BNWL-SA-5189

Conf-741086--7

TRANSURANIUM ELEMENTS IN THE ATMOSPHERE

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Work performed by Battelle, Pacific Northwest Laboratories, Richland, Washington for the U. S. Atomic Energy Commission under Contract AT(45-1)-1830.

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INTRODUCTION

The first significant injection of transuranium elements into the atmosphere was the result of the nuclear weapons test in Alamogordo, New Mexico on July 6, 1945. Between then and 1952 further nuclear detonations resulted in additional injections to the atmosphere, however most of this debris was confined to the troposphere. In 1952 the first thermonuclear device was tested resulting in a much higher energy yield and the injection of major amounts of debris into the stratosphere. The much higher yield of the fusion devices is accompanied by a much higher generation of neutrons. This in turn greatly increases the production of the transuranium elements. The majority of the transuranium elements and other nuclear debris which has been injected into the atmosphere was produced during the 1961-1962 U.S. and USSR nuclear weapons testing programs. Most of the studies of transuranium elements from nuclear weapons testing have been concerned with the measurement of Pu isotopes. However, the complete decay of the short-lived ^{241}Pu (15 yrs) will result in a greater residue of ^{241}Am activity than of ^{239}Pu. The operation of nuclear reactors also results in the production of transuranium elements and there exists the potential of release of some of these to the atmosphere. The modern nuclear power plants which are designed for the generation of electrical energy employ very

long fuel exposure periods. This results in successive neutron capture of the transuranium elements and production of very substantial quantities of higher mass elements. It has been estimated that approximately 10^9 curies of transuranium elements will be produced as a result of energy generation by the year 2000. Whereas Pu and Am isotopes are the main transuranium activities from nuclear weapons testing, ^{241}Am and the curium isotopes will be the principal activities resulting from nuclear power generation. A major injection of ^{238}Pu into the atmosphere occurred in April of 1964 when a satellite failed to achieve a stable orbit and disintegrated in re-entry into the atmosphere. The 17 kCi which was added to the atmosphere was relatively small compared with the 350 kilocuries of $^{239-240}$Pu which has been added by nuclear weapons testing. However, its point source injection has been useful in developing models describing global atmospheric mixing. Accidental releases of transuranium elements to the atmosphere have occurred both from nuclear plant operation and from the transport of nuclear weapons. The total amounts released to the atmosphere by these processes have been relatively minor, however such accidents can have rather serious local effects.

TRANSURANIUM ELEMENTS FROM NUCLEAR EXPLOSIONS

Prior to the detonation of the first thermonuclear device in 1952, atmospheric injections were confined mainly to the troposphere and most of the transuranic isotopes were of mass than about 243. In debris from the Ivy Mike shot on November 1 of 1952, elements of mass through 255 were observed. The much higher yield of the Ivy Mike device than that of earlier nuclear

devices permitted the multiple neutron capture by uranium, which allowed production of the very heavy elements. In the detonation process, multiple neutron capture by ^{238}U results in the production of extremely neutron-rich products, the beta decay of which produces nuclides along the line of greatest stability. Table I shows the relative abundance of the transuranium isotopes which were formed in the Ivy Mike test. The isobars of significant half-life are shown together with their decay properties and half-lives. Their total activities are normalized to ^{239}Pu to permit comparison of their relative production rates. It is evident that ^{241}Pu (a beta decay isotope) and in later years ^{241}Am are the most abundant activities. The production of large amounts of transuranium isotopes in the Ivy Mike test stimulated an interest in the use of nuclear explosions for the production of transuranium isotopes. In subsequent tests conducted by the Los Alamos Scientific Laboratory and the Livermore Laboratory, neutron fluxes as great as twentyfold those in the Ivy Mike device were achieved (See Table II) These very high fluxes produced the heavy isotope fermium concentrations of several orders of magnitude higher relative to ^{240}Pu than in the Ivy Mike test. From these tests it was calculated that the production of isotopes beyond mass 257 was not likely to occur unless very substantially higher neutron fluxes were achieved. It is believed that an island of stability near mass 298 and atomic No. 114 exists, but to produce such an isotope would require a substantially higher flux than has been realized in nuclear detonations.

