



**Defense Threat Reduction Agency
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TECHNICAL REPORT

Plutonium Bioassay Testing of U.S. Atmospheric Nuclear Test Participants and U.S. Occupation Forces of Hiroshima and Nagasaki, Japan

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14. ABSTRACT This report documents DTRA's study conducted from 1994 to 2004 to develop a urine bioassay program and investigate the feasibility of this method to complement the atomic veteran dose reconstruction under the Nuclear Test Personnel Review Program. Brookhaven National Laboratory (BNL) used fission track analysis (FTA) to estimate doses to the Marshallese Island population. In this study, BNL's FTA method was evaluated to determine if it can be applied to atomic veterans by measuring plutonium in urine samples. Guided by Congressional legislation (PL 105-85), DTRA conducted a pilot study involving 100 atomic veterans to see if expanded use on atomic veterans was feasible. The activities in this study consisted of establishing a calibration system for measuring plutonium content in urine samples; determining the accuracy, repeatability, and sensitivity of FTA; and measuring background plutonium levels in a representative U. S. population not involved with nuclear weapons testing or plutonium work. The results for the 100 atomic veterans were compared to those of the unexposed population, and veterans with results that were statistically above background were identified. Finally, bioassay results were compared to doses estimated by reconstruction.					
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UNIT CONVERSION TABLE

U.S. customary units to and from international units of measurement*

U.S. Customary Units	Multiply by Divide by †	International Units
Length/Area/Volume		
inch (in)	2.54 × 10 ⁻²	meter (m)
foot (ft)	3.048 × 10 ⁻¹	meter (m)
yard (yd)	9.144 × 10 ⁻¹	meter (m)
mile (mi, international)	1.609 344 × 10 ³	meter (m)
mile (nmi, nautical, U.S.)	1.852 × 10 ³	meter (m)
barn (b)	1 × 10 ⁻²⁸	square meter (m ²)
gallon (gal, U.S. liquid)	3.785 412 × 10 ⁻³	cubic meter (m ³)
cubic foot (ft ³)	2.831 685 × 10 ⁻²	cubic meter (m ³)
Mass/Density		
pound (lb)	4.535 924 × 10 ⁻¹	kilogram (kg)
atomic mass unit (AMU)	1.660 539 × 10 ⁻²⁷	kilogram (kg)
pound-mass per cubic foot (lb ft ⁻³)	1.601 846 × 10 ¹	kilogram per cubic meter (kg m ⁻³)
Pound-force (lbf avoirdupois)	4.448 222	Newton (N)
Energy/Work/Power		
electronvolt (eV)	1.602 177 × 10 ⁻¹⁹	joule (J)
erg	1 × 10 ⁻⁷	joule (J)
kiloton (kT) (TNT equivalent)	4.184 × 10 ¹²	joule (J)
British thermal unit (Btu) (thermochemical)	1.054 350 × 10 ³	joule (J)
foot-pound-force (ft lbf)	1.355 818	joule (J)
calorie (cal) (thermochemical)	4.184	joule (J)
Pressure		
atmosphere (atm)	1.013 250 × 10 ⁵	pascal (Pa)
pound force per square inch (psi)	6.984 757 × 10 ³	pascal (Pa)
Temperature		
degree Fahrenheit (°F)	[T(°F) – 32]/1.8	degree Celsius (°C)
degree Fahrenheit (°F)	[T(°F) + 459.67]/1.8	kelvin (K)
Radiation		
activity of radionuclides [curie (Ci)]	3.7 × 10 ¹⁰	per second (s ⁻¹ ‡)
air exposure [roentgen (R)]	2.579 760 × 10 ⁻⁴	coulomb per kilogram (C kg ⁻¹)
absorbed dose (rad)	1 × 10 ⁻²	joule per kilogram (J kg ⁻¹ §)
equivalent and effective dose (rem)	1 × 10 ⁻²	joule per kilogram (J kg ⁻¹ **)

* Specific details regarding the implementation of SI units may be viewed at <http://www.bipm.org/en/si/>.

† Multiply the U.S. customary unit by the factor to get the international unit. Divide the international unit by the factor to get the U.S. customary unit.

‡ The special name for the SI unit of the activity of a radionuclide is the becquerel (Bq). (1 Bq = 1 s⁻¹).

§ The special name for the SI unit of absorbed dose is the gray (Gy). (1 Gy = 1 J kg⁻¹).

** The special name for the SI unit of equivalent and effective dose is the sievert (Sv). (1 Sv = 1 J kg⁻¹).

DTRA-TR-15-058: DRAFT Plutonium Bioassay Testing of U.S. Atmospheric Nuclear Test Participants and U.S. Occupation Forces of Hiroshima and Nagasaki, Japan

Table of Contents

List of Figures ii

List of Tables ii

Executive Summary 1

Section 1. Introduction 2

 1.1 Requirements for an NTPR Plutonium Bioassay Program 2

 1.2 Approach to NTPR Plutonium Bioassay Testing 3

Section 2. Methods for Plutonium Measurement, Data Analysis, and Validation 6

 2.1 Technical Basis for Performing Urine Bioassay 6

 2.2 BNL FTA Analysis Method 6

 2.3 Methods for Analyzing Bioassay Data 7

 2.3.1. Linear Regression Approach 7

 2.3.2. Lognormal Regression Correction Approach 9

 2.3.3. Definition and Determination of Minimum Detectable Activity (MDA), Lower Limit of Detection (LLD), and Decision Level (DL) 10

 2.4 Measurement Validation of FTA Plutonium Bioassay 11

 2.4.1. Comparison of Results of Analyses of Plutonium in Urine Samples by Two Laboratories 11

 2.4.2. Analysis of Comparison Results 11

Section 3. Plutonium in the Urine Samples of U.S. General Population and U.S. Atomic Veterans 13

 3.1 Plutonium Measured in Urine of Selected Members of the U.S. General Population 13

 3.2 Results for Initial Phase of Measurements on Veterans 14

 3.3 Results of Resampling of 12 Selected Veterans 16

Section 4. Discussion of the Results 18

 4.1 Interpretation of the Results 18

 4.2 Comparison of Bioassay Measurements and Dose Reconstructions 18

Section 5. Conclusions 20

Section 6. References 22

Appendix A. Compilation of Project Bioassay Test Results 25

List of Figures

Figure 1. Typical linear regression calibration curve for FTA bioassay blanks and spikes.....	8
Figure 2. Spike and blank values used in calibration of the FTA method for calculation of ²³⁹ Pu activities in veterans' samples	9
Figure 3. Distribution of bioassay measurements in the U.S. general population group.....	14
Figure 4. Comparison of the distributions of bioassay results for the U.S general populaton and atomic veterans	15
Figure 5. Comparison of FTA and TIMS sample measurements for selected veterans with original FTA bioassay measurements above 100 aCi/sample.....	17

List of Tables

Table 1. Results of ²³⁹ Pu measurements by LANL and BNL	12
Table 2. Locations and numbers of subjects contributing urine samples for the general population group.....	13
Table 3. First and second split measurements for 11 selected bioassays with original readings above 100 aCi/sample	16
Table 4. Comparison of bioassays for veterans followed-up with TIMS	17
Table 5. Comparison of bioassay results with reconstructions of internal dose.....	19
Table A-1. Results of bioassay testing of the selected U.S. population group.....	25
Table A-2 . Results of bioassay testing of 100 eligible veterans.....	26

Executive Summary

This technical report describes the scientific and technical processes employed to conduct a pilot urine plutonium bioassay testing program on 100 atomic veterans using Brookhaven National Laboratory's (BNL's) Fission Track Analysis (FTA) technique. The report also discusses the methodology, collected data, scientific challenges, and test results of this pilot bioassay testing program. The Defense Threat Reduction Agency (DTRA) and its predecessor agencies conducted this program from 1994 to 2004 to determine whether it was feasible to use bioassay testing to supplement dose reconstruction techniques used to estimate atomic veterans' doses under the Nuclear Test Personnel Review Program.

Congressional action (PL 98-542, 1984, Sections 7.a.(2)(A), (B), and (C)) called for the evaluation of the reliability and accuracy of scientific and technical devices and techniques, and to consider their potential implementation for performing retrospective dose assessments on radiation-exposed veterans. National Research Council (NRC, 1985) also recommended bioassay testing of atomic veterans to assess the reliability of internal dose estimation. The results of the evaluation study (Boecker et al., 1991) recommended using the FTA technique already employed by BNL to assess doses to the Marshallese Islanders impacted by atmospheric nuclear testing. Following these findings, DTRA investigated the application of BNL's FTA technique to atomic veterans. Early test results showed unexplainable uncertainties in initial FTA tests on personnel with and without histories of Pu uptake. Further congressional action (PL 105-85, 1997) directed DTRA to conduct a pilot study involving 100 atomic veterans to better understand FTA uncertainties and repeatability difficulties, and to assess whether expanded use on veterans was feasible.

The pilot study of 100 veterans also included bioassay testing of a representative group of 58 persons from the general population who lived during the atmospheric nuclear test era but were not involved with the nuclear tests or radiation related occupations. The FTA technique is capable of measuring ^{239}Pu excreted in urine following intakes of plutonium in this population from worldwide fallout. It was essential that the technique be able to distinguish whether atomic veterans had plutonium intakes in excess of those from worldwide fallout estimates. This report shows that 85 of 100 veterans excreted low levels of ^{239}Pu similar to those detected in general population group. For the remaining 15 veterans, one result was confirmed as clearly positive. The other 14 veteran results raised concerns because of the large observed uncertainties in values ranging higher than those of the general population group. Subsequently, at DTRA's request, the Los Alamos National Laboratory used an alternate bioassay technique called thermal ionization mass spectrometry (TIMS) to independently replicate the tests on 12 of the 15 veterans. All were not available for resampling. The results of the TIMS tests showed that the ^{239}Pu results for the resampled atomic veterans were not above those for the general population group.

