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*Technical Memorandum*

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STRATOSPHERIC AIR CONCENTRATIONS OF PLUTONIUM ISOTOPES  
AND STRONTIUM-90, OCTOBER 1964 - FEBRUARY 1965

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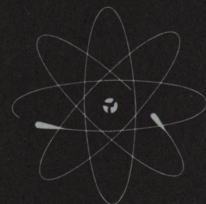
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Leonard P. Salter

June 11, 1965

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STRATOSPHERIC AIR CONCENTRATIONS OF PLUTONIUM ISOTOPES  
AND STRONTIUM-90, OCTOBER 1964 - FEBRUARY 1965

Leonard P. Salter

June 11, 1965

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United States Atomic Energy Commission  
New York, New York



Plutonium-238 from the SNAP-9A abort of April 1964 was first detected in high altitude balloon samples collected from 108,000 feet at 34°S in August 1964<sup>(1)</sup> and was found at 93,000 and 106,000 feet, 34°S, in September. This report presents the data on balloon and aircraft samples which were collected from October 1964 to February 1965 and analyzed under the supervision of the U. S. Atomic Energy Commission Health and Safety Laboratory (HASL), updating results reported previously<sup>(2)</sup>.

### Results

Tables I, II, and III present data for samples collected via balloon-borne filtering devices over Australia (34°S), Texas (31°N), Panama (9°N), and Alaska (65°N). The total gamma activity, Pu-238, Pu-239 + Pu-240<sup>a</sup> and Sr-90 concentrations are given, and the presence of SNAP-9A debris in the sample is deduced by the Pu-238/Pu-239+ ratio shown in the last column. The sampling equipment and the methods used to measure the altitudes, volumes, and total gamma activity for these samples have been described<sup>(2)</sup>.

Results of an extensive sampling of the lower stratosphere between 15°S and 47°S in early 1965 are given in Table IV. These samples were collected by WU-2 Aircraft Hatch Sampler<sup>(3)</sup> under the auspices of the U. S. Air Force Defense Atomic Support Agency. Two samples collected between 50°S to 60°S by RB-57F aircraft are included also in this Table.

Most of the radiochemical assays were performed by Isotopes, Inc. (II) under contract to the U. S. Atomic Energy Commission. A few samples were analyzed by Tracerlab, a Division of Laboratory for Electronics, Inc. (TLW),

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a. Pu-239 + Pu-240 is referred to as Pu-239+ throughout this report.



and others by HASL. Plutonium isotopes were assayed by alpha spectrometry and their recovery determined by the yield of a known amount of tracer Pu-236 added at the beginning of the chemical analysis. Many of the radiometric assays for plutonium in samples chemically analyzed at II were completed at HASL. Agreement on Pu-238 and Pu-239+ in plated plutonium samples counted at II and HASL indicates that the radiometric bias between laboratories is less than 5%.

The aircraft samples and a few of the balloon samples were split prior to radiochemical analysis. For these cases the radionuclide concentrations were calculated after correcting the total sample volume by the fraction of total gamma activity in the portion analyzed. This correction introduces an error of less than 5% in the concentrations.

Standard deviations due to counting (S.D.) are less than 10% except for data appearing with the following notation:

B: S.D. 10-20%  
C: S.D. 20-50%  
D: S.D. 50-100%

When the S.D. is greater than 100%, the data are reported as less than twice the S.D. ( $<2 \times \text{S.D.}$ ). Parentheses not otherwise explained by notes in the tables indicate data based on Pu-238 or Pu-239+ levels near the limit of detection. An asterisk (\*) after the Pu-238/Pu-239+ ratio denotes Pu-238 possibly attributable to SNAP-9A debris in the sample.

#### Quality Control

Included in Tables I and IV are results on 15 pairs of blind duplicate analyses which were performed on either two portions of the same sample or on two samples collected simultaneously. With the exception of the Pu-238 results on two samples in Table IV, which will be discussed later, these data suggest



that the reproducibility within laboratories and agreement between laboratories is satisfactory.

Additional analyses on samples spiked with known amounts of mixed fission products and standardized Pu-239+ are reported in Table V, and on blank samples in Table VI. These control samples were submitted and analyzed blind as regular stratospheric air filters. The spiked sample data show that accuracy is sufficient with only a slight bias of less than 5% relative to the standards. The blank data generally indicate a satisfactory level of sensitivity with the exception of the sporadically high Pu-239+ results reported by II. Since the samples range in volume from 500 SCF for collections above 100,000 feet to 5000 SCF for the fraction of aircraft samples analyzed, 1 dpm of Pu-239+ contamination would increase the apparent Pu-239+ concentration in the highest altitude samples by  $\sim 2 \text{ dpm}/10^3 \text{ SCF}$ , and in the lower stratosphere samples by  $\sim 0.2 \text{ dpm}/10^3 \text{ SCF}$ . It is quite likely, therefore, that the sporadic Pu-239+ contamination of some samples analyzed by II accounts for the results in Tables I - IV for which anomalously high Pu-239+/Sr-90 and Pu-239+/Total  $\gamma$  ratios were noted. These data have been appropriately marked.

