

# Radionuclides in Milk

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Monthly milk samples from the milk sheds serving Atlanta, Austin, Chicago, Cincinnati, Fargo-Moorhead, New York City, Sacramento, Salt Lake City, Spokane, and St. Louis were analyzed for Ca and K content,  $I^{131}$ ,  $Ba^{140}$ ,  $Cs^{137}$ ,  $Sr^{89}$ , and  $Sr^{90}$ . Analysis of variance applied to these data indicated geographical and seasonal variations. The average  $Sr^{90}$  and  $Cs^{137}$  content of milk samples from Sacramento was the lowest, while that from St. Louis was the highest. The remaining stations were grouped into one of two intermediate categories. Samples collected during early spring and fall showed higher  $Sr^{90}$  and  $Cs^{137}$  than at other times, probably associated with changes in seasonal feeding practices. No relationship was observed between the concentration of  $Sr^{90}$  and  $Cs^{137}$  in milk and publicly announced weapons tests. In contrast, a direct relationship was observed between the weapons tests and the concentration of  $I^{131}$ ,  $Ba^{140}$ , and  $Sr^{89}$ , which was influenced by the half life of the radionuclides, the meteorological conditions, and the geographical location of the sampling stations with respect to the test site. Observed concentrations of radionuclides were below maximum permissible concentrations. Impracticability of estimating concentration of  $Sr^{90}$  in milk, in terms of gross  $\beta$ -activity or  $Cs^{137}$ , is discussed.

SAMPLES representing a wide variety of environmental media have been analyzed to provide information on the amounts and kinds of radioactive materials found in air, water, biota, soil, etc. The contribution of radioactive material from the diet represented a major source of environmental exposure and was worthy of special attention in evaluating the effect of radioactive materials on man.

Inasmuch as milk is produced throughout the United States at all times of the year and is consumed extensively by all segments of the population, this food was selected as the first item for analysis. Even though milk has been studied most intensively, analyses have been made on other foods as time and circumstances permitted.

In mid-1957, five stations were selected for study within the milksheds serving the metropolitan areas of Sacramento, Salt Lake City, St. Louis, Cincinnati, and New York. The results of the first year of operation were published in 1959 (3). The Public Health Service study differs from other milk surveillance programs in that each sample taken from a milkshed is representative of the same production area, and radionuclides with short half lives, such as  $I^{131}$ ,  $Ba^{140}$ , and  $Sr^{89}$ , are determined in addition to  $Cs^{137}$  and  $Sr^{90}$ .

Early results indicated variations in specific radionuclide levels with respect to time and geographical location and emphasized the need for continuation of the sampling program to provide a better understanding of the transfer of radioactive materials from their source,

fallout, through the food chain of the domestic animal to man.

In mid-1958, the sampling points were increased to include portions of 10 milksheds. The new locations included the cities of Spokane, Fargo-Moorhead, Chicago, Austin, and Atlanta. Since the publication of the first year's results, reports have been submitted to the Public Health Service's Divisions of Radiological Health and Engineering Services, and have formed the basis of the monthly releases made by the Department of Health, Education, and Welfare.

Many laboratories in this country and abroad are currently engaged in the determination of  $Sr^{90}$  and other radionuclides in milk and other foods, and among those of special interest are included two surveys in 1958 and 1959 (4, 5). These samples represent the largest geographical coverage of the nations' milk supply on a short-term basis insofar as the  $Sr^{90}$  content is concerned. Their reports show that, on the average, the  $Sr^{90}$  content of market milk was slightly higher during July and August 1959, than in the corresponding period of 1958—i.e., the level increased from 8.0 to 8.8 strontium units, respectively.

Extensive measurements of  $Sr^{90}$  in milk and other environmental media have been made by the Health and Safety Laboratory of the Atomic Energy Commission as part of its program on radionuclides in the environment. Similar studies are now under way in certain state health departments including New York and Minnesota. Of

special note is the work by Anderson and associates (7, 2) on  $Cs^{137}$  in dried milk.

The purpose of this paper is to review the results of analyses from May 1957 through December 1959, and to extend our observations on seasonal and geographical variations in the radionuclide content of milk.