This study showed that FTA's ability to demonstrate positive or negative plutonium uptakes in atomic veterans was not credible. The conclusion was that FTA bioassay is not feasible to implement on a wide scale basis among atomic veterans or to complement the NTPR dose reconstruction program.

Section 1.

Introduction

This technical report describes the scientific and technical processes employed to conduct a pilot urine plutonium bioassay testing program on 100 atomic veterans¹. It summarizes the methodology, data that were collected during the course of the program, discusses scientific challenges that arose during its activities, reports the results of testing and provides an assessment of the applicability of the results to the atomic veteran population. This report concludes, consistent with National Research Council's findings (NRC, 2003), that urine ²³⁹Pu bioassay using FTA did not enhance the capability of DTRA's current scientific processes to perform accurate dose reconstructions for atomic veterans, and therefore, was not feasible for implementation on a wide scale basis for atomic veterans.

1.1 Requirements for an NTPR Plutonium Bioassay Program

In 1984, the Veterans' Dioxin and Radiation Exposure Compensation Standards Act (PL 98-542, 1984) called for the study of alternative scientific methods for performing retrospective dose assessments on radiation-exposed veteran populations. The National Research Council (NRC, 1985) also recommended bioassay testing of atomic veterans to assess the reliability of the internal dose estimates. The resulting study (Boecker et al., 1991) discussed plutonium bioassay as a potential means of retrospectively testing atomic veterans to detect and quantify uptakes of radionuclides from nuclear detonation debris and fallout. Of the several hundreds of possible inhaled or ingested radionuclides, ²³⁹Pu can be detected 35 or more years after intake in an excreted urine sample. The extremely long half-life and very low long-term excretion rate makes ²³⁹Pu the most suitable isotope of interest. The radionuclide ²³⁹Pu has been found in the environment since 1945 in extremely small quantities, which are orders of magnitude less than the quantities of natural uranium (including isotopes of interest such as ²³⁸U and ²³⁵U). Consequently, ²³⁹Pu identification by urinalysis was proposed as a marker for significant internal intakes of other associated radionuclides in nuclear weapons debris due to its low natural background. However, intakes of ²³⁹Pu in worldwide fallout can be measured in the urine of persons who lived during the atmospheric nuclear testing era and can confound the results of veteran-supplied bioassay samples if not appropriately considered.

In the mid-1990s, representatives from the Department of Veterans Affairs (VA) Veterans Health Administration and Defense Nuclear Agency² (DNA), the Defense Threat Reduction Agency's (DTRA's) predecessor agency, discussed the feasibility of applying techniques described in Boecker et al. (1991) to the atomic veteran population. VA ceded proponenty to DTRA for initiating a feasibility program because it was better suited to DTRA's mission under the Department of Defense (DoD) Nuclear Test Personnel Review (NTPR) Program. The technique found most promising was fission track analysis (FTA) being used by

¹ Atomic veteran is the term that refers to veterans who participated in U.S. nuclear tests and the post-World War II occupation of Hiroshima and Nagasaki, including liberated POWs.

² Hereinafter, DNA is referred to with the current agency's name, DTRA.

Brookhaven National Lab (BNL) to assess ^{239}Pu content in urine samples taken from the Marshallese Island population impacted by U.S. atmospheric nuclear testing (Sun et al., 1993). Department of Energy (DOE) was already investigating this technique as a possible way to estimate long-term population radiation doses of this population (Moorthy et al., 1988).

DTRA initiated tasks with BNL to enhance the sensitivity of FTA and adapt the experiences of the Marshallese Island urine ^{239}Pu bioassay sample collection methods to suit a nationally-distributed atomic veteran population that did not reside in one geographic location. By 1998, BNL had refined its FTA technique to detect low masses of Pu (equivalent to activities of tens of attocuries (aCi³) in a 24-hour urine sample from members of the general U.S. population exposed to worldwide fallout (Pietrzak et al., 1998). Based on this improvement in assay sensitivity, the NTPR Program selected BNL to measure ^{239}Pu levels in veteran urine samples using the FTA technique. Equally important was establishing the lowest level of detection benchmark to determine whether results of bioassay samples from the U.S. general population could be distinguished from results of synthetic urine blanks representing no plutonium level.

While the initial BNL work was underway, Congress expressed urgency to begin urine plutonium bioassay testing of atomic veterans. Congress learned that DTRA was contemplating curtailment of the BNL effort due to large anomalous uncertainties and difficulties with measurement repeatability being uncovered. The length of time to understand and resolve these issues was inestimable. Congress, responding by enacting legislation as part of the National Defense Authorization Act for Fiscal Year 1998 (PL 105-85, 1997), proposed a limited pilot test program as a way forward to ascertain the practical logistics of collecting samples from atomic veterans and to further investigate the technical problems. In addition, the limited program would permit assessing the feasibility of offering testing to a wider range of atomic veterans.

Consequently, bioassay testing was offered to the first 100 eligible veterans who contacted the NTPR Program after 1 June 1999 when the pilot program was publicly announced (DTRA, 1999). The NTPR program confirmed veteran eligibility by confirming their participation in U.S. atmospheric nuclear testing (1945 to 1962) or the post-war occupation of Hiroshima and Nagasaki, Japan. The VA assisted with collection and custody control of the bioassay samples sent to BNL for testing. DTRA, Science Applications International Corporation (now Leidos), and BNL formed a cooperative team to analyze, interpret, and report bioassay data and results.

1.2 Approach to NTPR Plutonium Bioassay Testing

The testing program encompassed two phases: the first being work conducted to fulfill the requirements under PL 98-542 and the second being work directed under PL 105-85 to perform a limited pilot testing of atomic veterans to determine the feasibility of continued testing on a wide scale basis. DTRA selected Pu bioassay as a potential method to support retrospective dose assessment for the following reasons:

- The radionuclide ^{239}Pu was the only remaining long-lived and very slowly-excreted isotope out of several hundred still being excreted by atomic veterans 35 to 50 years after an intake of nuclear test debris that could be measured;

³ 1 aCi = 10^{-18} Ci

- FTA was sufficiently sensitive to measure low levels of ^{239}Pu in urine samples of veterans collected 35 to 50 years after an intake;
- The BNL technique could be adapted to atomic veteran sampling based on the laboratory's experiences in collecting urine samples from Marshallese Islanders; and
- DTRA had already collected a wealth of dose reconstruction results on atomic veterans which could be used for comparisons with bioassay results to assess the feasibility of plutonium bioassay as a dose assessment tool.

Before embarking on an effort to sample urine of atomic veterans to analyze for low-level plutonium, DTRA found it essential to perform the following technical tasks in carrying out the charge of PL 105-85:

- Determine the lower limit of detection (LLD) for FTA detection of ^{239}Pu ;
- Determine that urine samples without plutonium can be measured as negative (zero level) quantities with reliability and repeatability;
- Establish a measurement benchmark by determining if samples of the U.S. general population living during the atmospheric test era could be measured as positive quantities above the FTA technique's LLD; and
- Determine if the ^{239}Pu content can be measured accurately in urine samples taken from persons already known to have well-characterized body burdens from occupational exposures.

After Congress directed the performance of bioassay sampling on a limited pilot test basis, the tasks were expanded to include the following:

- Establish and maintain standard measurement and calibration protocols to reliably determine plutonium content in bioassay samples collected from selected test populations;
- Determine by FTA the activity concentrations of ^{239}Pu excreted by a selected U.S. population having no history of working with plutonium or performing similar activities as veterans eligible for bioassay testing;
- Establish and schedule chemical, reactor irradiation, and counting procedures for batch processing urine samples in small groups and reporting results;
- Compare background and veteran sample measurements against the FTA technique's minimum detectable activity (MDA) and decide whether results are statistically positive;
- Interpret measurement results with respect to NTPR program-developed veteran dose reconstructions for possible correlations; and
- Corroborate measurements using other comparable urine ^{239}Pu bioassay testing techniques if bioassay test results require further investigation.

This technical report covers the FTA measurement process, the methods used to analyze the bioassay results, FTA measurement validation, the characterization of results from the U.S. general population samples, the results of the 100-sample atomic veteran population, the discussion of the results, and conclusions. The report authors considered the use of SI units throughout this technical report, but their decision was to remain with the legacy units, for

example attocurie (aCi) vs. microbecquerel (μBq). All of the working papers associated with the DTRA urine bioassay pilot program and some of the technical references supporting this technical report contain legacy units. Thus, the authors retain use of legacy units throughout the report for clarity and linkage to past work in this field of study.

Section 2.

Methods for Plutonium Measurement, Data Analysis, and Validation

This section summarizes the method BNL used to measure plutonium in urine samples collected from both veterans and the general population (Pietrzak et al., 1998, Kaplan, 1996, and Sun et al., 1993). The measurement of ^{239}Pu in urine from both populations is based on the possibility that there is sufficient plutonium contained in debris from nuclear detonations that can be inhaled or ingested, deposited in bodily tissues and organs, and retained there to be measured in urine samples collected many years later (Boecker et al., 1991).

2.1 Technical Basis for Performing Urine Bioassay

Intake models (ICRP, 1994a and b) formulate retention and excretion relationships that can be applied to internalized plutonium to predict the amount excreted in urine. Typically, about 1/100,000 of the original amount of a ^{239}Pu intake is predicted to be excreted daily 30–60 years after a detonation (Leggett, 1984 and Jones, 1985). The FTA methodology is sufficiently sensitive to measure amounts of plutonium as low as a few 10s of aCi (Sun et al., 1997). The specific plutonium isotope of interest for bioassay testing is ^{239}Pu . Although ^{240}Pu , another isotope in weapons grade plutonium, is important from a health perspective, its presence within a given urine sample being analyzed by FTA can only be inferred from ^{239}Pu content.

