Localization of Fallout in United States From May 1966 Chinese Nuclear Test

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BEFORE the May 1966 Chinese nuclear detonation, residual environmental fallout in the United States was principally due to the long-lived radionuclides resulting from the U.S. atmospheric nuclear tests of 1961–62 and from the two previous Chinese tests. The environmental levels of activity could be predicted fairly accurately from models developed from previous studies on the "effective" decay of radionuclides and the transfer of nuclides to foodstuffs (*1*).

One of the primary devices used to estimate levels of radioactivity in man's environment is the amount of radionuclides in milk (*2*). Approximately one-half of the radiostrontium and most of the radioiodine in a young child's diet comes from milk (*3*). As milk is the most direct vector for the transport of fresh fission products into man's food chain, the Public Health Service maintains an extensive surveillance program (Pasteurized Milk Network) for the determination of radionuclides in milk (*4*).

One of the first indications that the Southeastern Radiological Health Laboratory had of the presence in the U.S. environment of fresh fission products was a measurement by the Arkansas State Board of Health of 230 picocuries (pCi) per liter of iodine 131 in a sample of rainwater collected May 16 in Little Rock, Ark. Three days later, a peak level of 570 pCi per liter of iodine 131 was detected in cows' milk from this same general area. This level was much less, however, than the Protective Action Guide concentration of 84,000 pCi per liter established by the Federal Radiation Council for localized contamination (*5*).

Levels decreased rapidly, and a disappearance half-time of 4 days for iodine 131 was observed. Barium 140 and strontium 89 concentrations followed much the same pattern, having lower values and slightly longer half-times. (Disappearance half-time refers to the time required for the concentrations of a radionuclide to decrease by one-half following the maximum value.)

Data from the sampling program of the Pasteurized Milk Network (PMN) indicate the pattern of fallout (*6*). They show that the highest concentrations of iodine 131 in milk were in the Mississippi Valley region. Apparently the radioactivity was transferred from the atmosphere to vegetation through precipitation

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Dr. Raymond T. Moore, deputy director, National Center for Radiological Health (formerly program director for radiological health, Public Health Service Region VII), provided information and aided in coordinating the sampling program in Lonoke County, Ark.

Figure 1. Average iodine 131 concentrations (picocuries per liter) in samples from stations of the Pasteurized Milk Network in the conterminous United States, May 1966

