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health and safety laboratory

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ABSTRACT

Following the burn-up of a SNAP generator containing Pu-238 in the upper atmosphere of the Southern Hemisphere in April 1964, balloon and aircraft sampling successfully documented the atmospheric transport and inventory of the debris from this unexpected release. Attempts to measure the fallout rate on a continuous basis at sites in both the Northern and Southern Hemispheres were not successful. By the end of 1970, the stratospheric measurements indicated that 95 percent of the SNAP plutonium had deposited over the earth's surface. Integrated fallout in the form of soil samples collected at over 60 sites throughout the world provided the data required to assess the distribution pattern and inventory of the deposited SNAP debris. Of the 17 kilocuries of Pu-238 originally in the generator, 13.4 ± 2.2 kilocuries are globally deposited with 3.1 ± 0.8 kilocuries in the Northern Hemisphere and 10.3 ± 2.1 kilocuries in the Southern Hemisphere. This accounts for essentially all of the Pu-238 inadvertently released as a result of the satellite abort.

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INTRODUCTION

Plutonium isotopes, Pu-239,240 and Pu-238 in particular, are injected into the stratosphere as a result of atmospheric nuclear weapons tests. These isotopes are eventually globally dispersed and reach the ground as particulate fallout in a manner similar to many of the fission products. About 8 years ago, however, an accidental stratospheric injection of Pu-238 occurred which has resulted in almost a three-fold increase in the global fallout of this isotope. The purpose of this report is to describe the method used by the Health and Safety Laboratory to determine the cumulative fallout and geographical distribution which resulted from this unexpected radioisotopic release.

On April 21, 1964, a Transit navigational satellite was launched with a Thor Able Star rocket from Vandenberg Air Force Base in California. The payload included a Systems for Nuclear Auxilliary Power generator, SNAP-9A, containing 17 kilocuries or about 1 kilogram of Pu-238[1,2]. Because the rocket failed to boost the satellite into orbital flight, the payload re-entered the atmosphere in the Southern Hemisphere[2,3].

The SNAP-9A device, designed to convert heat developed by the Pu-238 into electrical energy, was not built for an intact re-entry. Based on subsequent stratospheric inventories[4-6] it was concluded that the generator completely burned up during re-entry and ablated into small particles at an altitude of about 46 km[4]. Concentrations of the SNAP Pu-238 in the stratosphere were measurable through the end of 1970 when it was estimated that less than a kilocurie remained above 12 km[7]. Attempts to measure the ground deposition rate on a continuous basis using the HASL fallout network, were unsuccessful[8]. We finally resorted to soil sampling to obtain integrated fallout samples. By measuring the Pu-238 content of soils collected in many areas of the world we were able to estimate the global distribution and inventory of the accumulated SNAP-9A fallout. The soil data form the basis of this report, but first we shall briefly discuss the stratospheric, surface air, and deposition rate measurements that have been made.