All urine samples collected from 100 veterans and the general population were prepared, measured, and analyzed by BNL under procedures approved by the NTPR Program. BNL used its FTA technique to characterize the ^{239}Pu activity in units of aCi (10^{-18} Ci) in a 24-hour urine sample or aCi/24-hr sample, hereinafter referred to as aCi/sample. This is an exceedingly small amount of radioactive material, because 1aCi is equal to 1.2 disintegrations per year. FTA is based on the high probability that ^{239}Pu will undergo nuclear fission in the presence of thermal neutrons.

DTRA, BNL, and VA collaborated on procedures to collect continuous 24-hour urine samples from the 100 veterans. DTRA's role was to communicate with veterans throughout the bioassay test process. VA's responsibility via its system of Veterans Affairs Medical Centers (VAMCs) was to receive the samples from veterans, perform a quality control check on the packaging, and maintain custody control of the sample in transport from the VAMC to BNL. BNL designed the packaging, handling, shipping and transport and urine collection procedures to insure intact samples were returned to BNL for bioassay measurement. BNL's experience with collecting and measuring Pu bioassay samples from Marshallese Islanders (Sun et al., 1993 and Sun et al., 1997) was adapted to atomic veterans to fully ensure successful sample collections.

2.2 BNL FTA Analysis Method

The FTA procedure entailed a two-step chemical process: first, the entire urine sample, collected from the individual and transferred to BNL, underwent co-precipitation and wet-ash digestion to oxidize organic matter and destroy protein complexes (Kaplan et al., 1995). At this point, the sample was split in half (called a split sample). The first split was prepared for FTA analysis and the second split was retained in case replication of the first split's measurement

should be necessary. Then, the first split underwent chemical separation of plutonium, using hyper-pure reagent resin exchange columns, from other fissionable materials such as uranium. The first split was then concentrated to a volume of a few microliters.

Each concentrated split was transferred to an ultra-clean quartz slide, evaporated, and irradiated in a nuclear reactor with a quantifiable thermal neutron flux. The ^{239}Pu fission fragments produced microscopic tracks in the slide's surface. Chemical etching was performed to make the tracks visible, and then the slides were viewed under a microscope and the magnified tracks counted. The number of tracks counted, which were directly related to the mass of ^{239}Pu irradiated, was proportional to the ^{239}Pu activity in the concentrated sample.

The FTA procedure was designed primarily to measure ^{239}Pu fissions. A primary source of potential interference was from the presence of ^{235}U , an isotope of uranium that also has a high cross section for fission from thermal neutrons. The resin exchange process was designed to maximize the separation of plutonium from uranium, although some low-level presence of ^{235}U in the samples was possible. The possibility of leakage was managed by monitoring for small increases in uncertainties of the FTA system calibration data collected for each bioassay batch (roughly 10 split samples per batch) over time. The resin columns were replenished for each new batch. If the small increases were severe enough to affect a particular batch of bioassay samples, the second split samples or new urine samples were obtained and processed as a new batch.

2.3 Methods for Analyzing Bioassay Data

2.3.1. Linear Regression Approach

Spikes and blanks provided calibration of the ^{239}Pu activity corresponding to the number of tracks deposited on the slides. The slides were prepared using synthetic urine, chemically equivalent to human urine, to which known activities of ^{239}Pu , traceable to National Institute of Standards and Technology (NIST), were added. The nominal activity levels of the spikes were 25, 50, 100, 150, 200, 300, and 400 aCi. For the blanks, the synthetic urine was not spiked. The spikes and blanks were irradiated with every batch, alongside samples to be measured, to ensure that uniform reactor irradiation geometries were maintained. Activation neutron flux monitors were used to assess the variability in neutron flux during the various irradiation batches to allow normalization of results. The sample array underwent preliminary testing to ensure there was no appreciable geometric flux inhomogeneity. These data allowed calibration of the FTA technique according to the following linear relationship:

$$T_c = mA_c + B \quad (1)$$

where:

T_c	=	observed number of tracks on the spike and blank slides;
A_c	=	known activities of the spike and blank slides (aCi);
m	=	slope of the best fit line (tracks/aCi); and
B	=	intercept of the best fit line (tracks).

The accumulated “spike and blank” calibration data were fit to equation (1) using linear least squares regression.

Once the relationship of equation (1) is established, the Pu activity of a test sample can be calculated using equation (2) as follows:

$$A_s = \left(\frac{fT_s - B}{m} \right) \quad (2)$$

where:

- A_s = activity (aCi) for bioassay sample having T_s tracks;
- f = scaling factor to correct for using a portion of the urine sample (typically around a value of 2), multiplied by a neutron flux normalization factor (ranging from 0.90 to 1.44), which is uniquely determined for each processed batch; and
- T_s = number of tracks counted on a sample slide.

Historical values for m and B were 1.06 and 19.8, respectively, and varied about 3% over time. The correlation coefficients of the regression lines ranged from 0.95 to 0.98, indicating a very consistent relationship between registered tracks on slides and known quantities of ^{239}Pu . The 95% confidence interval for the regression relationship averaged ± 36 aCi. New calibration data were checked for conformance with this regression relationship to validate the stability of the FTA method. Figure 1 shows a graphical representation of this linear calibration approach, also reported in the literature. The track values for spikes falling appreciably outside the uncertainty band (labeled as “95% limit”) shown in Figure 1 were rejected as outlier data (Schaeffer et al., 1999; Brodsky et al., 1999; and Schaeffer, 2002).

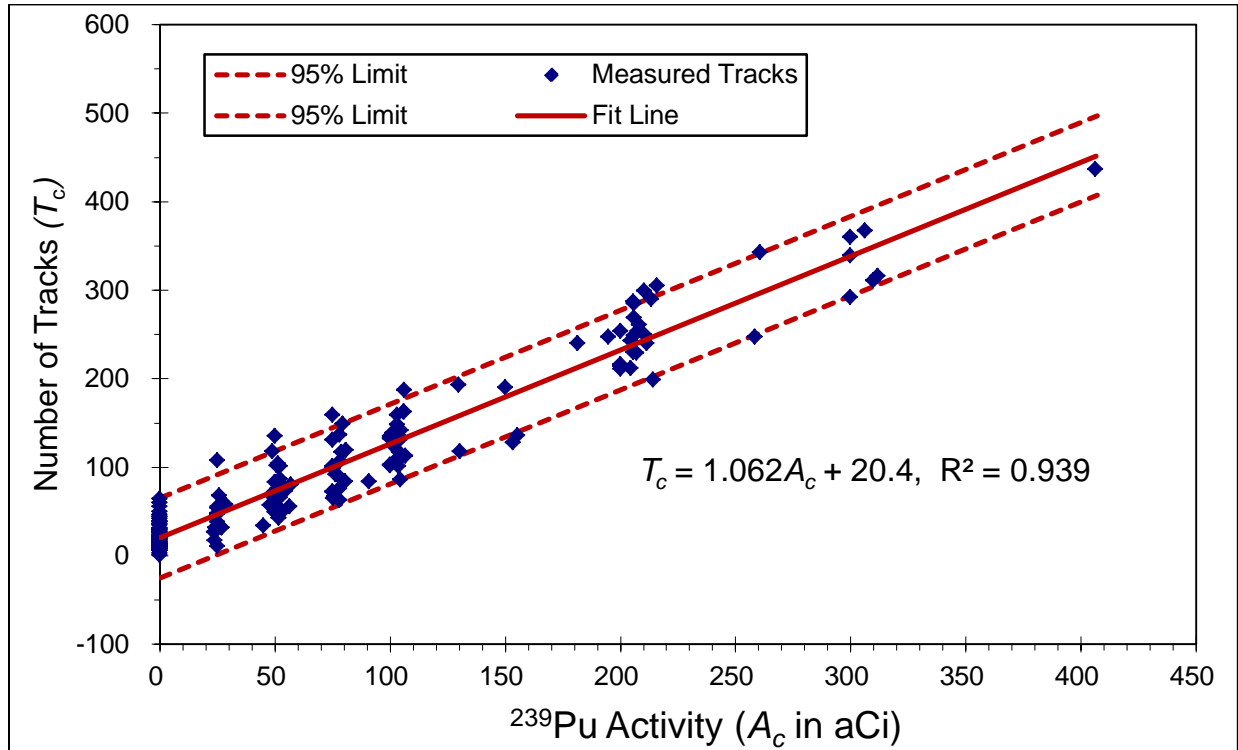


Figure 1. Typical linear regression calibration curve for FTA bioassay blanks and spikes

The “blank” data were analyzed separately to determine the lower limit of detection (LLD) of the FTA method. This analysis is crucial for taking into account the statistical uncertainties of measuring the “zero” activity of ^{239}Pu in the blanks. The LLD was determined by converting all of the blanks data (in number of tracks) to activity (aCi) by using equation (2). The data were averaged and the standard deviation computed. The LLD represented a value corresponding to trace ^{239}Pu present in the synthetic urine solution. The LLD established a level above which a bioassay result is distinguishable from the nearly zero activity of the blanks. This value corresponded to about 20 ± 36 aCi, roughly corresponding to the value of B intercept for blanks.

2.3.2. Lognormal Regression Correction Approach

The cooperative scientific team discovered over a series of five batches (consisting of about 10 bioassay samples per batch) that a considerable number of “spikes” data were being rejected as outliers in fitting calibration data using the linear regression approach. These outliers appeared to be commonly associated with samples of higher activity rather than at levels approaching the LLD. Figure 2 displays data used in the linear regression analysis along with the heretofore rejected outlier data. This realization led to a rejection of the linear regression approach outlined in section 2.3.1 and computation of new calibration curves based on lognormal regression. The bioassay values obtained with linear regression were replaced with new values computed using lognormal regression.

