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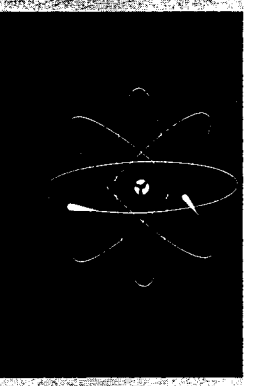
health and safety laboratory



FALLOUT PROGRAM
QUARTERLY SUMMARY REPORT

January 1, 1968

UNITED STATES ATOMIC ENERGY COMMISSION
NEW YORK, N. Y. 10014

DISCLAIMER

FALLOUT OF Pu-238 FROM THE SNAP-9A BURNUP-III

by Herbert L. Volchok, (HASL)

Plutonium-238 was released by the disintegration of a SNAP-9A power source upon re-entry into the atmosphere in April of 1964. It was estimated (1) that the re-entry took place at an altitude of about 46 kilometers, over the Indian Ocean. Krey (2) by integrating the concentrations of SNAP debris from samples obtained by balloon and aircraft sampling, was able to account for 15 kilocuries (kCi) in the stratosphere in the early part of 1966. Eighty percent of this total was found to be in the Southern Hemisphere, at that time.

In order to document the deposition of material from the SNAP-9A burn-up, the Health and Safety Laboratory (HASL) initiated a sampling and analysis program in 1966. Large area collections (about one square meter) of fallout are made each month at Melbourne, Australia and New York City. Brief descriptions of the samplers, analytical procedures and preliminary data were reported earlier, (3, 4).

Data from both sites through July of 1967 are now completed. At Melbourne, sampling was not started until May of 1966, in New York all of 1966 was sampled. Tables 1 and 2 summarize the results of the plutonium analyses and also lists the Pu-238 to Pu-239,240 ratios for each month. The plutonium isotope ratio is useful for indicating the presence or absence of SNAP-9A debris. Measurements of air samples obtained prior to the re-entry of the SNAP showed a fairly constant Pu-238/Pu-239 ratio averaging about 0.03 (5). Thus any sample with a ratio in excess of that may be presumed to contain Pu-238 from the SNAP-9A and the amount may be determined by subtracting out the indicated "background" amount. Tables 1 and 2 also list the amount of Pu-238 calculated by that method. Note that in Table 1, values for the fallout at Melbourne, Australia were estimated for the first four months of 1966, by first computing the

Pu-239 assuming that this is 1½ percent of the Sr-90 in all cases. Sr-90 has been measured at Melbourne as part of another program (6). Then by extrapolating the smoothly increasing ratio Pu-238/Pu-239 to 0.03, (the pre-SNAP value) the numbers listed in the Table were obtained.

Figure 1 shows the time variation of the Pu-238/Pu-239 for both sites. The Melbourne ratio was assumed to have initially exceeded the 0.03 background level at the beginning of 1966, since little or no Pu-238 from the SNAP-9A could be definitely identified in the surface air of the Southern Hemisphere before early 1966 (7). Thus the ratio at Melbourne must have risen rapidly after the first appearance of the debris, in contrast to the situation in New York where the increase was rather gradual until the spring of 1966. It was somewhat surprising to see the Southern Hemisphere ratio decrease from March through July of 1967. Presumably this is an indication of either large scale movements of air, or a seasonal pattern of diffusion in the lower stratosphere. As the Southern Hemisphere spring advances, this trend may reverse with the ratios at Melbourne increasing again.

Figure 2 shows the actual monthly deposition of the SNAP-9A Pu-238 at the two sites. It is interesting to note that even in the first year of deposition the seasonal effect was quite marked in the Southern Hemisphere in 1966 and recognizable in New York in 1967. Perhaps even more unexpected, the New York fallout of Pu-238 attributable to the SNAP-9A was greater than that at Melbourne for three out of the four months; April through July. In the subsequent months of 1967, advancing into the Southern Hemisphere spring, it is anticipated that the Melbourne concentrations will sharply increase to even higher levels than in 1966.

As in the prior reports (3, 4) a computation of the global deposit of the SNAP-9A debris has been made. For this calculation it is assumed that the total SNAP-9A fallout in each hemisphere is directly proportional to that at the single sampling site in that hemisphere, and further that these relationships are identical to those observed for Sr-90 fallout. For the Northern Hemisphere, nine years of Sr-90 data gives a value of 123 ± 10 kilocuries (kCi) deposited in the Northern Hemisphere for each millicurie per square kilometer (mCi/km^2) deposited in New York City. Eight years of measurements at Melbourne, Australia results in a value of 180 ± 15 for the Southern Hemisphere.

