PLUTONIUM CONCENTRATIONS IN TISSUES OF OCCUPATIONALLY EXPOSED NUCLEAR WORKERS

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ABSTRACT

Tissues from one hundred seventy-five autopsy cases of former workers at various nuclear facilities have been analyzed for their plutonium content. Not all the individuals had worked directly with Pu since many were secretaries, clerks, truck drivers and other support personnel. The results indicate that approximately two-thirds of these cases have Pu depositions not much different than fallout levels of Pu in the U.S. general population. Estimated whole body depositions in the remaining cases range from about 0.05-50 nCi. Data from the 12 highest deposition cases are compared with the in vivo estimates from urinalysis data. In general, the urinalysis data give estimates that are high by factors of 2-3, or more. The observed variation in the whole body distribution suggests that each exposure incident is unique and must be evaluated on the basis of the Pu exposure parameters including the chemical form, solubility, particle size, mode of entry and duration of exposure. Several of the highest depostion cases are presented to illustrate the variations observed. Estimates of Pu particle size distributions in the lung and tracheobronchial lymph nodes of one case are given.

I. Introduction

The Los Alamos National Laboratory established a program in 1959 to collect tissue specimens from routine post mortem examinations for radiochemical analyses of the tissues for their plutonium content. Among the samples collected since that time were tissue specimens from 125 former employees of the Los Alamos Laboratory. Not all employees had worked directly with plutonium since many were secretaries, clerks, truck drivers and other support personnel. In addition to the Los Alamos employees, our laboratory has analyzed 31 autopsy cases, including one whole body, for the U.S. Transuranum Registry (slide 1). These cases include participants in the Registry from all parts of the U.S. that have been identified as "Pu workers". Most had been associated with the Hanford Facility, Mound Laboratories, and the Savannah River Plant. Tissues from the U.S. T. R. members working at the Rocky Flats Plant are analyzed at that facility, although we have analyzed two cases for them. Fifteen cases, received from participating hospitals in the Augusta, Georgia area, and originally thought to be general population cases, have been identified as having worked for varying lengths of time at the Savannah River Plant. One case analyzed was identified as having worked at Oak Ridge and one member of the U.S. Armed Forces, stationed at the Nevada test site, was also analyzed.

Only the results of the Los Alamos workers will be discussed in this paper.

Materials and Methods

The radiochemical procedures used for analyzing the tissues for Pu have been reported elsewhere (Mc 79, BO 81). Briefly, the tissues are weighed, ashed, and dissolved in acid. Aliquots of the solutions are selected, run through ion exchange columns to isolate the Pu and the radioisotope is electrodeposited onto stainless steel planchets for alpha pulse height analysis. Each tissue solution is spiked prior to ion exchange with tracer quantities of ^{242}Pu to monitor the recovery of the Pu from that portion of the analytical procedure. Many years of experience from spiking tissues before ashing have provided data indicating that losses of Pu during the ashing and dissolution process are minimal. Recovery of the ^{242}Pu tracer has averaged $87 \pm 12\%$ for the last 2000 tissues analyzed.

The tissues collected during the autopsy include the lungs, tracheobronchial lymph nodes, liver, bone specimens (rib, sternum, vertebral body, and recently, patella), Kidney, thyroid, spleen, muscle, and blood. If the worker had a history of a contaminated puncture wound, tissue and lymph nodes from that area were obtained. If the individual had a tumor, this was also sampled and analyzed.

Sections of tissue from lung, lymph nodes and liver of the higher deposition cases were fixed in formalin solution and sent to a pathology laboratory for sectioning, staining and autoradiography. Techniques, using microscopic examination of the autoradiographs for alpha tracks, have provided information on the Pu particle size distribution in the lung and lymph nodes (Mc 76).