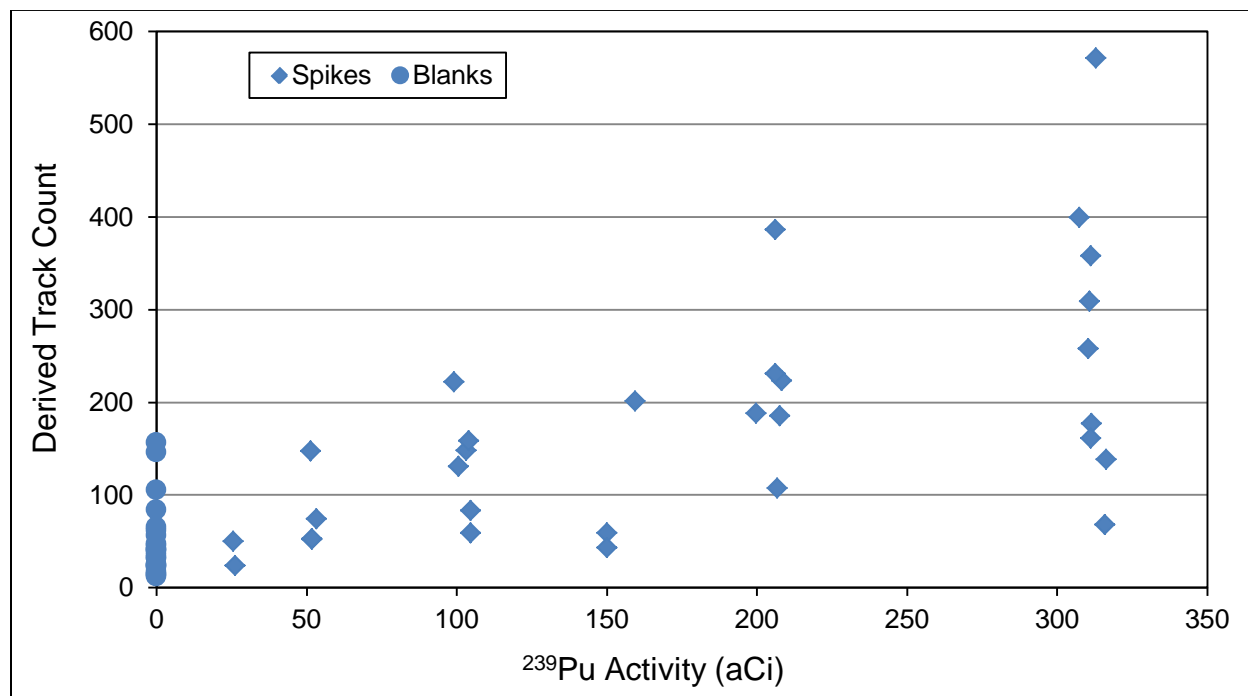


Figure 2. Spike and blank values used in calibration of the FTA method for calculation of ^{239}Pu activities in veterans’ samples

Changes in chemistry and neutron irradiation procedures over time resulted in a greater variability in track numbers at given plutonium levels, which became especially pronounced at

higher plutonium activities. An increasing non-homogeneous variance could be noticed with increasing plutonium levels. The linear regression approach (see Figure 1) showed a distribution of constant variance estimated to be ± 36 aCi. It was found using the data shown in Figure 2 data that the deviations from the mean were not consistent with a normal distribution as first thought (see section 2.3.1). It was found that the ratios of the number of tracks for a sample divided by the median (estimated by an applied fit to Figure 2 data) were lognormally distributed (Klemm et al., 2001 and 2003). The differences between the logarithm of observed number of tracks and logarithm of the median value were found to be normally distributed. The distribution of the number of tracks, corresponding to blanks, was also found to be lognormally distributed as well (Klemm et. al., 2001, 2002). Likewise, the differences between the logarithm of the observed number of tracks corresponding to the blanks and the logarithm of the blanks' median value were also found to be normally distributed.

This discovery resulted in the reanalysis of bioassay data to account for corrected distributions and calibration uncertainties noted in the data heretofore rejected in the linear regression analysis. The mathematical schemes for dealing with non-conventional uncertainty distributions are well documented (Brodsky et al., 1999; Brodsky, 2000, and Klemm et al., 2003). Klemm et al. (2003) demonstrated how to apply corrections to the calibration regression line used to fit blanks and spikes data to equation (1) and compute the activity in aCi of the bioassay samples per equation (2). All reported veteran bioassay data were computed in this manner. For example, equations (1) and (2) above become:

$$\text{Log}(T_c) = m'[\log(A_c)] + B' \quad (3)$$

where T_c and A_c are defined in equation (1) and m' and B' are slope and the intercept of the best fit linear regression line of equation (3). Then, the Pu activity in a sample is calculated with:

$$A_s = 10^{\{[\log(ft_s) - B'] / m'\}} \quad (4)$$

where A_s and T_s are defined in equation (2) and m' and B' are defined in equation (3) above.

2.3.3. Definition and Determination of Minimum Detectable Activity (MDA), Lower Limit of Detection (LLD), and Decision Level (DL)

It was particularly important to develop factors for the analysis and interpretation of all bioassay data collected for the pilot bioassay sample program. The useful factors are MDA, LLD, and DL. The most important factor was the MDA.

The MDA, derived from all the blanks and background population bioassay data, established the level above which a veteran bioassay sample measurement was judged to be statistically different from the 58 person background population. This factor was derived using the ANSI Standard N13.30 and other references (ANSI, 1996; Boecker et al., 1991; and Brodsky, 1992). The MDA was based on accounting for Type I and Type II errors at the 95th percentile (Boecker et al., 1991 and Brodsky, 1992). That is, the MDA computed to be 280 aCi/sample was the level above which there is at least a 95% chance that a veteran's bioassay sample measurement was truly positive and not a false negative.

The LLD is the level below which bioassay results can be considered indistinguishable from a "zero" level of activity. The LLD was derived as the median of the distribution of the

blanks. The median value was 18 aCi with the 95% percentile for Type I error value being ± 24 aCi and Type II error value being ± 75 aCi.

The DL was a term coined for a factor derived for simply characterizing the attributes of the distribution of the 58-sample U.S. general population results (see Section 3.2. and Figure 3). The DL was further broken down into DL_{95} and DL_{max} . DL_{95} defined the 95th percentile level below which 95% of the background population sample results were contained. DL_{max} corresponded to the highest result in the general population distribution. The DL_{max} was used to screen out values between it and the MDA of 280 aCi/sample for further study.

2.4 Measurement Validation of FTA Plutonium Bioassay

2.4.1. Comparison of Results of Analyses of Plutonium in Urine Samples by Two Laboratories

In 1996/1997, Dr. Payne S. Harris, the NTPR program's medical consultant, made arrangements with Los Alamos National Laboratory (LANL) to participate in a bioassay measurement comparison between BNL and LANL. LANL provided bioassay samples collected from LANL workers having prior ^{239}Pu intakes which LANL monitored and profiled historically for dose assessment and workers unexposed to plutonium, except for worldwide fallout during the atmospheric nuclear test era. The comparison was performed to test the ability of BNL's FTA technique to measure ^{239}Pu activity in the 1 to 10 femtocurie (fCi) (10^{-15} Ci) range. This was estimated to be the highest level ^{239}Pu excreted in urine for intakes that could be expected in atomic veterans. Other samples were included to assess the ability of FTA to measure samples at background levels. The collected bioassay samples were split between BNL and LANL to allow for independent analyses. The LANL results were not communicated to BNL until BNL processed their bioassay samples. The measurement techniques used by BNL and LANL for the comparison were FTA and Thermal Ionization Mass Spectrometry (TIMS). While the LLDs for both techniques differed somewhat, they were more than sufficient to not influence bioassay measurements in the 1–10 fCi range.

LANL reported (Harris, 1998 and Inkret and Efurd, 1996) the resulting measurements in Table 1. The measurements include LANL values determined from bioassay samples split between both laboratories, and BNL values determined from the other split sample which was, in turn, split again. Each sample represented a collection of all urine excreted during a period of 24 consecutive hours. Submitters collected the samples without supervision.

2.4.2. Analysis of Comparison Results

The 10 samples reported in Table 1 presented limited opportunities for measurement comparison. There appeared to be some general agreement between the BNL and LANL results. However, samples analyzed by LANL with reported results below 2 fCi had large fractional uncertainties. Two sets of measurements for each lab on samples 36726, 36737, and 36741 showed agreement within an order of magnitude. The three samples, identified as 36729, 36744, and 36745, for workers not occupationally exposed to ^{239}Pu , were consistently measured to be at or slightly above the LLD of the FTA technique. The measurements reflect similar levels of uncertainty as the blanks used to derive the LLD. The LANL TIMS technique, as opposed to the FTA technique, consistently appeared to yield results within its standard deviation, which was

almost an order of magnitude higher than the LLD of FTA. The advertised LLD of TIMS for this comparison was 0.130 aCi (Inkret et al., 1998) vs. 0.029 aCi for FTA (Kaplan et al., 1995).

Table 1. Results of ^{239}Pu measurements by LANL and BNL

Sample ID	BNL Activity (fCi/sample)	BNL Recheck Activity (fCi/sample)	LANL Activity (fCi/sample)	LANL Std Dev (fCi/sample)
36721	228	-	524.99	4.72
36723	lost	-	69.27	1.11
36726	0.756	0.108	1.13	0.95
36729*	0.108		1.73	0.95
36733	not sent	-	8.13	0.37
36737	0.07	-	0.04	0.49
36740	11.3	4.8	lost during analysis [†]	
36741	0.06	-	-0.06	0.8
36744*	<0.029	-	0.55	0.62
36745*	0.065	<0.029	1.57	1.03

* Worker was not occupationally exposed to ^{239}Pu

[†] Historical (LANL) measurements support values close to 11.3 fCi for this individual

DTRA's concern at the time of the comparison was the repeatability of the FTA technique. Samples 36726 and 36740 showed ratios of the results of the first split sample to the second split sample of 7 to 1 times and 2.4 to 1 times, respectively. The reasons for these seemingly high relative differences were not known at the time. When compared to the constant uncertainty limits shown in Figure 1, the split measurement repeatability was considerably out of the range of the limits. Figure 2 could suggest the emergence of a key insight when coupled with the discovery that the FTA system calibration curve and associated uncertainties followed more closely a lognormal than a linear relationship (Klemm et al., 2003). The lognormal calibration relationship shown in the Figure 2 data suggested that range of measurement uncertainty increased roughly by an order of magnitude with increasing measured activity level. In that context, BNL results for samples 36721, 36726, and 36740 could be viewed as roughly consistent with LANL results. The BNL split sample differences (7 to 1 and 2.4 to 1) might be suggestive of the FTA measurement uncertainty range increasing lognormally with increasing bioassay measurement levels. TIMS appeared to significantly overestimate the ^{239}Pu background levels in samples 36729, 36744, and 36745 for workers not occupationally exposed to plutonium.

