PLUTONIUM IN HUMAN LUNG IN THE HANFORD ENVIRONS

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1. INTRODUCTION

The release or potential for release of radioactive materials to the environs of nuclear facilities has been receiving considerable attention in the press and other news media. Frequently plutonium has been highlighted as a most serious inhalation hazard.

Plutonium has been produced, fabricated, transported, and stored in various forms at the Hanford complex for several decades and has been the subject of substantial research as to its occurrence in the environs.

To assure that programs are adequate, it is necessary to demonstrate the extent to which past and present practices have or have not limited public exposure to plutonium. Both measurements of plutonium in tissue samples from individuals potentially exposed and estimates of exposure based on measured concentrations of plutonium in air as discussed in this paper provide a basis for such demonstration.

2. SOURCES OF PLUTONIUM IN AIR

Potential sources of ^{239}Pu in near-surface air in the Hanford environs have included worldwide fallout from weapons testing and releases of contaminated air from plutonium production facilities and research laboratories and possibly from resuspension of plutonium from along waste ditches, ponds, and burial grounds located at the Hanford complex. The latter potential sources of plutonium may have been of little significance since they would amount to ground-level releases and were located about 32 km from inhabited areas. Since May 1973, waste streams containing plutonium have been routed to tanks for storage, thus eliminating most of that potential for release of plutonium to the Hanford environs (1).

The first thermonuclear device in the weapons testing program was detonated in 1952; however, it was not until 1961-1962 that large amounts of transuranic elements were injected into the atmosphere. An estimated 400 kCi $^{239,240}Pu$ were produced during weapons testing, of which about 325 kCi were globally dispersed (2). (^{239}Pu and ^{240}Pu are not distinguishable by alpha spectrometry, and where ^{239}Pu is cited, the sum of the two nuclides is meant.)

3. HANFORD MEASUREMENTS

At Hanford routine measurement of concentrations of ^{239}Pu in near-surface air began in late 1961 (4); since 1949, autopsy tissue samples have been radioanalyzed for plutonium. The point of near-surface air measurement is within the research laboratory area and about 32 km from the production facilities. Figure 1 shows the measured values beginning in 1962 and expressed as annual average concentrations of plutonium. Values of plutonium in air from 1952 through 1961 were inferred through comparison of ^{239}Pu and ^{137}Cs measurements at Hanford and ^{137}Cs measurements made at Chilton, England (5). Also shown for comparison are concentrations of plutonium in air at New York, N. Y., from 1964 forward and as inferred from 1954 through 1963 based on measurements of ^{90}Sr deposited on ground and ^{90}Sr in near surface air (2).

Also shown are annual average concentrations of ^{239}Pu measured in near-surface air in the USSR for the years 1969 through 1971 (3).

Figure 1 indicates that most of the data agree closely. Except for the year 1964, the measurements of plutonium in surface air at Hanford and New York are within a factor of two or less. The concentration of ^{239}Pu at Hanford was almost always less than that at New York. Although Hanford plutonium facilities may have contributed to the total, the total concentration of plutonium in air at Hanford appears indistinguishable from fallout-related concentrations measured elsewhere in mid-latitudes in the northern hemisphere.

4. THEORETICALLY ESTABLISHED CONCENTRATIONS OF PLUTONIUM IN LUNG

Expected concentrations of plutonium in lung were calculated using measured concentrations in air, Standard Man parameters, and the ICRP Task Force Lung Model (6,7). Class Y parameters were assumed since the oxide is the expected form of plutonium in fallout. An activity median aerodynamic diameter (AMAD) of 0.4 μm was assumed to characterize particulate fallout.(8)