### Discussion

The levels of SNAP-9A debris at the four balloon sites are summarized in Table VII. These data were obtained by applying to the observed Pu-238 concentration a small correction due to the Pu-238 background from nuclear weapons testing and normalizing to the total Pu-238 in the device. The correction for Pu-238 background was deduced from pre-August 1964 data<sup>(2)</sup>. A total of 17 kCi was used in converting to relative units. (The conversion factor is  $1 \text{ dpm Pu-238}/10^3 \text{ SCF} = 2.7 \times 10^{-18}$  parts of the device per  $10^3 \text{ SCF}$  of air.)

It is apparent from this distribution of SNAP-9A Pu-238 that burn-up into sub-micron particles did occur for a significant portion of the device. The observed Pu-238 levels are comparable to those predicted by Harley<sup>(4)</sup> and Machta<sup>(5)</sup>.

One quadrant from each of two samples collected in the lower stratosphere in early 1965 also indicated the possible presence of Pu-238 from SNAP-9A debris (Table IV, 15-23<sup>0</sup>S, 65,000 and 67,000 feet). The high Pu-238 results were not verified by assay of a second quadrant, which suggests that contamination during analysis may have occurred. However, a low concentration of larger particulates in the lower stratosphere could lead to the collection of a single or a few particles and also account for the irregular presence of Pu-238 in these samples. The diameter of a single spherical PuO<sub>2</sub> particle which would contain all the Pu-238 observed in each of these samples (~8 dpm) is ~0.34 μ.

#### References

1. Salter, L. P., "Measured Levels of Plutonium-238 and Plutonium-239 in the Stratosphere," USAEC Report No. HASL-149, p.143, October 1, 1964.
2. Salter, L. P., "High Altitude Balloon Sampling Program," USAEC Report No. HASL-158, p.214, April 1, 1965.
3. "The High Altitude Sampling Program," J. P. Friend, Editor, Defense Atomic Support Agency Report No. DASA-1300, Vol. I, August 31, 1961.
4. Harley, J. H., "Possible Pu-238 Distribution from a Satellite Failure," USAEC Report No. HASL-149, p.138, October 1, 1964.
5. Machta, L., "Calculations of Pu-238 Concentrations from the SNAP-9A Burn-up," USAEC Report No. HASL-155, p.297, January 1, 1965.



TABLE I

Pu-238, Pu-239+Pu-240, and Sr-90 Concentrations at 34°S, October 1964 - February 1965

Sampling Date	Altitude 10 <sup>3</sup> ft	Total $\gamma$ Activity cpm/10 <sup>3</sup> SCF	Lab <sup>a/</sup>	Activity Concentrations (dpm/10 <sup>3</sup> SCF)			Pu-238 <del>Pu-239+</del>
				Pu-238	Pu-239+	Sr-90	
Oct 28, 1964	68	208	II	0.11B	1.7	124	0.06B
Oct 27	82	176	II	(<0.11) <sup>b/</sup>	(1.7) <sup>b/</sup>	98	(<0.07) <sup>b/</sup>
Oct 19	92	137	II	5.8	0.8B	52	8B*
		144	½-II	7.3	(0.6C)	44	(13C)*
Oct 22	106	133	II	24	0.8B	30	30B*
		83	½-HASL	18	(0.2D)	In Process	(100D)*
Nov 20	62-69	199	II	(0.09C)	1.8	106	(0.05C)
Nov 17	84	128	II	(0.10B)	0.8	69	(0.13B)
Nov 10	94	121	II	4.3	0.5B	46	9B*
Nov 12	106	110	II	23	0.4B	26	52B*
Dec 16	67	209	II	(0.09B)	1.8	139	(0.05B)
Dec 10	79	149	II	0.14B	1.0	76	0.14B
Dec 9	87	119	II	16	0.7	61	24*
Dec 5	101	89	II	7.1	(0.3C)	35	(24C)*
Dec 15	102	78B	II	14	(0.8B) <sup>x/</sup>	19B	(19B) <sup>x/</sup> *
Jan 22, 1965	74	133	II	(0.07B)	1.0	93	(0.07B)
Jan 26	81	132	II	0.28	1.0	77	0.28*
Jan 19	88	126	II	4.3	1.0	59	4.3*
Jan 27	89	109	½-II	7.6	1.1B	51	7B*
			¼-TLW	7.2	0.7B	55	11B*
Jan 28	101	77B	II	20	(0.9) <sup>x/</sup>	28	(23) <sup>x/</sup> *
Feb 16	81	166	½-II	(0.14C)	1.8B	134	(0.08C)
Feb 4	82	137	II	(0.08C)	1.3	92	(0.06C)
Feb 9	88	87	II	2.6	0.5B	41	5B*
Feb 18	88	119	½-II	(0.25C)	1.1B	94	(0.22C)
Feb 11	102	(65) <sup>v/</sup>	II	(9.3) <sup>v/</sup>	(0.9B) <sup>vx/</sup>	(32) <sup>v/</sup>	(11B) <sup>x/</sup> *
Feb 25	104	86	½-II	15.6	(3.8) <sup>x/</sup>	19	(4) <sup>x/</sup> *

