PLUTONIUM FALLOUT IN UTAH

by E. P. Hardy, (HASL)
P. W. Krey, (HASL)
H. L Volchok, (HASL)

ABSTRACT

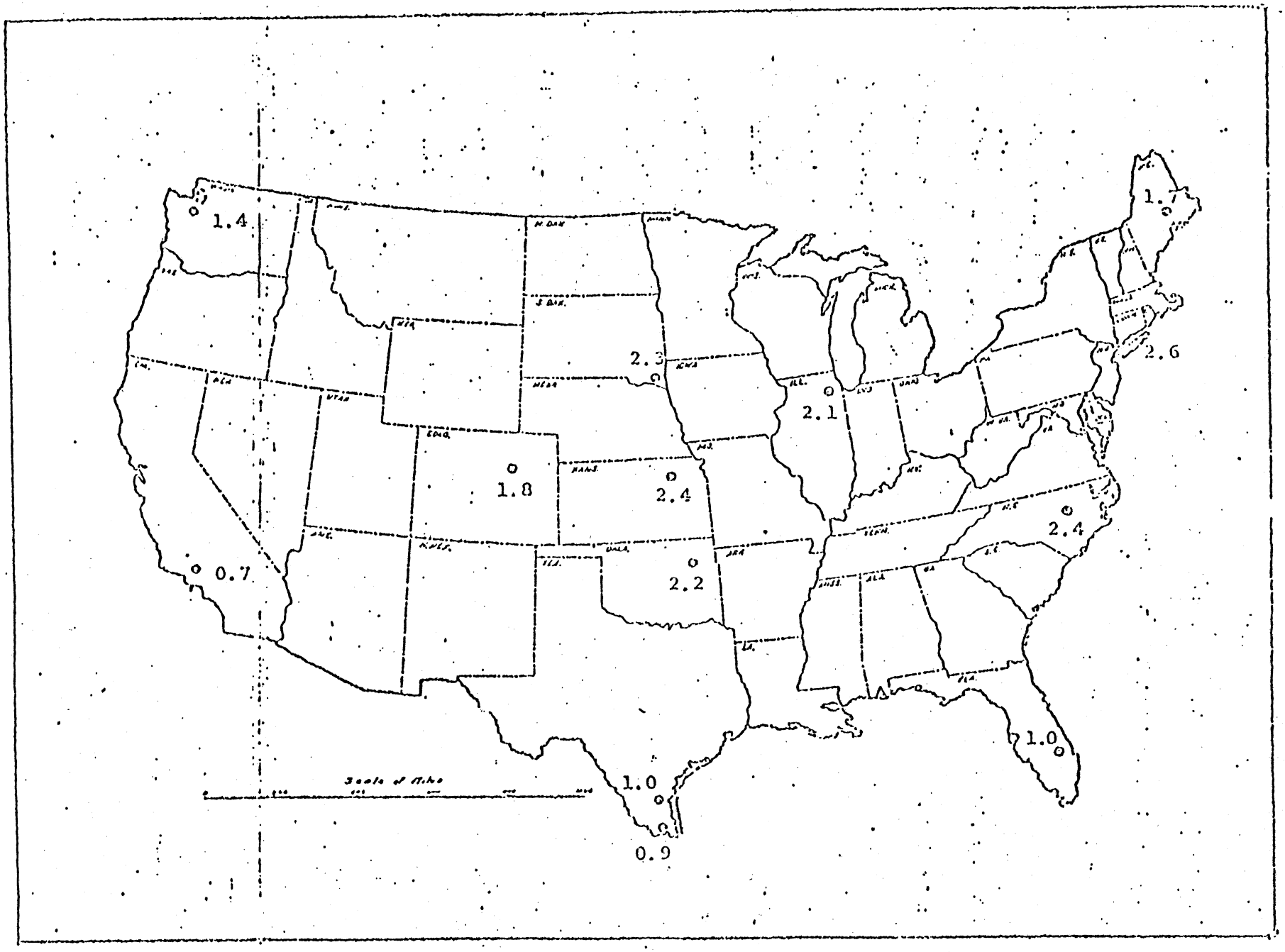
Accumulated plutonium fallout in the north central and southeastern sections of Utah was measured in soil samples collected in June 1971. The maximum values in Utah of the deposition (mCi Pu-239,240 per km^2) and deposition concentration (mCi per km^2 per cm of precipitation) were 2.2 and 3.8 times the maximum values found anywhere else in the United States. By comparing the Pu-239,240 to Sr-90 activity ratios of the Utah soils with the average activity ratio of 32 Northern Hemisphere soils collected in 1970-71, we estimate that up to 60 percent of the total Pu-239,240 activity deposited at some Utah sites was from a source other than the stratospheric reservoir. Soil samples taken at a site near the University of Utah from 1959 through 1971 revealed that the excess plutonium was delivered prior to 1959. Mass isotopic analyses indicate that the Nevada Test Site is the probable second source and that about 3.5 mCi Pu-239,240 per km^2 or twice the level expected from global fallout, deposited in the Salt Lake City area from NTS. Excess plutonium was not evident at sites north of Salt Lake City and due east in the valley regions beyond the Wasatch range.

