50

PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1972
TO THE USAEC DIVISION OF BIOMEDICAL
AND ENVIRONMENTAL RESEARCH
VOLUME II: PHYSICAL SCIENCES
PART 1. ATMOSPHERIC SCIENCES

Battelle

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Richland, Washington 99352

APRIL 1973

Prepared for the U.S. Atomic Energy
Commission under Contract AT(45-1):1830

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PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
U.S. ATOMIC ENERGY COMMISSION
Under Contract AT(45-1)-1830

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.95

PLUTONIUM CONCENTRATIONS IN SURFACE AIR AT RICHLAND, WASHINGTON

C. W. Thomas

The plutonium isotopes released by atmospheric nuclear testing are a combination of unreacted plutonium in the device and isotopes of plutonium formed by neutron capture in plutonium and any ^{238}U in the device. The most abundant isotope of plutonium is ^{239}Pu , with lesser amounts of ^{238}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu . The ^{238}Pu is produced by either the reaction of fast neutrons with ^{239}Pu or by the decay of ^{238}Np or ^{242}Cm . Plutonium-238 to ^{239}Pu ratios vary from detonation to detonation depending upon the burst conditions and the material used.

In April 1964, a transit navigational satellite was launched from Vandenberg containing a System for Nuclear Auxiliary Power (SNAP-9A) generator. The satellite failed to achieve a stable orbit and subsequently burned upon re-entry, releasing approximately 17 kCi of ^{238}Pu (~1 kg) at 46 km over the Indian Ocean. This ^{238}Pu almost tripled the global atmospheric ^{238}Pu inventory. Plutonium from this device was first detected in a sample collected by balloon at 33 km over Australia in August 1964. Since that time, it has been detected in stratospheric and tropospheric air from both hemispheres.

The atmospheric concentrations of ^{238}Pu and ^{239}Pu were measured in surface air samples collected at Richland, Washington from 1963 to 1972 as a part of a program to define the

rates of long term stratospheric processes in the northern hemisphere as well as the rates of interhemispheric mixing (Figure 1). The seasonal variations in the concentrations of ^{238}Pu and ^{239}Pu in surface air at Richland were similar to those of other nuclear-weapons-produced radionuclides of stratospheric origin; maximum concentrations occurred in the late spring and minimums occurred in the winter. The rate of decrease in the ^{239}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. Fission product yields indicate that the Chinese used ^{235}U as the fissile material in their high-yield tests of 1967 through 1970. However, large amounts of ^{237}U , ^{239}Np , and ^{239}Pu were produced, indicating that ^{238}U was used as a component part of the devices.

From 1962 through 1965 both the ^{238}Pu and the ^{239}Pu in surface air at Richland came primarily from the

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FIGURE 1. ^{238}Pu and ^{239}Pu Concentration in Surface Air at Richland, Washington

1961-62 U.S.-Russian test series (Table 1). The $^{238}\text{Pu}/^{239}\text{Pu}$ 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 $^{238}\text{Pu}/^{239}\text{Pu}$ 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 to 1969 the concentrations

TABLE 1. The Average Yearly Ratios of ^{239}Pu , ^{238}Pu , and ^{90}Sr in Surface Air at Richland, Washington

	dpm/KSCM					
	^{90}Sr	^{239}Pu	^{238}Pu		$^{239}\text{Pu}/^{90}\text{Sr}$	$^{238}\text{Pu}/^{239}\text{Pu}$
			Weapons at 1-1/2%	Other		
1962	90.6	1.15 ± 0.169	0.0166	0.0324	0.0127	0.044
1963	119	1.20 ± 0.02	0.0125	0.0015	0.010 ± 0.0043	0.019
1964	80.5	1.02 ± 0.015	0.0127	0.0005	0.0127 ± 0.012	0.015 ± 0.0015
1965	40.0	0.704 ± 0.012	0.0108	0.003	0.0176 ± 0.0072	0.019
1966	22.9	0.325 ± 0.007	0.0038	0.0069	0.0142 ± 0.0055	0.042
1967	7.08	0.125 ± 0.002	0.0023	0.0608	0.018 ± 0.0049	0.415
1968	10.3	0.230 ± 0.003	0.0027	0.0375	0.022 ± 0.0068	0.223
1969	11.4	0.310 ± 0.004	0.0028	0.0511	0.027 ± 0.0076	0.293
1970		0.284 ± 0.004	0.0023	0.0295		0.2066
1971		0.275 ± 0.003	0.0025	0.0073		0.063

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 radio-nuclides 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.