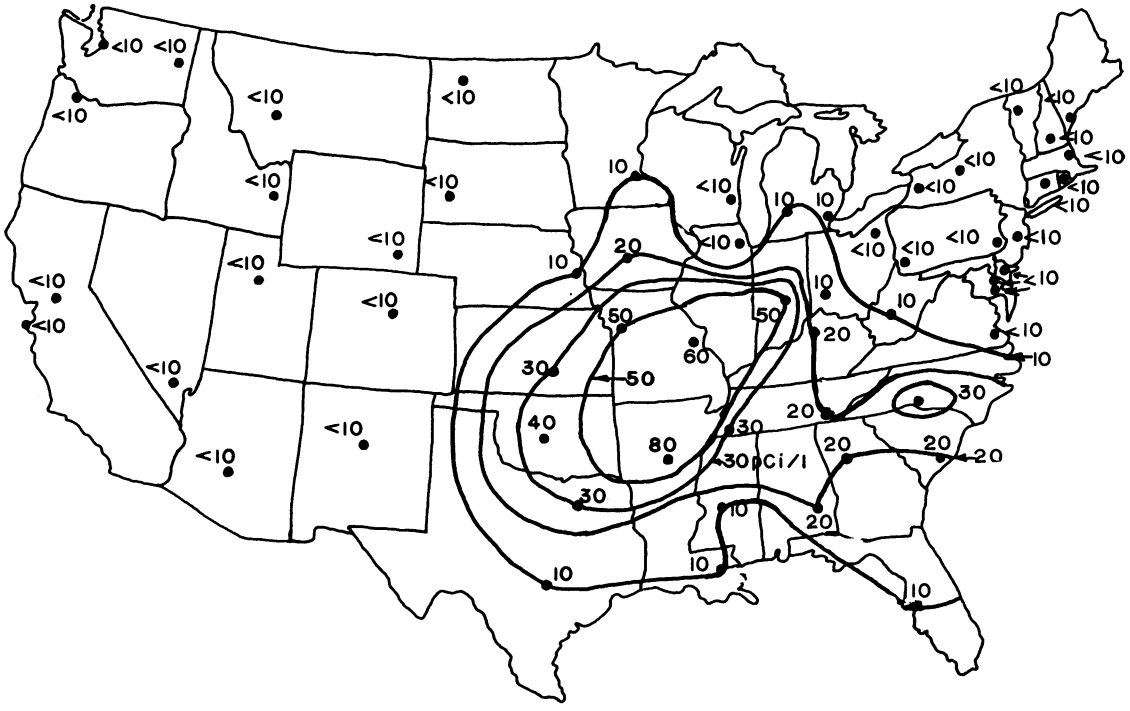
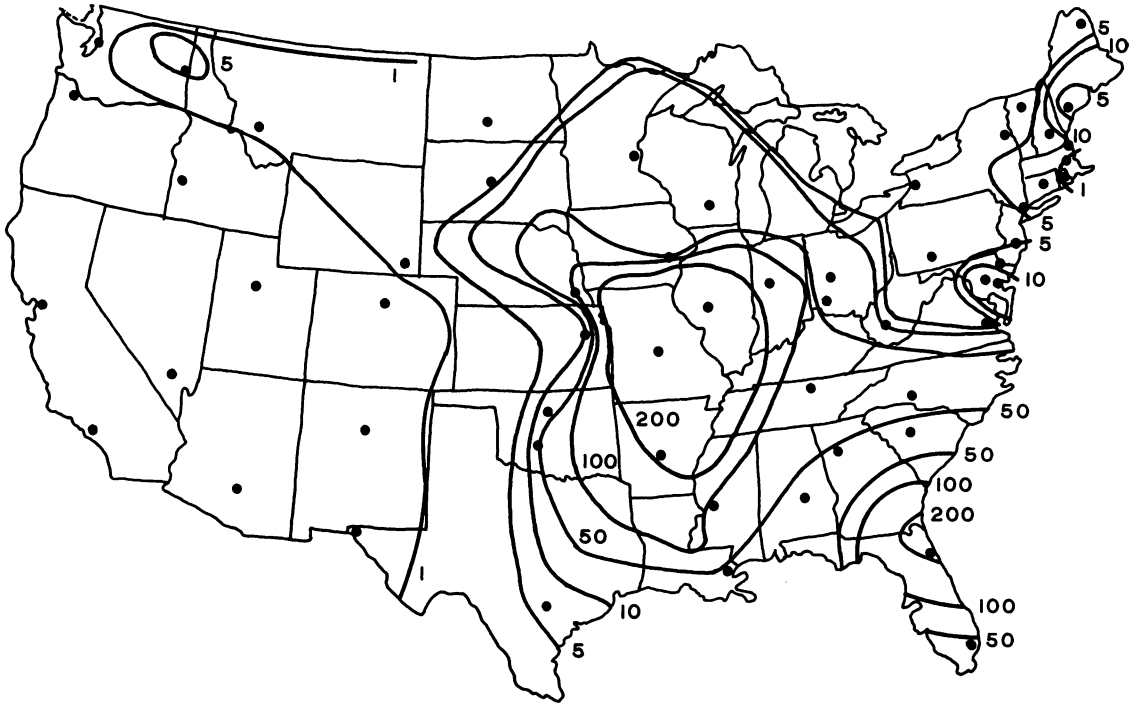


Figure 2. Deposition of radioactive material (nanocuries per square meter) in precipitation samples of the Radiation Surveillance Network, May 1966



occurring May 15–21 in those areas, for very little rise in radioactivity was noted in air samples in contrast to the substantial increases in rainwater samples.

Sampling and Analysis

Following the initial indication of high concentrations of fresh fission products in the Arkansas area, an expanded surveillance program was established by the Southeastern Radiological Health Laboratory and the Arkansas State Board of Health to supplement data of the Pasteurized Milk Network, which only provided biweekly sampling. In addition to the PMN samples, samples of fresh whole milk and ion-exchange cartridges through which fresh milk had been passed to collect iodine (*7*) were obtained from a dairy farm in Lonoke County, Ark., approximately 20 miles from Little Rock. These samples were collected by the Arkansas State Board of Health and airmailed to the Southeastern Radiological Health Laboratory for analysis. After the concentrations of iodine 131 dropped to less than 10 pCi per liter, the sampling frequency was reduced and collection of the ion-exchange cartridge samples was discontinued.

Gamma spectral analysis was performed on 3.5 liter samples of milk to determine the concentration of iodine 131, cesium 137, barium 140, and stable potassium (*8*). The ion-exchange cartridges were gamma counted in a well crystal for iodine 131 activity. Strontium 89, strontium 90, barium 140, and stable calcium were separated radiochemically by an ion-exchange procedure (*9*), and a low background beta counter was used to determine the amount of beta activity in the milk samples.