INTRODUCTION

Presently there is considerable interest on the part of the Atomic Energy Commission and affiliated laboratories in documenti current levels of plutonium in the environment. Most of the Pu-2 240 on the surface of the earth arrived as fallout from atmospher

nuclear weapons tests. The accidental destruction of a SNAP generator in April 1964 resulted in the dispersal of Pu-238 which nearly tripled the global deposit of this isotope by 1970 (1). Localized contamination by weapons grade plutonium has occurred in the Rocky Flats region near Denver, Colorado (2) and in the vicinity of Palomares, Spain (3) and Thule, Greenland (4). The Health and Safety Laboratory has measured the accumulated deposits of plutonium at many sites throughout the world through the collection and analysis of soil samples (1). Depending upon latitude and climatic conditions, the deposits of Pu-239,240 for example, vary from 0.7 mCi per km^2 to 2.6 mCi per km^2 in the conterminous United States as shown in Figure 1.

In June 1971 a special soil survey was made in the north central and southeastern sections of Utah to investigate unusually high Sr-90 fallout (5). The samples were also analyzed for Pu-239, 240 and Pu-238. These data showed an unexpectedly large variability in plutonium deposition among the Utah sites. Subsequent analyses of soil samples collected in Salt Lake City since 1959 indicated that plutonium from a source other than the stratospheric reservoir had significantly contributed to the plutonium fallout. In this report we discuss the available data on plutonium fallout in Utah and present the evidence to demonstrate this second source.