Section 3.

Plutonium in the Urine Samples of U.S. General Population and U.S. Atomic Veterans

3.1 Plutonium Measured in Urine of Selected Members of the U.S. General Population

To establish background levels in the U.S. population, a national cross section of individuals exposed only to worldwide fallout during the atmospheric nuclear testing era was characterized. Assessment of plutonium in urine samples from the general population group may be used to determine whether urine samples from veterans have significantly higher plutonium content than this general population group. BNL sampled the U.S. general population with the assistance of DOE occupational medical clinics (OMC) and VAMCs. With approval from BNL's Institutional Review Board for research on human subjects, BNL circulated the collection protocol to various OMCs and VAMCs nationwide. Urine samples were collected from 58 males over 40 years of age with no history of working with plutonium or performing activities similar to veterans eligible for bioassay testing. The 24-hour urine samples were taken in seven geographical locations from workers during routine physical examinations. The workers self-collected the urine sample over a continuous 24-hour period.

Table 2 describes the geographical locations and numbers of subjects contributing urine samples. Sampling of the general U.S. population overall was constrained by time and funding but considered technically and statistically adequate by BNL for determining background levels of Pu in the general population with sufficient geographic diversity.

Table 2. Locations and numbers of subjects contributing urine samples for the general population group

Location	Number of Samples
Golden, CO OMC	4
Berkeley, CA OMC	5
Cincinnati, OH VAMC	6
Rocky Flats, CO OMC	7
Oak Ridge, TN OMC	8
Pinellas, FL OMC	10
Upton, NY OMC	18
Total	58

Figure 3 shows the frequency distribution of the bioassay measurements (see Appendix A, Table A-1) for the background population group. About ninety-five percent of the population group samples measured lower than 84 aCi/sample (DL_{95}) as defined earlier in section 2.3.3. The highest measurement (DL_{max}) in the distribution was 118 aCi/sample. This distribution is important for determining whether or not the measurements for the 100 veteran samples are statistically different from the general U.S. background population group samples.

The 84 aCi/sample result for the general U.S. population group compared closely to the reported results of other general populations exposed only to worldwide nuclear testing fallout (Ibrahim et al., 1999 and Hamilton et al., 2007).

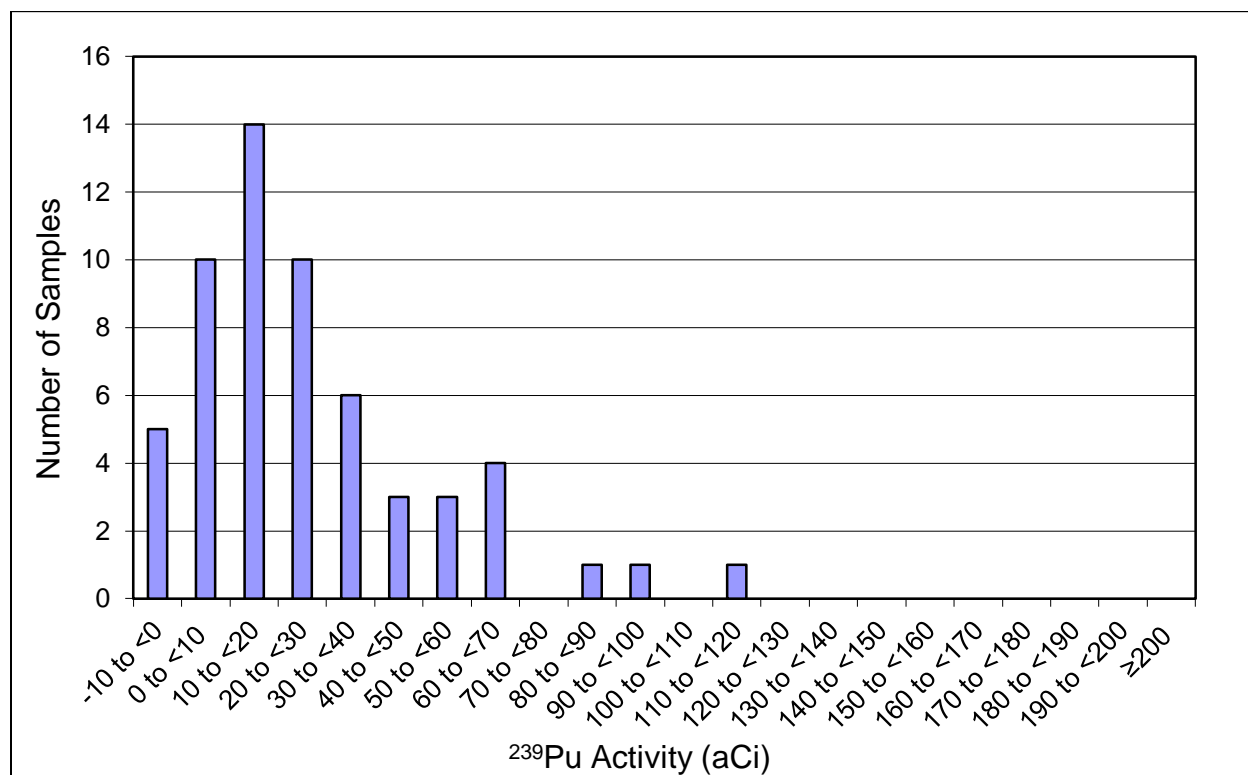


Figure 3. Distribution of bioassay measurements in the U.S. general population group

3.2 Results for Initial Phase of Measurements on Veterans

One hundred (100) confirmed atomic veterans volunteered to contribute urine bioassay samples taken over a continuous 24-hour period. DTRA publicly announced (DTRA, 1999) the pilot test program, including an explanation of the significance and extent of the effort constrained by the amount of Congressionally-directed funding (PL 105-85, 1997).

Figure 4 provides a composite of the U.S. Population and 100 veteran bioassay results. These results reflected all of the first split bioassay measurements. The veterans' results (see Appendix A, Table A-2) ranged from -2 to 2366 aCi/sample. There were four measurements above the FTA system MDA of 280 aCi/sample that could be considered true positives based on Type I and Type II statistics (Brodsky, 1992). The remaining 96 measurements were below the MDA.

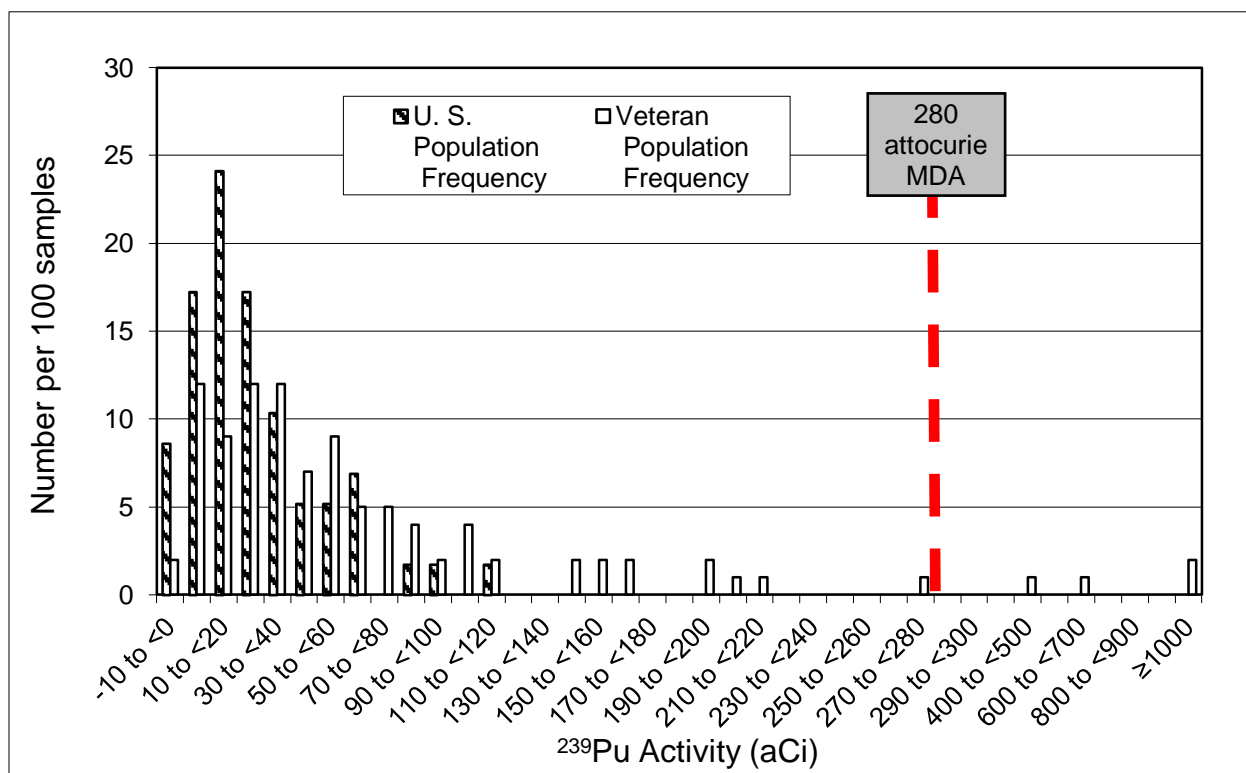


Figure 4. Comparison of the distributions of bioassay results for the U.S general population and atomic veterans

There were 11 measurements between the maximum background population measurement, DL_{max} and MDA of 118 and 280 aCi/sample respectively. Eighty five (85) of 100 veterans' samples measured below the 118 aCi/sample general population maximum. Ninety-five percent of the 85-veteran sample measurements were below 118 aCi/sample. This value is slightly higher than the DL_{95} (84 aCi/sample) for the general population group distribution. Within the limits of FTA measurement statistics, the 85-veteran distribution of measurements appears to be nearly similar to the 58-background population sample.