Table 3 summarizes the hemispheric and worldwide deposits of the Pu-238 from SNAP-9A by months, through July of 1967. From these data it can be seen that in 1966 a little more than 1.3 kCi was deposited on the earth's surface while almost as much came down in the first seven months of 1967. The total through July of 1967, 2.55 kCi, represents some 17% of the 15 kCi accountable in the stratosphere by Krey in early 1966 (2).

The deposition rate is seen to have increased substantially in 1967 and based upon the last five months a stratospheric half residence time of a little over three years was calculated. Presumably this value will decrease as the distribution of the SNAP-9A material in the stratosphere approaches that which currently exists for the nuclear weapons debris.

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Table 1

Plutonium in Melbourne, Australia Fallout[1]
(values in 10^{-3} mCi/km^2)

	Pu-239	Pu-239	Pu-238 / Pu-239	SNAP-9A Pu-238
Jan. 1966	1.35B	.06B	0.04B	0.01
Feb. "	1.05B	.08B	0.08B	0.04
Mar. "	1.80B	.22B	0.12B	0.16
Apr. "	1.20B	.24B	0.20B	0.16
May "	0.14	.05	0.36	0.04
Jun "	0.5B	.32	0.55	0.30
July "	1.35B	.96B	0.71B	0.91
Aug. "	0.49	.41B	0.84	0.39
Sept. "	0.51	.50	0.98	0.48
Oct. "	2.83	2.81	0.99	2.70
Nov. "	0.86	1.03	1.20	1.00
Dec. ")) Jan. 1967)	1.92	2.91	1.52	2.85
Feb. "	0.19A	.29	1.52	0.28
Mar. "	0.11A	.31	2.82	0.31
Apr. "	0.18A	.47	2.61	0.46
May "	0.21A	.48	2.29	0.47
June "	0.22A	.42	1.91	0.41
July "	0.50	.73	1.46	0.71

1 - Errors are less than ± 20% (1 sigma) except for "A" which indicate 20-50%.

B - Values were derived by extrapolation and from Sr^{90} levels.

Table 2

Plutonium in New York City Fallout[1]
(values in 10^{-3} mCi/km^2)

	Pu-239	Pu-238	Pu-238 / Pu-239	SNAP-9A Pu-238
Dec. 1965	3.23	0.09	0.03	0
Jan. 1966	3.47	0.12	0.04	0.02
Feb. "	3.18	0.30	0.09	0.20
Mar. "	2.02	0.12	0.06	0.06
Apr. "	4.59	0.18	0.04	0.04
May "	4.45	0.57	0.13	0.44
June "	2.23	0.17	0.08	0.10
July "	2.49	0.30	0.12	0.22
Aug. "	2.21	0.27	0.12	0.20
Sept. "	1.46	0.21	0.14	0.17
Oct. "	1.75	0.17	0.10	0.12
Nov. "	0.82	0.15	0.18	0.12
Dec. "	1.47	0.27	0.18	0.23
Jan. 1967	1.66	0.14	0.08	0.09
Feb. "	1.91	0.20	0.11	0.14
Mar. "	1.98	0.34	0.17	0.28
Apr. "	5.18	1.53	0.30	1.37
May "	3.06	0.91	0.30	0.82
June "	1.29	0.57	0.44	0.53
July "	2.57	2.44	0.95	2.36

1 - Errors are less than ±20% (1 sigma) for all data.

Table 3

Hemispheric and Worldwide SNAP-9A Pu-238 Deposition
(values in kilocuries)

	Northern Hemisphere	Southern Hemisphere	Worldwide
Jan. 1966	.002	.002	.004
Feb. "	.023	.008	.031
Mar. "	.007	.028	.035
Apr. "	.005	.028	.033
May "	.050	.008	.050
June "	.011	.053	.064
July "	.026	.165	.181
Aug. "	.022	.071	.093
Sept. "	.019	.086	.105
Oct. "	.014	.486	.100
Nov. "	.015	.179	.194
Dec. "	.026	.395*	.421
Total 1966	.220	1.509	1.319
Jan. 1967	.010	.118*	.120
Feb. "	.016	.050	.066
Mar. "	.032	.056	.088
Apr. "	.157	.083	.240
May "	.093	.085	.178
June "	.060	.074	.134
July "	.270	.128	.398
Sub Total	.638	.594	1.232
Total through July 1967	.858	2.103	2.551

*Derived from the December 1966 - January 1967 combined sample by the Sr-90 proportion.