TRANSURANIUM NUCLIDES FROM REACTOR OPERATION

With the use of very high exposure fuels for energy generation, the production of transuranium elements through the end of the century will be very substantial. It has been estimated that approximately one-quarter million tons of radioactive wastes will be generated which will include approximately 10^9 curies of the transuranium elements. In generation of energy by nuclear reactors, both ^{235}U and the ^{239}Pu will be burned to the highest degree possible. In the exposure of these fuels, very substantial amounts of Am and Cm isotopes will be formed. For example, as shown in Table II, the activity ratios of ^{241}Am and of $^{242-244}Cm$ will be hundreds to thousands of times greater than that of ^{239}Pu. The potential environmental impact of these transplutonium elements should therefore be of major concern relative to $^{239-240}Pu$. Because of their methods of production, the relative abundances of the transuranium elements are considerably different from nuclear detonations than from reactor operations. In nuclear detonations, neutron capture occurs in extremely rapid succession, producing very high mass uranium isotopes which rapidly decay to form the spectrum of transuranium elements. In this case there is no opportunity for the decay of the various uranium isotopes which could break the chain of successive neutron capture. In the reactor production of radionuclides, the neutrons are captured only one at a time and the resulting product may decay prior to additional neutron capture.

In Table IV the amounts of the various transuranium elements resulting from the Ivy Mike nuclear test are compared with those which result from nuclear power generation. It is immediately evident that the transuranium elements resulting from nuclear energy generation are much higher relative to ^{239}Pu in the region just above plutonium, than for those from nuclear weapons testing.

Reported releases of transuranium elements from nuclear plant operations indicate that in general these have been very small. Loss of material around the Rocky Flats plant has resulted in some environmental contamination. Accidents involving nuclear-weapons-carrying aircraft have resulted in the spread of plutonium over limited areas, however these appear to have resulted in rather minor injections of Pu into the atmosphere.

TRANSURANIUM NUCLIDES IN THE ATMOSPHERE

Since the testing of the first thermonuclear device in 1952, substantial amounts of fission products, as well as transuranium elements, have entered the stratosphere. These injections result in the long-term, relatively slow deposition of radioactivity over the entire surface of the earth. Empirical box models which describe the movement of radioactivity from the upper to the lower stratosphere, between hemispheres, from the stratosphere to the troposphere, and the deposition rate on the earth's surface have been developed and appear to be reasonably satisfactory. As a first approximation, the transuranium elements appear to behave in their atmospheric transport in essentially the same manner as other fission products. It has been shown [2], for example, that

the ratio of ^{90}Sr to $^{239-240}Pu$ was constant in fallout from several locations on the earth during several years following the U.S. and USSR nuclear tests of 1961 and 1962. In Figure 1 we have shown the $^{239-240}Pu$ to ^{137}Cs ratio for the past decade. This ratio appears to be constant within the accuracy of the measurements and thus tends to confirm the constancy of the transuranium behavior relative to that of the fission products. Figure 2 shows the concentrations of ^{238}Pu and ^{239}Pu from 1962 to the present. These measurements, which were made near our laboratory at Richland, Washington, show that seasonal variations in the $^{238-239}Pu$ were similar to those of other nuclear-weapons-produced radionuclides of stratospheric origin; maximum concentrations occurring the late spring and minimum occurring in the winter. The rate of decrease in the $^{239-240}Pu$ concentration from 1963 through 1967 corresponded to a stratospheric half-residence time of 10 to 11 months, which is the same as the half-residence times calculated from measurements of other radionuclides of stratospheric origin. The ^{239}Pu concentrations remained fairly constant from 1967 to 1972, primarily because of yearly injections of plutonium by thermonuclear tests conducted by the Chinese at Lop Nor (35° N); the contribution from the French tests in the South Pacific (23° S) may also have been significant. Relatively large amounts of the short lived ^{237}U and ^{239}Np were also observed following the individual tests.

From 1962 through 1965 both the ^{238}Pu and the ^{239}Pu in surface air at Richland came primarily from the 1961-62 U.S. and USSR series. The $^{238}Pu/^{239}Pu$ ratio averaged 0.015 in 1964. The ratio

increased slightly in 1965, and by the spring of 1966 it had increased to 0.042, indicating that ^{238}Pu from the SNAP-9A burnup was present. The amount of SNAP-9A ^{238}Pu present was calculated from the ^{238}Pu concentrations and the $^{238}Pu/^{239}Pu$ ratios, assuming that the ratio in debris from nuclear weapons tests was 0.015. These calculations indicate that the ^{238}Pu in Richland air from 1967 to 1971 came primarily from SNAP-9A. From 1967 through 1969 the concentrations of SNAP-9A plutonium at Richland remained fairly constant, indicating that the ^{238}Pu was being transferred across the equator into the northern hemisphere at a rate comparable to the rate at which ^{238}Pu was being deposited on the earth's surface. The ^{238}Pu concentrations showed seasonal variations typical of radionuclides of stratospheric origin, so the transfer was probably taking place primarily in the stratosphere. Concentrations of SNAP-9A plutonium at Richland have decreased rapidly since that time. These results indicate that the stratospheric debris injected into the high stratosphere of one hemisphere will produce high concentrations of the debris in ground level air in the other hemisphere in the spring 1 to 2 years later. These concentrations will then remain constant for about three years before beginning to decrease.