a. When less than whole sample was analyzed, portion of sample used is given.

b. Plutonium yield about 12%.

v. Concentration calculated from estimated volume data.

x. High Pu-239+/Sr-90 ratio and Pu-239+/Total  $\gamma$  ratio indicates possible Pu-239+ contamination of sample.

TABLE II

Pu-238, Pu-239+Pu-240, and Sr-90 Concentrations at 31°N, October 1964 - February 1965

Sampling Date	Altitude 10 <sup>3</sup> ft	Total $\gamma$ Activity cpm/10 <sup>3</sup> SCF	Lab	Activity Concentrations (dpm/10 <sup>3</sup> SCF)			Pu-238 Pu-239+
				Pu-238	Pu-239+	Sr-90	
Oct 14, 1964	65	870	II	0.25B	9.7	514	0.03B
Oct 20	79	369	II	0.23B	3.2	248	0.07B
Oct 15	91	256	II	0.18B	2.1	147	0.09B
Oct 6	102	147	II	(0.12B)	1.2	134	(0.10B)
Nov 6	66	666	II	0.24B	6.5	414	0.04B
Nov 20	79	264	II	(0.08C)	2.2	(137) <sup>a</sup>	(0.04C)
Nov 7	101	153	II	(0.11D)	1.1	79	(0.10D)
Nov 25	(105) <sup>b</sup>	(88B) <sup>v</sup>	II	(0.12C) <sup>v</sup>	(1.1) <sup>v</sup>	(34) <sup>v</sup>	(0.11C)
Nov 28 <sup>c</sup>	105	84	II	(0.27B)	0.5B	32	(0.55B)
Dec 14	65	595	II	0.20B	6.1	363	0.03B
Dec 15	79	248	II	(0.09C)	1.6	158	(0.06C)
Dec 12	80-88	(217) <sup>v</sup>	II	(0.12B) <sup>v</sup>	(1.7) <sup>v</sup>	(135) <sup>v</sup>	0.07B
Dec 7	91	210	II	(0.07C)	1.4	117	(0.05C)
Dec 16	105	(78B) <sup>v</sup>	II	(0.09C) <sup>v</sup>	(0.4) <sup>v</sup>	(22) <sup>v</sup>	(0.20C)
Jan 13, 1965	74	183	II	(0.15B)	2.0	131	(0.08B)
Jan 15	81	252	II	(0.15C)	2.3	127	(0.06C)
Jan 27	91	106	II	0.19B	0.8	32	0.22B
Jan 12	107	98B	II	3.3	(1.4) <sup>x</sup>	23	(2.3) <sup>x</sup> *
Jan 10 <sup>c</sup>	109	105	II	4.8	0.6B	35	8B *
Feb 14	79	188	II	(0.10C)	1.4	107	(0.07C)
Feb 22	89	98	II	0.81	0.7B	35	1.2B *
Feb 15	(104) <sup>b</sup>	(65) <sup>v</sup>	II	(3.7) <sup>v</sup>	(0.4C) <sup>v</sup>	(18) <sup>v</sup>	(8C) *

<sup>a</sup>. Sr-90 lost in analysis; Sr-90 dpm estimated as 0.67 x Cs-137 dpm.

<sup>b</sup>. Collection data suggests that sample may have been contaminated during descent by activity at lower altitudes.

<sup>c</sup>. Air ejector sample.

<sup>v</sup>. Concentration calculated from estimated volume data.