Results and Discussion

The liver concentrations of Pu in the Los Alamos nuclear workers were compared with the concentration observed in 700 cases analyzed from the U.S. general population from 1959-1976 (MC79). Ninty five percent of the general population cases, exposed primarily to atmospheric fallout, had liver concentrations less than 2.3 pCi/kg tissue wet weight. Sixty five percent of the worker cases in this study had liver concentrations that fell into this range. (slide 2). An additional 24%, (27 cases) had liver concentrations in the range of 2.4 to 36 pCi/kg. The remaining 12% of the cases analyzed had liver concentrations between 2000 and 4000 pCi/kg tissue, and all had well documented exposure histories.

One of the objectives of this study is to provide data on the tissue retention and distribution of Pu in nuclear workers and to compare the extrapolated whole body contents with _in vivo_ measurements, obtained primarily by urinalysis. Slide 3 compares the calculated Pu burdens in the five highest deposition cases analyzed. The urine estimate, based upon modifications of the Langham excretion curve (PUQFUA 2) tends to overestimate the systemic burden by factors of 1.1 to 3.5 (mean = 2.5 ± 0.9) but agrees fairly well with the estimated whole body burden (mean = 1.4 ∓ 1.0). Slide 4 shows similar data for the next seven highest deposition cases (range 0.5 - 9.1 nCi). The urine body burden estimates range from 0.8 to 8.1 nCi (mean = 3.4 $\pm$ 2.5) times the systemic burden and 0.8 to 6.5 times the whole body burden (mean = 2.9 $\pm$ 2.8). The computer code currently in use at Los Alamos has used the tissue data over the years to improve the estimates.

One obvious problem with the tissue analysis data for estimating whole body deposition is the small size and type of the bone specimens received. The skeleton represents a major deposition site for Pu in the body. Since Pu deposits primarily on the bone surface, the distribution of Pu throughout the skeleton is not uniform, being more heavily deposited in the trabecular bone. The bones specimens received from an autopsy are generally small (100-200g) and are usually highly trabeular bone. This makes it very difficult to extrapolate to the entire skeleton (10,000g in standard man). Currently there is almost no data available on the distribution factors for Pu in the various bones of the human skeleton. The U.S.T.R. has a study in which they are soliciting participants having significant depositions of Pu to donate their whole body for detailed analyses. We have recently finished the analysis of the first whole body donated to the registry. However, this individual was exposed primarily to ^{241}Am. It is hoped that several whole bodies with Pu depostions will be analyzed to provide the data necessary to make realistic estimates of skeletal depositions from the smaller anatomical specimens available from autopsy.

The relative distributions of Pu in the bodies of the four of the higher deposition cases are shown in slide 5.

Case 1-039 was a 39 year old male chemical technician that died shortly after his exposure during a criticality accident in 1958. This individual was exposed by chronic inhalation of moderate levels of airborne plutonium in various chemical forms from June 1946 to December 1958, while working as a plutonium chemical operator. He had four cuts or puncture wounds during this time with alpha counts detected on only one (August, 1948). The highest fraction (45%) of the retained Pu was estimated to be in the skeleton with the extrapolation made from anlyses of vertebral bodies (180g), a rib (21g) and the sternum (122g). The difficulties and uncertainties associated with estimates of skeletal burdens from small anatomical bone specimens has been discussed earlier in this paper. Thus, the total whole body burdens are in question and the relative percent distributions may be in some error. Not until several whole bodies of former plutonium workers have been analyzed to determine the deposition and retention of plutonium in the various bones of the body, will we be able to improve our estimates.

Of the soft tissues, the liver contained 40% of the whole body burden, the kidneys, spleen, muscle and heart collectively had about 1% of the body burden and the lungs and tracheobronchial lymph nodes had a total of 14% of the burden. Definition of the depostion between the lungs and the various plulmonary lymph nodes is also very difficult because of the inability to dissect all of the lymphatics from the lung. Therefore, all lung concentrations in this study, and all other reported human data that we are aware of, are not from just lung tissue alone but also include the many small pulmonary lymph nodes within this organ. The tracheobroncheal lymph nodes are usually relatively large and easily identified so that one or more can be excised and analyzed separately. However, seldom are all of these nodes removed and sent to us by the pathologist. Therefore, it is probably more reasonable to combine the lung and lymph node contents and identify the total as the "pulmonary" burden for the purpose of evaluating whole body distributions.