Table V shows the $^{238}Pu/^{239}Pu$ and $^{238}Pu;\ ^{239}Pu$ ratios in Richland, Washington and at Pt. Barrow, Alaska. These data show how the ratio of ^{238}Pu to ^{239}Pu increased at these sampling stations as a result of debris from the SNAP-9 device. This ratio increased by more than an order of magnitude and was substantially higher at Pt. Barrow, 71° N, than at Richland, 46° N. These data would suggest that interhemispheric circulation within the mesosphere may have been an

an important factor in ^{238}Pu transport toward the northern polar regions.

Although not directly formed in the nuclear weapons detonation, considerable amounts of ^{241}Am are present in fallout debris. This, of course, results from the decay of ^{241}Pu. Based on the mount of ^{239}Pu in the atmosphere and the ratio of ^{239}Pu to ^{241}Pu observed in the Ivy Mike test, one can calculate their approximate ratios as a function of time in the atmosphere. These ratios are plotted in Figure 5 together with the observed concentrations of ^{241}Am as measured from samples taken at our monitoring station in Richland, Washington. It is evident that the airborne concentrations are in reasonably good agreement with those which were calculated and that the ratio of ^{241}Am to $^{239-240}Pu$ does increase, as would be expected, as the debris ages. Based on the yields of transuranium elements which were observed in the Ivy Mike tests and the $^{239-240}Pu$ inventory established by HASL, the total amounts of the other transuranium elements which have entered the atmosphere can be estimated. These values are shown in Table VI. For isotopes of mass greater than 244, the total atmospheric injections appear to be in the range of hundredths to tens of curies.

Short-lived Radionuclides in Fallout from Nuclear Weapons Testing

Nuclear debris from the past three Chinese tests has been examined to estimate the radiation exposure resulting from individual short-lived radioisotopes. A typical analysis of these short-lived

radionuclides is shown in Table VII. It is evident from these data that the amounts of ^{237}U (6.75 days) and ^{239}Np (2.35 days) are very high compared with most of the short-lived fission products. It has been shown that these radionuclides will contribute a substantial fraction of the radiation exposure based on either submersion or the ground deposited radiation.

Relative abundance of heavy element produced during Ivy Mike Test

TABLE I

MASS ABUNDANCES AT TIME = 0

Mass No.	Isobar	Type Decay	t 1/2 (Yrs)	Relative Abundance, Atoms Mass 239 = 1	Relative Abundance, Activity Mass 239 = 1
239	Pu	α	2.44×10^{4}	1.0	1.0
240	Pu	α	6.54×10^{3}	.363	1.35
241	Pu	β^-	15	.039	63
242	Pu	α	3.87×10^{5}	1.9×10^{-2}	1.2×10^{-3}
243	Am	α	7.37×10^{3}	2.1×10^{-3}	6.9×10^{-3}
244	Pu	α	8.3×10^{7}	1.2×10^{-3}	3.5×10^{-7}
245	Cm	α	8.5×10^{3}	1.2×10^{-4}	3.6×10^{-4}
246	Cm	α	4.76×10^{3}	4.8×10^{-5}	2.4×10^{-4}
247	Cm	α	1.54×10^{7}	3.9×10^{-6}	6.2×10^{-9}
248	Cm	α	3.5×10^{5}	1.2×10^{-6}	8.4×10^{-8}
249	Bk	β^-	.852	1.1×10^{-7}	3.2×10^{-3}
250	Cm	sf	1.13×10^{4}	$\sim 3 \times 10^{-8}$	$\sim 6.5 \times 10^{-8}$
251	Cf	α	9.0×10^{2}	$\sim 1.4 \times 10^{-8}$	$\sim 3.8 \times 10^{-7}$
252	Cf	α	2.63	1.0×10^{-9}	9.3×10^{-6}
253	Cf	β^-	.049	5.0×10^{-10}	2.5×10^{-4}
254	Cf	β^-	.164	5.0×10^{-11}	7.4×10^{-6}
255	Es	β^-	.107	4.0×10^{-11}	9.1×10^{-6}