Equations for determining the lung burden as a function of time were developed as follows: According to the model, two components remain in the lung, one with a one-day half-time and one with a 500-day half-time. For present purposes, the component with the long half-time predominates in determining the quantity in the lung at a given time. The change in lung burden (referred to as the pulmonary region in the ICRP model), Q_p, with time may be given by

$$\frac{dQ_p}{dt} = f_e D_1 D_5 - \lambda_p Q_p$$

where λ_p is based on the 500-day half-life, $f_e = 0.6$ the fraction of the deposited quantity D_5 removed with a 500-day half-time. D_5 is about 0.35 for 0.4-μm AMAD aerosols. D_1 is the quantity of the inhaled aerosol. Solution of the above equation yields

$$Q_p = \frac{D_1 f_e D_5}{\lambda_p} (1 - e^{-\lambda_p t})$$

which gives the quantity of plutonium in the lung at the end of a period of constant intake. Division of Q_p by the mass of the lung (1000 g) yields the concentration of plutonium in lung.

Using this equation and the measured and inferred concentrations of plutonium in surface air, the concentration in lung was calculated for individuals who may have resided in the Hanford environs for 1 to 22 years. A breathing rate of 20 m^3/d and a constant level of plutonium in air over one year's time was assumed. The calculated values are tabulated in Table 1. For a given year the total lung burden at the end of the year was determined from the average Pu concentration in air for that year using the above equation plus the total at the end of the previous year decayed for one year with a 500-day half-life. Although exposure begins at birth, adult breathing parameters were used in the calculations.

Figure 2 presents these results graphically for 5-year intervals beginning in 1953. As shown in the figure, concentrations of plutonium to be expected in lung over a 22-year period ranged from about 0.01 to a maximum of 1.1 fCi/g of lung. The average for the period was about 0.4 fCi/g lung. Regardless of the year of initial exposure, the estimated concentration at the end

TABLE 1. Cumulative Concentrations in Lung At End of Period, fCi/g

Exposure Beginning	1953	1954	1955	1956	1957	1958	1959	1960	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970	1971	1972	1973	1974
1953	0.06	0.10	0.28	0.40	0.50	0.66	1.08	0.76	0.60	0.82	0.82	0.75	0.70	0.50	0.34	0.27	0.22	0.21	0.19	0.16	0.11	0.11
1954		0.07	0.26	0.39	0.50	0.65	1.08	0.76	0.60	0.82	0.82	0.75	0.70	0.50	0.34	0.27	0.22	0.21	0.19	0.16	0.11	0.11
1955			0.22	0.38	0.48	0.64	1.08	0.75	0.60	0.82	0.82	0.75	0.70	0.50	0.34	0.27	0.22	0.21	0.19	0.16	0.11	0.11
1956				0.23	0.40	0.60	1.05	0.73	0.58	0.81	0.81	0.74	0.70	0.50	0.34	0.27	0.22	0.20	0.19	0.16	0.11	0.11
1957					0.26	0.51	1.00	0.70	0.57	0.81	0.81	0.74	0.69	0.50	0.34	0.27	0.22	0.20	0.19	0.16	0.11	0.11
1958						0.35	0.90	0.65	0.53	0.78	0.80	0.74	0.69	0.49	0.34	0.27	0.22	0.20	0.19	0.16	0.11	0.11
1959							0.69	0.52	0.45	0.74	0.77	0.72	0.68	0.49	0.34	0.26	0.22	0.20	0.19	0.16	0.11	0.11
1960								0.10	0.20	0.59	0.68	0.66	0.65	0.47	0.32	0.26	0.22	0.20	0.19	0.16	0.10	0.11
1961									0.14	0.55	0.66	0.65	0.64	0.46	0.32	0.25	0.22	0.20	0.19	0.16	0.10	0.11
1962										0.46	0.60	0.62	0.62	0.45	0.31	0.25	0.21	0.20	0.18	0.15	0.10	0.11
1963											0.32	0.45	0.52	0.39	0.28	0.23	0.20	0.19	0.18	0.15	0.10	0.11
1964												0.24	0.40	0.32	0.23	0.20	0.18	0.18	0.18	0.15	0.10	0.10
1965													0.25	0.22	0.18	0.17	0.16	0.17	0.17	0.14	0.10	0.10
1966														0.08	0.09	0.12	0.13	0.15	0.16	0.14	0.09	0.10
1967															0.04	0.09	0.12	0.14	0.15	0.13	0.09	0.10
1968																0.06	0.10	0.13	0.14	0.13	0.09	0.10
1969																	0.06	0.11	0.13	0.12	0.08	0.10
1970																		0.07	0.11	0.11	0.08	0.09
1971																			0.06	0.08	0.06	0.08
1972																				0.04	0.04	0.07
1973																					0.01	0.05
1974																						0.04