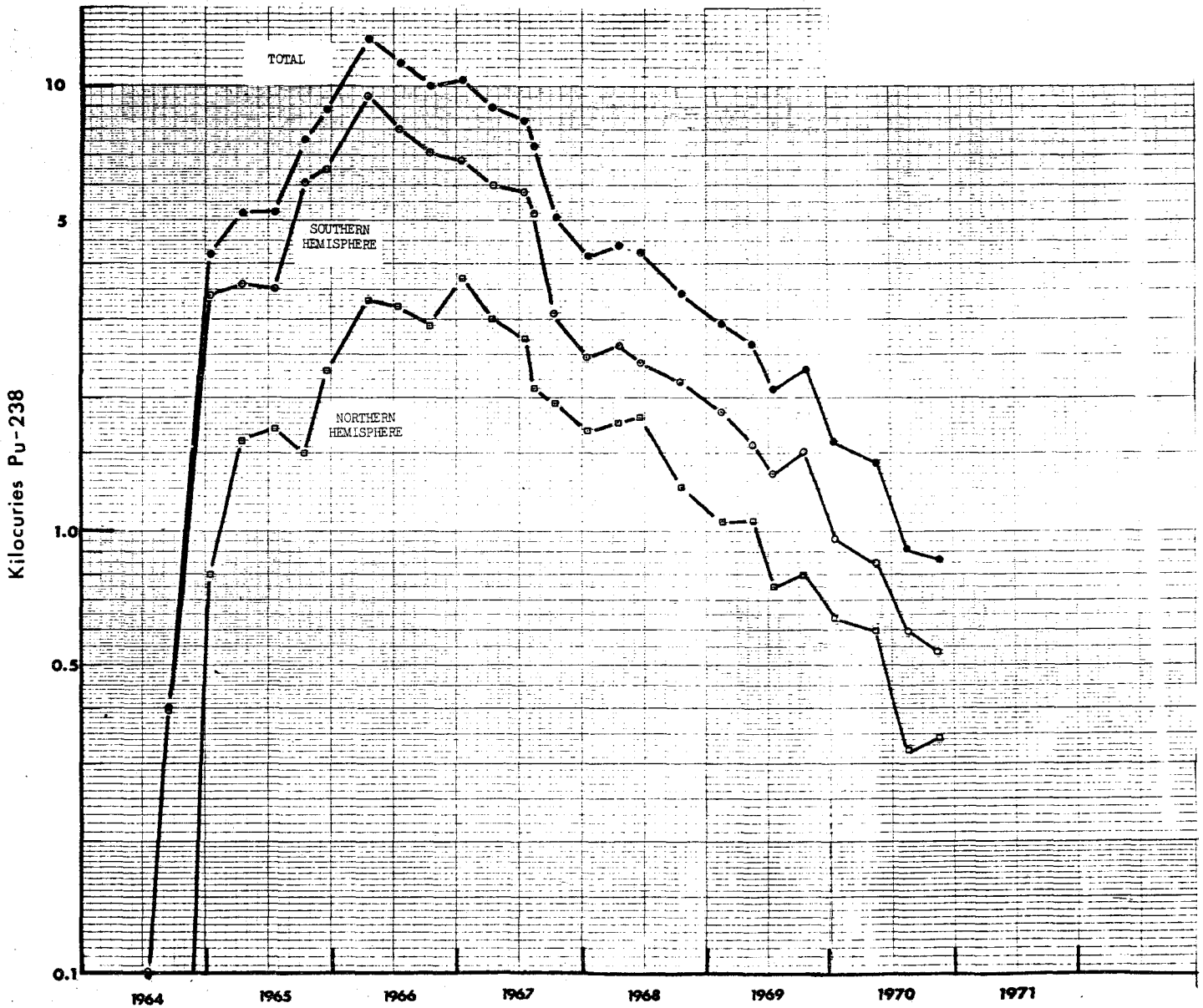
AIR AND FALLOUT MEASUREMENTS

The SNAP-9A Pu-238 was first identified at 33 km over latitude 34° south, four months after the satellite failure[9]. The debris was collected on filters under the AEC's High Altitude Balloon Sampling Program. By January 1965 the SNAP plutonium was detected at 33 km in the Northern Hemisphere at latitude 31° [10]. The subsequent dispersion and subsidence of the debris was followed under Project Stardust, an aircraft sampling program sponsored by the Defense Atomic Support Agency[6] and the AEC's continuation of the aircraft sampling under Project Airstream[4,11-13]. Figure 1 shows the stratospheric inventories at quarterly intervals from 1964 through 1970. Peak stratospheric concentrations occurred in 1966 and the stratosphere was subsequently depleted with a half-residence time of 14 months[7].

Measurements were made of the SNAP Pu-238 particle sizes by radioautography of the balloon-borne filters bearing the debris collected between 20 and 30 km[14]. Although individual particles of SNAP debris could not be isolated, the size distribution of equivalent Pu-238 oxide spheres was log normal with a mode at about 0.013 μm (particle diameter) and a size range of 0.010 and 0.040 μm. As these small individual Pu-238 oxide particles entered the atmosphere they were expected to agglomerate with the larger natural aerosol particles quite rapidly.

Based on experiences with other high altitude injections of unique radioisotopes, predictions were made as to when ground level sampling systems would first detect fallout from the SNAP-9A[2,15]. Those predictions were eventually borne out. Ground level air measurements at Ispra, Italy first detected the SNAP plutonium in early 1966[16]. Shortly thereafter it was identified in monthly fallout collections in New York[17] and Ispra[16]. The peak surface air concentration and deposition rate at Ispra occurred during the spring of 1967[16]. By this time, large area fallout collection systems in New York and Melbourne were discovered to be faulty due to contamination problems at HASL and incomplete removal of the plutonium from the collector at

Figure 1. Stratospheric Inventory of SNAP-9A Pu-238



Melbourne[8]. We began analyzing monthly fallout samples from Seattle, Honolulu, Rio de Janeiro, Salisbury, and Durban in 1968, but by this time the levels were too low. Ispra is the only site where continuous fallout measurements have been made. Consequently, it was neither possible to inventory the ground deposit nor determine the geographical distribution of the SNAP-9A fallout.

GLOBAL SOIL SAMPLING

The stratospheric inventories clearly indicated that by mid-1970, 95 percent of the SNAP Pu-238 had deposited on the earth's surface[7]. It was important to find out how it was distributed and to inventory the ground deposit since there was little information on which to base predictions. The isotope, mode and altitude of release, and the fact that the burn-up occurred in the Southern Hemisphere were conditions which had not been experienced. Knowledge of the fate of this debris is pertinent to studies of nuclear explosions in the upper atmosphere and the use of nuclear power sources in space.

Under the circumstances, soil sampling and analysis was the only way to measure the accumulated deposit. From October 1970 through January 1971, 65 sites around the world were sampled by HASL personnel and scientists from other countries (see appendix listing of people who assisted in sampling). The samples were sent to our laboratory where they were prepared for analysis and then submitted to two non-government laboratories, Teledyne Isotopes in Palo Alto, California (IPA) and Trapelo Division West in Richmond, California (TLW) for the actual chemical separation and counting of plutonium. A small number of the samples were analyzed for plutonium at HASL. The sampling[18a] and analytical methods[18b] were prescribed by HASL.