1.4
2.3
2.1
1.8
2.4
2.6
2.4
0.7
2.2
1.0
1.0
0.9

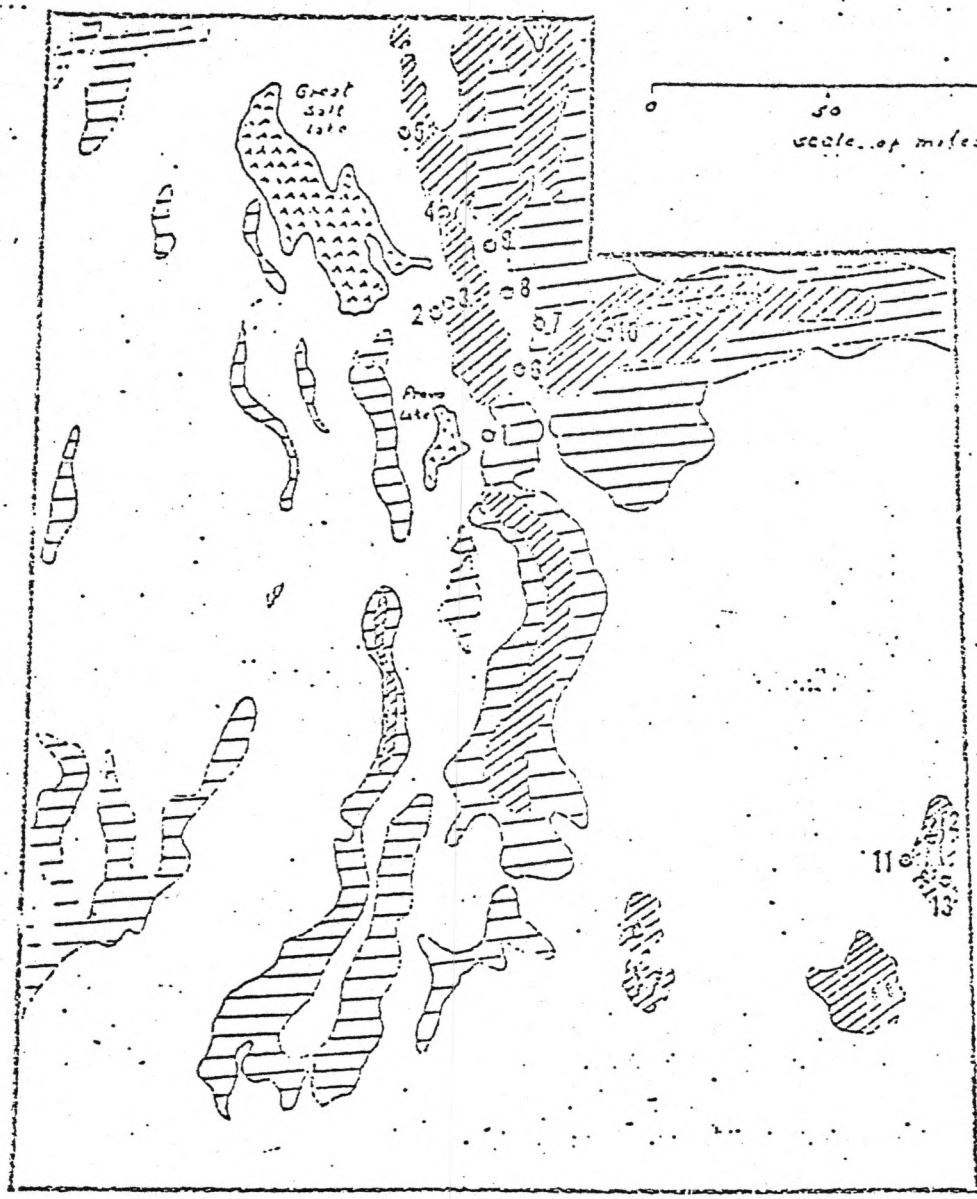
1971 SOIL SAMPLES

Thirteen sites were sampled in Utah during June 1971. Figure 2 shows the locations of these sampling sites. The predominant topographic features of the state are illustrated by the shaded altitude contours. Five sites were sampled west of the Wasatch range (Nos. 1 through 5), four in valley areas east of Salt Lake City (Nos. 6 through 9), and one at relatively high elevation in the Uinta Mountains (No. 10). Samples were also collected near Moab in southeastern Utah about 200 miles from Salt Lake City. Site No. 11 is in the dry Spanish Valley, site No. 12 is at higher elevation in the Manti-Lasal National Forest, and site No. 13 is in a valley area to the south east of the mountains.

The sampling method has been described in detail elsewhere (6) One thousand gram aliquots of the samples were acid leached using Pu-236 as a tracer and following separation and electrodeposition, the Pu-239,240 and Pu-238 were determined by alpha spectrometry(7). It is not possible to distinguish between the activities of Pu-239 and Pu-240 by alpha pulse height analysis because their energies cannot be resolved. The large aliquot size was necessary to measure the activities with counting errors of 5 percent or less.

Figure 2

1971 SOIL SAMPLING SITES IN UTAH



1. Provo
2. Salt Lake City (Lib. Pk.)
3. Salt Lake City (Univ. Ut.)
4. Ogden
5. Brigham
6. Heber
7. Marion
8. Wanship (Rockport Lk.)
9. Menefer
10. Uinta Mtns. (Trial Lk.)
11. Moab (BLM)
12. Manti Lasal N.F. (Geyser Pass
13. Manti Lasal N.F. (GS)

The plutonium analyses were carried out by Teledyne Isotopes, Palo Alto, California. We sampled to a depth of 30 cm to insure that all of the deposited plutonium and Sr-90 was collected (8).

Duplicate soil samples were taken at site 3, the University of Utah in Salt Lake City to reflect the overall sampling and analytical reproducibility. Table 1 shows that the cumulative Pu-239,240 and Pu-238 deposits were measured with precisions of about 2 and 6 percent, respectively.

Table 1

DUPLICATE SOIL SAMPLES
- University of Utah Site -
1971

Sample	mCi per km^2 Pu-239.240	Pu-238
1	5.2	0.17
2	5.3	0.16
difference	1.9%	6.1%

From a conservative standpoint then, we have assumed that the error terms associated with all the measurements made in this study are on the order of 10 percent.

The plutonium data for the 13 sites are presented in Table 2. The isotope ratios, Pu-239,240 to Sr-90 and Pu-238