TABLE III

TESTS FOR PRODUCING TRANSURANIC ISOTOPES

Test Name	Laboratory	Date	Exposure*
Ivy Mike	LASL	11-52	2.0
Anacostia	LRL	11-62	2.5
Kennebec	LRL	6-63	4.7
Anchovy	LASL	11-63	2.0
PAR	LRL	10-64	11
Barbel	LASL	10-64	11
Tweed	LRL	5-65	12
Cyclamen	LASL	5-66	18
Kankakee	LRL	6-66	12
Vulcan	LRL	6-66	12
Hutch	LRL	7-69	40

*Equivalent 20 KeV time integrated neutron flux in moles of neutron/cm^2.

TABLE III

ACTIVITY OF HEAVY ELEMENTS IN ACCUMULATED WASTE FOR ENTIRE NUCLEAR INDUSTRY

Normalized to 1 for ^{239}Pu

	1972	1974	1976	1978	1980	1982	1986	1990	2000
^{237}Np	0.22	0.21	0.22	0.21	0.20	0.18	0.15	0.14	0.076
^{239}Np	11.5	11.2	11	10.8	14.5	26.5	43.1	31.7	7.84
^{236}Pu	0.00092	0.00076	0.00075	0.00067	0.00056	0.00061	0.0036	0.010	0.012
^{238}Pu	36.1	44.1	49	50	55.6	80.6	167	283	361
^{239}Pu	1	1	1	1	1	1	1	1	1
^{240}Pu	1.54	1.65	1.7	0.58	1.89	2.55	4.45	5.33	3.14
^{241}Pu	315	306	300	287	295	337	403 3	367	255
^{242}Pu	0.0043	0.0043	0.0044	0.0042	0.0047	0.0065	0.0099	0.011	0.011
^{243}Pu	--	--	--	--	--	--	--	--	--
^{241}Am	96.1	95.3	96.5	93.6	103	137	178	141	77.2
^{242m}Am	5.69	5.64	5.5	5.4	7.23	12.2	19.5	14.2	5.68
^{242}Am	5.69	5.64	5.5	5.4	7.23	12.2	19.5	14.2	5.68
^{243}Am	11.5	11.2	11	10.8	14.5	26.5	43.1	31.7	7.84
^{242}Cm	5637	3887	3020	2255	2624	3876	3183	953	278
^{243}Cm	2.31	2.23	2.25	2.16	2.34	3.16	4.03	3.00	2.16
^{244}Cm	1561	1511	1510	1427	2441	5671	9813	6464	919
^{245}Cm	0.23	0.22	0.23	0.22	0.50	1.43	2.64	1.92	0.33
^{246}Cm	0.046	0.045	0.046	0.046	0.072	0.26	0.49	0.35	0.063
^{247}Cm	--	--	--	--	--	--	--	--	--
TOTAL CURIES	7.2×10^{5}	3.9×10^{6}	1.4×10^{7}	3.0×10^{7}	7.9×10^{7}	2.7×10^{8}	9.3×10^{8}	1.2×10^{9}	1.8×10^{9}

TABLE IV

RELATIVE COMPOSITIONS OF TRANSURANIUM ELEMENTS FROM POWER REACTORS AND IVY MIKE SHOT

(Values normalized to ^{239}Pu)

Mass No.	Isotope	Power Reactors	Ivy Mike Shot
238	Pu	44.1	.015
239	Pu	1	1
240	Pu	1.65	1.35
241	Pu	306	63
	Am	95	
242	Pu	4.3×10^{-3}	1.2×10^{-3}
	Am	5.64	
243	Pu	--	6.9×10^{-3}
	Am	11.2	
244			3.5×10^{-7}
	Cm	15.11	
245	Cm	.22	3.6×10^{-4}
246	Cm	4.5×10^{-2}	2.4×10^{-4}
247	Cm		6.2×10^{-9}
248	Cm		8.4×10^{-8}
249	Bk		3.2×10^{-3}
250	Cm		$\sim 6.5 \times 10^{-8}$
251	Cf		$\sim 3.8 \times 10^{-7}$
252	Cf		9.3×10^{-6}
253	Cf		2.5×10^{-4}
254	Cf		7.4×10^{-6}
255	Es		9.1×10^{-6}

*Assuming 30,000 MWD/ton exposure

Table 13

Accumulative ^{239}Pu and ^{238}Pu Concentrations in Air at Richland, Washington and Pt. Barrow, Alaska

