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TRANSURANIC ELEMENTS IN THE ENVIRONMENT

A Summary of Environmental Research on Transuranium
Radionuclides Funded by the U. S. Department of Energy
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Wayne C. Hanson, Editor
Pacific Northwest Laboratory

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Worldwide Fallout

R. W. PERKINS and C. W. THOMAS

Since the first nuclear weapons test at Alamogordo, N. Mex., on July 16, 1945, approximately 360,000 Ci (1360 kCi) of $^{239,240}\text{Pu}$ has been injected into the atmosphere. In addition, 17,000 Ci (17 kCi) of ^{238}Pu entered the atmosphere in April 1964 as a result of the high-altitude burnup of a SNAP-9 satellite power source. Since most of the plutonium from nuclear weapons testing, as well as that from the SNAP-9 burnup, entered the stratosphere, fallout has been worldwide. The deposition is influenced by meteorological conditions and also by topographical features of the earth's surface. Residence time in the stratosphere is about 10 to 11 months; however, because of the high-altitude burnup of the SNAP-9 device, it was 2 yr before significant amounts of this debris reached the earth's surface.

In addition to plutonium, substantial amounts of ^{241}Am are formed from the decay of the weak beta emitter ^{241}Pu and are an important constituent of fallout.

The majority of radioactivity entering the stratosphere during this past decade has been a result of the Chinese nuclear weapons testing. The ratio of plutonium to ^{137}Cs has been relatively constant throughout the nuclear weapons period, and thus a measurement of ^{137}Cs permits a reasonable estimate of the plutonium deposition. The ratio of transuranic elements in fallout is substantially different from that in power reactor wastes, which contain far more americium and curium relative to plutonium. Fresh fallout from thermal nuclear weapons contains large amounts of short-lived ^{237}U and ^{239}Np , and these may contribute substantially to the radiation exposure at the earth's surface.

The first significant injection of transuranium elements into the atmosphere occurred as the result of the nuclear weapons testing in Alamogordo, N. Mex., on July 16, 1945. Between then and 1952 further nuclear detonations resulted in additional injections to the atmosphere, however, because of their relatively low yield, most of this debris was confined to the troposphere. On Nov. 1, 1952, the first thermonuclear device was detonated. This 14-Mt explosion injected large amounts of debris into the stratosphere. The relatively high energy yield of this fusion device, together with a much higher integrated neutron flux, greatly increased 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 and 1962 United States (U. S.) and Union of Soviet Socialist Republics (U.S.S.R.) nuclear testing programs. A nuclear weapons test-ban agreement between the United States, United Kingdom, and Soviet Union in early 1963 suspended atmospheric testing. However, in late 1964 the Chinese exploded their first atmospheric nuclear test, and since that time they have continued testing in the northern hemisphere. France was not a member of the test-ban agreement, and in mid-1966 they began atmospheric testing in the southern hemisphere. The test-ban agreement in 1963 did not rule out underground tests, which do not vent to

reasonably constant in fallout since the U.S. and U.S.S.R. nuclear tests of 1961 and 1962. These data are based on measurements beginning in 1962 at Richland, Wash. (46°N), and in later periods in New York, N.Y. (41°N), and Harwell, England (52°N). The $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratio appears to be constant in most cases within the accuracy of the measurements and thus tends to indicate a general constancy of the transuranium production and atmospheric behavior relative to that of the fission products.

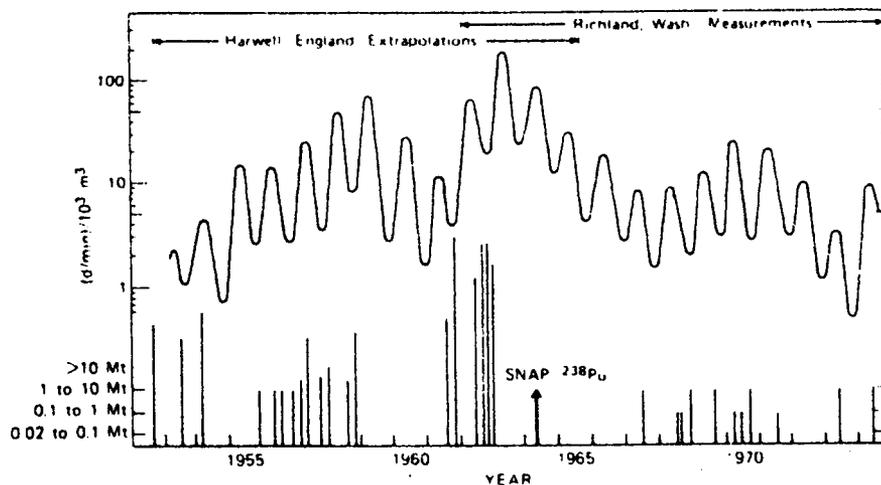


Fig. 5 Concentrations of ^{137}Cs in surface air at 46°N latitude since 1953. The concentrations prior to 1962 were estimated by normalizing concentrations measured at Harwell, England.

