Mathematical Model of Transport Mechanisms Influencing Strontium 90 Levels in Milk

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AS A PART of its radiation surveillance program, the Public Health Service maintains a network of milk sampling stations throughout the United States. Measurements of radionuclide concentrations in milk have been reported since 1957. The original network consisted of five stations (St. Louis, Sacramento, New York City, Cincinnati, and Salt Lake City) reporting the concentration of strontium 90 in raw milk. Since 1960 a processed milk network consisting of 60 stations has been in operation, and values are reported monthly in *Radiological Health Data.* Strontium 90 values in general have been slightly lower for the processed than the raw milk, particularly in the St. Louis area. The raw milk network has been in existence for a much longer time, and fluctuations in deposition rates during the different testing situations have influenced the observed raw milk levels quite markedly. Because of the longer timespan covered by the raw milk measurements, it is convenient to use these numbers as baseline data to test any assumptions about the effects of the different factors influencing levels of strontium 90 in milk. Once the model incorporating the effects of these factors is developed, predictions for future levels in milk can be made.

In general, determination of the levels of strontium 90 in milk is important because of the extent to which milk contributes to total dietary intake, which in turn determines levels of strontium 90 in bone. Accurate predictions for strontium 90 content of milk should lead to more accurate predictions for strontium 90 in bone and the resultant dose to the bone marrow. In this paper, the 5 years of data from the raw milk sampling network are used to derive a model representing the mechanisms involved in the movement of strontium 90, from deposition on the plant to incorporation into the milk. Using variables derived from experimental observations, predictions are made for the levels to be expected in milk for each station during the spring and summer of 1962. An attempt is made to document the validity of each assumption made in the chain of reasoning.

Other attempts based on empirically reasonable relationships have been used to fit past milk data and to predict future levels. Dr. Harold Knapp of the Atomic Energy Commission (*1*) has attempted to relate the strontium 90 level in milk to deposition rates and cumulative deposition. He assumed that the proper function of the two factors should be a linear one.

Average strontium 90 in milk expressed in micromicrocuries per liter $= a_1x_1 + a_2x_2$
where

x_1 = average cumulative strontium 90 level in U.S. soil expressed in millicuries per square mile

x_2 = average strontium 90 deposition for the preceding month expressed in millicuries per square mile per month

The relative contributions from soil and current deposition can be deduced by estimating the scale factors a_1 and a_2 from the experimental

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data. Unfortunately, these results were derived by using data from only 2 months in the entire sampling period, even though the 2 months selected seem to be particularly significant. Critics have pointed out that the estimates for a_1 and a_2, which turned out to be 0.11 and 2.4 respectively, vary quite widely according to the particular 2 months picked for solution. In any case, if one has feelings, subjective or otherwise, about the relative effects of different months, weighted least squares is the appropriate estimation technique. Knapp's figures are about 30 percent too low during a period of about 9 months (October 1959 to July 1960), an anomaly which he attributes to winter barn feeding of cows in northern areas of the country.

Harris and co-workers (*2*) have introduced an empirical model for short-lived radionuclides which includes the effects of barn feeding. In predicting future milk levels, Kulp (*3*) has used the same general model as Knapp with different time periods. In his formulation x_1 refers to the cumulative deposit of strontium 90 measured at the midpoint of the growing season and x_2 is the average rate of deposition per 6 months of a particular growing season. In this case, only 5 years of data yielding five points can be used to estimate the 2 parameters a_1 and a_2. Thus, it is not surprising that a predicted level should agree with an observed level no matter what the true relationship or estimation procedure should happen to be. In fact, increasing the number of parameters to 5 would lead to a perfect fit regardless of the input variables. If one looks critically at these basic models, one is inclined to wonder why the milk levels should be related to the previous 1-month, 6-month, or any other period of deposition, and if so, why linearly. Harris (*2*) has pointed out that the apparent lag time from spring deposition peaks to milk level peaks is probably an artifact due to letting the cows out to pasture before the second peak.

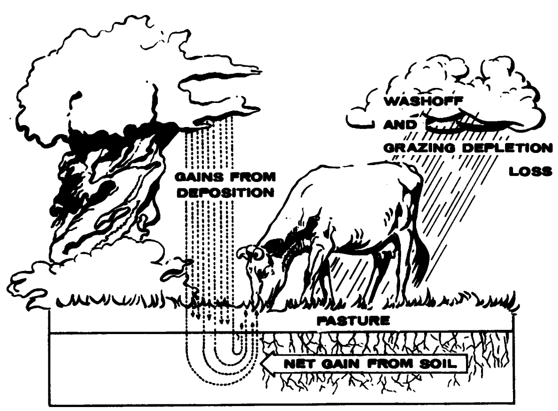


Figure 1. Schematic representation of strontium 90 movement

The preceding statements indicate a need for developing meaningful prediction models which apply not only to average U.S. levels over a yearly period but to specific area values which may prevail during particular time periods.