TABLE 2

CUMULATIVE DEPOSIT OF Pu-239,240 AND Pu-238 IN 1971 UTAH SOILS

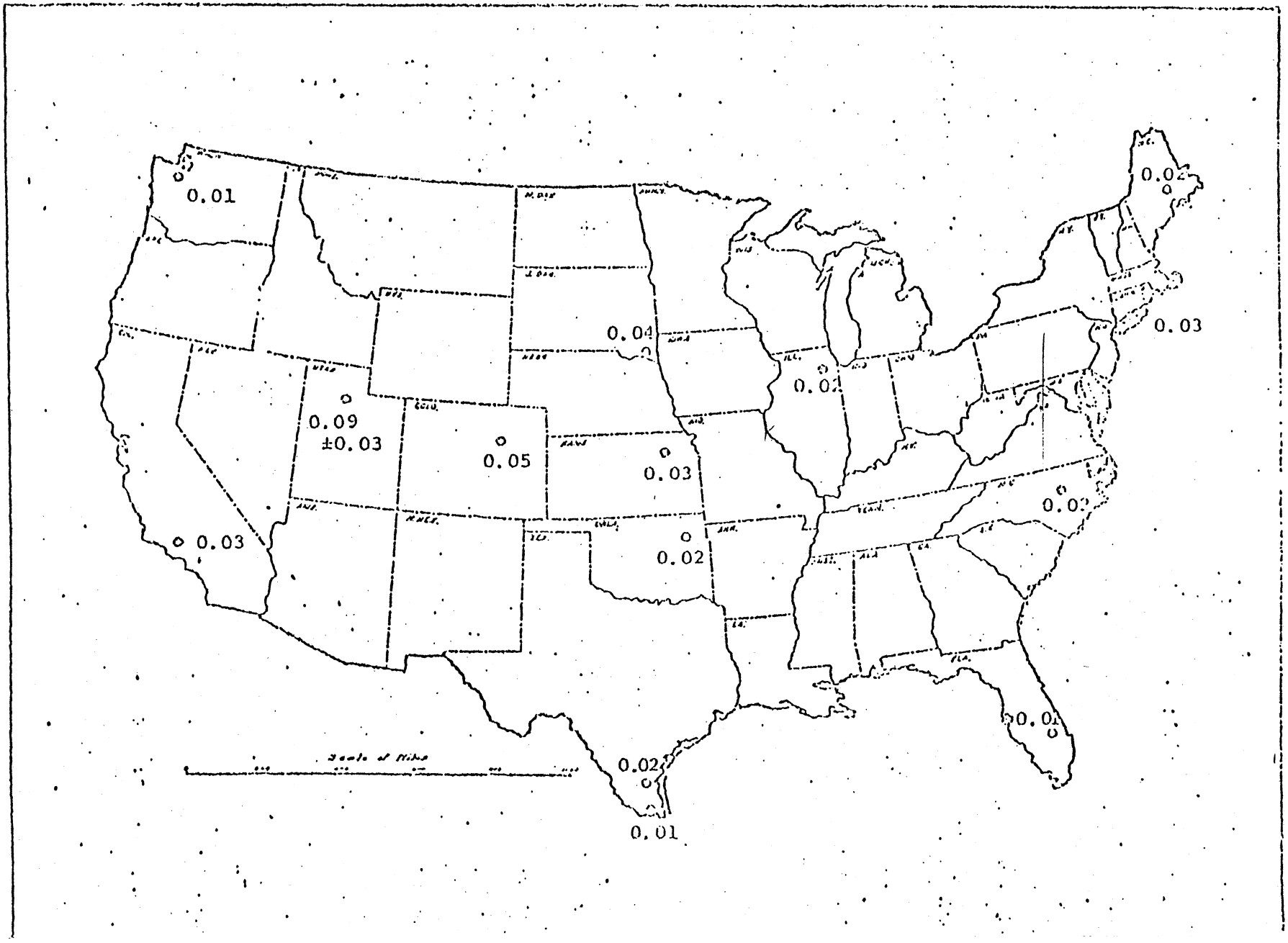
Map Site No.	Location	Site	Mean Ann. Precip. (cm)	Alt. (ft.)	mCi per km²		Ratios		mCi Pu-239,340 per km² per cm
					Pu-239,240	Pu-238	Pu-239,240 / Sr-90	Pu-239 / Pu-239,240	
		Sites West of the Wasatch Mtns.							
1	Provo	Utah State Hospital	39	4600	5.8	0.14	0.062	0.024	0.15
2	Salt Lake City	Liberty Park	39	4300	4.1	0.11	0.048	0.027	0.11
3	Salt Lake City	Univ. of Utah	41	4800	5.2	0.16	0.047	0.031	0.13
4	Ogden	Cache Nat'l. Forest	53	5200	5.5	0.20	0.030	0.036	0.10
5	Brigham	Tabernacle	46	4500	2.9	0.14	0.032	0.048	0.06
		Valley Areas East of Salt Lake City							
6	Heber	Tabernacle	43	5600	3.0	0.094	0.042	0.031	0.03
7	Marion	cemetery	63	6600	4.6	0.15	0.031	0.033	0.07
8	Wanship	Rockport Lake	42	6100	2.9	0.12	0.031	0.041	0.07
9	Henefer	meadow	38	5500	2.9	0.12	0.035	0.041	0.08
		Mountain Area							
10	Uinta Mtns.	Trial lake	76	9500	2.7	0.11	0.034	0.041	0.04
		Moab Area							
11	Spanish Valley	B.L.M. area	20	5300	3.0	0.084	0.045	0.028	0.15
12	Manti Lasal N.F.	Geyser Pass	81	9500	3.3	0.11	0.040	0.033	0.04
13	Manti Lasal N.F.	Lasal G.S.	41	7400	2.3	0.093	0.031	0.040	0.06