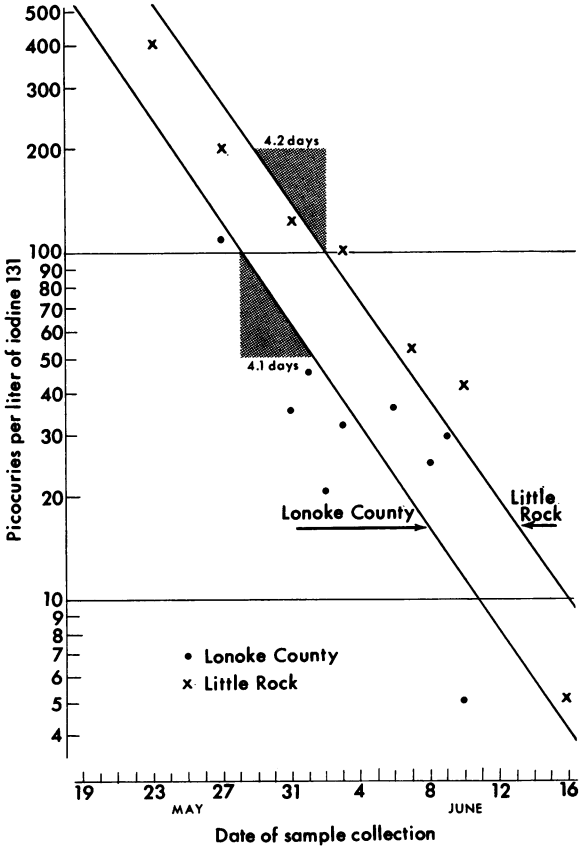
As the possibility of the expansion of environmental radioactivity southeast from Little Rock became apparent, additional samples were collected from northern and central Louisiana by the Louisiana State Board of Health. The purpose of this collection was to monitor the area between Little Rock and New Orleans, which was not covered by a PMN sampling station.

Results and Discussion

The average concentrations of iodine 131 in pasteurized milk during May 1966, based on samples collected weekly or twice weekly from Pasteurized Milk Network stations, are shown in figure 1. Previous 1966 levels of iodine 131 had been less than 10 pCi per liter at all PMN sampling locations. The highest average monthly concentration, 80 pCi per liter of iodine 131, was reported at Little Rock for May 1966. "Isoconcentration" lines have been drawn on figure 1 to show the approximate shape and size of the areas in which various levels of iodine 131 were found in milk.

Analysis of air data for May from the Radiation Surveillance Network (RSN) indicates that fresh radioactive material, mostly at high altitudes, began crossing the United States on about May 13, 1966, and reached the east coast by May 15 (*10*). Until May 20, however, concentrations of airborne activity of more than 1.0

Figure 3. Disappearance half-time for iodine 131 in milk samples collected in May and June 1966 from a Lonoke County (Ark.) dairy farm and from the Little Rock (Ark.) station of the Pasteurized Milk Network



Results of analysis of 3.5 liter samples of milk and of ion-exchange

Item and data collected	Gamma analysis				Radiochemical analysis			
	Picocuries per liter			Potassium (grams per liter)	Picocuries per liter			Calcium (grams per liter)
	Iodine 131 [1]	Barium 140 [1]	Cesium 137		Strontium 90	Strontium 89 [1]	Barium 140 [1]	
May 19 [2]								
Sample	570	70	55	1. 38	39	55	50	1. 07
May 21								
Sample	400	50	65	1. 62	26	60	40	. 98
Cartridge	350							
May 27								
Sample	110	80	60	1. 01	40	75	80	1. 27
Cartridge	110							
May 31								
Sample	40	40	55	1. 31	32	60	30	1. 28
Cartridge	30							
June 1								
Sample	40	30	40	1. 45	28	50	30	1. 26
Cartridge	50							
June 2								
Sample	20	30	40	1. 65	33	55	30	1. 22
Cartridge	20							
June 3								
Sample A	20	20	40	1. 74	32	30	20	1. 04
Sample B	20	20	40	1. 70	36	20	20	1. 06
Sample C [3]	50	30	50	1. 72	26	30	30	. 94
Cartridge A	50							
Cartridge B	50							
Cartridge C	40							
June 6								
Sample	40	20	45	1. 75	27	30	20	. 96
Cartridge	30							

pCi per cubic meter were widely scattered and probably resulted from local atmospheric disturbances which brought material down from high altitudes. The largest part of the significant deposition of radioactive material by precipitation, however, occurred before May 20. Each "hot" rain was preceded or accompanied by a slight rise in airborne activity.