of 1974 ranged from 0.04 to 0.1 fCi/g lung. The total Pu intake by inhalation over the 1953-1974 period is estimated to be 30 pCi. The average lung burden for this period was estimated to be 0.4 pCi. Bennett estimated a 42.6 pCi cumulative intake using the data obtained at New York for the period 1954-1975 (2). An average lung burden of 0.7 pCi was obtained from the New York data.

5. TISSUE ANALYSIS

Since 1949, tissue samples obtained at autopsy from residents (non-Hanford workers) in the Hanford environs have been radioanalyzed for plutonium. Figure 3 shows graphically the concentration of plutonium in lung of individuals sampled. One problem associated with these measurements has been the lack of sensitivity commensurate with the very small quantities in the lung. As a consequence, many sample results were reported only as less than the detection limit; this limit has varied over the years and with sample size. In Figure 3, the average ($\bar{x}$) shown is the average assuming each sample result in the set had been as much as the detection limit. Although it too may have been a "less than" value, the maximum reading of the set is also indicated in the figure. The lower value measured in a set of samples is indicated by the letter L.

In 1974, our laboratory converted from an autoradiography measurement technique, which had the advantage of rather good sensitivity but an overriding disadvantage of uncertainty in radiochemical yield, to a process of alpha spectrometry. Improved knowledge of chemical yield through use of ^{236}Pu and ^{242}Pu tracers and alpha spectrometry yields more reliable data at a slight loss in sensitivity at reasonable counting times.

6. CONCLUSIONS

As shown in Figure 4, the measured and theoretical results compare closely. In most cases, the average concentration of the set was within a factor of 5 of the theoretical estimate. The marked decrease in measured concentrations of plutonium in lung from 1966 to 1970 coincided with an approximate tenfold increase in tissue sample size. That period was also one in which fallout concentrations decreased. Caution should be used in developing conclusions from these results because many of the sample results during this time and earlier were "less thans", and their true value may have been substantially less than that shown. The apparent increase in measured concentrations of plutonium in lung in the period 1971-1974 is not understood. The new alpha spectrometry procedure was used on these samples, and some systematic error in earlier work may be suggested. The possibility of a small contribution from Hanford facilities cannot be absolutely ruled out.

The average concentration in lung over the period covered (assuming a "less than" value to be at detection limit) was 0.5 fCi/g of lung, and the average of the maximum values noted was 0.8 fCi/g. For comparison, the theoretical average value where it is assumed that each year since 1954 an individual is autopsied was 0.4 fCi/g. If Hanford facilities have contributed to the plutonium content of lungs in individuals residing in the environs, that contribution would appear at most to be about that from worldwide fallout. But more likely the plutonium in lung derives almost totally from fallout, and the Hanford contribution is negligible by comparison.

Regardless of source, a sustained lung burden of plutonium of 0.5 fCi/g of lung would relate to a dose of 0.5 mrem/year. Over the 22-year period ending in 1974 a total dose to lung of 10 mrem was estimated from these data.

Numerically, this is trivial compared to the approximately 1600-2000 mrem (1) the individual would likely have received from naturally occurring sources in the Hanford area during this same period.