METHODS

The soils were collected from areas that had been undisturbed for at least 15 years. Each sample consisted of ten 8.89 cm diameter cores taken to 30 cm depth, representing a surface area of 622 cm^2. The samples were analyzed for Pu-238, Pu-239,240 (hereafter referred to as Pu-239), and Sr-90. The measured Pu-238 included that from weapons testing plus the SNAP-9A contribution. The Pu-239 was assumed to be entirely from weapons testing as we avoided areas known to be locally contaminated such as at Rocky Flats, Colorado[19], Palomares, Spain[20], and Thule, Greenland[21].

Strontium-90 analyses were carried out at HASL to compare with previous soil studies of this fission product[22] and to insure that the samplings included all of the accumulated fallout. Depth profiles taken at Brookhaven National Laboratory in September 1970 and reported in Table 1 indicated that 30 cm would be a suitable sampling depth for both nuclides. Table 1 shows that Sr-90 penetrated deeper than Pu-239 with 42 percent of the total Sr-90 deposit and 57 percent of the total Pu-239 deposit being contained in the top 7 cm. These data on vertical movement of Sr-90 and plutonium are in agreement with other studies[23-25]. At four other sites in the United States we sampled below 30 cm (see appendix Table A) and found less than 5 percent of the total Pu-239.

One thousand gram aliquots of soil were required to measure the total Pu-238 with one standard deviation of the counting error less than or equal to ± 10 percent. An acid leaching procedure developed at HASL[26] was used to accommodate this large size sample. This procedure was compared to complete dissolution and two other extraction methods and found to be comparably efficient (see appendix Table B). Strontium-90 was extracted from 100 gram soil aliquots using a base-acid leaching method developed by the USDA Soil Conservation Service[18c].

TABLE 1

DEPTH DISTRIBUTION OF Pu-239 AND Sr-90 IN SOIL

Depth (cm)	mCi per km^2		Percent of Total	
	Pu-239	Sr-90	Pu-239	Sr-90
	BROOKHAVEN SOIL (September 1970)			
0-7	1.18±3%	30±9%	57	42
7-11	0.57±4%	21±6%	27	30
11-15	0.23±6%	11±12%	11	15
15-21	0.07±20%	5.8±7%	3	8
21-25	0.02±40%	1.4±20%	1	2
25-30	0.01±50%	1.9±50%	1	3
	FRILFORD, BERKS. SOIL (October 1970)			
0-5	0.53±2%	15±8%	55	43
5-30	0.44±3%	20±14%	45	57

SITE INTEGRITY

We have previously demonstrated in our soil samplings for Sr-90 that the accumulated deposits at nearby sites are within 10 percent of one another on the average[27,28]. Table 2 shows the data for five areas in this most recent global survey. The average percent difference between nearby sites was 15 percent for Pu-239, 23 percent for total Pu-238, and 23 percent for Sr-90. These differences, larger than previously encountered, can be explained by the relatively few areas where two sites were sampled in this survey and the greater difficulty in finding undisturbed sites in 1970 as compared to five years ago. Furthermore, almost half of the samples were collected by people who had little experience in this type of sampling. For inventory purposes, however, these differences are quite tolerable.

ANALYTICAL QUALITY CONTROL

As is customary at HASL, the soil analyses were checked by submitting blanks, reference soils, and duplicate unknowns on a blind basis to the HASL and contractor laboratories. A soil collected before 1945 was used as a blank. Only once was a measurable Pu-239 value reported for the blank and even on this occasion, the activity level was an order of magnitude lower than that encountered in most samples (see appendix Table C).

Four reference soils were analyzed at random during the course of the analyses. The Brookhaven soil was sampled on the grounds of the Brookhaven National Laboratory on Long Island, New York. One hundred cores were taken to a depth of 5 cm and composited. The "Black" and "Red" soils were sampled 14 years ago near Raleigh, North Carolina and McHenry County, Illinois, respectively, and have been used extensively in other Sr-90 in soil studies. The Woodcliff

TABLE 2

COMPARISON OF Pu AND Sr-90 IN SOILS AT NEARBY SITES
(1970 SAMPLINGS, 0-30 cm)

Location	Distance Between Sites	mCi per km^2					
		Pu-239		(total) Pu-238		Sr-90	
		Site 1	Site 2	Site 1	Site 2	Site 1	Site 2
Oslo, Norway	meters	1.4	1.5	0.041	0.052	53	45
Wantage, U. K.	5 km	1.2	1.0	0.055	0.035	42	35
Melbourne, Australia	meters	0.61	0.56	0.079	0.099	22	22
New York, USA	100 km	2.4	2.6	0.098	0.097	-	-
Lourenco Marques, Moz.	meters	0.25	0.18	-	-	9	5

Lake, New Jersey sample was collected in March 1970 at a depth below 90 cm with the intention of using it as a blank. Before processing, the sample was slightly contaminated and so aliquots taken from the blended reference samples contain low but measurable amounts of Pu-239. Analytical results for these reference samples are shown in the appendix Table D. The error term associated with each value is that due to counting. Averages were calculated for each reference sample with an error term representing one standard deviation of the mean. In those sets of replicate soil aliquots where the counting errors were less than 10 percent, the maximum standard deviation of the mean was ± 9 percent for Pu-239 and ± 18 percent for Pu-238. Using the same criterion for the Sr-90 analyses, the maximum standard deviation of the mean was ± 7 percent (see appendix Table E).