to Pu-239,240 are also included as well as the mean annual precipitation and altitude for each site.

REGIONAL VARIATIONS IN PLUTONIUM FALLOUT

The most striking observation is that the accumulated deposits of Pu-239,240 at the 13 Utah sites range up to 2.2 times the highest plutonium fallout measured at other sites in the United States less than a year earlier (see Figure 1). The highest deposit, 5.8 mCi Pu-239,240 per km^2 was found at Provo (Site 1) and the lowest deposit, 2.3 mCi Pu-239,240 per km^2 at site 13 in southeastern Utah.

The comparison of plutonium fallout in Utah with other areas of the United States becomes even more dramatic when we consider the fact that precipitation is the predominant process by which nuclear debris particles are brought down to the earth's surface[9]. Dividing by the mean annual precipitation for each site, the average Pu-239,240 deposition concentration for sites through 9 (last column of Table 2) is 0.09 ± 0.03 mCi per km^2 pe cm. Figure 3 shows that the deposition concentrations in other areas of the United States range from 0.01 to 0.05 mCi per km^2 per cm. In the present samples, only the high mountain sites (Nos. 10 and 12) approach the values found outside of Utah.


0.01
0.04
0.09
±0.03
0.05
0.03
0.02
0.03
0.02
0.03
0.02
0.03
0.02
0.01
0.04
0.03
0.02
0.04
Scale of Miles

ANOTHER SOURCE OF PLUTONIUM FALLOUT

Comparison of the Pu-239,240 to Sr-90 ratios measured in the Utah soils (Table 2) with the average ratio of 0.028 ± 0.003 found for 32 soils collected in the Northern Hemisphere in 1970-71 indicates that another source of plutonium has contributed to the deposit at some of the Utah sites.

The contributions from global fallout and this other source were calculated for each site as a first approximation by the following equations and are reported in Table 3:

$$(Pu)_F = \frac{0.028}{R} (Pu)_T$$

and

$$(Pu)_2 = (Pu)_T - (Pu)_F$$

where $(Pu)_F$, $(Pu)_2$, and $(Pu)_T$ equal the Pu-239,240 activity from global fallout, from the additional source, and the total measured activity, respectively, at each site. The symbol R is the measured Pu-239,240 to Sr-90 ratio.

It is clear from Table 3 that significant amounts of excess plutonium fallout occurred at Provo and Salt Lake City (sites 1 through 3) and probably at Heber (site 6) as well as sites 11 and 12 in the Moab area. It appears that little if any excess plutonium fallout occurred further north of Salt Lake City (sites 4

TABLE 3

COMPONENTS OF PLUTONIUM FALLOUT IN 1971 UTAH SOILS

Map Site No.	Location	Site	mCi Pu-239,240 per km^2 Global	Enc
		Sites West of Wasatch Mtns.		
1	Provo	Utah State Hospital	2.6±0.4	3.2
2	Salt Lake City	Liberty Park	2.4±0.3	1.7
3	Salt Lake City	Univ. of Utah	3.1±0.4	2.1
4	Ogden	Cache Nat'l. Forest	5.0±0.7	0.5
5	Brigham	Tabernacle	2.6±0.4	0.3
		Valley Areas East of Salt Lake City		
6	Heber	Tabernacle	2.0±0.3	1.0
7	Marion	cemetery	4.2±0.6	0.4
8	Wanship	Rockport Lake	2.6±0.4	0.3
9	Henefer	meadow	2.3±0.3	0.6
		Mountain Area		
10	Uinta Mtns.	Trial Lake	2.2±0.3	0.5
		MOAB Area		
11	Spanish Valley	B.L.M. area	1.9±0.3	1.1
12	Manti Lasal N.F.	Geyser Pass	2.3±0.3	1.0
13	Manti Lasal N.F.	Lasal G.S.	2.1±0.3	0.2

and 5) or in the valley areas due east of Salt Lake City (sites 7 through 10). Table 3 indicates that at some sites this excess plutonium fallout contributes up to 60 percent of the total measured deposition.