Strontium 90 Levels in Pasture Feed

Consider first the concept of "pasture level," which is defined as the concentration of strontium 90 per kilogram of dry feed taken from that pasture by grazing. This net level of strontium 90 in the pasture is dependent largely on the following factors:

1. Rate of deposition expressed in millicuries per square mile per month.
2. Net rate of gain from other pasture areas expressed in micromicrocuries per kilogram per month.
3. Net rate of absorption from soil expressed in micromicrocuries per kilogram per month.
4. Net rate of loss by washing off of strontium 90 particulates and grazing depletion of pasture grasses expressed in micromicrocuries per kilogram per month.

We may represent the pasture condition schematically as in figure 1.

The milk level is the result of sampling from the dairies in the milkshed and represents average transport factors which might prevail over a large number of farms as a whole, but not on any farm in particular. We would not expect as large an area as a county to have an appreciable net gain or net loss of strontium 90 particulates from other pasture areas. Thus, this factor is neglected in figure 1. In making computations average gain and loss factors will be used which will undoubtedly vary from farm to farm or from pasture to pasture. This, again, is justified by the observation that milk samples are derived from an averaging procedure over a large area and should be influenced by changes in average rate factors over that same area. In particular, we may make the assumption that the net rate of loss of foliar-deposited strontium 90 by washoff and grazing depletion is proportional to the amount of the radionuclide present on the foliage at any particular time. This means that during times when the foliage is highly contaminated more strontium 90 is lost. This assumption is consistent with the large drops observed in milk values following the spring peaks. That is, the quick buildup in strontium 90 is followed by an even more rapid plunge reflecting the increased amount available on the leaf for washoff. From the preceding, the rate of change equation for the amount of strontium 90 in the plant, exclusive of that contributed by the soil, may be written

$$\frac{dL(t)}{dt}=D(t)-\lambda L(t) \qquad [1]$$

where $D(t)$ is the deposition rate at time t, $L(t)$ the pasture level, and λ the depletion constant. The deposition rates expressed in millicuries per square mile have been measured by the Atomic Energy Commission and are taken from the Health and Safety Laboratory report (*4*). In conformity with Knapp, deposition rates for Tulsa were used for St. Louis, West Los Angeles for Sacramento, and Pittsburgh for Cincinnati. Solving equation 1, then gives

$$L(t)=C_1e^{-\lambda t}+e^{-\lambda t}\int e^{\lambda t}D(t)dt \qquad [2]$$

where C_1 is an appropriate constant and uptake from soil has yet to be considered.

Uptake from soil during any month is regarded as proportional to the amount present in the soil during that month. Soil measurements are available but their inherent variability seems to preclude any definitive quantitative specification. Knapp has presented national average soil levels of strontium 90. He shows that soil levels increased fairly slowly and uniformly over the 3-year period May 1957 to August 1960. We shall assume that local soil levels are proportional to the national average soil levels. This assumption is substantiated further by observing from figure 2 that local deposition rates are proportional to national deposition rates. Thus, equation 2 for the amount present in the pasture may be extended to

$$L(t)=C_1e^{-\lambda t}+e^{-\lambda t}\int e^{\lambda t}D(t)dt+C_2S(t) \qquad [3]$$

where $S(t)$ represents the level in the soil at time t.

The depletion constant λ, which is a measure of the rate of strontium 90 loss from the pasture, is unknown and must be estimated from the data. It is more convenient to work with $0.693/\lambda$ which is the half-residence time in the pasture. An upper bound for this value is the radioactive half-life of strontium 90, which is about 28 years. Values ranging for ¼ month to 28 years were tried using a computer. Best results were obtained for residence times between ¼ month and 2 months.