Five of the soils collected in this survey were analyzed in duplicate for the Pu isotopes. The analytical laboratories were unable to identify the samples. Average percent deviations between aliquots were 4 percent for Pu-239 and 6 percent for Pu-238 (see appendix Table F). Twenty-two sets of blind duplicates were analyzed for Sr-90 with an average percent deviation of 8 percent (see appendix Table G). These data convinced us that sample contamination was not a problem and that analytical reproducibility and accuracy were adequate for the objectives of this study.

LATITUDE DISTRIBUTION

The soil data are summarized in Table 3. The activity levels of the two plutonium isotopes and Sr-90 are expressed in the units, millicuries per square kilometer (mCi/km^2). Two isotope ratios, Pu-239 to Sr-90 and Pu-238 to Pu-239 are also given. The Pu-239 to Sr-90 ratio has been reasonably constant in stratospheric air[28] and in some areas, in fallout[30]. Anomalously high ratios have occasionally been observed in surface air since 1968, however, but the reasons are not clear[31]. The Pu-238 to Pu-239 ratio reflects the contribution of weapons plutonium and the SNAP-9A Pu-238. The weapons

Table 3

Pu Isotopes and Sr^{90} in Soil

Collected 1970-71

Lat. band	Country	Site	Lat.°	Long.°	Avg. annual precip. (cm)	mCi per km² Sr^{90}	mCi per km² Pu^{239}	mCi per km² Pu^{238}	Pu^{239}/Sr^{90}	Pu^{238}/Pu^{239}
) - 70 N	Greenland	Thule	76.6N	69.0W	<15	9	0.33		0.037	
	U. S. A.	Barrow, Alaska	71.3N	156.8W	14	18	0.40	0.010	0.022	0.025
70 - 60 N	U. S. A.	Fairbanks, Alaska	64.8N	147.9W	28	34	0.85	0.040	0.025	0.048
	Iceland	Reykjavik	64.1N	22.0W	86	55	1.5	0.052	0.028	0.034
	U. S. A.	Palmer, Alaska	61.6N	149.3W	41	32	0.92	0.044	0.029	0.048
	Norway	Bergen	60.4N	5.3E	192	92	3.1	0.123	0.034	0.039
60 - 50 N	Norway	Oslo	59.9N	10.8E	76	53	1.5	0.046	0.028	0.032
	U. K.	Wick, Caith.	58.4N	3.1W	80	56	1.4	0.048	0.025	0.034
	Denmark	Roskilde	55.6N	12.1E	59	48	1.2	0.038	0.025	0.032
	U. K.	Wantage, Berks	51.6N	1.4W	68	39	1.1	0.045	0.028	0.041
50 - 40 N	Germany	Munich	48.1N	11.6E	87	82	2.8	0.088	0.034	0.031
	U. S. A.	Puyallup, Wash.	47.2N	122.3W	109	58	1.4	0.046	0.024	0.032
	Italy	Ispra	45.8N	8.6E	145	84	2.5	0.085	0.030	0.034
	U. S. A.	Orono, Maine	44.9N	68.7W	94	63	1.7	0.057	0.027	0.033
	Japan	Sapporo	43.1N	141.4E	118	62	1.9	0.072	0.030	0.038
	U. S. A.	Vermillion, S. D.	42.8N	96.9W	65	86	2.3	0.104	0.027	0.046
	U. S. A.	Argonne, Ill.	41.8N	88.0W	84	74	2.1	0.069	0.028	0.033
	U. S. A.	New York, N. Y.	40.8N	73.9W	90	98	2.6	0.097	0.026	0.037
40 - 30 N	U. S. A.	Denver, Colo.	39.8N	104.9W	38	67	1.8	0.077	0.027	0.043
	U. S. A.	Manhattan, Kan.	39.2N	96.6W	77	88	2.4	0.087	0.027	0.036
	Portugal	Sacavem	38.8N	9.1W	73	58	1.5	0.048	0.026	0.032
	U. S. A.	Tulsa, Okla.	36.2N	95.9W	94	73	2.2	0.091	0.030	0.041
	U. S. A.	Raleigh, N. C.	35.8N	78.6W	116	91	2.4	0.105	0.026	0.044
	Japan	Tokyo	35.7N	139.8E	152	50	1.5	0.054	0.030	0.036
	U. S. A.	Burbank, Calif.	34.2N	118.4W	26	22	0.73	0.021	0.033	0.028
	Pakistan	Lahore	31.6N	74.4E	49	67	1.6	0.056	0.024	0.035
30 - 20 N	U. S. A.	Kingsville, Tex.	27.5N	97.9W	63	36	0.99	0.032	0.028	0.032
	U. S. A.	Ft. Pierce, Fla.	27.5N	80.7W	142	40	1.0	0.034	0.025	0.034
	U. S. A.	Weslaco, Tex.	26.2N	97.9W	61	28	0.88	0.037	0.031	0.042
20 - 10 N	U. S. A.	Papaikou, Hawaii	19.8N	155.1W	400	129	4.0	0.123	0.031	0.031
	Venezuela	Maracay	10.3N	67.5W	96	8	0.24	0.009	0.030	0.038
10 - 0 N	Colombia	Bogota	4.6N	74.1W	106	4	0.13	<0.002	0.032	
0 - 10 S	Kenya	Muguga	1.2S	36.6E	96	21	0.52	0.023	0.025	0.044
	Brazil	Belem	1.5S	48.5W	268	20	0.42	0.029	0.021	0.069
	Ecuador	Guayaquil	2.2S	79.9W	110	5	0.18	0.008	0.036	0.044
	Angola	Luanda	8.8S	13.3E	43	4	0.093	0.008	0.023	0.086
10 - 20 S	Peru	Lima	12.1S	77.0W	4	4	0.12	0.018	0.030	0.15
	Australia	Darwin	12.4S	130.2E	158	9	0.28	0.081	0.030	0.29
	Mozambique	Nampula	15.2S	39.2E	102	7	0.15	0.030	0.021	0.20
	Rhodesia	Salisbury	17.7S	31.0E	83	7	0.16	0.026	0.023	0.16
	Australia	Townsville	19.2S	146.8E	119	6	0.18	0.050	0.032	0.28
	Mozambique	Beira	19.8S	34.9E	151	5	0.17	0.040	0.034	0.24
20 - 30 S	Australia	Port Hedland	20.3S	118.6E	31	6	0.20	0.022	0.035	0.11
	Brazil	Angra dos Reis	23.0S	44.3W		26	0.60	0.112	0.023	0.19
	Australia	Alice Springs	23.7S	133.9E	24	10	0.35	0.079	0.035	0.23
	S. Africa	Pretoria	25.8S	28.2E	76	10	0.32	0.060	0.032	0.19
	Mozambique	Lourenco Marques	26.0S	32.6E	80	7	0.25	0.049	0.036	0.20
	Australia	Brisbane	27.4S	152.9E	108	19	0.40	0.071	0.021	0.18
30 - 40 S	Australia	Perth	33.1S	115.9E	102	16	0.41	0.066	0.025	0.16
	Chile	Santiago	33.5S	70.7W	36	9	0.22	0.048	0.024	0.22
	S. Africa	Stellenbosch	33.9S	18.8E	70	12	0.26	0.037	0.022	0.14
	Australia	Sydney	34.8S	150.7E	144	20	0.47	0.061	0.023	0.13
	Argentina	Buenos Aires	34.8S	58.5W	99	18	0.49	0.093	0.027	0.19
	Australia	Adelaide	35.2S	138.7E	88	14	0.33	0.086	0.024	0.26
	New Zealand	N. Auckland	35.8S	174.2E	181	16	0.43	0.096	0.027	0.22
	Australia	Melbourne	38.2S	145.9E	103	21	0.55	0.078	0.026	0.14
40 - 50 S	Chile	Puerto Montt	41.5S	73.0W	260	7.3	0.20	0.062	0.027	0.31
	Australia	Hobart	41.5S	147.1E	83	12.	0.26	0.051	0.021	0.19
	New Zealand	Greymouth	42.5S	171.2E	250	24.	0.67	0.142	0.028	0.21
	New Zealand	S. Canterbury	44.4S	171.2E	57	12	0.28	0.055	0.023	0.19
50 - 60 S	Chile	Punta Arenas	53.2S	70.9W	22	7				