CHRONOLOGY OF PLUTONIUM DEPOSITION

We were fortunate to have in storage, aliquots of soils sampled at the University of Utah site from 1959 through 1965. These were sent to Teledyne Isotopes in Palo Alto for plutonium analysis. The data are presented in Table 4. The Pu-239,240 to Sr-90 and the Pu-238 to Pu-239,240 ratios are also given. The Sr-90 data for this site were reported in another publication (5).

Table 4

CUMULATIVE DEPOSIT OF PLUTONIUM
University of Utah: 1959 - 1971

Sampling Time		mCi per km^2		Ratios	
Year	Month	Pu-239,240	Pu-238	Pu-239,240 / Sr-90	Pu-238 / Pu-239,240
1959	Oct.	4.2	0.094	0.11	0.022
1960	Oct.	3.8	0.091	0.10	0.024
1962	Oct.	4.2	0.11	0.086	0.025
1963	Aug.	4.5	0.11	0.060	0.024
1964	Sept.	4.9	0.11	0.048	0.023
1965	Sept.	4.7	0.12	0.045	0.026
1971	June	5.2	0.16	0.048	0.030

Table 4 shows that the Pu-2 9,240 deposit at the University of Utah site in 1959 was as high as 80 percent of the deposit measured in 1971. The Pu-239,240 to Sr-90 ratios which drop fro 0.11 in 1959 to 0.048 in 1971 are all significantly greater than the average value of 0.028 ± 0.003 found for 32 Northern Hemisph soils in 1970-71. These data indicate that the excess plutonium fallout was delivered prior to 1959. It is plausible to assume that this second source of plutonium fallout is the Nevada Test Site. The test site is about 350 miles southwest of Salt Lake C and has occasionally been a source of short-lived fission produc in Utah following atomic tests (10, 11).

The excess plutonium for this site reported in Table 3 assu that all the Sr-90 deposited was from global fallout. However, has been concluded that in 1959 about 16 mCi Sr-90 per km^2 were present in Salt Lake City from a second source independent of gl fallout(5). By 1971 this second source component would have dec to 12 mCi per km^2 which, when subtracted from the total measure deposit in 1971, yields 109 − 12 = 97 mCi per km^2 as the integr deposit in Salt Lake City from all previous global fallout.

Multiplying 97 mCi Sr-90 per km^2 by the average Pu-239,240 to Sr-90 ratio in 1970-71 for integrated global fallout (0.028)

2.7 mCi per km^2 as the global fallout component of the Pu-239 240 deposit. The difference from the total measured plutoniu deposit is 2.5 mCi per km^2 which is the contribution from the second source. This second source may well be the Nevada Tes Site. Similar calculations can be made from the monthly Sr-9 deposition data at Salt Lake City from 1959-1971[12] to give reasonably comparable estimates of 2.3 and 2.9 mCi Pu-239,240 km^2 as the global fallout and second source components, respe tively.

It has been demonstrated that mass spectrometric analysi of plutonium in soil can differentiate between global fallout plutonium and plutonium from another source[13]. Table 5 gi the Pu-240 to Pu-239 mass ratios of the 1971 University of Uta the global fallout reference sample collected at the Brookhave National Laboratory and three surface soil samples from areas Nevada Test Site. The comparison of the University of Utah Pu to Pu-239 ratio with the global fallout reference ratio is fur strong evidence that the plutonium deposited in Salt Lake City could not be entirely from global fallout.

Table 5

MASS ISOTOPIC COMPOSITION

Soil Sample	Pu-240/Pu-239
Salt Lake City: Univ. of Utah	0.0845 ± 0.44%
Global Fallout Reference[a]	0.1801 ± 0.24%
Surface Sample No.1 from NTS[b]	0.0497 ± 0.4%
Surface Sample No.2 from NTS[b] (Project '57 area)	0.0628 ± 0.2%
Soil Sample No.3 from NTS[c]	0.0539 ± 0.4%

(a) collected at Brookhaven National Laboratory, 1970

(b) samples supplied by Mr.G. Hamada, Reynolds Electric & Engineering Co., Las Vegas, Nevada