## Methods

The sampling procedures are the same as described in the previous report (3). All the  $\gamma$ -emitting radionuclides are determined according to the method described by Hagee *et al.* (6), while  $Sr^{89}$  and  $Sr^{90}$  are determined by radiochemical methods (10, 13).

Since the initiation of this project several modifications in techniques have been developed which have resulted in substantial savings in manpower and expense. The most significant improvement in the determination of  $Sr^{90}$  is the elimination of the ashing step (12). A nonspectrometric method for the determination of  $I^{131}$  in milk has also been developed and may be used in conjunction with  $Sr^{90}$  analysis (17).

## Results

The results of analysis of milk samples collected in the Public Health Service study during the period May 1958 through December 1959 for Ca and K content,  $I^{131}$ ,  $Ba^{140}$ ,  $Cs^{137}$ ,  $Sr^{89}$ , and  $Sr^{90}$  are presented in Table I.

In considering the results here, however, the radionuclides have been divided into groups based on their half lives. The first group consists of  $Sr^{90}$

**Table I. Concentration of Various Radionuclides, Ca, and K in Monthly Milk Samples—May 1958 to December 1959**

	Concentration, Micromicrocuries per Liter																			
	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
	Strontium-90																			
New York	3.6	9.9	10.5	3.3	5.6	9.5	8.8	7.9	6.9	6.5	7.6	12.0	14.0	14.6	12.3	4.2	8.3	9.3	9.2	
Cincinnati	8.6	11.6	13.0	7.5	9.4	8.6	15.2	9.6	11.7	12.2	16.5	18.2	17.4	10.5	12.4	6.6	9.3	12.4	13.8	
St. Louis	10.0	14.8	18.7	14.1	15.4	12.2	20.1	15.6	18.6	19.6	37.3	34.6	11.2	17.6	19.6	15.0	25.1	24.0	21.8	
Salt Lake City	3.3	7.1	4.7	4.2	5.0	3.1	5.2	6.1	4.5	4.6	5.4	9.8	11.8	9.4	7.5	5.1	6.5	5.4	6.4	
Sacramento	3.8	7.1	3.3	6.8	3.8	1.4	6.1	4.2	4.3	3.9	7.5	8.6	6.4	4.3	3.0	1.0	3.6	6.1	3.7	
Atlanta	..	13.7	10.2	11.9	13.8	10.4	8.8	10.2	13.4	7.3	20.8	22.8	19.9	15.9	14.3	13.8	13.2	15.7	14.3	
Austin	..	2.2	3.5	3.4	3.9	5.1	3.7	2.6	5.7	6.3	8.9	6.5	8.2	6.3	4.2	2.7	3.8	5.3	7.0	
Fargo-Moorhead	..	15.6	16.3	10.2	14.1	15.0	11.5	12.3	12.3	11.8	12.2	16.2	20.6	22.1	15.5	8.1	11.4	14.5	15.8	
Chicago	..	..	7.1	9.3	7.9	6.0	8.4	7.0	6.7	6.3	7.2	12.5	12.1	11.7	9.3	5.2	10.1	8.4	8.1	
Spokane	..	..	..	6.9	8.2	8.6	11.9	8.6	9.1	6.3	10.9	22.6	22.1	18.3	5.9	5.6	9.8	12.1	13.1	