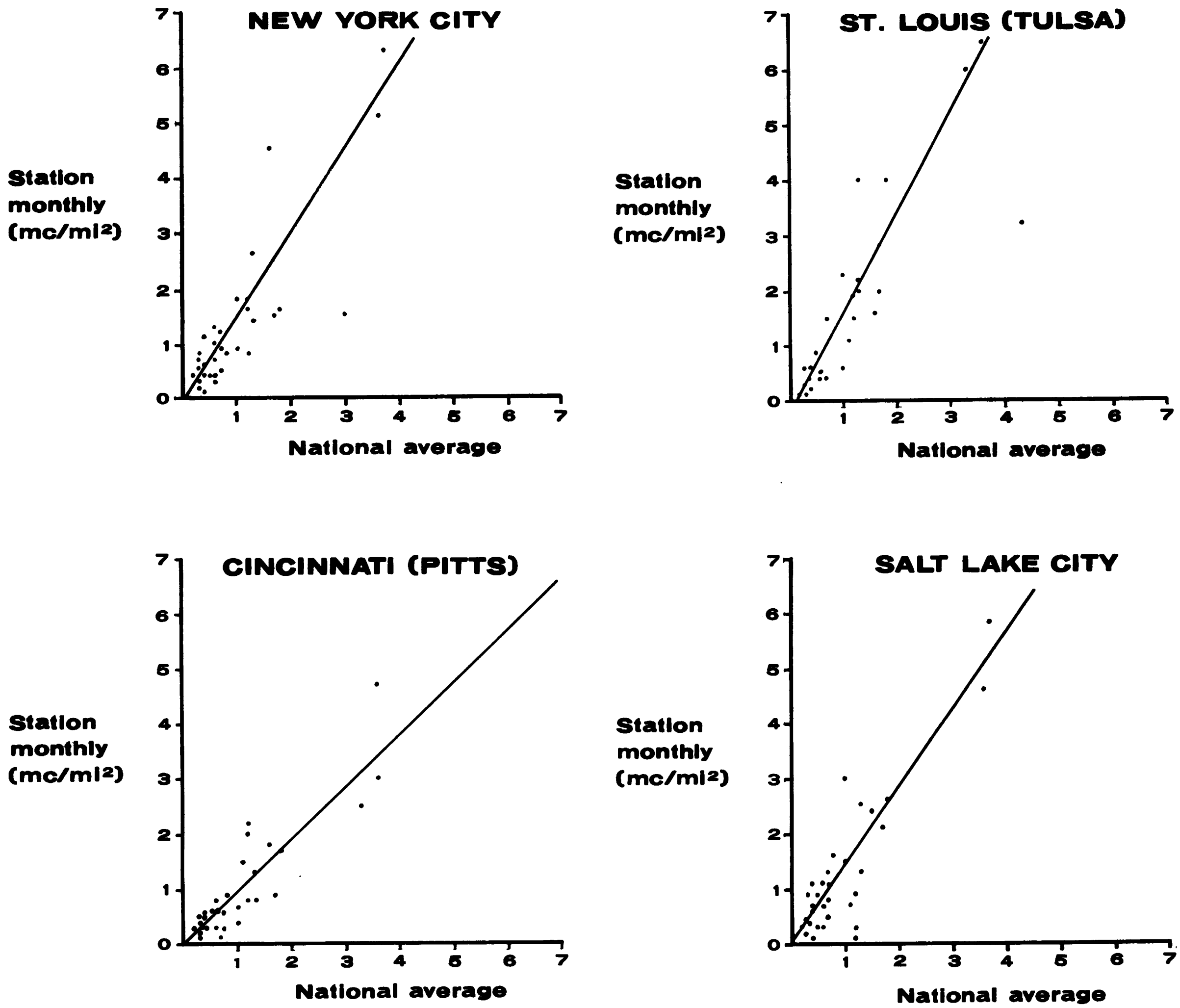
One might expect that rainfall would be a factor influencing the milk level, but trials of models using rainfall weighted by the strontium 90 inventory instead of deposition rates were unsatisfactory. The rate of major concern is that of deposition on the pasture. Climatic changes will reflect themselves as changes in these deposition rates. Therefore, $D(t)$ in equation 3 was replaced by a term proportional to monthly rainfall and stratospheric inventory of strontium 90.

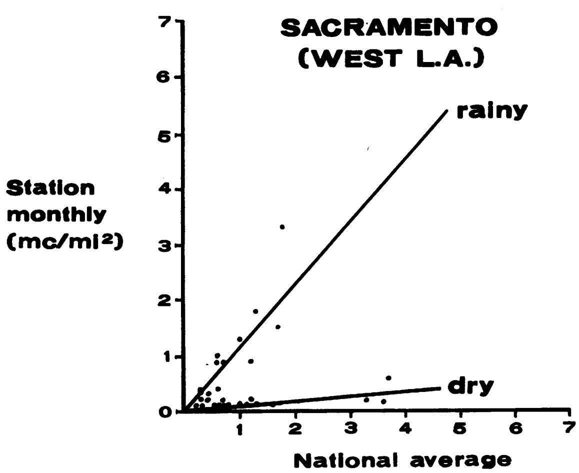
Strontium 90 Levels in Barn Feed

While the cattle are in the barn during the winter months they are using feed which was probably obtained during the previous harvest season. Thus, the strontium 90 levels in milk when a majority of the cattle were in the barn should reflect the pasture levels during the previous harvest season. This would be true only if the cows in the milkshed, as a whole, incorporated amounts of strontium 90 into their milk in direct proportion to the levels in the feed they were using. To establish this assertion, an observed ratio is defined as the ratio of strontium 90 in the milk to the strontium 90 in the feed, that is:

$$\text{Observed ratio}=\frac{\mu\mu\text{c/kg. of Sr}^{90}\text{ in milk}}{\mu\mu\text{c/kg. of Sr}^{90}\text{ in feed}} \qquad [4]$$