	Richland			Alaska		
	^{239}Pu	^{238}Pu	$^{238}Pu/^{239}Pu$	^{239}Pu	^{238}Pu	$^{238}Pu/^{239}Pu$
1962	10.32	0.2367	.02292			
1963	6.423	0.1698	.02644			
1964	5.646	0.1886	.03340			
1965	4.740	0.07114	.01501	1.3142	0.0256	.01948
1966	1.719	0.1073	.06271	0.6003	0.0576	.09595
1967	0.9629	0.3672	.3722	0.1622	0.0856	0.5277
1968	1.3667	0.3361	.2459	0.4300	0.1289	0.2998
1969	1.3920	0.4983	.3580	0.3941	0.0987	0.2504
1970	1.5744	0.2268	.1441	0.3024	0.4976	0.1646
1971	1.6357	0.0952	.0582			
1972	0.8924	0.0735	.0823			

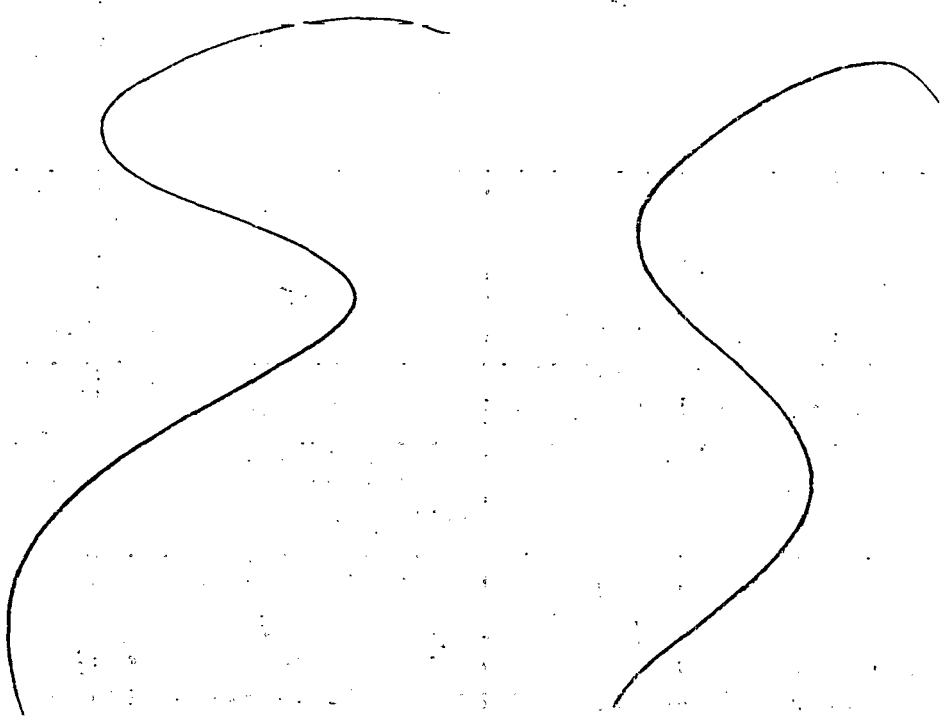


TABLE VI

RELATIVE ABUNDANCE OF TRANSURANIUM ELEMENTS BASED ON IVY MIKE AND ESTIMATED ATMOSPHERIC INJECTION

	Activity Abundance	Total Injection, KCi*
^{239}Pu	1	150
^{240}Pu	1.35	203
^{241}Pu	63	9450
^{241}Am	--	327**
^{242}Pu	1.2×10^{-3}	.18
^{243}Am	6.9×10^{-3}	1.04
^{244}Pu	3.5×10^{-7}	5.1×10^{-5}
^{245}Cm	3.6×10^{-4}	5.4×10^{-2}
^{246}Cm	2.4×10^{-4}	3.6×10^{-2}
^{247}Cm	6.2×10^{-9}	8.6×10^{-7}
^{248}Cm	8.4×10^{-8}	1.3×10^{-5}
^{249}Bk	3.2×10^{-3}	.48
^{250}Cm	6.5×10^{-8}	8.6×10^{-6}
^{251}Cf	3.8×10^{-7}	5.6×10^{-5}
^{252}Cf	9.3×10^{-6}	1.4×10^{-3}
^{253}Cf	2.5×10^{-4}	3.8×10^{-2}
^{254}Cf	7.4×10^{-6}	1.1×10^{-3}
^{255}Es	9.1×10^{-6}	1.4×10^{-3}

*Assumes 350 KCi $^{239-240}Pu$ atmospheric injection

** ^{241}Am formed on total decay of ^{241}Pu

TABLE VII

ATMOSPHERIC CONCENTRATION OF FALLOUT DEBRIS FROM CHINESE TEST OF 3-18-72 IN SURFACE AIR AT RICHLAND, WASH.