The 15 veteran samples with results greater than 118 aCi/sample were singled out for further investigation. Nine (9) of the 15 samples were chosen for reanalysis of the second split sample. The highest one of the 15 measurements was confirmed as a true positive by comparison with an available employment occupational ^{239}Pu bioassay measurement voluntarily submitted by the veteran and was eliminated from reanalysis. Only 9 of the remaining 14 and 2 additional samples measuring closest to 100 aCi/sample were chosen for analysis of the second split. These 11 results were produced in the fifth and higher-numbered batches, which were suspected of being impacted by external trace low environmental levels of plutonium from other on-site BNL work.

Table 3 contains the results of the 11 samples showing first and second split measurements. The second split measurements are lower than the first split measurements, except for one. The results seemed to be confounding in that 4 of the 11 second split

measurements are considerably lower than the first split measurements. Also, 6 of the 11 second split measurements were roughly comparable to the first result. In one case, the second split measurement was higher than the first.

Table 3. First and second split measurements for 11 selected bioassays with original readings above 100 aCi/sample

Bioassay Number	First Split Measurement (aCi/sample)	Second Split Measurement (aCi/sample)
P-85	1059	358
P-120	627	32
P-86	436	74
P-64	274	237
P-83	197	111
P-104	167	131
P-17	165	128
P-31	158	35
P-105	151	97
P-53	106	65
P-99	99	209

Most of the above-MDA (>280 aCi) results and all results between 100 and 280 aCi/sample occurred during times when FTA measurements exhibited higher than expected calibration uncertainties. BNL reported that apparent elevated results were concurrent with fuel removal from a reactor decommissioning in a neighboring building, causing a temporary shutdown of FTA analyses to replace all of the FTA reagent chemicals. Twelve (12) veterans with the higher urine results were selected for re-sampling based on the availability and willingness of a veteran to provide a second 24-hour urine sample. This resampling provided a possible way to better understand the results in Table 3. To address potential background contributions and other experimental factors, the NTPR Program employed LANL to conduct follow-up ²³⁹Pu measurements using TIMS (Lyons, 1999).

3.3 Results of Resampling of 12 Selected Veterans

The follow-up bioassay test measurements, reported by LANL (Glover et al., 2003), are listed in Table 4 and displayed in Figure 5, which includes 10 of the 11 measurements reported in Table 4 for comparison. As mentioned before, not all veterans with measurements in Table 4 could be re-sampled and re-analyzed with TIMS. Two additional veterans with measurements of about 100 aCi/sample were available for re-sampling and those results were included in the comparison.

Table 4. Comparison of bioassays for veterans followed-up with TIMS

ID Number	FTA Activity (aCi/sample)*	TIMS Activity (aCi/sample)*,†
P-85	1059/358	27±12/34±30
P-120	627/32	24±16/9±18
P-86	436/74	28±16/47±26
P-64	274/237	47±52/-2±24
P-83	197/111	23±24/64±174
P-104	167/131	32±28/31±26
P-17	165/128	51±24/64±20
P-31	158/35	30±20/44±76
P-105	151/97	7±32/-56±76
P-96	146	-7±28/31±20
P-106	144	68±30/18±16
P-99	99/209	17±40/41±20

* Cells with two entries reflect that both split samples were analyzed

† ±2 standard deviations

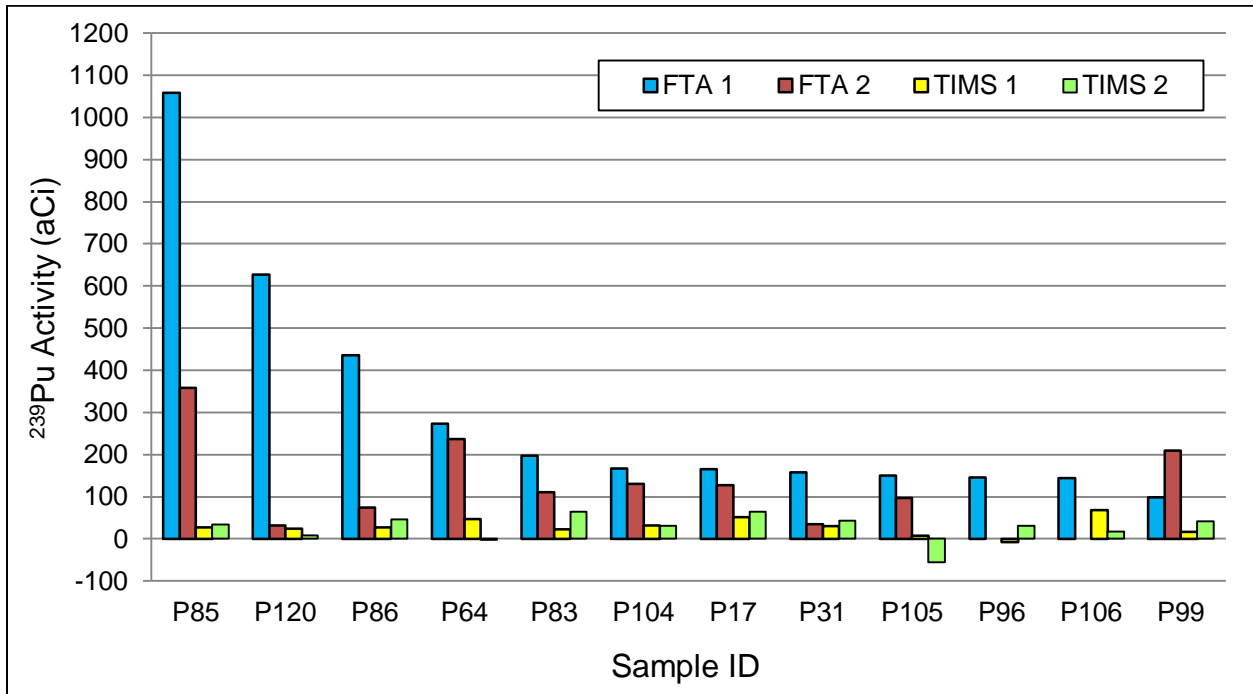


Figure 5. Comparison of FTA and TIMS sample measurements for selected veterans with original FTA bioassay measurements above 100 aCi/sample

Section 4.

Discussion of the Results

4.1 Interpretation of the Results

A review of the data (Figure 4 and Appendix A, Table A-2) indicates that 4 of the 100 veterans' sample results were above the ^{239}Pu MDA of 280 aCi/sample. Eleven (11) of 100 results measured between 118 and 280 aCi/sample. The highest measurement in the general population background distribution was 118 aCi. All 58 general population samples measured below the MDA. Discounting the 15 measurements above 118 aCi/sample, the veteran sample distribution closely resembled the general population sample distribution, noting a small shift to slightly higher, within range, background values.

Second split samples for 12 of the 15 veterans whose results ranged from 100 to < 280 aCi/sample were measured. See Table 4 for a summary of the measurements. Review of the values shown in Figure 2 suggested that uncertainties between the two splits increased dramatically with increasing measured activity in the sample. This trend was also observed and reported as a finding to NRC/NAS (Schaeffer, 2002) and as suggested earlier in Section 2.4.2.

The corroborating TIMS measurements (Table 4), showing excellent repeatability in their split sample measurements, were consistently much lower in value than the BNL-measurement of paired samples. LANL demonstrated tight calibration controls and excellent measurement repeatability on the TIMS measured samples (Lewis and Guilmette, 2002 and Glover et al., 2003). In addition, TIMS at LANL was a Department of Energy Laboratory Accreditation Program accredited process (DOE, 1998) used in its occupational exposure monitoring program to document internal doses from ^{239}Pu exposures. Thus, the TIMS measurements appear to be more credible than the corresponding BNL measurements.

That being the case, it is highly likely that only one of the veteran sample results was greater than the DL_{max} of 118 aCi/sample. The one result from sample P-6 (see Table A-2) was corroborated by the results provided by the veteran of an earlier ^{239}Pu bioassay performed at LANL. The LANL result, 2300 aCi/sample, was consistent with the BNL-measured result of 2366 aCi/sample.

4.2 Comparison of Bioassay Measurements and Dose Reconstructions

Dose reconstruction reports in the NTPR case files of all 100 veterans were examined. The expected ^{239}Pu excretion levels were estimated for veterans who had positive values of internal dose. Some veteran's dose reconstruction reports did not show potential for accrual of internal dose. Table 5 shows a summary of the comparison, enumerating the veteran bioassay measurements above the MDA with dose reconstruction predicted values of ^{239}Pu excretion and those below.

Table 5 indicates that 52 veterans' dose reconstructions yielded positive values for internal dose that bioassay would be expected to predict. However, only 1 of 52 veteran samples showed a correlation to a positive internal dose reconstruction. In contrast, there were 48 dose reconstructions that did not predict a potential for internal dose. Forty-seven (47) of 48 sample measurements were consistent with a zero-valued dose reconstruction prediction. However, there was only one bioassay result above MDA, which did not correlate with the zero-valued dose

reconstruction prediction. Fifty-one (51) samples corresponding to positive internal dose reconstruction predictions did not correlate with above MDA bioassay results. Table 5 indicates that there was no discernable correlation that the predicted and measured results agreed.