Figure 2. Comparison of monthly deposition for each milkshed and national average deposition (collection station in parentheses)





The relative constancy of this ratio can be demonstrated by considering data collected by the Public Health Service in Brainerd, Minn. (*5*), and St. Louis, Mo. (*6*). In these studies measurements were made for the concentrations of strontium 90 in both feed and milk for a number of cows on farms located within the milksheds. The data in table 1 show the computed average observed ratios and their associated 95 percent confidence limits.

The variability in this ratio is very small, the mean observed ratios in both milksheds lying approximately between 0.03 and 0.05. It is interesting to note that a ratio calculated using

Table 1. Mean observed ratios of strontium 90 in milk to strontium 90 in feed and their associated variation

Station	Mean observed ratio	Confidence limits (95 percent)
St. Louis	.046	.035–.057
Brainerd (overall)	.039	.035–.043
Brainerd farm 2L	.038	.028–.048
Brainerd farm 10L	.035	.025–.045
Brainerd farm 12H	.040	.031–.049
Brainerd farm 14H	.044	.037–.051

micromicrocuries of strontium 90 per gram of calcium (strontium units) exhibited more variability in the estimates. However, the basic result is that

$$\text{Level in feed} = k \text{ (level in milk)}$$

so that the levels in the barn feed should be proportional to the previous harvest season's milk values. As an empirical verification of this last statement, average milk levels during the barn-fed periods in 1958, 1959, and 1960 were compared with average milk levels during the previous harvest season in each of the five stations. The results are shown in figure 3. The level in the winter seems to be somewhere between 0.9 and 1.1 times the level during the previous harvest season (June–September). Errors are present because of the impossibility of stating which part of the harvest used in barn feed comes from which part of the harvest season. Despite the errors introduced, an approximately linear relationship

$$B(t) = k\overline{M}_{hs} \quad [5]$$

can be used for the barn level at time t where $\overline{M}_{hs}$ is the average milk level over the previous harvest season.

Strontium 90 Levels in Milk

The average levels in milk will depend upon the relative numbers of cattle on pasture and in the barn since the feed levels in the two locations will usually be quite different. The proportion of cows on pasture, $p(t)$, for the five original milk stations has been taken from Harris and co-workers and is given in table 2. The average milk level at time t, $M(t)$, will then be related to the average of the barn and pasture feed levels weighted by the proportion of cows in the barn and pasture respectively during that time.

$$M(t) = a_1 p(t) L(t) + a_2[1 - p(t)] B(t) \quad [6]$$

Combining equations 3, 5, and 6 yields the final model

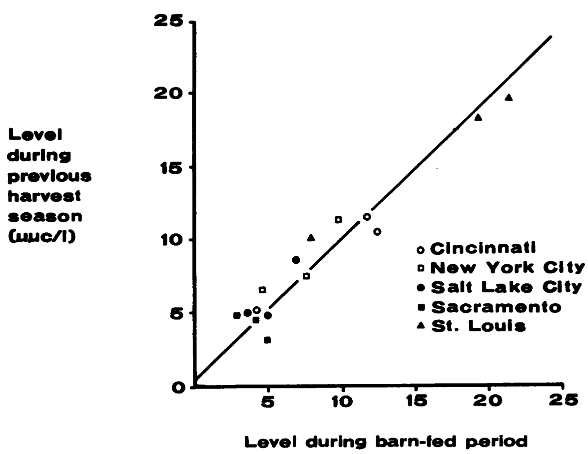
$$M(t) = \alpha_1 p(t) S(t) + \alpha_2 p(t) e^{-\lambda t} \int e^{\lambda t} D(t)\, dt + \alpha_3 [1 - p(t)] \overline{M}_{\mathrm{hs}} + \alpha_4 p(t) e^{-\lambda t} \quad [7]$$

Some estimate can be made of the half-residence time in advance because of the sporadic nature of the milk fluctuations. That is, half-residence times of more than a year produced predicted levels which did not correspond well with observed values. The sharp plunges which follow peak milk levels without preliminary drops in the deposition rates also indicate that strontinum 90 leaves the pasture fairly rapidly. Studies conducted by the Atomic Energy Commission (*1*) have also indicated that milk levels are greatly influenced by short-term deposition rates. Therefore, trial values for $T_{1/2} = 0.603/\lambda$ ranging from ¼ month to 3 months were used in the final formulation.

