Health Physics Pergamon Press 1972. Vol. 22 (June), pp 925–930. Printed in Northern Ireland

PLUTONIUM IN AUTOPSY TISSUE SAMPLES*

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(*Presented by* I. C. NELSON)

Abstract—Tissue samples have been obtained at autopsy from former plutonium workers, other employees and residents in the environs of the Hanford Atomic Project since 1949. These samples, which included lung, liver, bone, sometimes tracheobronchial lymph nodes and occasionally other tissue samples, were analyzed radiochemically for plutonium. Data obtained since 1965 and a recapitulation of results since 1949 are presented. A discussion of these results relative to plutonium work exposure and plutonium from worldwide fallout is also presented.

INTRODUCTION

A PROGRAM of collection and radioanalysis of tissue samples obtained postmortem from plant workers and residents in the plant environs has been maintained at Hanford since 1949. The significance of this effort lies primarily in developing the capability for assessing the overall long-term effectiveness of operating controls, safety procedures and engineering safeguards incorporated into plutonium facilities which control the release of radioactive materials to the Hanford environs. The data obtained in this program can also be important for assessing the overall impact of occupational exposure on workers and evaluating the applicability of internal deposition prediction models through correlation of exposure conditions and resulting body burdens. In addition, such data may permit assessment of possible causative association of radioactive materials *in situ* and observed pathological conditions in cases involving large amounts of radioactive material.

Results obtained in this program through early 1965 were summarized by NEWTON *et al.*[1] It is the purpose of this paper to update those program results for selected tissue samples from the approximately 350 cases obtained since program inception.

PROGRAM DESCRIPTION

At present, tissue samples obtained consist of portions of lung, liver, bone and the tracheobronchial lymph nodes from non-contageous disease autopsy cases as selected by the pathologist. In addition, blood, pancreas, prostate, seminal vesicles and occasionally spleen samples have been taken. Because of the infrequency of measurable plutonium in these samples, they are no longer obtained on a routine basis. In cases where significantly measurable plutonium was anticipated because of known occupational exposure, the sampling of tissues was more extensive.

Sample sizes have varied over the years, but large samples are now sought in an effort to improve the sensitivity of the measurement and to improve confidence in estimates of whole organ content where non-uniform distributions of plutonium within the organ may exist. When practical, the left lung and approximately 200 g of liver and 100 g of bone are taken for radioanalysis. Bone samples usually consist of sternum, although rib and vertebra have been sampled also. All prominent tracheobronchial lymph nodes in the vicinity of the carina are sampled routinely.

Samples are dry ashed and then brought into solution with nitric acid. Because of problems associated with relatively large amounts of mineral salts, bone samples are usually taken into solution and a portion is passed through an ion exchange column and finally through the remainder of the process. Other tissue samples are processed in total. The plutonium is isolated by lanthanum fluoride coprecipitation,

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* This paper is based on work performed under U.S. Atomic Energy Commission Contract AT(45-1)-1830.

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TTA (thenoyltrifluoroacetone) extraction, and prepared for counting by electrodeposition on stainless steel disks. The final preparation may be counted electronically for alpha particles if sufficient plutonium is present or analyzed by autoradiography and alpha track counting after the method of SCHWENDIMAN *et al.*[2] for increased measurement sensitivity. Although reporting is usually given in terms of ^{239}Pu, the process yields total plutonium alpha.

Detection capabilities for plutonium in tissue samples have varied over the years and have been particularly sensitive to the alpha tracks in the film emulsion due to background sources. Prior to 1954, the detectable amount of plutonium was nominally taken to be 0.3 disintegrations per minute (d/m) or about 0.1 pCi. From 1954 to 1958 a detection limit of 0.05 d/m per sample was used. During the period 1958 through 1964, 0.025 d/m per sample was considered detectable. Since that time a nominal minimum reporting limit of 0.05 d/m per sample has been adopted.