On the basis of observed ^{137}Cs concentrations at 46°N latitude since 1962 and an extrapolation back to 1953 by normalizing Harwell, England, to Richland, Wash. ^{137}Cs air concentrations during the period 1962 to 1964 (as indicated in Fig. 5), it should be possible to obtain a good estimate of the airborne plutonium concentrations during this entire period. Such extrapolations are, of course, subject to some uncertainty.

An atmospheric sampling program using high altitude aircraft has been conducted since 1959 (Hardy, 1973). Sampling aircraft normally operate at four latitudes -- 70°N, 35°N, 10°N, and 40°S. Sampling altitudes normally range from 15,000 to 70,000 ft. Figure 6 shows the ratios of ^{240}Pu to ^{239}Pu , ^{241}Pu to ^{239}Pu , and ^{242}Pu to ^{239}Pu in air at 70°N latitude as a function of time. It is evident that there is considerable variation in these ratios which is undoubtedly associated with the type and energy of the weapon responsible for the plutonium isotope production. There is a substantial increase in the heavy-to-light plutonium isotopes immediately following the 1961 and 1962 U.S.-U.S.S.R. test series.

Figure 7 shows the concentrations of ^{238}Pu and $^{239,240}\text{Pu}$ from 1962 to the present. These measurements, which were made near Richland, Wash., show that seasonal variations in the $^{238,239,240}\text{Pu}$ were similar to those of other nuclear-weapons-produced radionuclides of stratospheric origin; maximum concentrations occur in the late spring,

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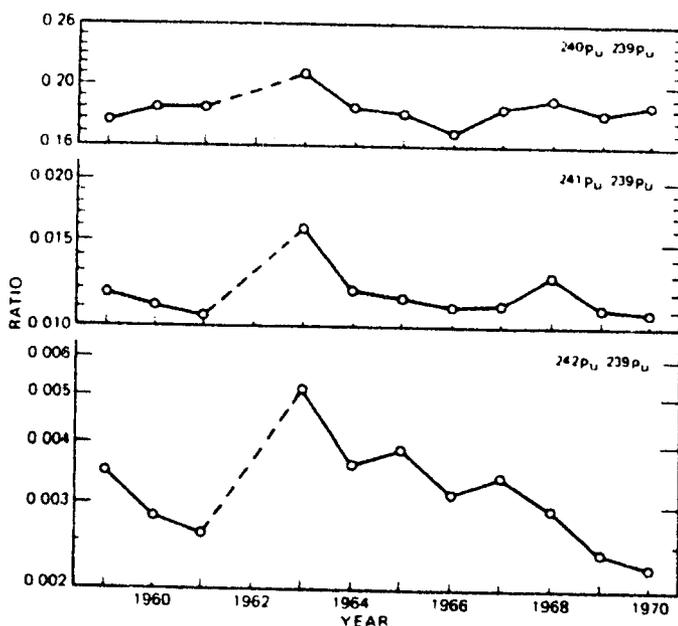


Fig. 6 Atom ratios of plutonium isotopes from an air column (15,000 to 17,000 ft high) at 70°N latitude.

and minimum concentrations occur in the winter. The rate of decrease in the $^{239,240}\text{Pu}$ concentrations from 1963 through 1967 corresponded to a stratospheric half-residence time of 10 to 11 months, which is similar to the half-residence times calculated from measurements of other radionuclides of stratospheric origin. The $^{239,240}\text{Pu}$ 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 (44°N); the contribution from the French tests in the South Pacific (23°S) may also have significance.