Computations

For values of $T_{1/2}$ (half-residence time) on the order of 1 month, equation 7 can be written appproximately

Figure 3. Comparison of levels of strontium 90 in milk during the barn-fed period and levels during the previous harvest season



$$M_n=\alpha_1 P_n S_n+\alpha_2 P_n e^{-\lambda n}\sum_{k=1}^{n} D_k e^{\frac{\lambda(2k-1)}{2}} +\alpha_3 \overline{M}_{hs}(1-P_n) \quad [8]$$

where

M_n =milk level in the n^{th} month expressed in micromicrocuries per liter

P_n=proportion of cows on pasture in the n^{th} month

S_n =cumulative level of strontium 90 in the soil in the n^{th} month expressed in millicuries per square mile

λ =pasture depletion constant, the reciprocal of the half-residence time

D_k =deposition during the k^{th} month expressed in millicuries per square mile per month

$\overline{M}_{hs}$ =average milk level over the closest preceding harvest season expressed in micromicrocuries per liter

This equation has the form of a multiple regression

$$M_n=\alpha_1 X_{1n}+\alpha_2 X_{2n}+\alpha_3 X_{3n} \quad [9$$

with independent and dependent variables as indicated. The basic time period will be taken as 1 month with $n = 1, 2, 3, \ldots, 63$ months as the total time covered extending from July 1957 to September 1962. The values of the variables X_{1n}, X_{2n}, and X_{3n} were computed and the parameters α_1, α_2, and α_3 were determined by the least squares analysis using a computer. Computation times involved are on the order of ½ hour per station. Values for deposition and soil levels, when available, were extracted from the Health and Safety Laboratory report made by the Atomic Energy Commission.

Unfortunately, there were a few missing monthly deposition values in each of the stations. However, if the particular monthly station values are compared with the overall average deposition rates by month, a constant relation is observed. These results are plotted in figure 2 for the five stations. As an example, suppose that the value for New York is missing for June, but that the corresponding overall average is 4 millicuries per square mile. Then, the estimated station value is 6 millicuries per square mile. A conservative estimate for the spring of 1962 is that the deposition rates will be the same as during the spring of 1959. The maximum deposition rates which would occur under the worst possible assumptions are assumed to be about twice the expected rates. The average soil levels are based on those estimated by Knapp.

The estimated milk levels do not differ greatly with different values for $T_{1/2}$, although they get progressively worse with the longer half-times. The optimum half-time for each station was

Table 2. Average proportion of cows in pasture during each month

Station	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
Cincinnati	0. 20	0. 20	0. 22	0. 36	1. 00	1. 00	1. 00	1. 00	1. 00	0. 98	0. 78	0. 37
New York City	. 00	. 00	. 00	. 00	. 62	1. 00	1. 00	1. 00	1. 00	. 87	. 01	. 01
Sacramento	. 08	. 10	. 47	. 81	. 84	. 84	. 84	. 84	. 83	. 72	. 32	. 12
St. Louis	. 01	. 02	. 13	. 94	. 97	. 97	. 97	. 95	. 93	. 45	. 09	. 03
Salt Lake City	. 00	. 00	. 00	. 00	. 61	. 71	. 75	. 75	. 78	. 45	. 02	. 00

Table 3. Model parameters yielding best fit for each station

Station	$T_{1/2}$	α_1	α_2	α_3	Percent variation accounted for
Cincinnati	0. 75	0. 10	3. 7	1. 1	66
New York City	. 75	. 10	2. 7	. 9	47
Sacramento	2. 00	. 02	3. 2	1. 0	22
St. Louis	. 25	. 21	13. 7	1. 0	60
Salt Lake City	2. 00	. 04	1. 1	. 8	46

chosen by maximizing the percentage variation for which the model could account. The percentage variation accounted for is defined by

$$V=1-\frac{\sum_{n=1}^{r}(M_n-\alpha_1X_{1n}-\alpha_2X_{2n}-\alpha_3X_{3n})^2}{\sum_{n=1}^{r}(M_n-\bar{M})^2} \quad [10]$$

where M_n is the observed milk level in the n^{th} month and $\bar{M}$ is the average observed milk level for r months. The parameter values for the optimal model are shown in table 3. Note that the α's differ from region to region, reflecting the differing relative emphasis on the soil level and deposition rate factors.

The percentage variations accounted for are smaller in stations where there is less variability in the milk levels. From inspecting the variability of the milk levels during the barn-fed period when one would expect them to be essentially constant, we can see that one could hardly hope to account for more than 50 percent of the variation in some of the stations. The results from the best model for each station are shown graphically in figure 4, where solid and dotted lines represent observed and expected (calculated) values respectively. The average predicted values are based on the assumption that the deposition rates will be about what they were in 1959.