	Cesium-137																			
New York	..	..	43	36	32	37	46	41	45	58	46	55	39	44	36	17	20	35	40	
Cincinnati	..	..	65	35	..	24	54	39	47	29	54	57	47	34	14	12	15	40	35	
St. Louis	..	..	88	61	58	51	62	49	63	51	90	160	107	81	75	35	45	40	45	
Salt Lake City	..	..	24	28	30	21	26	17	26	34	..	63	62	..	31	30	35	35	35	
Sacramento	..	..	13	22	16	15	19	17	21	42	43	33	20	16	6	10	15	15	10	
Atlanta	..	..	55	52	39	45	27	43	61	60	90	102	74	58	..	40	30	40	70	
Austin	..	..	26	20	20	15	29	28	22	41	55	52	46	28	22	15	15	25	20	
Fargo-Moorhead	..	..	87	71	74	53	53	52	48	56	57	65	81	67	44	24	20	40	70	
Chicago	..	..	46	38	28	32	41	56	47	59	54	65	63	54	36	30	25	40	35	
Spokane	..	..	..	53	28	57	54	57	64	70	71	119	86	67	42	38	45	50	85	
	Strontium-89																			
New York	23	63	84	41	73	85	55	11	10	6	7	21	29	9	15	0	0	0	0	
Cincinnati	70	106	194	97	98	56	108	37	33	14	19	53	32	16	9	5	0	0	0	
St. Louis	107	122	306	263	283	139	168	84	77	84	130	140	77	27	13	10	14	0	6	
Salt Lake City	26	68	26	44	44	26	13	5	6	5	6	32	25	16	3	0	0	0	0	
Sacramento	38	40	20	24	17	11	18	10	15	12	58	25	10	5	3	0	0	0	0	
Atlanta	..	37	39	114	72	76	30	27	62	62	100	94	42	19	16	10	4	0	5	
Austin	..	26	15	40	32	35	49	32	27	37	68	38	24	7	11	0	0	0	0	
Fargo-Moorhead	..	80	142	53	140	161	53	30	27	17	12	20	44	31	19	10	5	0	0	
Chicago	..	..	59	64	94	53	64	14	7	1	6	57	19	18	12	6	3	0	0	
Spokane	..	..	..	64	43	53	35	21	20	16	15	90	54	24	20	11	0	0	0	
	Barium-140 <sup>a</sup>																			
New York	5	20	64	28	15	14	30	0	3	0	0	0	0	0	0	0	0	0	0	
Cincinnati	16	36	112	84	13	8	10	5	4	0	0	0	0	0	0	0	0	0	0	
St. Louis	14	44	229	113	48	14	36	8	14	4	5	0	0	0	0	0	0	0	0	
Salt Lake City	8	21	16	20	14	5	4	0	5	0	0	0	0	0	0	0	0	0	0	
Sacramento	12	6	7	0	0	1	14	6	0	0	0	0	0	0	0	0	0	0	0	
Atlanta	..	6	12	50	7	4	8	8	8	0	0	0	0	0	0	0	0	0	0	
Austin	..	1	5	18	6	0	30	12	2	3	0	0	0	0	0	0	0	0	0	
Fargo-Moorhead	..	18	47	14	53	16	28	0	1	0	0	0	0	0	0	0	0	0	0	
Chicago	..	..	36	18	10	1	3	8	0	0	0	0	0	0	0	0	0	0	0	
Spokane	..	..	..	13	5	8	52	7	3	0	0	0	0	0	0	0	0	0	0	
	Iodine-131 <sup>a</sup>																			
New York	32	0	67	69	10	61	35	9	0	5	0	0	0	0	0	0	0	0	0	
Cincinnati	0	0	107	42	5	31	177	8	2	4	1	0	0	0	0	0	0	0	0	
St. Louis	0	0	624	61	19	35	123	23	7	0	4	0	0	0	0	0	0	0	0	
Salt Lake City	0	0	156	77	14	11	33	8	2	1	2	0	0	0	0	0	0	0	0	
Sacramento	0	240	45	22	9	12	17	14	0	3	1	0	0	0	0	0	0	0	0	