RESULTS AND DISCUSSION

Analytical results for samples obtained through late 1970 have been tabulated in detail by NELSON *et al.*,[3] for lung, liver, bone and tracheobronchial lymph nodes. Cases were segregated according to whether the individual had a history of employment in Hanford plutonium facilities, other Hanford employment, was a local resident for more than 3 yr, was a local resident for less than 3 yr or was a non-resident. Those with a history of employment in Hanford plutonium facilities are, elsewhere in this paper, also referred to as Plutonium Workers. In reality, this group includes those having a significant potential for exposure to plutonium such as process operators, maintenance personnel and radiation monitors, and in addition includes patrolmen, engineers, office workers and others engaged in non-plutonium work who nevertheless had access to the plutonium facilities and could have been exposed to plutonium aerosols.

Detailed work histories, including incidents of known exposure to plutonium and results of bioassay sampling, are available for many of the plutonium workers. As older and sometimes vague work histories continue to be investigated, it is possible that a few individuals may be reclassified. It may also be necessary to reclassify some non-occupationally exposed individuals if investigation reveals early Hanford employment. Computerized record keeping in recent years has greatly simplified the search for evidence of Hanford employment. Investigation for possible employment prior to that time is laborious.

Since only portions of organs were sampled and in most cases the entire organ was not weighed, it was considered more appropriate to report the results of the radioanalyses in terms of concentration, i.e. fCi ^{239}Pu/g. Sample weights were obtained by weighing the frozen specimens after thawing. Although imprecise due to differences in organ weights among individuals and other factors, the use of standard man organ weights, together with the reported concentrations, permits some indication of plausible total organ burdens.

The data presented by NELSON *et al.*[3] reveal a large number of small positive results, a larger number of values reportable only as less than some variable amount, and a few larger measured values. While a large number of small or not significantly measurable results support a conclusion of adequate controls on releases of plutonium, they are cumbersome to summarize and present adequately. Further, division of the analytical results by highly variable sample weights results in reported values having apparently grossly different detection limits. Selected cases have been taken from the larger tabulation of data for presentation in this report. Such cases were included if the sample weight had been measured and if one of the tissue sample analyses was greater than an arbitrarily selected 5 fCi Pu/g. Analytical results fitting those criteria for data reduction are presented in Tables 1 and 2. There, cases have been categorized as to Plutonium Workers, Other Hanford Workers, Local Residents and Non-Residents. No activity entry indicates that no sample was taken. It may be noted that a sustained concentration of 5 fCi Pu/g in soft tissue will result in a calculated dose of about 5 mrem/yr to that tissue and the same concentration of Pu in bone will result in a calculated dose of about 25 mrem/yr to bone.

Table 1 reveals that there were 16 cases out

Table 1. Concentration of plutonium in tissue samples from Hanford workers

Case No.	Years Potential Pu exposure	Sex	Age at death	Cause of death or major disease	Plutonium concentration, fCi/g: Tracheo-bronchial lymph nodes	Lung	Liver	Bone
				Hanford plutonium workers (selected* from 71 cases)				
1450	4	M	50	Tuberculosis, pericarditis		<6	5.9	
0256	7.5	M	63	Pulmonary emphysema		11	34	<2
4858	13	M	56	Coronary heart disease		9.2	7.6	<2
2661	0.5	M	54	Coronary heart disease	9.0	0.78	4.8	<4
3162	10	M	54	Skin burns	60	34	8.3	<2
5362	6	M	51	Coronary heart disease	7.8	25	8.0	<3
1963	4	M	62	Pulmonary emphysema	<3	0.6	7.9	<2
H100	25	M	55	Coronary heart disease	23,000	170	42	220
				Others with Hanford work experience (selected* from 89 cases)				
0158		M	52	Carcinoma of prostate		9.1	24	4.4
2459		M	65	Postoperative—prostatectomy	<2	0.91	5.0	<3
3660		M	62	Amyotrophic lateral schlerosis	<4	0.84	<0.4	6.1
4963		M	71	Acute myocarditis	7.4	0.33	0.48	<1
2164		F	57	Coronary heart disease	5.9	0.84	0.28	14
2264		M	63	Cerebral thrombosis	30	0.17	2.4	<2
2665		M	69	Ruptured aortic aneurysm	4.1	1.1	0.46	125
1269		M	56	Coronary heart disease	9.5	<0.05	0.34	<7

* See text for selection criteria.