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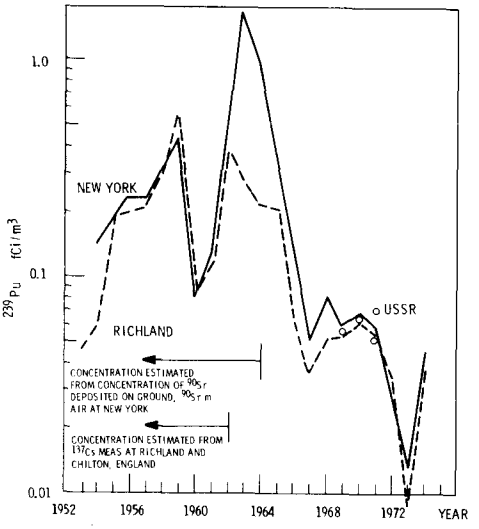

FIGURE 1. Concentration of Plutonium in Surface Air at New York, NY and Richland, WA

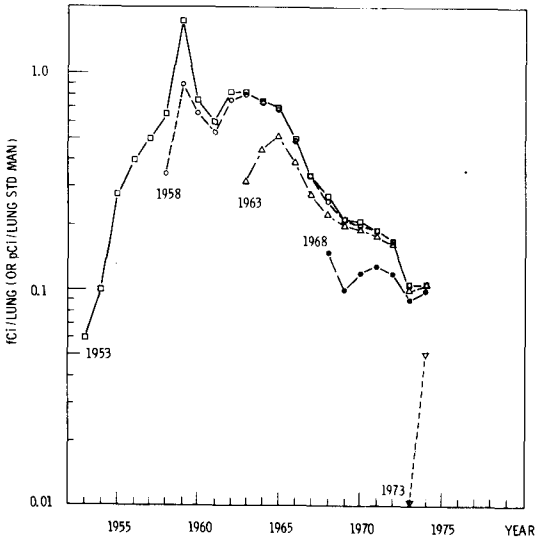

FIGURE 2. Theoretical Concentration of Plutonium in Lung in the Hanford Environs (by year of initial exposure)

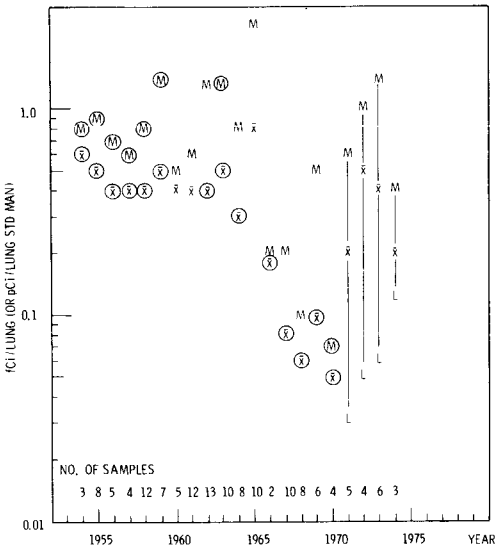

FIGURE 3. Concentrations of Plutonium in Lung

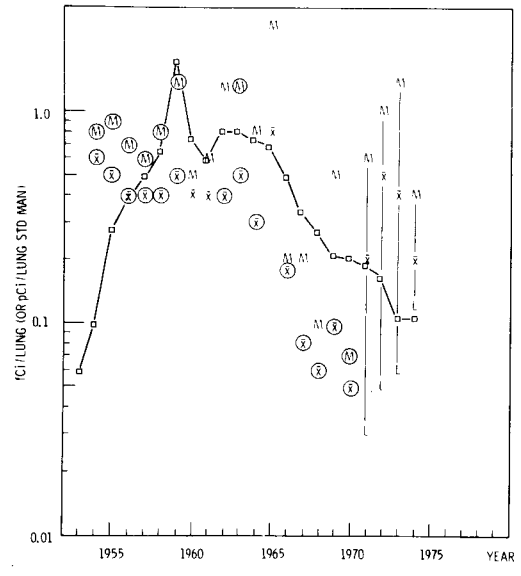

FIGURE 4. Comparison of Measured and Theoretical Concentrations of Plutonium in Human Lung