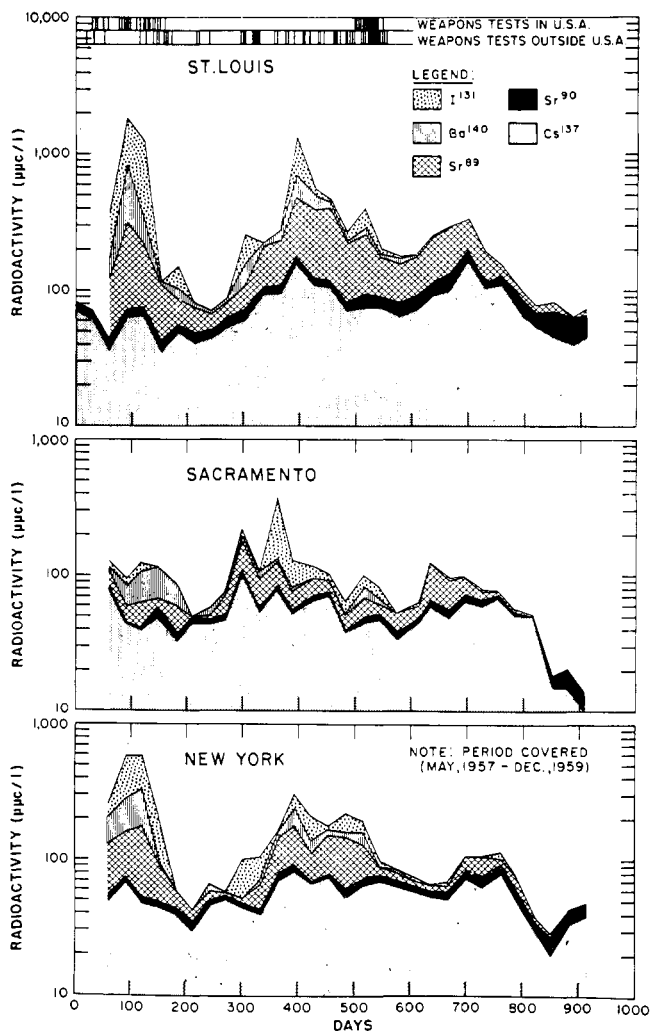


Figure 1. Radionuclide content of milk samples

the variation in levels of this radionuclide was similar to that of  $\text{Sr}^{90}$ . The milk from Sacramento had the lowest  $\text{Cs}^{137}$  content while that of St. Louis had the highest. The remaining areas fell into one of two intermediate groups as indicated in the table. Samples produced during the spring of 1959 contained significantly more  $\text{Cs}^{137}$  than samples produced at other times. Similarly, no clear association was observed between the concentration of  $\text{Cs}^{137}$  and weapons tests.

In contrast to the findings with  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$ , a correlation between weapons test activity and the occurrence in milk of radionuclides with short half lives was observed in all areas.

In Figure 1 the radioactivity contributed by each radionuclide determined (other than  $\text{K}^{40}$ ) in the samples received from the St. Louis, New York, and Sacramento milksheds has been summed and plotted as a function of time since May 1957. At the top of the graph the number of weapons tests conducted inside and outside the continental United States have been marked on the same time scale. The  $\text{Cs}^{137}$  values employed in this figure were derived from radio-

chemical analysis (13) to be consistent with the first year's results. The highest levels of radioactivity coincide with periods of weapons tests and were largely due to the presence of the shorter half-lived nuclides,  $\text{I}^{131}$ ,  $\text{Ba}^{140}$ , and  $\text{Sr}^{89}$ . This relationship is not precise, however, since the number of tests do not reflect the fission yield of the explosion and the collection of samples was made independent of the testing program. This is especially evident during the last period of tests in which most of the explosions were very small.

The pattern of activity observed for the New York and St. Louis samples appears to be true for all of the areas studied except Sacramento, in which the levels of activity are relatively nonresponsive to continental weapon tests. This difference is presumably related to the location of the milkshed with respect to the test area and the prevailing west to east movement of air.

In all locations  $\text{I}^{131}$  and  $\text{Ba}^{140}$  had decayed to negligible values by the end of 1958 or about 3 months after the last weapons tests. The half life of  $\text{Sr}^{89}$  is sufficiently long that measurable concentrations of this radionuclide were