(c) G.M. Leies, AFTAC, Dept. of the Air Force, Personal Communication, May 1972

Krey and Krajewski[13] derived an equation to calculate the relative mass contribution of a second source of plutonium to global plutonium. Modifying this equation to give the

relative activity contribution yields:

$$\frac{\text{Pu activity from a 2nd Source}}{\text{Pu activity from global fallout}} = R = \frac{C_1 - C_2}{C_2 - C_3}\left(\frac{1 + 3.60\ C_3}{1 + 3.60\ C_1}\right)$$

where:

C_1 = Pu-240 to Pu-239 mass ratio from global fallout

C_2 = Pu-240 to Pu-239 mass ratio in Univ. of Utah soil

C_3 = Pu-240 to Pu-239 mass ratio from second source

We can calculate the relative plutonium activity contributions of the two source from the above equation, provided that the constants are evaluated. From Table 5: $C_1 = 0.1801$, $C_2 = 0.0846$, and $C_3 \cong 0.05$. This approximate assignment of C_3 assumes that the second source was either debris from some of the safety tests in which plutonium was physically dispersed or tests in which little fission occurred. The three soil samples in Table 5 were collected in areas contaminated mainly by debris from safety tests. From the assignments $R \cong 2$. Since the measured plutonium deposit from Table was 5.2 mCi per km^2, the global fallout plutonium = 1.7 mCi per km^2 and plutonium from the second source = 3.5 mCi per km^2. This estimate of the second source contribution is only about 1/3 greater than those calculated from the Sr-90 measurements which is reasonable agreement under the assumptions of the computation.

PLUTONIUM FALLOUT IN OTHER AREAS

We showed in Figure 1 and Table 2 that the accumulated Pu-239,240 deposit in other areas of the United States is les than in the Salt Lake City area. Soil samples collected in C City, Utah in 1963, 64 and 65, and previously analyzed for Sr- were subsequently analyzed for Pu-239,240. Cedar City is abo 225 miles south-southwest of Salt Lake City and about 175 mil east of the Nevada Test Site. It is in a lower rainfall area than Salt Lake City and the cumulative plutonium fallout was about ¼ of that observed at Provo and the University of Utah can be seen from Tables 6 and 2. The Pu-239,240 to Sr-90 rat however, indicates that some plutonium from the Nevada Test S must have fallen out here also. On the other hand, Bozeman, Montana (340 miles north of Salt Lake City), Rapid City, South Dakota (500 miles northeast), and Denver, Colorado (360 miles east) showed no evidence of a second source of plutoniu The Pu-239,240 to Sr-90 ratios for these last three regions were essentially the same as we found for other areas of the

world where only global fallout had occurred. These data emphasize our earlier suggestion that plutonium from the Nevada Test Site did not deposit north of the Salt Lake City area or further east than the Utah - Colorado border.

Table 6

CUMULATIVE DEPOSIT OF PLUTONIUM AT OTHER SITES

Sampling Time Year	Month	mCi $Pu^{239,240}$ per km^2	$Pu^{239,240}/Sr^{90}$
	Cedar City, Utah annual precip.: 23 cm		
1963	Sept.	1.4	0.044
1964	Sept.	1.5	0.043
1965	Sept.	1.4	0.038
	Bozeman, Montana annual precip.: 43 cm		
1965	Sept.	1.9	0.025
	Rapid City, South Dakota annual precip.: 45 cm		
1965	Sept.	2.5	0.026
	Denver, Colorado annual precip.: 37 cm		
1965	Sept.	1.5	0.024
1970	Oct.	1.8	0.027

CONCLUSIONS

Plutonium isotopes, injected into the stratosphere follow atmospheric nuclear weapons tests, are dispersed globally and eventually reach the ground by a variety of meteorological and climatological processes. The predominant source of plutonium out then, in most locales, is the stratospheric reservoir. The cumulative deposits of Pu-239,240 as measured by soil samples c lected in the United States in 1970-71 range from 0.7 to 2.6 mC km^2.

Analyses of soils collected in north central and south eastern Utah in June 1971, however, revealed cumulative Pu-239, deposits that were twice expected levels at some sites. Furthe more, the Pu-239,240 to Sr-90 ratios at these sites were signif cantly higher than we found in 32 soils sampled in the Northern Hemisphere in 1970-71. Excess plutonium fallout was clearly de strated at Provo, Salt Lake City, and Heber but not at Ogden an Brigham City further north. We could find no evidence of exces plutonium in the valley areas due east of Salt Lake City or at higher elevation in the Uinta mountains. Two sites in the Moab area also showed evidence of excess plutonium fallout.