Discussion

It is interesting to note that the estimates for the parameters in areas of similar environmental conditions are close to one another. This is particularly reflected in the pasture half-residence time $T_{1/2}$. The areas of low rainfall, Sacramento and Salt Lake City, have a longer residence time than New York City and Cincinati, probably because of a much slower washout. Also, the α's are very similar in the similar areas.

Since the deposition rates contain a large element of uncertainty, some of the minor peaks are missed occasionally. In periods of low deposition, one would not expect to predict fluctuations of minor magnitude. The predicted line remains essentially constant during the months of barn feeding so that the deviations of the observed values during this period measure the sampling variation. The sharp peak which sometimes appears in October, November, or December, and is not fitted well by the theoretical line might be a result of using feed from an early spring harvest. This factor might well account for the fall peak sometimes observed and should be investigated. It seems reasonable on the basis of figure 4, however, to assume that the model is capable of predicting large fluctuations if the correct deposition input is known.

Obviously, we have considered quite a range of deposition inputs. The future milk values will probably be a little less than the predicted line shown as "expected." Estimates in May 1962 place the expected deposition from 0 to 20 percent less than in 1959. The milk levels of the five stations were averaged to arrive at national average observed and expected milk levels. Figure 5 shows the observed milk levels along with the levels calculated by Knapp and the model presented here. He predicts an average of 15 micromicrocuries of strontium 90 per liter for 1962, whereas our model predicts an average of 11 micromicrocuries of strontium 90 per liter. However, if the fallout deposition should be twice what is expected at every station, our model predicts an average value of 15 micromicrocuries per liter in the U.S. milk supply for 1962. Then, on the basis of fallout deposition predictions and transport assumptions, it becomes possible to predict milk levels for any given future time period in segments of the country where the required measurements are available. This leads to a much better estimate of the average yearly bone marrow dose in any region.

Summary

A mathematical model has been developed for the purpose of predicting strontium 90 concentrations in milk. It is the successor of several other models designed to predict the activity of radionuclides in milk. A relatively large yield of strontium 90 is deposited on the surface of the earth as a result of nuclear weapons tests. This radionuclide then finds its way through the food chain to human beings. Milk is one of the most important contributors to the total dietary intake.

Figure 4. Observed and expected concentrations of strontium 90 in samples of milk from specified milksheds, July 1957–September 1962