Pu-238 to Pu-239 ratio should not vary greatly. A ratio of 0.03 is considered characteristic of weapons test debris[28,32]. The hemispheric differences found from the soil analyses are due to the unique geographical distribution of the SNAP debris.

There are not enough sites to warrant plotting these cumulative deposition data on a constant area world map and drawing isolines as was done for previous Sr-90 inventories[22]. Instead, we grouped the sites into ten degree latitude bands and averaged the values. We plotted these averages against the sine of the latitude in order to make straight line extrapolations to zero at the poles. The weapons Pu-238 contribution was obtained by multiplying the Pu-239 values by 0.024. This factor is the average weapons Pu-238 to Pu-239 ratio found for six soils collected prior to fallout from the SNAP-9A. Table 4 shows the data for these pre-SNAP 9A soils. This average ratio has a standard deviation of ± 12 percent. The SNAP-9A Pu-238 is simply the difference between the measured Pu-238 and the weapons Pu-238. The average activities per km^2 in each 10 degree latitude band are given in Table 5.

Results for one soil sample, reported in Table 3, were not included in the averaging. The site at Papaikou, Hawaii is in an exceptionally high precipitation region (400 cm per year). The measured deposition is reasonable for this amount of rainfall but the site is highly non-representative of the latitude band.

The error term associated with the average for each ten degree latitude band is simply the standard deviation when two or more sites are located in the same band. In those cases where only one site is represented or the value was derived by extrapolation, an average error for other bands in the hemisphere was applied.