From 1962 through 1965 the ^{238}Pu and $^{239,240}\text{Pu}$ in surface air at Richland, Wash., came primarily from the 1961 and 1962 U.S. and U.S.S.R. series. The $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity ratio averaged about 0.020 in 1964. The activity ratio stayed almost the same in 1965, but, by the spring of 1966, it had increased to 0.042, which suggests that ^{238}Pu from the SNAP-9A burnup was present. The amount of SNAP-9A ^{238}Pu present was determined from the ^{238}Pu concentrations and the $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity ratios; the activity ratio in debris from nuclear weapons tests was assumed to be 0.020. These considerations indicate that the ^{238}Pu in Richland air from 1967 to 1971 came largely from SNAP-9A. From 1967 through 1969, the concentrations of SNAP-9A plutonium at Richland remained fairly constant, which indicates that the ^{238}Pu was being transferred into the northern hemispheric lower stratosphere at a rate comparable to the rate at which ^{238}Pu was being deposited on the earth's surface. This suggests that a substantial amount of ^{238}Pu was retained in the upper stratosphere, and its slow movement into the lower stratosphere maintained a nearly constant level for about 2 yr. The fact that the ^{238}Pu concentrations showed the usual seasonal variations typical of radionuclides of

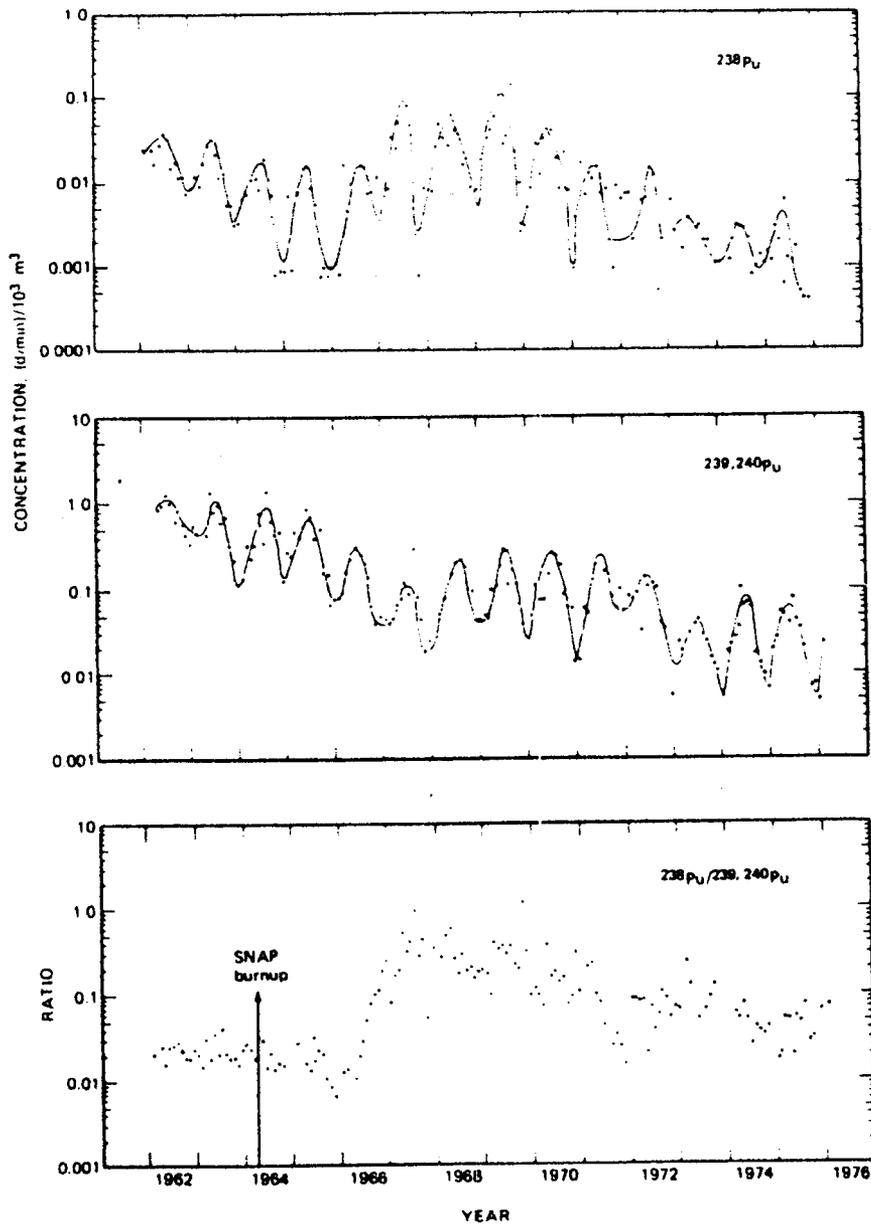


Fig. 7 Concentrations of ^{238}Pu and $^{239,240}\text{Pu}$ in surface air at Richland, Wash.