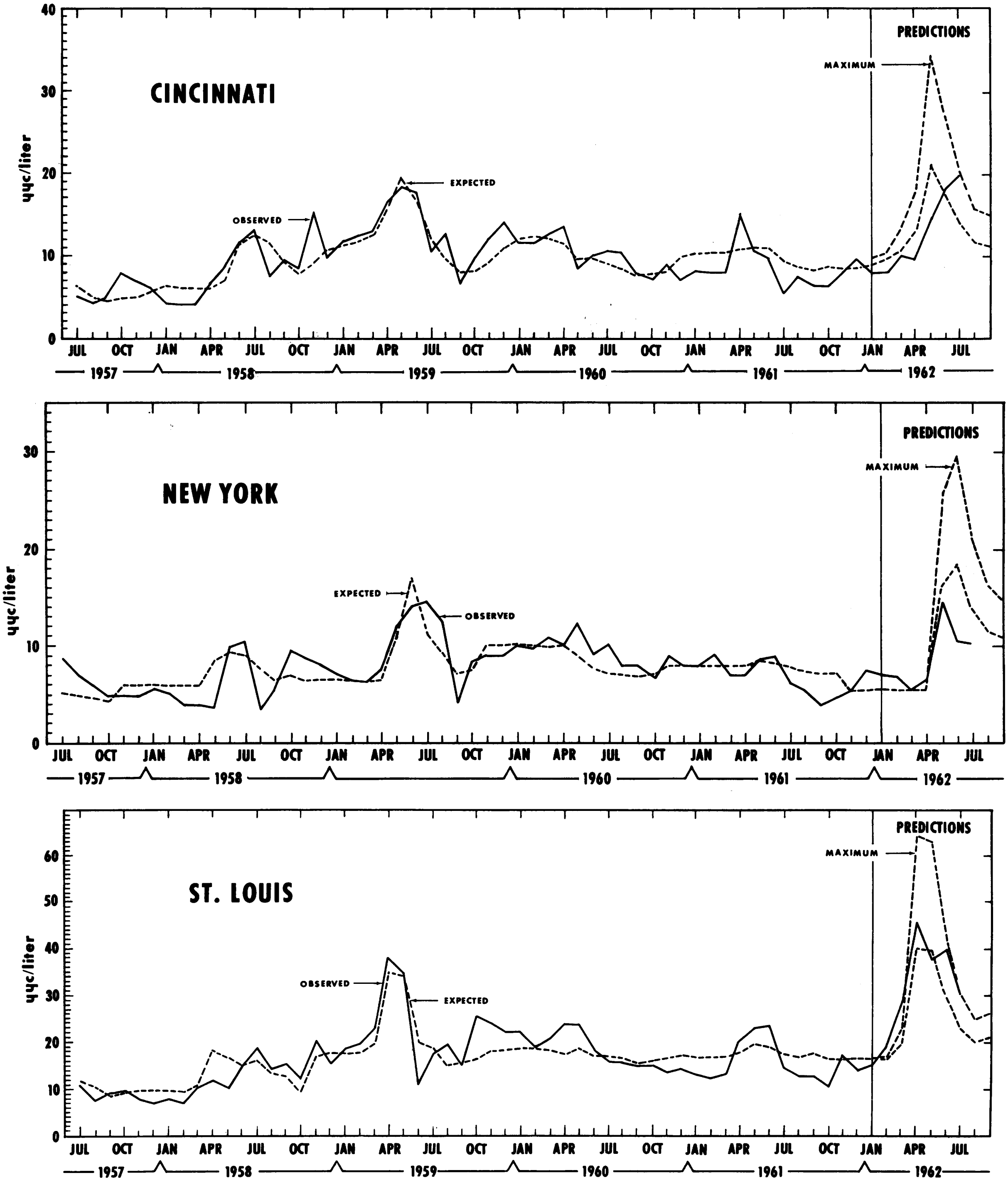
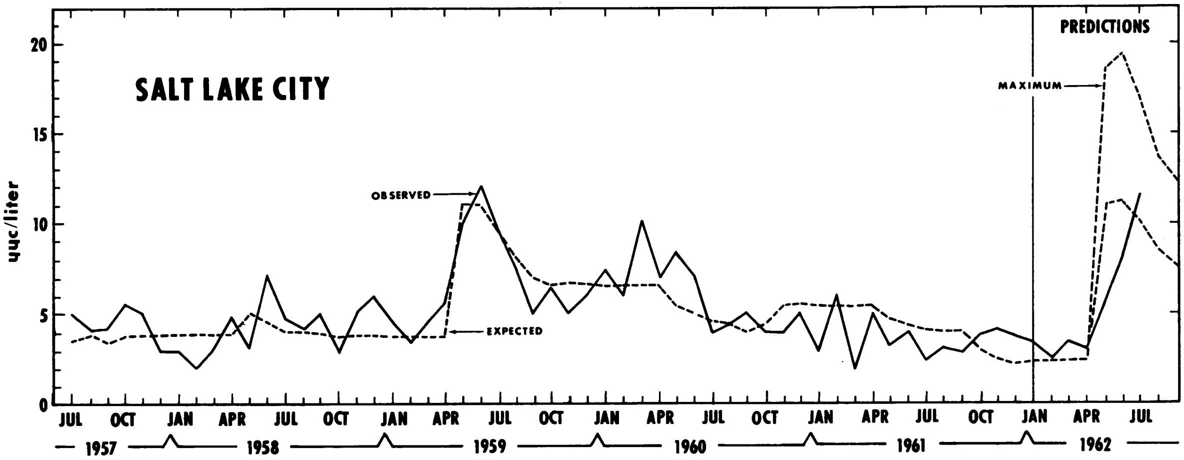


Figure 4. Observed and expected concentrations of strontium 90 in samples of milk from specified milksheds, July 1957–September 1962—Continued