Table 2. Concentrations of plutonium in tissue samples from non-occupationally exposed persons

Case No.	Sex	Age at death	Cause of death or major disease	Plutonium concentration, fCi/g: Tracheo-bronchial lymph nodes	Lung	Liver	Bone
			Local residents of more than 3 yr (selected* from 146 cases)				
0258	M	66	Coronary heart disease		<0.2	0.54	7.7
1959	F	12	Drowning	13	<0.3	0.77	<4
3461	M	40	Cerebral hemorrhage	5.4	0.63	<0.7	<3
2165	F	67	Bronchogenic carcinoma	140†	<0.2	0.68	<9
3967	F	61	Pulmonary emphysema		<0.05	1.0	5.2
4167	F	53	Mitral stenosis		<0.04	0.18	5.7
4367	M	49	Acute liver failure		<0.04	0.20	9.3
			Individuals not residing locally (selected* from 50 cases)				
1050	M	34	Trauma		<4	6.5	
1657	F	59	Generalized lymphosarcoma		1.8	<0.8	5.7
0764	M	82	Chronic lymphocytic leukemia	68	<0.1	<0.4	<2
3264	F	70	Carcinoma of kidney	6.3	0.31	0.70	<0.5
3664	F	50	Carcinomatosis	6.0	0.69		
4867	F	36	Scleroderma	6.2	0.63	<0.08	<7

* See text for selection criteria.

† Result is questionably high due to erratic laboratory background at time of analysis and small sample size (0.2 g). Under usual conditions the minimum reporting limit for this sample size would be 110 fCi/g.

of 160 or 10% of the occupationally-exposed individuals having at least one tissue sample for which a concentration of 5 fCi Pu/g was measured. Of the 16 occupationally-exposed individuals, one-half had no confirmed exposure to plutonium. Further investigation into work histories for these individuals may reveal a potential for exposure to plutonium not presently known. Unrecognized laboratory contamination could possibly account for some positive results. It is not considered likely that these results are due to fallout from weapons testing since for the period of interest, expected concentrations of plutonium in lung would amount to less than 0.5 fCi Pu/g as calculated by NEWTON *et al.*[1] The conspicuously large results for Case H100 will be discussed later.

As shown in Table 2, there were 13 individuals out of 193 having no known occupational exposure to plutonium for whom a concentration of 5 fCi/g or more in at least one tissue sample is reported. Again, it is possible that some of these individuals will be shown eventually to have worked in a plutonium facility at some time. The reason for the relatively frequent positive measurements of plutonium in bone, assuming that these individuals were indeed not exposed to occupational sources of plutonium, is unknown. Contamination of the samples may be indicated, since the four results reported for 1967 were all processed on the same slide at the same time.

Case H100 is unique in that it represents the first case for which significantly measurable amounts of plutonium were anticipated and a thorough sampling was undertaken. This case is associated with long-term experience in plutonium facilities and the individual was involved in a number of incidents involving exposure to plutonium or to the potential for exposure. In addition to a detailed work history, considerable bioassay data exist for this individual. Because of the extensive work history of this case, a much more detailed analysis will be made of exposure, bioassay results and ultimate body burden and presented elsewhere. Only a few highlights of this case will be presented here for comparison.

Some pertinent detailed data for case H100 are presented in Table 3. There the concentrations of plutonium in various tissue samples are presented in descending order of concentration. At least in terms of total plutonium per gram of tissue, the importance of the lymph nodes in an autopsy sampling program is strikingly confirmed in this case. Based on these results, plutonium occurs in relatively small amounts in the lung parenchyma. The importance of lung and liver samples, however, is substantiated in Table 1, where significantly measurable quantities of plutonium in these tissues occur frequently. Positive measurements found in the usual lung samples may be due to the presence of plutonium in the pulmonary lymph nodes, which are not usually removed and analyzed separately. The occurrence of plutonium in the liver and/or bone, but without measurable plutonium in the lung or tracheobronchial lymph nodes, is suggested as due to entry via contaminated puncture wounds or lacerations.