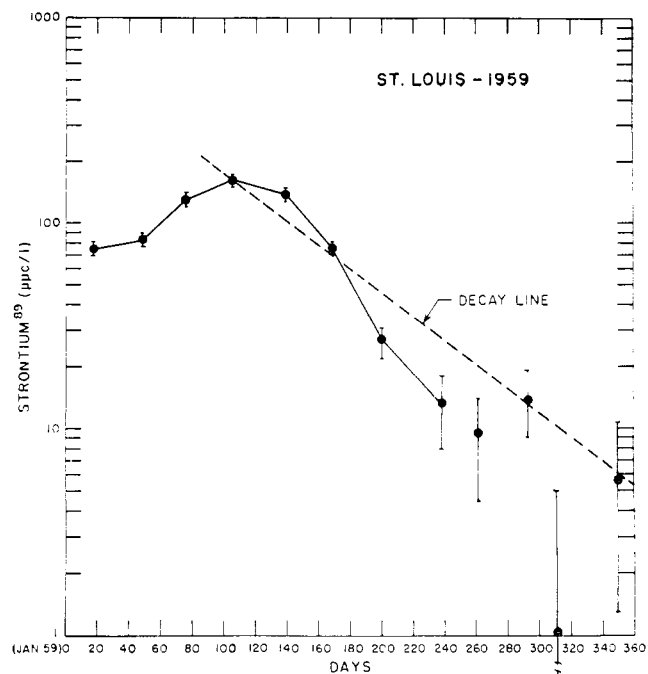


Figure 2. Concentration of  $\text{Sr}^{89}$  in milk samples from St. Louis area, January 1959 to December 1959

observed until the fall of 1959 or about a year after the last tests. At the present time, with the exception of  $\text{K}^{40}$ , most of the radioactivity in milk is due to  $\text{Cs}^{137}$  and  $\text{Sr}^{90}$ .

The variations corresponding to seasons through 1957 and 1958 also coincided in a general way with weapons tests activity; thus, it was not until the cessation of testing that the seasonal character of the variation in  $\text{Sr}^{90}$ ,  $\text{Cs}^{137}$ , and  $\text{Sr}^{89}$  in milk could be confirmed. This phenomenon appears to be generally true for all geographical areas studied.

In order to gain more insight into the influence of season on the level of radioactivity in milk, the concentration of  $\text{Sr}^{89}$  in the samples received from the St. Louis area from January 1959 through December 1959 has been plotted in Figure 2. During this period no weapons tests were conducted, but levels of  $\text{Sr}^{89}$  increased steadily from January to reach a peak in April, which coincided with the maximum concentration of  $\text{Sr}^{90}$ . The value obtained in May also reflected an increase in the  $\text{Sr}^{89}$  content since this value is above that which would have been expected as a result of the decay applied to the April sample, thus indicating that additional  $\text{Sr}^{89}$  was deposited during this period or was otherwise made available to the cattle. The June levels are falling at a rate faster than that evidenced by decay alone, while the July, August, and September values approximately follow the rate of decay.

The calcium and potassium contents of milk samples shown in Table I are provided so that strontium and cesium

units may be calculated. An analysis of variance applied to these data indicates a significant difference in the concentration of calcium both by month and geographic location. The samples from New York and Chicago had the lowest calcium content while those from St. Louis were the highest. Seasonally, samples received during July and August had the lowest calcium values. In contrast to calcium, the potassium content of milk samples from St. Louis was significantly lower than that from other milksheds with the samples from New York being highest.

In view of the inherent difficulties associated with the radiochemical determination of Sr<sup>90</sup>, any procedure which will provide evidence concerning the concentration of this radionuclide but not requiring the tedium associated with the direct analysis would be welcome. One approach to this problem has been the estimation of Sr<sup>90</sup> from the gross  $\beta$ -activity of the sample. This approach was not satisfactory, however, since the errors involved in the measurement of gross  $\beta$ -activity were usually greater than the observed concentration of Sr<sup>90</sup>. Furthermore, the gross  $\beta$ -activity of a sample may vary considerably with respect to time if radionuclides with short half lives are present. Correction of gross  $\beta$ -activity for K<sup>40</sup> did not appreciably improve its usefulness.

The second approach to estimating Sr<sup>90</sup> concentration may be found in the relation between Cs<sup>137</sup> and Sr<sup>90</sup>. Van Dilla and Anderson (14) and Low and Edvarson (9) have observed a relationship between these two radionuclides in which samples containing high concentrations of Cs<sup>137</sup> contain high levels of Sr<sup>90</sup>. In a general way similar observations may be made from the data presented here. However, the data presented in Table III for all samples from August 1958 through December 1959 show that the strength of the relationship between Cs<sup>137</sup> and Sr<sup>90</sup> is variable and in no case sufficiently great to be particularly useful in closely estimating the concentration of one radionuclide from an observed value of the other.

