest level of 7.5 pCi ^{90}Sr/g-Ca was in August, 1967. Remarkable decrease in annual average levels has been observed since 1967, which phenomena coincide with the fact that atmospheric nuclear tests have been prohibited. For the determination of powdered milk, samples were taken from the following companies: Dai Han Food Ind. Co., Seoul Dry Milk Co., Namyang Dairy Products Co., Young

Tae Food Co., Pusan U Up Sa, Morinaga Milk Industry Co., LTD, Japan and Sam Yang Food Co. The average contamination level in powdered milk of 14.2 pCi ^{90}Sr/g-Ca is the same level as liquid milk. Fig. 1 shows the annual variations of strontium-90 content in milk collected in Seoul and New York city which is located on the same latitude as Seoul. The tendencies of the variation in both areas are similar, but the contamination level in Seoul is actually about two times as high as in New York city. Although the contaminated level in Seoul area is far below the maximum permissible level of 296 pCi ^{90}Sr/g-Ca[4] for strontium-90 content in foodstuffs, the calculation of the maximum permissible level is based on the value of 2 uCi[3] for bone recommended by International Committe on Radiation Protection. The routine observation on food contamination should be continued on the following purposes;

(1) Determination of whether a given level in the diet is reached.

(2) Estimation of mean and maximum dietary levels as may be related to geographical considerations or dietary habits of special groups.

(3) Provision of informations for policy decisions and for the public.

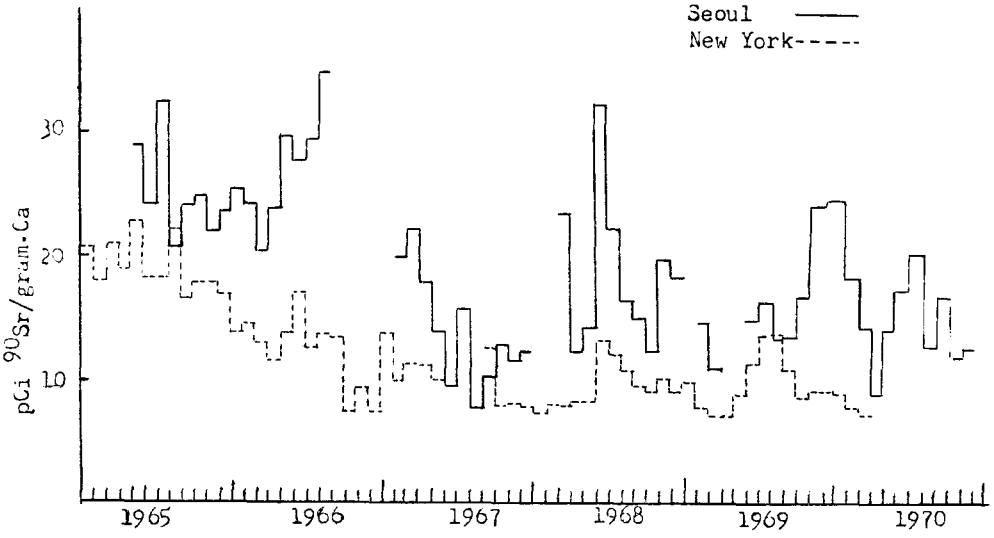

Fig. 1. Strontium-90 in Liquid Milk

References

1) HASL AEC, NYO-4700, p. E-Sr-01-01. (1967).
2) HASL AEC, HASL-227 APP., N. Y. 10014, p. E-4. (1970)
3) I.C.R.P., Report of Committee II on Permissible Dose for Internal Radiation., Pergamon Press, London. (1959).
4) Douyoyiannis, L., and Mimi kos,N. Health Physics 12, p. 1620. (1966).
5) K. R. Yang, J. of the Korean Chem. Society, vol. 13, No.2, p. 181~186, (1969).
6) K. R. Yang, J. of Nuclear Science(Korea), vol. 9, No. 1, part 1, p. 47~59, (1969).