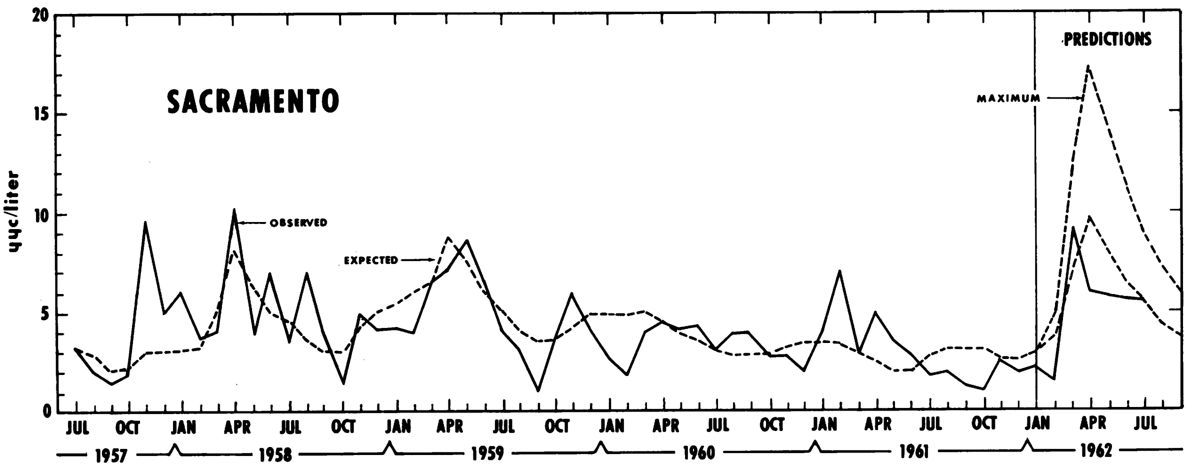
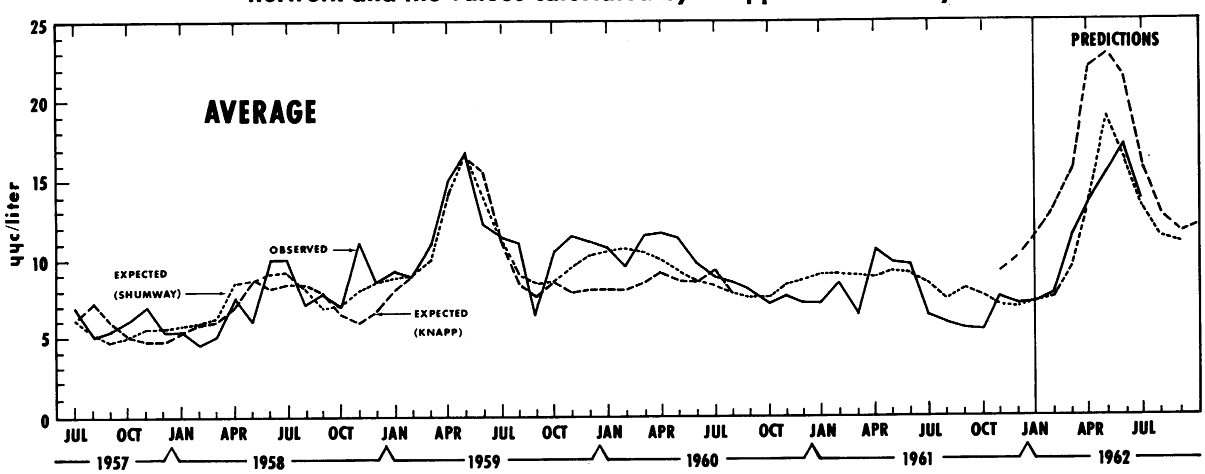


Figure 5. U.S. average observed concentrations of strontium 90 in milk sampled in the original network and the values calculated by Knapp and Shumway



SOURCE: Milk levels calculated by Dr. Harold A. Knapp from November 1961–October 1962 were obtained through personal communication.

The strontium 90 is deposited on the pasture. Some is retained on the foliage and some is taken up by the soil. Precipitation causes some of the strontium 90 to be washed off and additional amounts are depleted by grazing. The cattle, however, may be in the barn during a portion of the year consuming feed obtained during the previous harvest season. The level of strontium 90 in milk, therefore, reflects the concentration of strontium 90 in the pasture feed and in the barn feed. Based on these assumptions, the model includes the following factors: deposition rate, rate of absorption from the soil, the residence time of strontium 90 on the plant, and the proportion of cows on pasture feed and of cows on barn feed. The derived equation allows values for specific areas to be considered for a particular time period.

The model is fitted to approximately 5 years of observed milk measurements from five metropolitan areas comprising the original raw milk sampling network. Deviations of the observed values from those given by the model are interpreted as fluctuations arising from experimental error or secondary perturbations or both. Calculated values were generally found to be reasonable. A higher degree of reliability in predictions of deposition may result in more accurate predictions of strontium 90 concentrations in milk. Modifications of the transport assumptions may be necessary.

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Biological Serial Record Center

The Biological Serial Record Center, a depository for bibliographic information on the world's biological journals, is being established in Washington, D.C., by the American Institute of Biological Sciences.

When its files are ready, the new center will enable biological and other scientists to find in one place titles usually recorded either in major libraries or in special libraries, collections, or prepared lists. It will provide scientists with complete information on journals pertinent to their work. This information will include addresses, editor's name, nature of contents, price, language and frequency of publication, coverage by abstracting agencies, and format.

Identifying and recording of approximately 20,000 serial publications began in October 1962. In addition to making library searches, the center's staff is requesting serial titles from publishers and scientific organizations and asking biologists traveling to foreign countries to obtain specific information on serial literature.

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The work of the center, which is an activity of the Biological Sciences Communication Project, is supported by a 3-year grant from the Division of General Medical Sciences, National Institutes of Health, Public Health Service.