The distribution patterns for Sr-90, Pu-239, and weapons Pu-238 are very similar showing heaviest deposition in the Northern Hemisphere temperate latitudes and minima in the polar and equatorial regions. There is a slight rise in the Southern Hemisphere temperature zone which, at its peak, is about one-fifth of the Northern Hemisphere maximum.

Table 4

Pre-SNAP-9A Soils

Country	Site	Lat°	Long°	Date Sampled	mCi per km²* Sr^{90}	Pu^{239}	Pu^{238}	Pu^{239}/Sr^{90}	Pu^{238}/Pu^{239}
U. S. A.	Barrow, Alaska	71.3N	156.8W	Aug 1964	13	0.33	0.0082	0.025	0.025
Norway	Bergen	60.4N	5.3E	Jul 1964	74	2.8	0.073	0.038	0.026
U. S. A.	New York, N.Y.	40.8N	73.9W	Dec 1964	58	1.8	0.047	0.031	0.026
Panama	Canal Zone	9.0N	79.6W	Nov 1963	13	0.31	0.0083	0.024	0.027
Kenya	Muguga	1.2S	36.6E	Nov 1963	13	0.32	0.0074	0.025	0.023
Argentina	Buenos Aires	34.8S	58.5W	Dec 1963	7.8	0.24	0.0046	0.031	0.019

*as of Aug. 1971

Table 5

Average Latitudinal Distributions of Cumulative Sr^{90}, Pu^{239}, and Pu^{238} Fallout

Hemisphere	Lat. band	Sr-90	Pu-239	Pu^{238} weapons	Pu^{238} SNAP-9A
		mCi per km^2		μCi per km^2	
Northern	90 - 80	(3±1)	(0.10±0.04)	(2.5±1.0)	(0.1±0.06)
	80 - 70	14± 6	0.36±0.05	8.7±1.2	0.4±0.2
	70 - 60	53±28	1.6 ±1.0	38. ± 25.	26 ± 15
	60 - 50	49±7	1.3 ±0.2	31 ± 4	13 ± 4
	50 - 40	76±14	2.1 ±0.5	52 ± 11	26 ± 12
	40 - 30	64±22	1.8 ±0.6	42 ± 14	25 ± 15
	30 - 20	35±6	0.96±0.07	23 ± 2	11 ± 4
	20 - 10	8±4	0.24±0.10	5.8±2.3	3.2±1.8
	10 - 0	4±2	0.13±0.06	3.1±1.2	0
Southern	0 - 10	12±9	0.30±0.20	7.1±4.6	9.9±6.8
	10 - 20	6±2	0.18±0.05	4.2±1.3	36 ± 21
	20 - 30	13±8	0.35±0.14	8.4±3.2	57 ± 27
	30 - 40	16±4	0.40±0.12	9.4±2.7	61 ± 20
	40 - 50	14±7	0.35±0.21	8.4±5.1	69 ± 38
	50 - 60	7±3	(0.20±0.09)	(5.0±2.2)	(44 ± 23)
	60 - 70	(3±1)	(0.10±0.04)	(2.3±1.0)	(22 ± 12)
	70 - 80	(1±0.5)	(0.03±0.01)	(0.9±0.4)	(8 ± 5)
	80 - 90	(0.5±0.2)	(0.01±0.004)	(0.2±0.1)	(4 ± 2)

NOTES: results in parentheses were derived by extrapolation
error terms are standard deviations

The SNAP-9A Pu-238 has an entirely different distribution pattern. Most of the SNAP debris has deposited in the Southern Hemisphere where the maximum fallout is $2\frac{1}{2}$ times that in the Northern Hemisphere. As in the case of weapons debris there is a low in the equatorial region and decreasing deposition toward the poles.

ISOTOPE RATIOS

Table 6 shows the ratios of Pu-239 to Sr-90 and Pu-238 to Pu-239 found in the soil samples averaged by 10 degree latitude bands. The Pu-239 to Sr-90 ratio shows no significant hemispheric difference and averages on a global basis 0.028 ± 15%. The relatively small standard deviation of the mean indicates that in most cases the soils sampled included the total Sr-90 that had fallen out. The Pu-238 to Pu-239 ratio for the Northern Hemisphere averaged 0.036 ± 17% and for the Southern Hemisphere from 10° - 50°S, 0.20 ± 25%. Again the standard deviations from the means are reasonably small indicating little fractionation within hemispheres. In the equatorial region of the Southern Hemisphere the Pu-238 to Pu-239 ratio is much lower probably reflecting interhemispheric mixing.

GLOBAL INVENTORY

The average and extrapolated activities per km^2 for each ten degree latitude band were multiplied by the area of the latitude band to calculate the total deposit in kilocuries. These values were then summed to obtain the inventories in Table 7. The errors were propagated by conventional means. For example, the square of the individual errors in each ten degree latitude band were summed and the square root taken to give the error of the hemispheric average.