<sup>x</sup>. High Pu-239+/Sr-90 ratio and high Pu-239+/Total  $\gamma$  ratio indicates possible Pu-239+ contamination of sample.



TABLE III

Pu-238, Pu-239+Pu-240, and Sr-90 Concentrations at 9°N and 65°N, October 1964 - February 1965

<u>Latitude</u>	<u>Sampling Date</u>	<u>Altitude 10<sup>3</sup> ft</u>	<u>Total <math>\gamma</math> Activity cpm/10<sup>3</sup> SCF</u>	<u>Lab</u>	<u>Activity Concentrations (dpm/10<sup>3</sup> SCF)</u>			<u>Pu-238</u>
					<u>Pu-238</u>	<u>Pu-239+</u>	<u>Sr-90</u>	<u>Pu-239+</u>
9°N	Oct 14, 1964	78	782	II	0.23	7.2	(500) <sup>a</sup>	0.03
	Oct 10	(90) <sup>b</sup>	613	II	0.25B	5.1	355	0.05B
	Oct 12	102	182	II	(0.09B)	1.4	126	(0.06B)
9°N	Jan 15, 1965	84	601	II	0.26B	6.4	379	0.04B
	Jan 12	93	359	II	(0.11B)	3.2	264	(0.03B)
	Jan 17	(105) <sup>c</sup>	(235) <sup>v</sup>	II	(0.61) <sup>v</sup>	(2.3) <sup>v</sup>	(138) <sup>v</sup>	0.27*
	Feb 23	79	573	II	0.22B	6.7	436	0.03B
	Feb 21	88	250	II	(0.13C)	2.5	253	(0.05C)
	Feb 15	103	110	II	0.76B	1.1	58	7B*
65°N	Nov 5, 1964	77	288	II	0.14B	1.9	154	0.07B
	Nov 8	88	125	II	(0.05C)	0.7	46	(0.07C)

a. Sr-90 lost in analysis; Sr-90 dpm estimated as 0.67 x Cs-137 dpm.

b. 85% of sample collected at 90,000 feet, 15% collected between 84,000 and 90,000 feet.

c. Collection data suggests that sample may have been contaminated during descent by activity at lower altitudes.

v. Concentration calculated from estimated volume data.

TABLE IV

Pu-238, Pu-239+Pu-240, and Sr-90 Concentrations in Fractions of Aircraft Samples  
 Collected from the Southern Hemisphere, January 28 and February 2, 1965

Latitude, Sampling Date, 1965	Altitude 10 <sup>3</sup> ft	Total $\gamma$ Activity cpm/10 <sup>3</sup> SCF	Lab	Activity Concentrations (dpm/10 <sup>3</sup> SCF)			Pu-238 Pu-239+	
				Pu-238	Pu-239+	Sr-90		
15-23°S Feb 2	55	29	II	(0.02C)	(1.2) <sup>X/</sup>	25	(0.01C) <sup>X/</sup>	
			TLW	(0.01C)	0.3	21	(0.04C)	
	60 <sup>a/</sup>	132	II	(0.03C)	2.0	139	(0.02C)	
	65 <sup>a/</sup>	173	II	1.8	2.2	161	0.83*	
			II	(0.07C)	2.8	In Process	(0.02C)	
67 <sup>a/</sup>	178	II HASL	(0.06C) 1.4	2.4 2.5	190 In Process	(0.03C) 0.56*		
23-30°S Feb 2	55	85	II	(0.03C)	1.0	67	(0.03C)	
			TLW	0.03B	0.8	65	0.04B	
	60	157	II	0.06B	1.9	142	0.03B	
	65 <sup>a/</sup>	171	II	(0.06C)	2.1	172	(0.03C)	
69 <sup>a/</sup>	220	II	0.11B	2.9	223	0.04B		
30-37°S Feb 2	55	97	II	(0.04C)	1.2	78	(0.03C)	
			II	(0.04C)	1.5	88	(0.03C)	
	60	153	II	(0.04C)	1.3	145	(0.03C)	
			II	(0.06C)	1.5	115	(0.04C)	
65	207	II	(0.07C)	2.2	172	(0.03C)		
		II	(0.08C)	2.3	196	(0.03C)		
38-41°S Jan 28	55	145	TLW	0.07B	1.4	116	0.05B	
			TLW	0.06B	1.6	115	0.04B	
	60	193	II	(0.06C)	2.0	122	(0.03C)	
	65	196	II	(0.09C)	2.2	107	(0.04C)	
67 <sup>a/</sup>	181	II	(0.07C)	2.3	126	(0.03C)		
41-44°S Jan 28	55	166	II	(0.05C)	1.9	114	(0.03C)	
			TLW	(0.03C) <sup>b/</sup>	(1.8) <sup>b/</sup>	134	(0.02C)	
	60	190	II	(0.03C)	1.8	122	(0.02C)	
	65	191	TLW	(0.07C)	1.4	120	(0.05C)	
TLW			(0.08C)	1.4	112	(0.06C)		
67 <sup>a/</sup>	170	II	(0.05C)	2.0	110	(0.02C)		
44-47°S Jan 28	55	166	II	(0.05C)	1.8	148	(0.03C)	
			II	(0.05C)	2.0	129	(0.02C)	
	65	187	II	(0.08C)	1.4	115	(0.06C)	
	70	173	II HASL	<0.04 (0.06C)	1.4B 1.2	82 In Process	<0.04 (0.05C)	
50-55°S Jan 23	Unknown		II	Pu-239+ Sr-90 : 0.10		0.04		
55-60°S Jan 23	Unknown		II	Pu-239+ Sr-90 : 0.10		0.04		