### Discussion

Adequate interpretation of the health significance of the data from a surveillance program for milk presents some formidable obstacles. The contribution of radioactivity from any one item of the diet or other environmental media does not reflect total exposure. Since maximum permissible concentrations (M.P.C.) for water and air were established primarily in terms of acceptable body burdens of the various radiation sources, a concept based on total intake appears to be a feasible approach for evaluating potential exposure. For

example, from the Recommendations of the International Commission on Radiological Protection (7), the permissible daily intake calculated from the M.P.C. for Sr<sup>90</sup> in water amounts to 2200  $\mu\text{c.}$  per day and for air, 2000  $\mu\text{c.}$  per day, giving an average value of 2100. This level of intake will not exceed a body burden of 2  $\mu\text{c.}$  over a 50-year period. This is, however, an occupational exposure for radiation workers and is based on a 168-hour week. If it is divided by 30, the factor recommended by I.C.R.P., one obtains an average general population permissible level (8) of exposure, or 70  $\mu\text{c.}$  of Sr<sup>90</sup> per day. This may be used as a point of reference in evaluating the total amount of Sr<sup>90</sup> received by man irrespective of source. (The Federal Radiation Council has been instructed to review the whole problem of radiation exposure and permissible levels and to advise the President of their conclusions.) A sample calculation of total intake is shown in Table IV based on analysis of samples made in the Cincinnati area during 1958-59. If this concept is accepted, these levels of Sr<sup>90</sup> activity, including that from milk, do not exceed the reference value. Recognition of the metabolic relationships of Ca and Sr<sup>90</sup> may be accomplished through the calculation of strontium units based on total Sr<sup>90</sup> and Ca intake. Although this concept is useful in estimating exposure levels, certain significant parameters are not included in the calculations of the M.P.C. Some of these include the nature of the chemical compounds with which the radionuclides are associated and the concentration of the stable elements corresponding to or very similar to the radionuclide. Furthermore, M.P.C. values have been developed on the basis of standard man and are not in fact directly applicable to certain segments of the population of great concern—namely, fetal life and the very young.

Until sufficient factual information on the biological effects of low levels of radiation is available, the interpretation of data from surveillance programs will be of only limited usefulness for evaluating the health effects of radiation exposure. Meanwhile, the need exists to continue the surveillance programs, to investigate the factors influencing the concentration of fallout products in foods, and to develop practical methods for the control, reduction, or removal of these substances from man's environment.

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**Table III. Statistical Relationships between Sr<sup>90</sup> and Cs<sup>137</sup>**

Milkshed	Correlation Coeff., $r$	Error Variance, $100(1 - r^2)$ %
Spokane	0.77	40.7
Austin	0.71	49.6
Cincinnati	0.69	52.4
Atlanta	0.68	53.8
Salt Lake City	0.67	55.1
Sacramento	0.65	57.8
Fargo-Moorhead	0.57	67.5
St. Louis	0.51	74.0
Chicago	0.31	90.4
New York	0.29	91.6

**Table IV. Estimated Ca and Sr<sup>90</sup> Intake from Environmental Sources, Cincinnati, 1958-59**

Source	Estimated Weekly Intake	Ca, G./Week	Sr <sup>90</sup> , $\mu\text{c.}/\text{Week}$
Air	140 cu. meters	..	5.6
Water	7 liters	0.4	5.6
Food			
Milk	3.5 kg.	4.2	42.0
Dairy products <sup>a</sup>	0.3 kg.	1.0	12.4
Vegetables	2.6 kg.	1.0	23.7
Meats, sea-foods and eggs	2.6 kg.	0.7	5.9
Cereals and pastries	0.9 kg.	0.4	9.5
Other	0.9 kg.	0.3	3.0
	Total per week	8.0	107.7
	Total per day	1.1	15.4

<sup>a</sup> Other than milk.