Table 6

Isotope Ratios from Soil Analyses

Hemisphere	Lat. Band	No. of Sites	Pu^{239}/Sr^{90}	No. of Sites	Pu^{238}/Pu^{239}
Northern	90 - 80	0	-	0	-
	80 - 70	2	0.030±0.011	1	0.025
	70 - 60	4	0.029±0.004	4	0.042±0.007
	60 - 50	4	0.027±0.002	4	0.035±0.004
	50 - 40	8	0.028±0.003	8	0.036±0.005
	40 - 30	8	0.028±0.003	8	0.037±0.006
	30 - 20	3	0.028±0.003	3	0.036±0.005
	20 - 10	2	0.031±0.001	2	0.035±0.005
	10 - 0	1	0.032	0	-
Avg.	80 - 0	32	0.028±0.003		
Avg.	80 - 10			30	0.036±0.006
Southern	0 - 10	4	0.026±0.007	4	0.061±0.021
	10 - 20	6	0.028±0.005	6	0.22 ±0.06
	20 - 30	6	0.030±0.007	6	0.18 ±0.04
	30 - 40	8	0.025±0.002	8	0.18 ±0.05
	40 - 50	4	0.025±0.003	4	0.22 ±0.06
	50 - 60	0	-	0	-
	60 - 70	0	-	0	-
	70 - 80	0	-	0	-
	80 - 90	0	-	-	-
Avg.	0 - 50	28	0.027±0.005		
Avg.	10 - 50			24	0.20 ±0.05

Table 7

INVENTORY SUMMARY

kilocuries deposited

	Sr-90	Pu-239	Pu-238 Weapons	Pu-238 SNAP-9A
Northern Hemisphere	9,000±1,100	253±33	6.1±0.8	3.1±0.8
Southern Hemisphere	2,600±600	67±14	1.6±0.3	10.3±2.1
Global Deposit	11,600±1,250	320±36	7.7±0.9	13.4±2.2

error terms are standard deviation

We estimate that 11,600 ± 1,250 kCi of Sr-90 has accumulated on the earth's surface from weapons tests. Within the limits of error, this is in agreement with the cumulative deposits derived from the 1965-67 soil survey[33], the HASL monthly fallout network[34] and the fallout sampling program operated by the United Kingdom Atomic Energy Authority[35]. The total Pu-239 deposit is 320 ± 36 kCi and the weapons Pu-238, 7.7 ± 0.9 kCi. Our estimate of the SNAP-9A Pu-238 deposit, 13.4 ± 2.2 kCi, is in reasonable agreement with the 16 kCi we would expect to have fallen out based on the stratospheric inventory.

CONCLUSIONS

A global inventory and distribution pattern of Pu-238 fallout from the SNAP-9A satellite were derived from analyses of soils collected at over 60 sites throughout the world in 1970-71. As a result of this unique injection in the high stratosphere of the Southern Hemisphere, the SNAP-9A debris has a different distribution pattern than Pu-238 and fission products from nuclear weapons tests. Close to 75 percent of the total SNAP Pu-238 deposit is in the Southern Hemisphere whereas only about 20 percent of the total weapons Pu-238 fallout occurred in that hemisphere. In the Northern Hemisphere, 34 percent of the total Pu-238 fallout is attributed to SNAP-9A while in the Southern Hemisphere, 86 percent is from the generator. This accidental release of Pu-238 which occurred in April 1964, almost tripled the global deposit of this plutonium isotope by 1970. We can account for 13.4 ± 2.2 kilocuries deposited as compared to 17 kilocuries of Pu-238 which comprised the generator prior to burn-up. Since stratospheric inventories indicate slightly less than 1 kCi still in the atmosphere at the end of 1970, the fallout inventory derived from soil analyses, within the limits of error, provides a material balance of the SNAP-9A debris.

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It was only through the cooperation of scientists in other countries that we were able to obtain soil samples from so many regions of the world. Many other people, both in the United States and South America made it possible for us to find suitable sites and assisted us in the sampling. The appendix lists the names and affiliations of those to whom we are indebted for their cooperation.

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A P P E N D I X

Table A

Pu-239 and Sr-90 Below 30 cm

Site	depth (cm)	mCi per km^2 Pu-239	mCi per km^2 Sr-90	Percent of total below 30 cm Pu-239	Percent of total below 30 cm Sr-90
Brookhaven, N.Y.	0-30	2.1 ± 2%	71 ± 5%		
	30-60	0.01 ± 100%	1 ± 100%	0.5	1.4
Denver, Colorado	0-30	1.8 ± 2%	62 ± 6%		
	30-46	0.050 ± 13%	7 ± 33%	2.8	10.
Raleigh, N. C.	0-30	2.4 ± 3%	91 ± 2%		
	30-41	0.12 ± 15%	5 ± 25%	4.8	5.2
Vermillion, S. D.	0-30	2.3 ± 7%	86 ± 2%		
	30-41	0.063 ± 18%	2 ± 33%	2.7	2.3
Weslaco, Texas	0-30	0.85 ± 2%	20 ± 11%		
	30-41	0.039 ± 38%	8 ± 10%	4.4	28.