Case 7-138 died in a automobile accident at the age of 46. He had been exposed to chronic inhalation of moderate levels of airborne plutionium of various chemical forms from October, 1948 to March of 1973 while employed as a health physics surveyor in a plutonium processing facility (2 years) and as a plutonium chemistry technician (22 years). Two plutonium contaminated wounds were excised on February 11, 1969 and on June 3, 1970. He was in a room that had a plutonium drybox fire without a respirator (February, 1969) but subsequent nose counts from nasal smears were low. He was restricted from plutonium work in March of 1970 because in vivo estimates of his lung burden by chest counting was high (28 nCi). He was permitted to work with plutonium again in July of 1971 after his chest counts were less than the minimum detectable level. The largest deposition of plutonium measured following his death was in the lungs and lymph nodes (81% of total). The skeleton was estimated to contain 11%; the liver 7%; and the remaining soft tissue (muscle, spleen, thyroid, brain, and testes) contained 1% of the whole body deposition of plutonium.

Case 3-016 died at the age of 51 from a brain tumor. He had been exposed from chronic inhalation of low-to-moderate levels of airborne plutonium of various chemical forms from 1946 to 1965 while working as an analytical chemist in a plutonium laboratory. No plutonium wounds were recorded. The largest deposition of plutonium was found in the liver (54%). The skeleton contained an estimated 12%; the lungs and tracheobroncheal lymph nodes contained 33% and the kidney had less than 1% of the plutonium burden.

Case 18-026 is of special interest because this individual was a member of the military assigned to the Manhattan Project at Los Alamos in 1945. He received his only known exposure to plutonium during his one-year employment at Los Alamos where he was involved in plutonium fluoridation. He later entered medical school and began practicing medicine in 1950. He was included in a long term study of 26 early Los Alamos plutonium workers (V079). These subjects, who worked with plutonium during World War II under extraordinarily crude working conditions, were followed medically for a period of 32 years.

This individual died at the age of 50 from injuries sustained in an automobile-pedestrain accident. The primary route of his exposure was probably by inhalation. This hypothesis was supported by the finding of about 75% of the whole body deposition of plutonium in the lungs and tracheobronchial lymph nodes. Ten percent of the plutonium was in the skeleton and 10% was in the liver. The remaining 5% was found in the other soft tissues analyzed (spleen, muscle, thyroid, testes, brain, blood and skin, in order of decreasing concentrations). Of note is the relatively high concentration of plutonium in a piece of the calvarium (skull cap) as compared to the usually received bone specimens (vertebral wedge, rib, sternum and femur). This may prove significant in the in vivo estimates of skeletal deposition by counting the activity in the head and extrapolating the results to the whole skeleton. Again, the importance for obtaining whole bodies so that skeletal distributions of plutonium can be determined is emphasized.

Autoradiographs made from sections of the lung and tracheobroncheal lymph nodes from case 7-138 have been used to estimate the particle size distribution of Pu in these tissues (MC76). The activity median diameter of the particles in the lung was 0.16 um (GSD = 1.4); in the lymph nodes, it was 0.32 um (GSD = 1.5). The larger size in the lymph nodes was probably due to aggregation of the particles during translocation from the lung.

Summary

Data, using the liver concentration of plutonium as an index of exposure, indicate that the majority of personnel working at the Los Alamos National Laboratory do not have depositions of plutonium that greatly exceed that of the U.S. general population. Current in vivo estimates of the whole body deposition of plutonium, based upon urinalysis, are relatively close to the whole body content estimated from tissue analysis results. The observed variation in the distribution of plutonium among the various tissues analyzed suggests that each exposure incident is unique and must be evaluated on the basis of the plutonium exposure parameters including the chemical form, solubility, particle size, mode of entry and duration of exposure.