a. Assay of additional fraction in process.

b. Plutonium yields estimated from average yield for other plutonium assays.

c. High Pu-239+/Sr-90 ratio and high Pu-239+/Total  $\gamma$  ratio indicates possible Pu-239+ contamination of sample.

TABLE V

Analysis of Spiked Filter Paper Samples

<u>HASL No.</u>	<u>Lab</u>	<u>Pu-239+</u>			<u>Sr-90</u>		
		<u>Added (dpm)</u>	<u>Found (dpm)</u>	<u>Found / Added</u>	<u>Added (dpm)</u>	<u>Found (dpm)</u>	<u>Found / Added</u>
A1556	II	6.9	7.7	1.12	65	60	0.92
A1558	II	21.0	22.6	1.08	288	283	0.98
A1561	II	8.7	8.8	1.01	199	187	0.94
A1555	TLW	5.5	6.0	1.09	73	71	0.97
A1557	TLW	30.0	31.2	1.04	259	255	0.98
A1559	HASL	8.7	8.8	1.01	In Process		

TABLE VI

Analysis of Blank Filter Paper Samples

<u>HASL No.</u>	<u>Lab</u>	<u>Pu-238 (dpm)</u>	<u>Pu-239+ (dpm)</u>	<u>Sr-90 (dpm)</u>
A1276	II	0.08C	0.8	1.1C
A1318	II	0.09B	0.12B	1.4C
A1344	II	<0.03	<0.04	5.8B
A1385	II	0.04D	0.06C	5.0C
A1427	II	0.04D	0.03D	2.4C
A1450	II	<0.04	0.16B	0.7C
A1453	II	0.02D	0.52	1.8C
A1467	II	0.07C	0.08C	1.5C
A1469	II	0.04D	0.20B	In Process
A1525	II	0.05D	0.25B	<1.2
A1529	II	0.06C	1.6	<1.1
A1592	II	0.11B	1.9	<1.1
A1526	TLW	0.05D	<0.10	<1.0
A1527	TLW	<0.10	<0.06	<1.0
A0742	HASL	<0.02	<0.02	In Process
A1448	HASL	<0.03	<0.03	In Process



TABLE VII

Relative SNAP-9A Pu-238 Concentrations  
 (10<sup>-18</sup> parts of device per 10<sup>3</sup> SCF)

ALT (10 <sup>3</sup> ft)		1964								1965		ALT (km)
		May	June	July	Aug	Sept	Oct	Nov	Dec	Jan	Feb	
65°N	110			<3								35
	100		<3		<3							30
	90	<3	<3	<3	<3			<3				25
	80	<3	<3	<3	<3			<3				20
	70											
	60											
31°N	110	<3	<3	<3	<3			<3, <10	<3	130 90	90	35
	100						<3	<3				30
	90	<3	<3	<3	<3	<3	<3		<3	4	20	25
	80	<3	<3	<3	<3	<3	<3	<3	<3	<3 <3	<3	20
	70	<3	<3	<3	<3		<3	<3	<3			
	60											
9°N	110									15	20	35
	100						<3			<3		30
	90						<3			<3	<3	25
	80						<3				<3	20
	70											
	60											
34°S	110	<3	<3	<3	30	300	600*	600			400	35
	100					15		120	200,400	500	250	30
	90	<3	<3	<3	<3		160*			120,200*	70, <10	25
	80	<3	<3	<3	<3	<3	<3	<4	400		<3*	20
	70					<3	<3			5 <3	<3*	
	60	<3	<3	<3	<3			<3	<3		<3*	

\* Average of two analyses.