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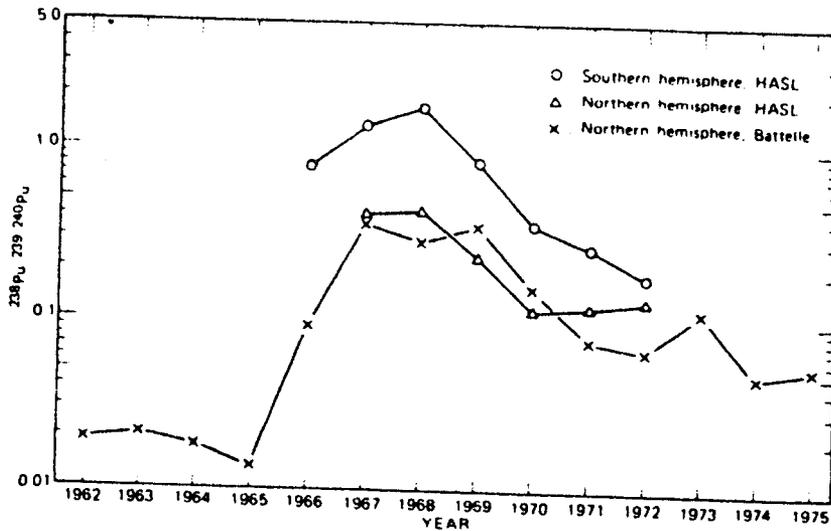


Fig. 8 Average yearly activity $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios in surface air.

stratospheric origin indicates that the transfer involved movement into the northern stratosphere and then to the troposphere. The hemispheric yearly averages compared with the yearly average at Richland, Wash., are shown in Fig. 8. Concentrations of SNAP-9A ^{238}Pu in the northern hemisphere and at Richland have decreased rapidly since 1968 and 1969, respectively.

Similar changes in ground-level air concentrations were observed at other locations in the northern and southern hemispheres (Hardy, 1976). These results indicate that the stratospheric debris injected into the high stratosphere may not produce high concentrations of the debris in ground-level air until 2 yr later. These ground-level concentrations may in some regions remain nearly constant for about 2 yr before they begin to decrease.

Although not formed directly 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 . On the basis of the amount of $^{239,240}\text{Pu}$ in the atmosphere and the ratio of ^{239}Pu to ^{241}Pu observed in the Mike test, one can calculate the ^{241}Am as a function of time in the atmosphere. The ^{241}Pu and the ^{241}Am ratios are plotted in Fig. 9 together with the observed concentrations of ^{241}Am as measured from samples taken at a monitoring station in Richland, Wash. It is evident that the airborne concentrations are in reasonably good agreement with those calculated. Also, the ratio of ^{241}Am to $^{239,240}\text{Pu}$ does increase, as would be expected, as the debris ages. On the basis of the yields of transuranium elements, which were observed in the Mike tests, and the $^{239,240}\text{Pu}$ updated inventory established by the Environmental Measurements Laboratory (EML), the total amounts of the other transuranium elements that have entered the atmosphere can be estimated. These values are shown in Table 8. For isotopes of mass greater than 244, the total atmospheric injections are in the range of hundredths to tens of curies, and the total alpha-decay activity of all the transuranium elements of mass greater than 241 is only about 1% of the $^{239,240}\text{Pu}$.

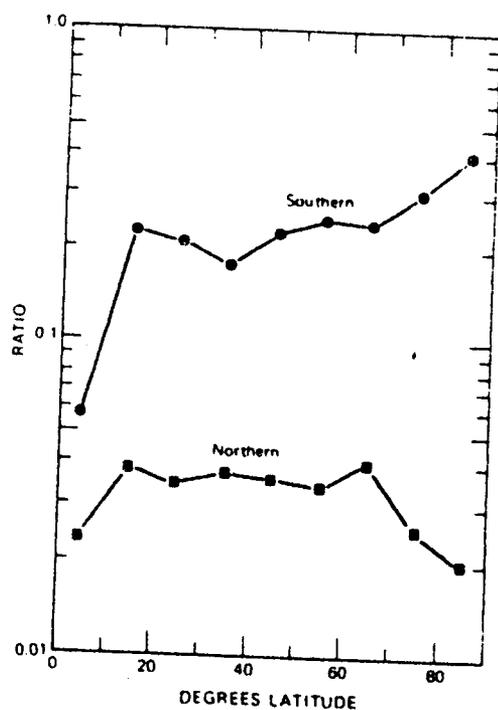


Fig. 15 Activity ratio of $^{239}\text{Pu}/^{240}\text{Pu}$ as a function of latitude in integrated soil sample.

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