RSN data show that on May 20 a large mass of air, bearing fission products, arrived over the western third of the country and that, for the rest of May, atmospheric levels, particularly in the southern half of the country, were generally elevated. Levels of airborne activity, however, never exceeded 15 pCi per cubic meter, which are much lower than levels resulting from some earlier tests. National Air Sampling Network data also show an increase of activity and the presence of fresh fission products after May 20 (personal communication from Byron Branson, Radiological Health Research Activities, National Center for Radiological Health, Public Health Service, Cincinnati, Ohio).

RSN precipitation samples, especially those collected May 16–18 in the central Mississippi Valley, exhibited a significant amount of radioactivity. The pattern of deposition corresponds to data from the U.S. Weather Bureau (*11*) showing that the heaviest rainfall during this time occurred in the central Mississippi Valley. Figure 2 shows isodeposition curves derived from RSN precipitation data. The values used to plot these curves are the sums of deposition (nanocuries per square meter) at each station—the depositions resulting from precipitation showing more than 200 pCi per

cartridges collected in Lonoke County, Ark., May 19–August 4, 1966

Item and data collected	Gamma analysis				Radiochemical analysis			
	Picocuries per liter			Potassium (grams per liter)	Picocuries per liter			Calcium (grams per liter)
	Iodine 131 [1]	Barium 140 [1]	Cesium 137		Strontium 90	Strontium 89 [1]	Barium 140 [1]	
June 7								
Sample	10	10	40	1.55	32	10	10	1.08
Cartridge	20							
June 8								
Sample	20	30	45	1.26	58	30	30	1.46
Cartridge	30							
June 9								
Sample	30	<10	50	1.74	30	15	<10	1.13
June 10								
Sample	<10	10	60	1.73	16	10	<10	1.04
July 20								
Sample A	<10	<10	45	1.62	23	20	<10	1.12
Sample B	10	<10	30	1.52	20	<5	<10	1.18
Sample C	10	<10	45	1.44	19	10	<10	1.00
July 21								
Sample	20	15	35	1.62	21	10	<10	1.08
July 28								
Sample	<10	<10	30	1.71	22	<5	<10	1.12
August 4								
Sample	<10	<10	20	1.64	23	<5	<10	1.06

[1] Corrected for decay to date of collection.
[2] No cartridge.
[3] Milk in sample C same as in cartridges A, B, and C. Samples A and B were not mixed with sample C before analysis.

liter of gross beta activity. No RSN precipitation data are available from Little Rock during this period. Figure 2 and the previously mentioned Arkansas Board of Health observation for May 16, however, indicate that most of the State was in the deposition area of more than 200 nanocuries per square meter.

A comparison of figures 1 and 2 shows the rough correlation of iodine 131 activity in milk with radioactivity deposited by precipitation, especially in the Mississippi Valley. Other areas where relatively high values of radioactivity in depositions were noted, such as Jacksonville, Fla., could not be correlated, as there is no PMN station in that area.

The higher concentrations of radioactivity detected in milk, although there was almost no rise in radioactivity in the air, are attributed to the fact that precipitation in general contributed 90 percent or more of the total fallout reaching the earth (*12*). This greater radioactivity in connection with precipitation was also noted during the Chinese nuclear test of October 1964 (*13*).

The absence of fallout deposition in the western United States may be explained by the relative aridity of that area (*1*) and by the little rainfall on the west coast at the time the cloud of radioactivity was passing over (*13*).

Analysis of samples of rainwater collected in Lonoke County on May 16, 1966, revealed the following concentrations of radioactive nuclides (picocuries per liter): cerium 144—200, iodine 131—230, ruthenium 106—120, cesium 137—less than 5, zirconium 95—20, zinc 65 and potassium 40—each less than 10, and barium 140—240. The iodine 131, the zirconium 95, and the barium 140 values were corrected for decay to date of collection.

Results of analysis of the milk samples collected in Lonoke County are shown in the table. Studies have shown that it takes approximately 3 to 4 days for milk to exhibit maximum iodine 131 radioactivity following localized fallout (*14, 15*). This observation indicates that the value of 570 pCi of iodine 131 per liter of milk collected on May 19 (3 days after collection of the precipitation sample) was probably close to the peak activity in Lonoke County, as all samples collected following that date showed decreased radioactivity.