Table 3. *Concentrations of plutonium in selected tissue samples from case* H100

Tissue	Plutonium concentration, fCi/g	Fresh wt, g
Carina lymph node	61,000	1.10
Intrapulmonary lymph nodes	20,000	2.2
Hilar lymph nodes, right	12,000	1.2
Other tracheobronchial lymph nodes and adjacent connective tissue	2400	1.44
Pleura and sub-pleura left lung	520	148*
Vertebrae (lumbar)	340	103.7
Hepatic lymph node	180	2.2
Rib	99	113.3
Liver	42	259.3
Parenchyma left lung	8.8	562*

* Formalin fixed weight.

Except for Case H100, the only case for which the bioassay program was found to yield positive results was Case 5362. As reported by NEWTON *et al.*,[1] three positive urine analyses for plutonium were obtained for this individual (Case #28/532 in that work) which suggested a body burden of 0.4 nCi. They also reported that, based on autopsy results, the individual's total body burden was calculated to be less than

0.06 nCi. The infrequent occurrence of cases in our autopsy series with a record of both occupational exposure and positive bioassay results, emphasizes the importance of maximizing the number of autopsies and the need to maintain such a program over many years.

Using standard man parameters and 0.02 d/m plutonium/10^3 ft^3 of air as representative of plutonium in air due to fallout, NEWTON *et al.*[1] calculated a lung burden of 1 d/m for continuous inhalation. An airborne concentration of 0.02 d/m/10^3 ft^3 is equivalent to about 0.35 fCi/m^3. Fallout since 1964 has tended toward smaller values of plutonium in air and in late 1970 was on the order of 0.05 fCi/m^3 according to the Environmental Protection Agency.[4] As a result of this decline, no new calculation of expected lung burden was made and a much rounded concentration of 0.5 fCi Pu/g of lung is taken as attributable to fallout.

The two principal routes for occupational exposure to plutonium are inhalation of plutonium aerosols and absorption of plutonium through contaminated wounds. In the former case, when the aerosol consists largely of relatively insoluble plutonium dioxide particles, the highest concentrations of plutonium might be expected in the tracheobronchial lymph nodes and lesser concentrations in lung, liver and bone, in that order, based on beagle dog data of PARK *et al.*[5] In the case of a predominantly plutonium nitrate aerosol, dog data obtained 30 days after exposure indicates nearly equal plutonium concentrations in lungs and tracheobronchial lymph nodes, with smaller concentrations in liver and bone.[6] Dogs sacrificed 100–300 days after exposure to a plutonium nitrate aerosol showed highest concentrations of plutonium in the tracheobronchial lymph nodes and lesser concentrations in lung, liver and bone, in that order.[7] In the case of wounds contaminated with plutonium nitrate-like material, intravenous injection studies with plutonium nitrate in dogs suggest that liver and spleen might contain the highest concentrations of plutonium followed by tracheobronchial lymph nodes, bone and lung, in that order.[6] Because an individual may be exposed to different compounds and different modes of intake and at different times during his work experience, it is not surprising that the order of concentration of plutonium in tissues, as suggested by animal experiments, is followed in only a few human cases.

CONCLUSIONS

The reported measurements of plutonium in tissue samples obtained at autopsy from former Hanford employees and residents in the plant environs continue to demonstrate that, while for some workers measurable plutonium does occur in the body, the majority of workers and residents have not received significantly measurable amounts of plutonium. In no case was plutonium found in tissue samples in an amount that would suggest an annual dose to the tissue exceeding the recommendation of the National Council on Radiation Protection and Measurements of 15 rem/yr to specific organs for those occupationally exposed and 0.5 rem/yr to non-occupationally exposed individuals in the general population.[8]

In cases of significant occupational exposure to plutonium, the results of analyses at autopsy dramatically identify preferential sites of plutonium retention in the body and provide a basis for evaluating the precision of bioassay and *in vivo* counting as tools for estimating amounts of plutonium internally deposited.

Acknowledgements—The authors wich to express their gratitude to the many individuals who have assisted in this program over the years. In particular, recognition is due to W. D. NORWOOD for continuing advice and counsel, to J. F. PARK for special tissue preparations, to S. MARKS for many of the autopsy samples in early years, to J. A. NORCROSS and C. E. NEWTON, JR. for assistance from the U.S. Transuranium Registry, to V. W. THOMAS, JR. for radiochemical analyses, and to C. W. KIRKLIN for medical record assistance.

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