Figure 1

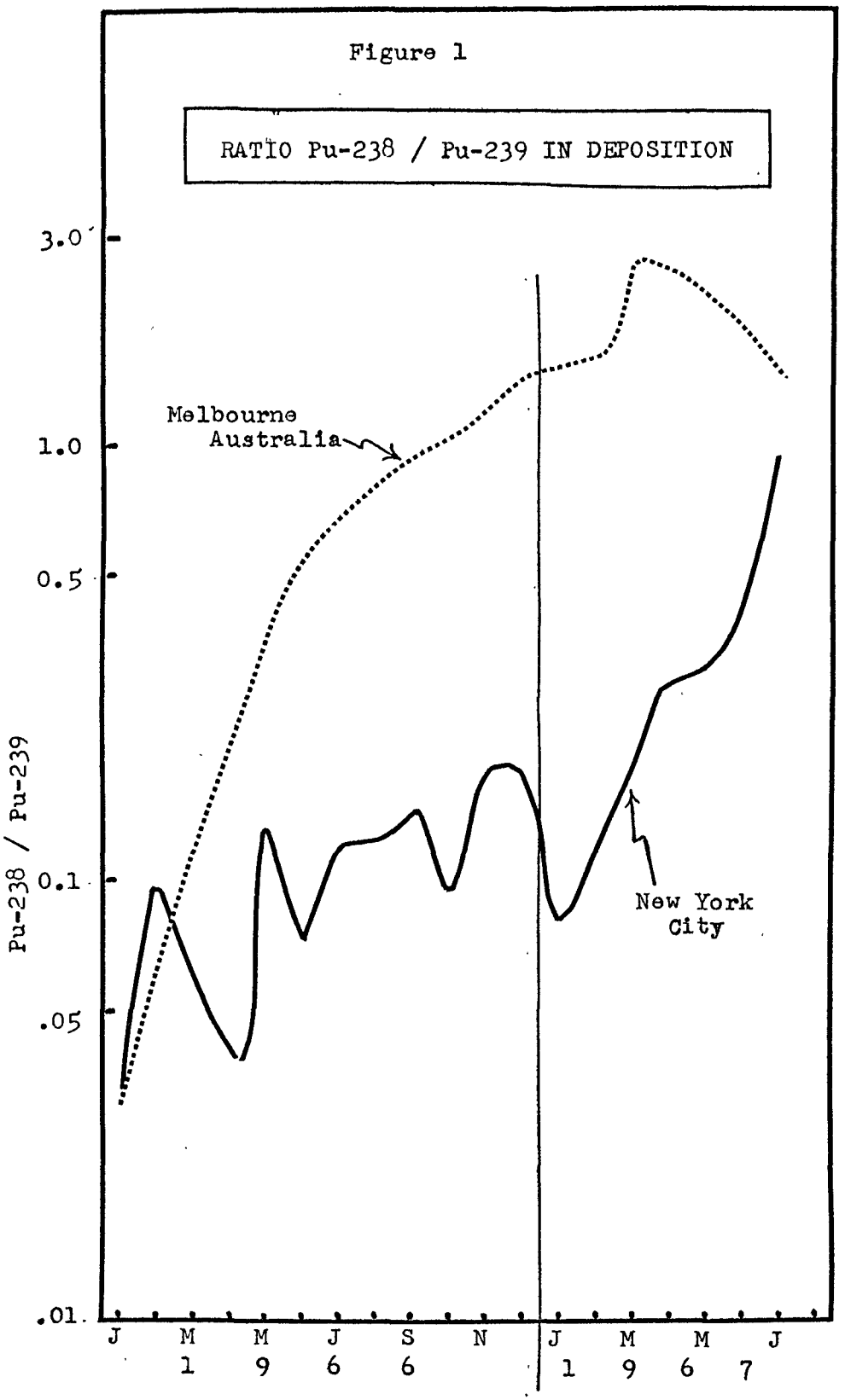
RATIO Pu-238 / Pu-239 IN DEPOSITION
3.0
1.0
0.5
0.1
.05
.01
Pu-238 / Pu-239
Melbourne
Australia
New York
City
J
M
M
J
S
N
J
M
M
J
1
9
6
6
1
9
6
7

Figure 2

MONTHLY DEPOSITION OF Pu-238 FROM SNAP-9A