From the data in the table it should be noted that values obtained for iodine 131 by gamma analysis of the 3.5 liter sample, as compared with those obtained by gamma analysis of the ion-exchange resin cartridge, agreed within one standard deviation analytical error of their mean; results from gamma spectral analyses for barium 140, as compared with those for radiochemical analyses, were also within one standard deviation of their mean.

In figure 3, the values for Lonoke County for iodine 131 show a disappearance half-time of 4.1

Figure 4. Disappearance half-times for strontium 89 and for barium 140 in milk samples collected from a Lonoke County (Ark.) dairy farm, May and June 1966

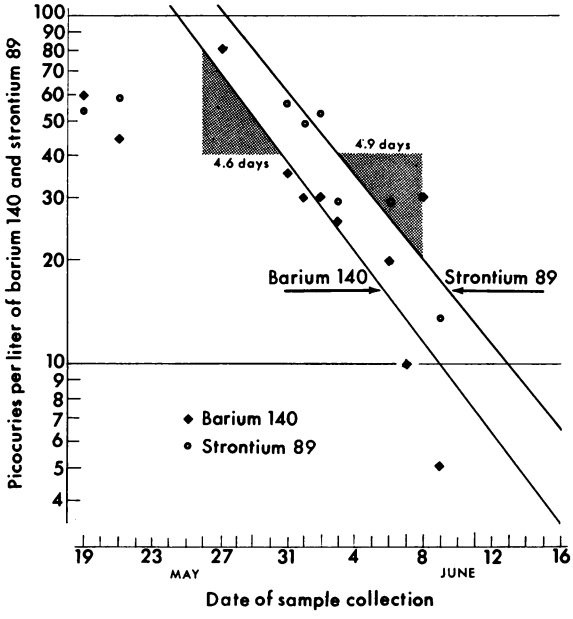
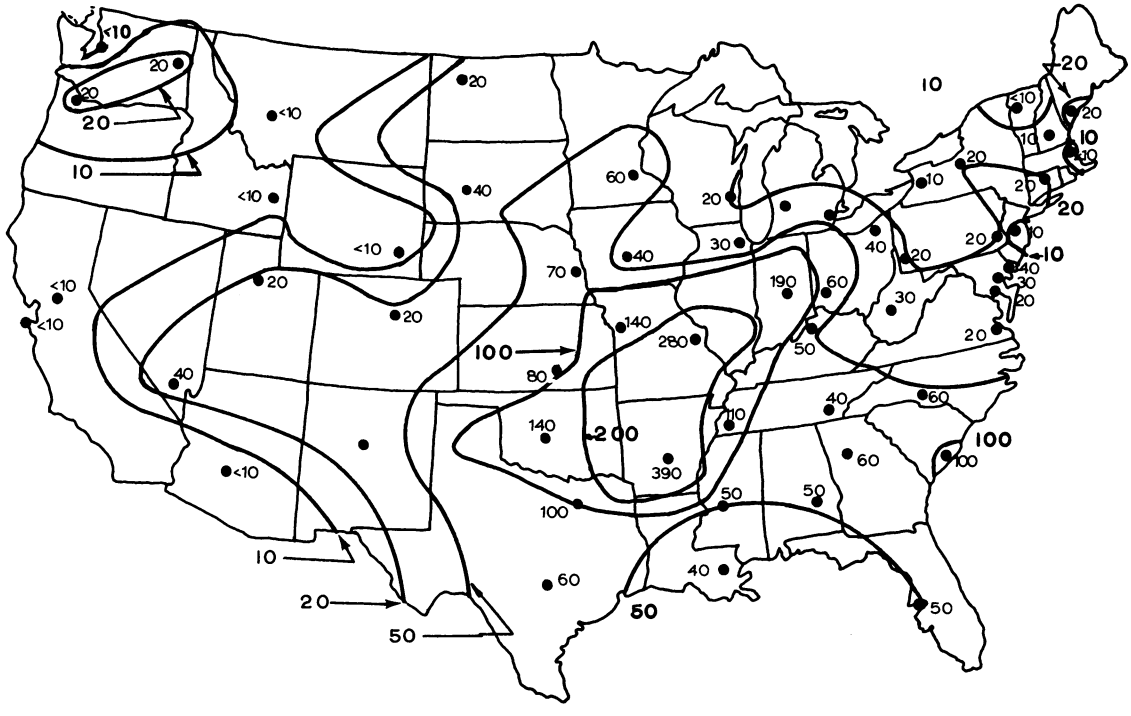


Figure 5. Peak values for iodine 131 (picocuries per liter) in samples from the Pasteurized Milk Network stations in the conterminous United States, May 23–June 7, 1966