ON		OFF		Air Volume	dpm/10^3 scm										
Date	Time	Date	Time	$10^3 m^3$	^{147}Nd	^{99}Mo	^{141}Ce	^{237}U	^{239}Np	^{143}Ce	^{131}I	^{97}Zr	^{132}I	^{95}Zr	^{140}Ba
3-16-72	1015	3-21-72	1445	73.6	<0.38	<0.09	0.45	<0.20	<0.30	<0.07	<0.09	<0.05	<0.05	2.89	<0.12
3-21-72	1505	3-22-72	0925	10.8	968	3681	213	4138	74086	7929	303	16315	506	430	87
3-22-72	0947	3-23-72	1000	14.5	19	106	9.3	175	3605	181	20	85	50	11	15
3-23-72	1030	3-24-72	1005	14.1	100	655	49	876	14155	740	87	1618	215	38	60
3-23-72	1030	4-5-72	1315	187	33	186	34	638	10362	---	61	---	---	23	73
4-5-72	1330	4-20-72	1005	215	37	---	28	369	---	---	32	---	---	31	35

Corrected for decay to 3-18-72 at 0600

Figure I

Ratio of ^{239}Pu to ^{137}Cs in Fallout debris since 1962

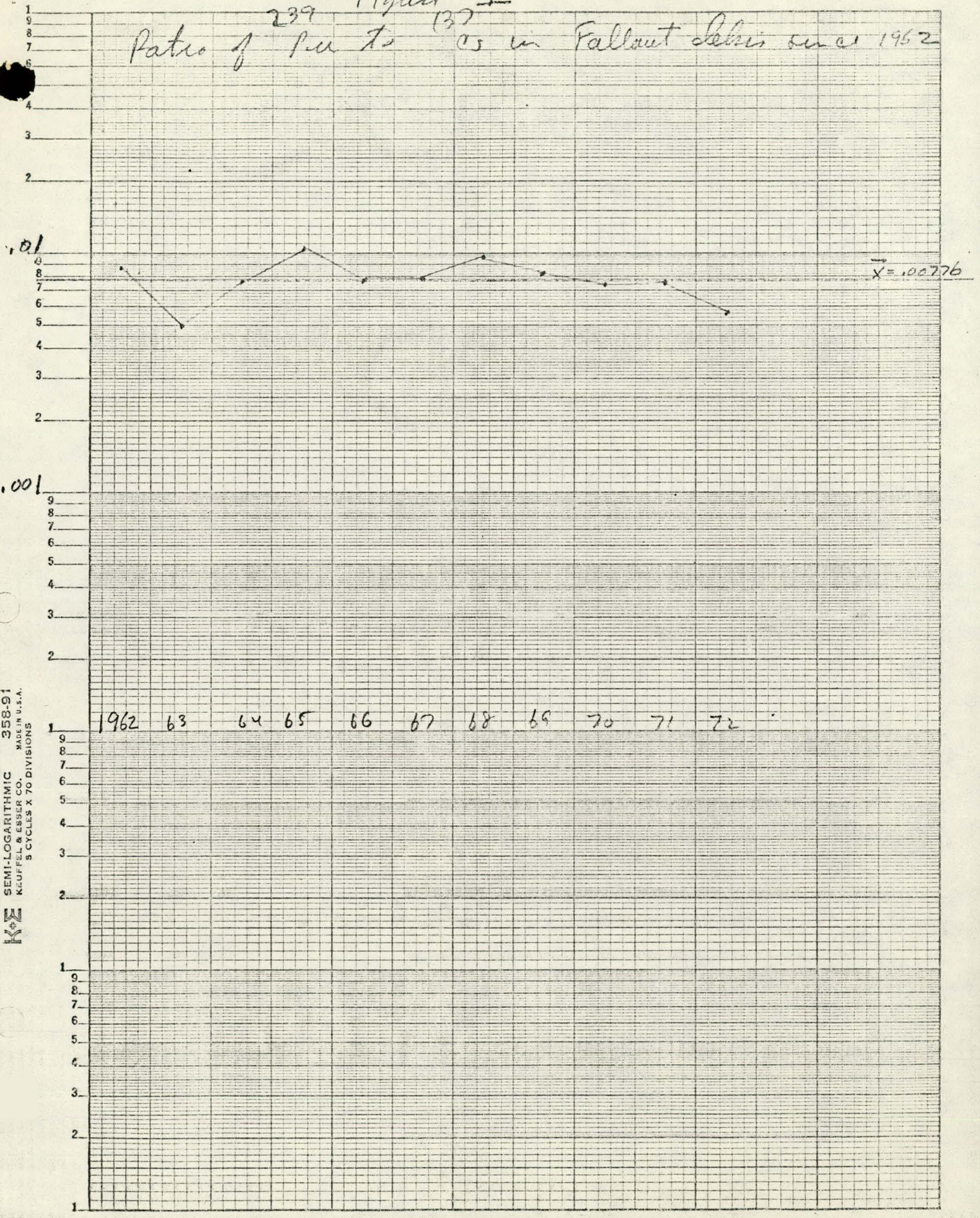
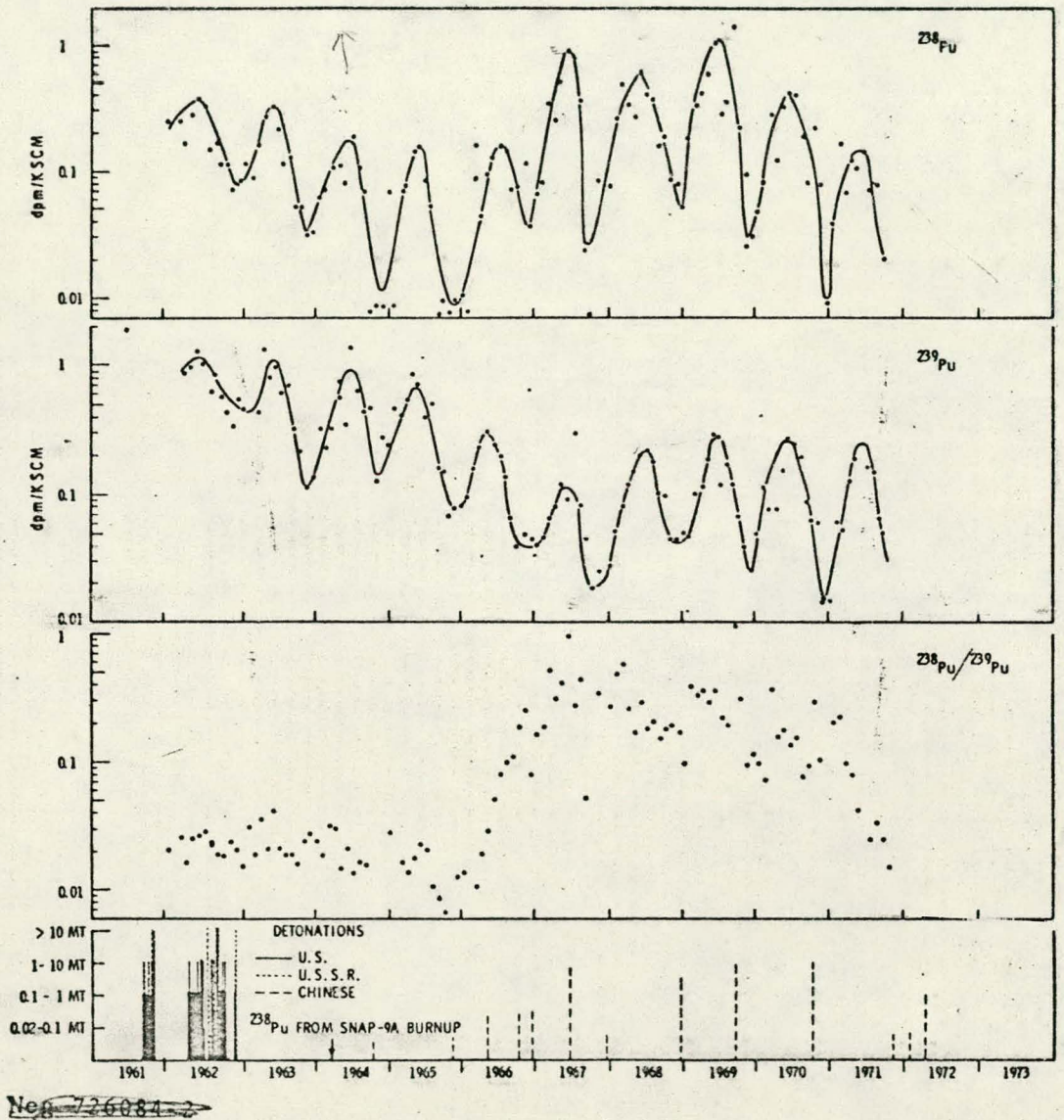