Table B

Comparison of Methods for Pu in Soil

(BROOKHAVEN SOIL, 1970)

(100 g aliquots)

Method	Lab	fCi Pu^{239} per gram
HNO_3-HCl leach	HASL	18.6 ± 5%
	HASL	17.7 ± 6%
	IPA	22.1 ± 3%
	IPA	18.9 ± 3%
	IPA	19.1 ± 5%
Δ to 500°C then HNO_3-HCl leach	HASL	19.2 ± 5%
	HASL	18.2 ± 5%
HNO_3-HCl - HF leach	IPA	18.4 ± 4%
Complete dissolution	IPA	18.5 ± 5%
	IPA	18.9 ± 5%

Table C

Pu in Blank Soil
(soil collected before 1945)

(1000 gram aliquots)

Lab	femtocuries per gram (fCi/g) Pu-239	Pu-238
IPA	0.009 ± 100%	0.004 ± 100%
IPA	0.20 ± 7%	0.001 ± 100%
IPA	0.003 ± 100%	0.003 ± 100%
TLW	0.009 ± 100%	0.01 ± 100%
TLW	0.02 ± 100%	0.04 ± 100%

Sr^{90} in Blank Soil
(soil collected before 1945)

Lab	aliq. wt. (g)	fCi per g
IPA	100	1.4 ± 100%
HASL	100	4.5 ± 100%
HASL	100	4.5 ± 100%
HASL	200	9.0 ± 100%

Table D

Plutonium in Reference Soils
(1000 gram blind aliquots)

Lab	femtocuries per gram (fCi/g) Pu-239	Pu-238
	BROOKHAVEN (1970)	
HASL	19.5 ± 0.5%	0.86 ± 5%
"	19.0 ± 0.5%	0.76 ± 6%
"	18.7 ± 0.5%	0.81 ± 6%
"	18.6 ± 1%	0.47 ± 7%
IPA	19.8 ± 2%	0.86 ± 4%
"	19.0 ± 2%	0.72 ± 4%
"	19.2 ± 3%	0.76 ± 5%
"	18.3 ± 2%	0.72 ± 6%
"	18.7 ± 3%	0.75 ± 5%
TLW	18.4 ± 3%	0.72 ± 7%
"	18.7 ± 2%	1.0 ± 5%
Avg.	18.9 ± 3%	0.77 ± 18%
	"BLACK SOIL" (1958)	
HASL	2.5 ± 10%	-
IPA	2.4 ± 2%	0.054 ± 15%
TLW	2.4 ± 5%	0.032 ± 88%
Avg.	2.4 ± 3%	0.043 ± 37%
	"RED SOIL" (1958)	
HASL	1.6 ± 10%	-
IPA	1.9 ± 2%	0.058 ± 15%
TLW	1.8 ± 6%	0.076 ± 36%
Avg.	1.8 ± 9%	0.067 ± 19%
	WOODCLIFF LAKE (1970)	
IPA	0.58 ± 4%	0.014 ± 50%
TLW	0.68 ± 16%	0.081 ± 99%
Avg.	0.63 ± 11%	-

Table E

Sr^{90} in Reference Soils
(HASL analyses)

Aliq. wt (g)	fCi/g
BROOKHAVEN (1970)	
100	504 ± 2%
200	468 ± 1%
200	459 ± 1%
100	423 ± 1%
100	423 ± 1%
100	473 ± 2%
100	473 ± 2%
100	514 ± 2%
Avg.	467 ± 7%
"BLACK SOIL" (1958)	
100	54 ± 8%
100	55 ± 8%
100	53 ± 8%
Avg.	54 ± 2%
"RED SOIL" (1958)	
100	40 ± 11%
100	50 ± 9%
Avg.	45 ± 16%

Table F

Pu Blind Duplicate Analyses of Soils Sampled in 1970
(Lab: IPA, 1000g aliq.)

Site	aliq.	fCi per g Pu-239	Pu-238
Argonne, Illinois	A	4.6	0.15
	B	4.4	0.14
Angra dos Reis, Brazil	A	1.6	0.29
	B	1.7	0.32
Roskilde, Denmark	A	2.9	0.10
	B	3.0	0.09
Ispra, Italy	A	11.7	0.39
	B	11.4	0.38
Tokyo, Japan	A	9.4	0.33
	B	9.2	0.33

Table G

Sr^{90} Blind Duplicate Analyses of Soils Sampled in 1970

Lab: HASL, 100g aliquots

Site	fCi per g aliq. A	aliq. B
Argonne, Illinois	162	158
Angra dos Reis, Brazil	76	68
Ispra, Italy	374	396
Tokyo, Japan	311	306
Brookhaven, New York	194	203
Fairbanks, Alaska	90	81
Palmer, Alaska	117	99
Burbank, California	58	58
Denver, Colorado	171	144
Papaikou, Hawaii	757	748
Manhattan, Kansas	234	225
Orono, Maine	167	149
Vermillion, S. D.	230	243
Puyallup, Washington	261	212
Bergen, Norway - 1	234	203
Bergen, Norway - 2	1099	1072
Oslo, Norway - 1	162	144
Oslo, Norway - 2	131	131
Muguga, Kenya	63	58
Samford, Australia	54	45
Warragul, Australia - 1	54	58
Warragul, Australia - 2	76	72

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(An asterisk, *, indicates that the soil was collected under the direction of the individual cited. Otherwise HASL personnel collected the sample with the cooperation of the person listed).

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