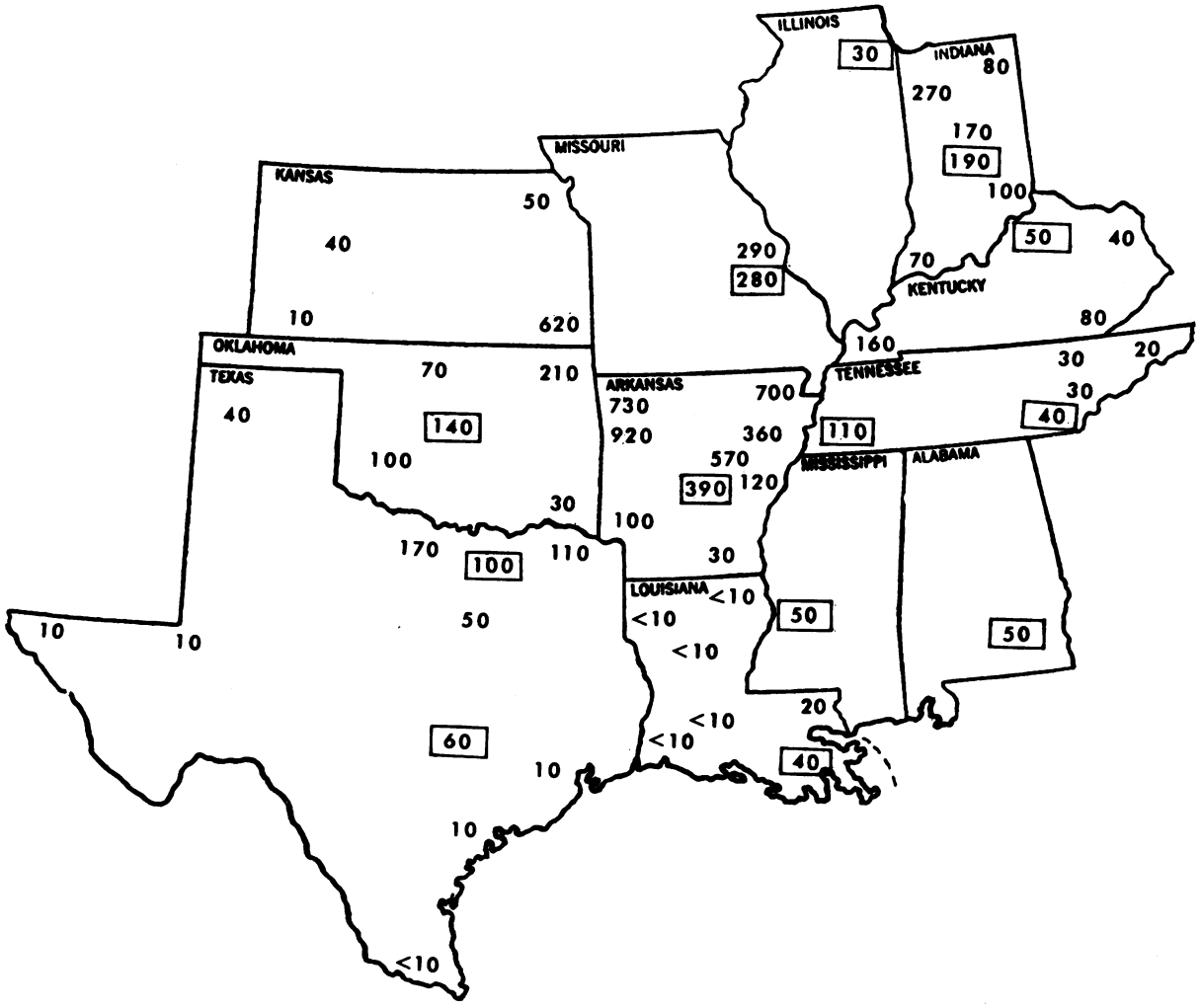


days, whereas data from the composite Pasteurized Milk Network sample from Little Rock indicate a half-time of 4.2 days. These values correspond to data collected following nuclear tests at the Nevada test site (*16*) and to data published by the Federal Radiation Council (*5*).

Data in figure 4 indicate disappearance half-times of 4.6 days for barium 140 and 4.9 days for strontium 89. In figures 3 and 4, the lines were fitted to the points by the method of least squares (*17*).