Fig II

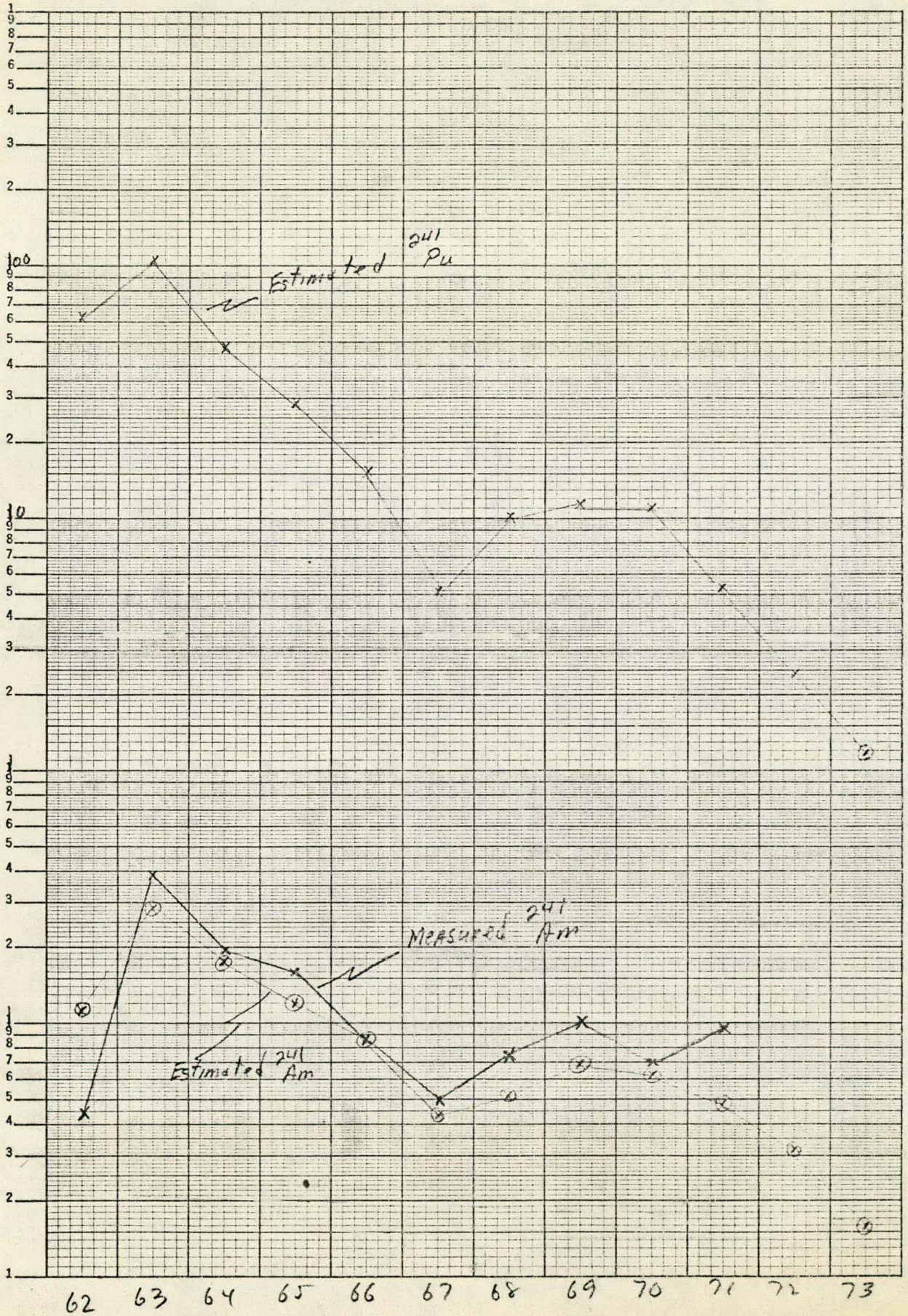
^{238}Pu and ^{239}Pu Concentration in Surface Air
at Richland, Washington



Neg. 726084-2

Fig III

Estimated concentration of ^{241}Pu and ^{241}Am as compared to the measured concentration of ^{241}Am in surface air since 1962 at Ashland



DPM/KSCM