Table 5. Comparison of bioassay results with reconstructions of internal dose

Internal Dose Reconstruction Prediction	Bioassay Result		Total
	Number of Results \leq MDA	Number of Results $>$ MDA	
Dose $>$ 0 [*]	51	1 [†]	52
Dose \leq 0 [†]	47	1	48

* Dose reconstruction reported an internal dose commitment.

† Dose reconstruction reported no internal dose commitment.

‡ Does not include the 2 samples (P-120 and P-86) (see Appendix A) with first split bioassay measurements above the MDA and second split measurements well below the MDA.

The one measurement of particular interest was sample P-64 (274 aCi/sample) which was coincidentally close to the value of the FTA system MDA. The corresponding veteran was determined to be a participant in the post-war occupation of Nagasaki. The Nagasaki weapon, being a plutonium device, presented opportunities for inhaling and ingesting fallout containing low levels of ²³⁹Pu (McRaney and McGahan, 1980).

Examining the activities of U.S. occupation troops at Nagasaki, the NTPR Program found that troops stationed around the Nishiyama reservoir, which was assumed to have received an appreciable amount of fallout from rainfall, could possibly excrete Pu levels as high as 470 aCi/sample at about 45 years after exposure. Comparatively, occupation troops assigned to the Artillery Group could be expected to excrete about 100 aCi/sample. Considering the 95% confidence level (DL₉₅) of 84 aCi/sample, sample P-64 could be attributable to worldwide fallout in addition to possible Nagasaki activities. The veteran could have had a sufficient intake of Pu to produce a urine level between 185 and 555 aCi/sample.

The BNL results for the two split samples for P-64 were in close correspondence (see Table 3). Similarly, so were the two split TIMS measurements, but they were well within the measurement range of the background sample distribution. Determination of whether or not sample P-64 represented a likely positive bioassay measurement was confounded by the MDA level of the FTA system, and the two split TIMS measurements at values that were consistently at low values of the background population distribution. Thus, in consideration of the uncertainties in the FTA and TIMS analyses and the dose reconstruction for the veteran represented by sample P-64, it was not discernable whether or not the veteran received a dose from plutonium consistent with his activities at Nagasaki.

Section 5.

Conclusions

The proposal (Boecker et al., 1991) that FTA could measure ^{239}Pu activity concentrations in urine samples that could be used to retrospectively quantify internal doses in eligible veterans has not been demonstrated in the bioassay results described herein. In all but a few cases, veteran bioassay samples did not measure statistically higher than samples submitted by 58 persons representing a sample of the U.S. population in seven geographic locations nationwide.

Plutonium uptake in veteran participants of atmospheric nuclear testing may be confirmed provided there is a sufficient potential to produce a positive bioassay result 35 years after weapon detonation. Uptake, retention and excretion of ^{239}Pu are highly variable among individuals over any period of time. Given that more than 35 years had lapsed from exposure to measurement, FTA results confirmed intakes in a small fraction of individuals in this sample cohort. At these low measurement levels (around a few hundred aCi), repeatability and collective analyses were difficult due to observed high uncertainties. Therefore, FTA may be used to confirm presence of ^{239}Pu in urine bioassays, but not to quantify ^{239}Pu activity with an acceptable degree of confidence.

This finding is supported by the 10 elevated measurements in 99 veteran bioassay samples that were not confirmed by follow-up TIMS analyses. (One of the 100 veterans was a confirmed positive). Additionally, FTA did not demonstrate a ^{239}Pu level above the MDA level for 51 veterans for whom NTPR Program dose reconstruction reports indicated a potential for internal dose. Additionally, the bioassay results for 47 veterans correctly correlated to indications of no potential for an internal dose. The FTA technique provided one potential indicator of plutonium presence (see Table 5). However, due to the individual variances with respect to location, complexity of specific response to exposure, and confounding environmental effects over the elapsed time since exposure, it was difficult to find a correlation between dose reconstruction predictions and bioassay measurements.

In conclusion, bioassay testing using FTA is not a reliable quantitative indicator of internal dose potential for the sampled veteran population. Other available analytical methods, developed since FTA, such as TIMS, appeared to provide quantitative bioassay results with higher accuracy and consistency than FTA. FTA did not reliably provide an indication of participation in atmospheric nuclear testing (in the absence of other information). It is important to note that, in the 10 of the sampled veteran population where internal doses were suggested by initial bioassay testing, follow-up testing at a corroborating laboratory (LANL) was unable to confirm the positive (i.e., above MDA) FTA-determined results for veterans who submitted second samples. (See section 3.3.2). Performing ^{239}Pu bioassay measurements to correlate with veteran dose reconstructions is scientifically daunting, given the uncertainties and technique variations at the levels of ^{239}Pu being detected. Nevertheless, NTPR dose reconstructions with internal dose components do provide positive dose results for this population. Urine bioassay measurements of ^{239}Pu utilizing FTA tend to be so highly uncertain due to large variances in measurement results as well as uncertainties in assumed parameter values used for biokinetic modeling in dose calculations (NRC, 2003)— that they cannot be considered as useful predictors of atomic veteran internal doses.

This report concludes, consistent with National Research Council's findings (NRC, 2003), that urine ^{239}Pu bioassay using FTA did not enhance the capability of DTRA's current scientific processes to perform accurate dose reconstructions for atomic veterans, and therefore, was not feasible for implementation on a wide scale basis for atomic veterans.

Section 6.

References

- ANSI (American National Standards Institute), 1996. *Performance Criteria for Radiobioassay*, HPS Publication N13.30-1996. Health Physics Society, McLean, VA. May.
- Boecker, B., Hall, R., Inn, K., Lawrence, J., Ziemer, P., Eisele, G., Wachholz, B. and Burr, W. Jr., 1991. "Current Status of Bioassay Procedures to Detect and Quantify Previous Exposures to Radioactive Materials," *Health Phys Supplement 1*, 60:45–100.
- Brodsky, A., 1992. "Exact Calculation of Probabilities of False Positives and False Negatives for Low Background Counting," *Health Physics* 63(2):198–204.
- Brodsky, A., Schaeffer, D. M., O'Toole, S.A., Kaplan, E., Barss, N., Dancz, J., Klemm, W. J., Raine, D. A., and Stiver, J., 1999. "Statistical Model for Fission Track Analysis of Plutonium in Human Samples". Philadelphia, PA: Proceedings of the 44th Annual Health Physics Society Meeting. June.
- Brodsky, A., 2000. "Mathematically Analytic Distributions Can Explain Radioanalytic Results", Seattle, WA: Proceedings of the 46th Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry. November.
- DOE (Department of Energy), 1998. *The Department of Energy Laboratory Accreditation Program for Radiobioassay*, DOE Standard, DOE-STD-1112-98. Washington, DC. December.
- DTRA (Defense Threat Reduction Agency), 1999. *One-time Plutonium Bioassay Service for Nuclear Test Personnel Review Program*, DTRA Fact Sheet, Defense Threat Reduction Agency, Alexandria, VA. April.
- Glover, S.E., Wagner, S.E., Steiner, R.E., Boone, S., Duffy, C., 2003. *Bioassay Analysis for the Determination of Plutonium in Atomic Veterans*. Report LA-UR-03-3245, Los Alamos National Laboratory, Los Alamos, NM. May.
- Hamilton, T.F., Kehl, S.R., Brown, T.A., Martinelli, R.E., Hickman, D.P., Jue, T.M., Turner, S.J., and Langston, R.G., 2007. *Individual Radiological Protection Monitoring of Utrok Atoll Residents Based on Whole Body Counting of Cesium-137 (¹³⁷Cs) and Plutonium Bioassay*, LLNL Marshall Islands Program Field Operations Report UCRL-MI-231678. Lawrence Livermore National Laboratory, Livermore, CA. June.
- Harris, P.S., 1998. *The LANL/BNL Comparison*, Logicon IT Memo (unpublished), Alexandria, VA. March 15.
- Ibrahim, S.A., Whicker, F.W., Reuss, S.K., Whicker, R.D., Chapman, P.L., and Krahenbuhl, M.P., 1999. "Plutonium Excretion in Urine of Residents Living Near the Rocky Flats Environmental Technology Site," *Health Physics* 76:368–379.
- ICRP (International Commission on Radiation Protection) 1994a. "Dose Coefficients for Intakes of Radionuclides by Workers," ICRP Publication 68, *Ann ICRP 24(4)*. Pergamon Press, Elmsford, NY.