A series of 7 soil samples collected between 1959 and 1971 at the University of Utah showed that the cumulative deposit of Pu-239,240 was higher than expected from global fallout throughout the entire period. This indicated that prior to 1959, plutonium from a second source-presumably the Nevada Test Site - contaminate the Salt Lake City area. A component analysis of the Pu-239,240 fallout demonstrated that a chronic contamination through resuspension could not have taken place after 1959. We estimate that the excess plutonium fallout at the University of Utah is equal to or perhaps a factor of two greater than the global plutonium. This estimate is supported by the mass ratio of Pu-240 to Pu-239 measured in the University of Utah soil compared with the mass ratios of global fallout and debris from the Nevada Test Site.

It is possible that our estimates of the second source plutonium fallout are low because the soils were acid leached. Some fraction of the plutonium in Nevada Test Site debris is not acid extractable although we have no supporting data for the Utah soils. Nevertheless, global fallout plutonium can be acid extracted from soil[3] and so our estimates of plutonium from this source are probably reliable.

We could find no evidence that excess plutonium had deposited in Bozeman, Montana; Rapid City, South Dakota, or Denver, Colorado which verified our observation that plutonium contamination from the Nevada Test Site had not occurred north of Salt Lake City or further east than the Utah - Colorado border.

ACKNOWLEDGEMENTS

We are grateful to those who made it possible for us to obtain the soil samples in June 1971. Mr. William Wertz of the Forest Service in Ogden, Utah arranged the contacts with Forest Service and State Parks and Recreation Commission personnel. Dr. T. B. Hutchings, State Soil Scientist, arranged the Soil Conservation Service contacts. Mr. Warren L. White, Superintendent of Parks, Salt Lake City and Mr. David Lofgren, Superintendent of General Services, University of Utah, made it possible to collect samples in Liberty Park and the University golf course respectively.

We appreciate the cooperation and interest of Dr. Russel Hendricks, Western Area Occupational Health Laboratory Salt Lake City in maintaining the monthly fallout collector for the past 15 years.

Dr. L. T. Alexander, consultant to AEC, collected most of the soil samples prior to 1971 and provided valuable advice as to sampling locations for the 1971 investigation.

Interesting that everyone but the Health [illegible] has been involved in a matter that [illegible] be of concern to them because of [illegible] extensive radiological monitoring etc.

REFERENCES

1. Hardy, E. P., P. W. Krey and H. L. Volchok
Global Inventory and Distribution of Pu-238 from SNAP-9
USAEC Report HASL-250, March 1, 1972

2. Krey, P. W. and E. P. Hardy
Plutonium in Soil Around the Rocky Flats Plant
USAEC Report HASL-235, August 1, 1970

3. Iranzo, E., and S. Salvador
Inhalation Risks to People Living Near a Contaminated A
Second Int'l. Congress of the Int'l. Rad. Prot. Assoc.
Brighton, England (May 3-8, 1970)

4. Aarkrog, A.
Radioecological Investigations of Plutonium in an Arcti
Environment
Health Physics, Vol. 20, pp. 31-47, January 1971

5. Hardy, E. P., H. L. Volchok, and P. W. Krey
Strontium-90 Fallout in Utah
USAEC Report HASL-257, pp. I-71 to I-94, July 1, 1972

6. Harley, J. H. (Editor)
Manual of Standard Procedures
USAEC-HASL Report NYO-4700, pp. D-04-01 to 07, August 1

7. Harley, J. H. (Editor)
Manual of Standard Procedures
USAEC-HASL Report NYO-4700, pp. E-Pu-06-01 to 03,
August 1971

8. Hardy, E. and P. W. Krey
Determining the Accumulated Deposit of Radionuclides by
Soil Sampling and Analysis
Proceedings of Environmental Plutonium Symposium
LA-4756, 1971

9. Hardy, E. and L. T. Alexander
Rainfall and Deposition of Strontium-90 in Clallam County, Washington
Science, 136, No. 3519, pp. 881-882, June 8, 1962

10. Pendleton, R. C. and R. D. Lloyd
Environmental Levels of Radioactivity in Utah Following Operation Pinstripe
Rad. Health Data and Reports, 11, No. 2, pp. 65-67, February 1970

11. Pendleton, R. C. and R. D. Lloyd
Zirconium-95 in Utah Vegetation Produced During the 1966 Growing Season
Rad. Health Data and Reports, 11, No. 6, pp. 277-28, June 1970

12. Fallout Program Quarterly Summary Report
USAEC Report HASL-257
Appendix Section A, pp. 83-84, July 1, 1972

13. Krey, P. W. and B. T. Krajewski
Plutonium Isotopic Ratios at Rocky Flats
USAEC Report HASL-249, pp. I-67 to I-94, April 1, 1972