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## Radionuclides in Man from Nuclear Tests

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The results of recent experimental work in the determination of the stratospheric inventory of fission products—i.e., the fraction of  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$  taken up directly from rain—and new measurements of the concentration of  $\text{Sr}^{90}$  in human bone at this laboratory make possible a more accurate prediction of the future radiation doses from these isotopes to the world population. It is concluded that the U. S. diet has passed its peak concentration of  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$ , that  $\text{Sr}^{90}$  probably will remain the largest contributor to the radiation dose to an individual, and that  $\text{Cs}^{137}$  measurement can be used to monitor  $\text{Sr}^{90}$  in milk in emergency situations. These results also suggest that in the event of large-scale nuclear warfare, the general radioactive contamination would not preclude the existence of large populations if short time (6 to 12 months) survival were possible.

IN 1953, when the potential seriousness of world-wide fallout was first clearly identified, virtually nothing was known of the mechanisms, or rates of movement of nuclear debris from the point of detonation to the human population. In the intervening years, as a result of a major research effort at many laboratories, the larger aspects of the fallout problem have been solved. The previous speakers and their associates have played a large role in this development and their papers have treated many key problems in the movement of fission products to and through the food chains. It is my lot to summarize briefly the present status of the situation from these papers and other sources, and to report on some of the work at the Geochemical Laboratory of Columbia University on the levels of some of the critical radionuclides in man.

### Production and Distribution of Nuclear Debris

A comprehensive study of the stratospheric reservoir has been operative for the past 3 years. By using a meridional net woven by flights of manned aircraft and supplemented by the balloon program of the Atomic Energy Commission, results were obtained which now make it possible to state the rate and mechanism of removal of the debris from the stratosphere (8). Figure 1 shows the tropical and polar tropopauses, the discontinuity, and the ground level. Debris injected into the polar stratosphere has a half residence time of about

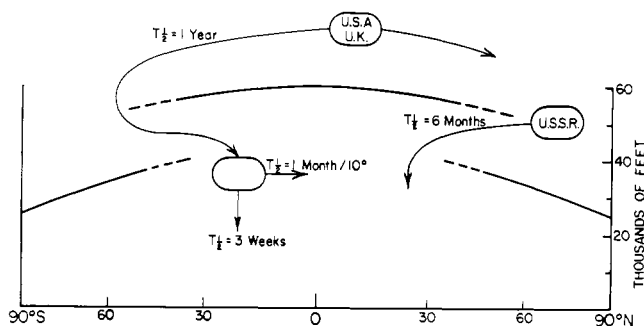


Figure 1. Transfer of fission product debris from stratosphere to surface of earth

6 months and essentially all comes out into the Northern Hemisphere. Debris injected by megaton weapons at the equator has a half residence time of about 1 year. These values were determined by actually measuring the change in the stratospheric reservoir with time.

The dominant transfer mechanism of stratospheric debris is through the break between the polar and equatorial tropopauses. The quantity of debris which passes through this gap is greater during the winter than at any other time of year, as this is the period of greatest activity of the jet stream which is located in this discontinuity. Once in the troposphere, stratospheric debris is removed by precipitation with a 3-week half life. The debris also moves laterally at the rate of about  $10^\circ$  per month. Since the polar regions have very low precipitation, this volume of the troposphere merely acts as a storage space,

and thus, the average specific activity of rain is essentially constant from  $30^\circ$  to  $90^\circ$  N or S. Toward the equator, however, the washout is effective so that a minimum in the deposition occurs in the  $0^\circ$  to  $10^\circ$  S latitude belt.

Figure 2 shows that the specific activity of rain increases in the spring of each year, following the winter stratospheric transfer, and then reaches a minimum in the fall. The fact that  $\text{W}^{185}$ , a tracer introduced in the U. S. Hardtack series at  $10^\circ$  N in the summer of 1958, also shows a spring peak in 1959, proves that this seasonal pattern has a meteorological origin and is not primarily due to Russian testing.

The distribution of the  $\text{Sr}^{90}$  on the surface of the earth is also fairly well known. The more recent estimate is shown in Figure 3 (78) where the ratio of specific activity on the ground between the latitude zones  $40^\circ$  to  $50^\circ$  N to