- ICRP (International Commission on Radiation Protection) 1994b. "Age Dependent Doses to Members of the Public from Intake of Radionuclides: Compilation of Ingestion and Inhalation Dose Coefficients," ICRP Publication 72, *Ann ICRP 26(1)*, Pergamon Press, Elmsford, NY.
- Inkret, W.C. and Efurud, D.W., 1996. *Applying Mass Spectrometry to the Detection of Plutonium Intakes*, LANL Progress Report (unnumbered), Los Alamos, NM. May.
- Inkret, W.C., Efurud, D.W., Miller, G., Rokop, D.J., and Benjamin, T.M., 1998. *Applications of Thermal Ionization Mass Spectrometry to the Detection of ²³⁹Pu and ²⁴⁰Pu Intakes*, LANL Draft Report (unnumbered), Los Alamos, NM. January
- Jones, S.R., 1985. "Derivation and Validation of a Urinary Excretion Function for Plutonium Applicable over Tens of Years Post-uptake, *Radiation Protection Dosimetry 11*:19–27.
- Kaplan, E., Pietrzak, R., Doty, R., Moorthy, A., and Baum, J., 1995. *Plutonium Bioassay Program Review*, briefing presented to DTRA, Brookhaven National Laboratory, Upton, NY. December 5.
- Kaplan, E., 1996. *Plutonium Bioassay Program Review*, briefing presented to DTRA, Alexandria, VA. October 8.
- Klemm, W. J., Brodsky, A., Schaeffer, D. M., 2001. "Radioanalytical Data Interpretation-When the Ratio Reading/Median Is Lognormally Distributed," Proceedings of the 46th Annual Health Physics Society Meeting. Cleveland, OH. June.
- Klemm, W. J., Brodsky, A., Schaeffer, D. M., 2003. "Radioanalytical Data Interpretation-When the Ratio Reading/Median Is Lognormally Distributed," *Health Physics*. 85(6):721–732
- Leggett, R.W., 1984. *Bioassay Data and a Retention-Excretion Model for Systemic Plutonium*, NRC Publication NUREG/CR-3346. Oak Ridge National Laboratory, Oak Ridge, TN.
- Lewis, D. and Guilmette, R., 2002. *Acceptance Criteria for Bioassay Data*, LANL Report HSR-12-DAC-02.01. Los Alamos National Laboratory, Los Alamos, NM.
- Lyons, D., 1999. *Los Alamos Technology Detects Plutonium at Lower Levels in Radiological Workers*, Los Alamos News Release. Los Alamos National Laboratory, Los Alamos, NM. September 23.
- McRaney, W. and McGahan, J., 1980. *Radiation Dose Reconstruction U.S. Occupation Forces in Hiroshima and Nagasaki, Japan, 1945-1946*. Technical Report DNA 5512F, Defense Nuclear Agency, Washington, DC. August 8.
- Moorthy, A.R., Schopfer, C.J., and Banerjee, S., 1988. "Plutonium from Atmospheric Weapons Testing: Fission Track Analysis of Urine Samples," *Analytical Chemistry* 60:857–860.
- NRC (National Research Council), 1985. *Review of the Methods Used to Assign Radiation Doses to Service Personnel at Nuclear Weapons Tests*. National Academies Press, Washington, DC.
- NRC (National Research Council), 2003. *A Review of the Dose Reconstruction Program of the Defense Threat Reduction Agency*. National Academies Press, Washington, DC.

- Pietrzak, R., Doty, R., Finfrock, C., Moorthy, A., Kaplan, E., and Thind, K., 1998. “²³⁹Pu in Urine by ICP-MS and FTA Methods and Their Application to Occupational Exposure Measurements,” Proceedings of the 44th Bioassay Analytical Environmental Radiochemistry (BAER) Conference, Albuquerque, NM. November 15.
- PL 98-542 (Public Law 98-542), 1984. *Veterans’ Dioxin and Radiation Exposure Compensation Standards Act*. Washington, DC. October.
- PL 105-85 (Public Law 105-85), 1997. *National Defense Authorization Act for Fiscal Year 1998, Section 244, Bioassay Testing of Veterans Exposed to Ionizing Radiation during Military Service*”. Washington, DC. November.
- Schaeffer, D. M., 2002. *Bioassay Testing of U.S Atmospheric Nuclear Test Participants*. Briefing given to the National Research Council (NRC) Committee to Review the Dose Reconstruction Program of the Defense Threat Reduction Agency (DTRA) and Paper presented to the Baltimore-Washington Chapter of the Health Physics Society. Washington, DC and Bethesda, MD. Feb and May.
- Schaeffer, D. M., O’Toole, S. A. and Kaplan, E., 1999. *Calibration and Interpretation of Fission Track Analysis of Plutonium in Urine*. Proceedings of the 44th Annual Health Physics Society Meeting, Philadelphia, PA. June.
- Sun, L.C., Meinhold, C.B., Moorthy, A.R., Kaplan, E., and Baum, J.W., 1997. “Assessment of Plutonium Exposure in the Enewetak Population by Urinalysis,” *Health Physics* 73(1):127–132.
- Sun, L.C., Clinton, J.H., McDonald, J., Moorthy, A.R., Kaplan, E., and Meinhold, C.B., 1993. “Urine Collection Protocol in the Republic of the Marshall Islands,” *Radiation Protection Management* 10(4):64–72.

Appendix A.

Compilation of Project Bioassay Test Results

Table A-1. Results of bioassay testing of the selected U.S. population group

No.	Location	²³⁹Pu Activity (aCi)	No.	Location	²³⁹Pu Activity (aCi)
1	Rocky Flats, CO	-10	30	Oak Ridge, TN	20
2	Upton, NY	-8	31	Pinellas, FL	20
3	Pinellas, FL	-2	32	Golden, CO	21
4	Berkeley, CA	-2	33	Rocky Flats, CO	21
5	Upton, NY	-1	34	Rocky Flats, CO	24
6	Pinellas, FL	0	35	Pinellas, FL	24
7	Berkeley, CA	0	36	Upton, NY	27
8	Cincinnati, OH	3	37	Cincinnati, OH	27
9	Rocky Flats, CO	3	38	Upton, NY	28
10	Pinellas, FL	3	39	Upton, NY	29
11	Berkeley, CA	3	40	Golden, CO	33
12	Oak Ridge, TN	6	41	Upton, NY	33
13	Rocky Flats, CO	7	42	Oak Ridge, TN	34
14	Oak Ridge, TN	8	43	Pinellas, FL	35
15	Pinellas, FL	8	44	Rocky Flats, CO	37
16	Cincinnati, OH	10	45	Upton, NY	38
17	Cincinnati, OH	10	46	Pinellas, FL	40
18	Berkeley, CA	10	47	Rocky Flats, CO	47
19	Golden, CO	10	48	Upton, NY	47
20	Upton, NY	10	49	Upton, NY	50
21	Upton, NY	12	50	Upton, NY	54
22	Berkeley, CA	13	51	Upton, NY	59
23	Cincinnati, OH	15	52	Upton, NY	61
24	Cincinnati, OH	15	53	Oak Ridge, TN	64
25	Upton, NY	17	54	Oak Ridge, TN	65
26	Pinellas, FL	17	55	Upton, NY	65
27	Golden, CO	18	56	Upton, NY	84
28	Pinellas, FL	19	57	Oak Ridge, TN	94
29	Oak Ridge, TN	19	58	Upton, NY	118

Table A-2. Results of bioassay testing of 100 eligible veterans

ID No.	²³⁹Pu Activity (aCi)*	Test Series†	ID No.	²³⁹Pu Activity (aCi)*	Test Series
P-6	2366	GH, BJ, IV,UK, CA, TP, PB	P-149	43	Hiroshima/Nagasaki (N)
P-85	1059/358	TS	P-89	41	IV
P-120	627/32	UK	P-92	41	Nagasaki (MC)
P-86	436/74	CA,TP	P-9	39	GH
P-64	274/237	Nagasaki (MC)	P-23	38	Nagasaki (N)
P-55	213	Nagasaki (MC)	P-60	36	UK
P-109	204	CA	P-117	36	Nagasaki (MC)
P-83	197/111	Nagasaki (MC)	P-35	35	CR
P-71	196	CR	P-12	33	CR
P-104	167/131	IV	P-5	33	CA, IV
P-17	165/128	Hiroshima	P-84	33	PB
P-31	158/35	RW	P-38	32	DO-1
P-105	151/97	RW, HT-1	P-42	31	CR
P-96	146	DO-1	P-97	31	Nagasaki (MC)
P-106	144	HT-1	P-27	30	Nagasaki (MC)
P-152	114	CA	P-63	29	CR
P-127	112	Nagasaki (N)	P-135	27	CR
P-76	109	Nagasaki (N)	P-146	27	CR
P-11	109	TS	P-3	25	Nagasaki (N)
P-125	108	BJ	P-7	25	CR
P-53	106	Hiroshima/Nagasaki (N)	P-121	25	TS
P-99	99/209	RW	P-101	24	BJ, TS
P-142	94	Nagasaki (MC)	P-1	22	Nagasaki (MC)
P-116	88	SS	P-25	22	HT-1
P-103	83	Hiroshima	P-36	21	CR
P-39	80	CR	P-50	21	Nagasaki (N)
P-140	80	Nagasaki (MC)	P-100	20	CR
P-79	79	CR	P-4	19	UK
P-136	76	Nagasaki (N)	P-51	19	Hiroshima
P-78	74	DO-1	P-107	19	Nagasaki (N)
P-26	73	CA	P-123	19	CR
P-57	70	pHT-1	P-102	18	BJ
P-87	69	Nagasaki (N)	P-113	18	UK
P-98	67	CR	P-150	14	Nagasaki (N)
P-134	67	CR	P-93	13	UK
P-80	64	BJ	P-54	10	BJ
P-138	62	CA	P-46	9	Hiroshima
P-94	59	BJ,TS	P-8	8	GH
P-13	57	CR	P-40	8	RW
P-28	57	GH	P-88	8	Nagasaki (N)

Table A-2. Results of bioassay testing of 100 eligible veterans (cont.)

ID No.	²³⁹Pu Activity (aCi)*	Test Series[†]	ID No.	²³⁹Pu Activity (aCi)*	Test Series
P-118	56	TS	P-49	6	RW
P-18	55	Hiroshima	P-81	6	Nagasaki (A)
P-110	54	CR	P-62	5	CA, GH
P-139	53	TS	P-32	5	DO-1
P-144	53	RW	P-61	2	Nagasaki (N)
P-65	52	Hiroshima/Nagasaki (N)	P-37	0	HT-1
P-58	47	RW	P-73	0	Hiroshima
P-148	46	UK	P-72	0	Hiroshima
P-108	45	Nagasaki (MC)	P-75	-2	SS
P-30	45	BJ	P-128	-2	Hiroshima

*Results for two splits are reported in the order—first split/second split; activity is on 24-h sample basis.

[†] Key to Test Series Entries

Nagasaki (A) – Army participant

Nagasaki (MC) – Marine Corps participant

Nagasaki (N) – Navy participant

CR – Crossroads

SS – Sandstone

GH – Greenhouse

BJ – Buster-Jangle

TS – Tumbler-Snapper

IV – Ivy

UK – Upshot-Knothole

CA – Castle

TP – Teapot

RW – Redwing

PB – Plumbbob

HT-1 – Hardtack-1

pHT-1 – post Hardtack-1

DO-1 – Dominic-1