Peak values for iodine 131 at stations of the Pasteurized Milk Network are depicted in figure 5. Figure 6 shows the peak values registered at various locations in Arkansas, Oklahoma, Texas, Missouri, Illinois, Indiana, Kansas, Kentucky, Tennessee, Alabama, and Mississippi in May 1966, following the May 9 detonation. At Fort Smith, Ark., a peak of 920 pCi of iodine 131 per liter was detected—the highest value recorded in the United States. The peak recorded at the Lonoke County collection station was 570 pCi per liter. (The peak values for some of these States were supplied in personal communications to the authors by radiological health officials of the Public Health Service and of the State health departments. These sources were Dr. Raymond

Figure 6. Peak values for iodine 131 (picocuries per liter) in milk samples collected in Arkansas and the surrounding States, May 1966



NOTE: PMN values are boxed to differentiate them from the data obtained from the States represented. All of these samples were collected in May 1966 following the Chinese nuclear detonation.

T. Moore, deputy chief, National Center for Radiological Health—data on Texas; Frank Wilson, Arkansas State Board of Health; Robert L. Craig, Oklahoma State Department of Health; Dr. Gerald Jacobson, Public Health Service, Region VI—data on Missouri; Dr. John E. Frank, Public Health Service, Region V, and Perry E. Miller, Indiana State Board of Health—data on Indiana; Jerome A. Halperin, Kansas State Department of Health; Richard M. Fry, Kentucky State Department of Health; and Bill Graham, Tennessee Department of Public Health.)

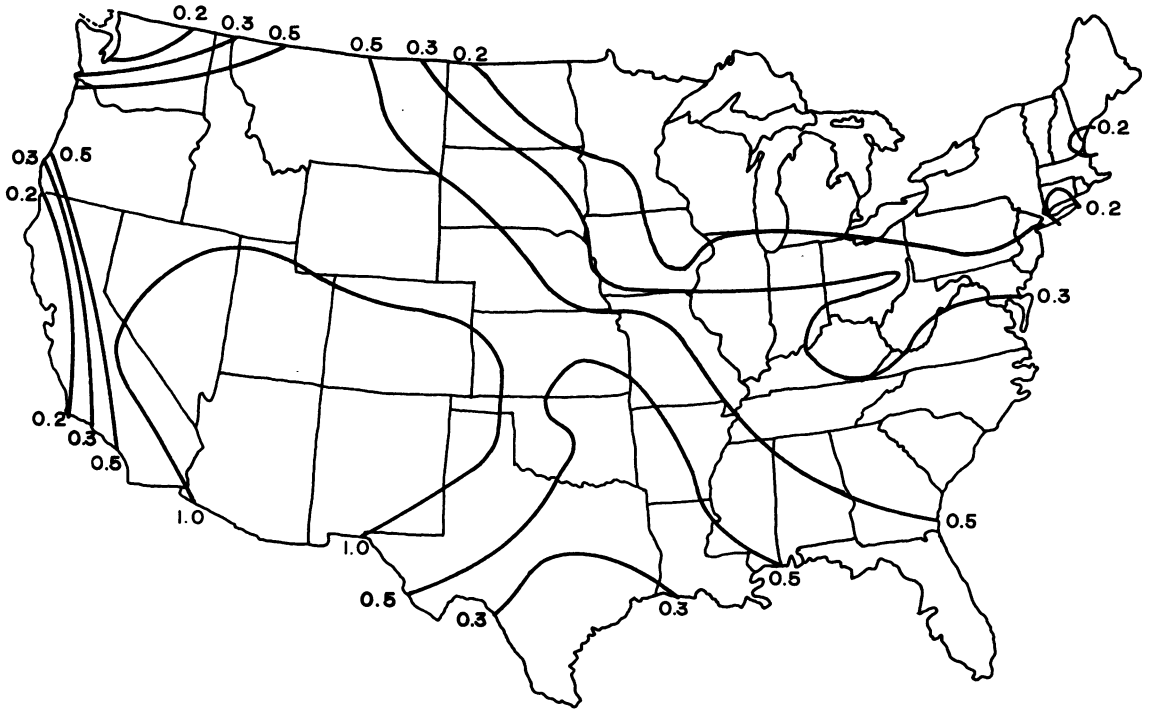
Isoconcentration lines have also been plotted on figure 5. Every high milk reading corresponds to a high point in precipitation deposition (fig. 2) except in several southwestern States, where slightly elevated values correspond to the highest average concentrations of airborne radioactivity in the country (according to a personal communication from William P. Kirk, chief, Radiation Surveillance Network, National Center for Radiological Health, Public Health Service). The concentrations of radioactivity in the air remained relatively low during May 1966; the highest concentrations of fresh fission products were observed in the southwest (fig. 7).

It is evident from the data available that Arkansas was the focus for the fallout in the United States from the third Chinese nuclear detonation. States surrounding Arkansas were also affected, but the radionuclide levels in milk in these States measured considerably less than those in milk from Arkansas. Concentrations of nuclides in special samples collected in Louisiana indicated that the higher levels of radioactivity did not extend southward from Arkansas (fig. 6). Precipitation on and around May 16 was responsible for depositing a major portion of the radionuclides (*11*).

Summary

The Chinese nuclear detonation of May 9, 1966, provided fresh fission products, which were deposited in the United States during the week of May 15–21. An examination of milk data from both the Pasteurized Milk Network of the Public Health Service and from the pasteurized milk networks of the individual States

Figure 7. Average concentrations of gross beta radioactivity (picocuries per cubic meter) in air samples from the Radiation Surveillance Network, May 1966



revealed that the heaviest concentration of fallout was in the State of Arkansas; lesser amounts were measured in a number of surrounding States. This localization in the Arkansas area is believed to have been caused by the heavy rainfall in the Mississippi Valley during the time the cloud of radioactivity was passing over the United States.

An expanded milk sampling program was established in Arkansas, which provided for the collection of daily samples from a Lonoke County dairy farm near Little Rock. A maximum value of 570 picocuries of iodine 131 per liter was detected at the Lonoke collection station, while the highest value recorded in the United States was 920 picocuries per liter at Fort Smith, Ark.

Disappearance half-times for the fresh fission products in milk were determined to be 4.1 days for iodine 131, 4.6 days for barium 140, and 4.9 days for strontium 89.

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To Develop Standard Radiological Methods

Development of standard radiological methods is proceeding according to plans of a new subcommittee of the Coordinating Committee on Laboratory Methods of the American Public Health Association. The Subcommittee on Radiological Methods for Biological and Environmental Samples consists of an advisory board, nine referee committees, and collaborating investigators in local, State, Federal, industrial, and institutional laboratories. The subcommittee will include in its activities the updating of radiological methods currently given in the 12th editions of "Standard Methods for the Examination of Water and Wastewater" and "Standard Methods for the Examination of Dairy Products."

Each referee committee consists of a chairman and two to four members. Seven referee committees are responsible for drafting and pretesting selected procedures for specific types of samples. These committees, listed on pages 332 and 333 of the February 1967 issue of the *American Journal of Public Health*, are designated A on air, B on water, C on milk, D on food, E on feed and soil, F on industrial wastes, and G on biological samples. Referee committee H is responsible for in vivo gamma spectrometry, and referee committee I is responsible for instructions on instrument counting and calibration.

It is the joint responsibility of the chairman of the subcommittee, the advisory board, and the chairmen of the referee committees, with the approval of the Coordinating Committee on Laboratory Methods, to limit the number of techniques and the duplication of effort, to secure a broad spectrum of professional competence, and to coordinate the development of radiological methods with other professional organizations. Coordination is obtained in part by having technical representation on similar task groups of other associations concerned with the development of radiological methods.

The referee committees plan to prepare drafts of suitable methods by July 1967 and subject these methods to ruggedness tests to assure the validity of the test procedures. As soon as each method is drafted, potential participants will be invited to conduct round robin tests of the methods with reference samples at three levels.

These studies will begin during July 1967. By January 1968 all methods should be drafted and have successfully passed ruggedness tests. By August 15, 1968, all collaborative testing, including statistical analyses, should be complete so that the standard methods developed by each committee may be submitted to the Coordinating Committee on Laboratory Methods for clearance by January 1969. The projected publication date for the radiological methods is October 1969.

A critical aspect in developing standard methods is the nationwide collaborative testing of reference samples by investigators who rigidly adhere to the detailed procedures of the referee committees. To obtain the desired proficiency, subsequent to the distribution of reference samples the plans include distributing standard solutions of radionuclides along with the draft of the procedures to be used. Investigators will be asked to prepare appropriate test samples for use in conducting a limited number of tests to calibrate their instruments and become familiar with the test procedures. Reports of these preliminary studies will be used to guide the referee committees in evaluating the results on the subsequent reference samples and assigning the degree of precision, sensitivity, and accuracy of the tests.

The subcommittee has a list of 85 investigators representing 85 laboratories that have agreed to participate in the testing of reference samples. Additional volunteers are urged to share in the responsibility of developing uniform methods; the results promise to be mutually beneficial to all those working in radiological health.

For further information write to Dr. Lloyd R. Setter, National Center for Radiological Health, Public Health Service, 1901 Chapman Ave., Rockville, Md. 20852.