# The Evolution of Internal Dosimetry Bioassay Methods at the Savannah River Site

# George A. Taylor

#### **Abstract**

Throughout the 50-year history of the Savannah River Site (SRS), its employees have engaged in many activities that potentially could result in an accidental intake of radioactive material. For that reason, radiation workers have always been monitored for internal deposits of radioactive material. An overview of *in vitro* and *in vivo* monitoring methods are provided, as well as computational methods used to calculate radiation dose.

#### Introduction

The Savannah River Site (SRS) has monitored radiation workers for internal deposits of radioactive material since the Site's inception. This monitoring is part of a comprehensive internal dosimetry program that also includes workplace and personal contamination monitoring, air monitoring, bioassay, dose evaluation, administrative scheduling and reporting, and regulation and oversight. Program objectives are to detect and assess intakes (quantity of radioactive material that passes through the nares, the mouth, of the skin) of radioactive material in the workplace. This paper will concentrate on the progression of the bioassay and dose evaluation programs at SRS.

Two methods of quantifying how much radioactive material is in the body exist. In the first method, an inference is made as to how much radioactive material is in the body from how much is excreted in the urine or feces. This is referred to as in-vitro bioassay. In the second method, an inference is made as to how much radioactive material is in the body from the amount of photon radiation (a particle of electromagnetic energy) emitted from the body. This is referred to as in-vivo bioassay, which includes chest and whole body counting (WBC). The method of choice depends on the biokinetics of the radioactive material in the body; in particular, the fraction of the intake excreted through the urine and the intensity and energy

of any photons emitted.

# In-Vitro Bioassay

Prior to completion of whole body counting facility in 1960, only *in-vitro* bioassay, primarily urinalysis (analysis of a urine sample), was performed. *In-vitro* urine bioassay for nuclear by-products performed during this period was typically a gross beta activity analysis. Since that time *in-vitro* bioassay analysis has become more radionuclide specific, and may be subdivided into the following categories:

- plutonium urinalysis
- tritium urinalysis
- uranium urinalysis
- trivalent actinide urinalysis
- neptunium urinalysis
- fission product urinalysis
- · fecal bioassay

# **Plutonium Urinalysis**

The first procedure for plutonium urinalysis (Sanders 1956) was implemented in 1954 and used until 1959. This procedure used bismuth phosphate and lanthanium fluoride coprecipitations to separate plutonium from 1500 mL of urine. The precipitate was resuspended in acid and electroplated on stainless steel disks. These disks were placed for one week on glass plates coated with Kodak film emulsions. The plates were developed and the

alpha tracks were counted to quantify the plutonium. This analysis for plutonium had a minimum detectable amount (MDA) on the order of 0.035 disintegrations per minute (dpm) per liter of urine, which is comparable with detection capabilities available today.

The coprecipitation procedure was labor intensive. In 1959 (Sanders 1961), an ion-exchange technique was adopted that cut processing time in half and reduced the amount of urine used to 250 mL, while keeping the MDA about the same. This procedure, which also employed electrode position and alpha track counting, was used until about 1966. A reporting level of 0.05 dpm per 1.5 liters of urine was used for plutonium urinalysis reported during the 1954-1966 period (i.e., the "less-than" level was 0.05 dpm per 1.5 liters). Results were expressed per 1.5 liters of urine because Standard Man was assumed to excrete 1.5 liters of urine per day, which permitted (in theory) a direct conversion from concentration to excretion per day. Standard Man represented a set of agreed-upon values for the many characteristics of man that are needed for internal dose calculation.

In the mid 1960s, two significant changes were made to the plutonium urinalysis. First, in 1964, solid-state surface barrier alpha detectors (Butler 1966) were introduced to replace Kodak film emulsions. These gross-alpha detectors reduced the time for counting a sample from one week to one day and greatly simplified the counting procedure. Second, around 1966, the ion-exchange method was replaced with a liquid ion-exchange method, utilizing the organic extractant trifluoro thionylacetone or TIOA (Butler 1968, Butler 1965). The reporting level for the TIOA gross-alpha method was 0.1 dpm per 1.5 liters of urine. With few changes, this method was used for plutonium urinalysis until 1988, a remarkable span of over 20 years.

In the early 1980s, experimentation began with a coprecipitation technique developed by Kressin (1981). This method used alpha spectroscopy, which permits the addition of Pu-242

tracer into a sample to determine radiochemical recovery of plutonium and detector counting efficiency. With gross alpha counting, only approximate "batch" estimates are made of these parameters. Up until about 1988, only routine urine samples were analyzed by the alpha spectrometry method and the TIOA ion exchange method was used for special urine samples. Reporting levels for alpha spectrometry during this period were 0.05 dpm per 1.5 liters for Pu-238 and 0.07 dpm per 1.5 liters for Pu-239. Around 1988, the coprecipitation step of Kressin's procedure was dropped to give the procedure that is in use today.

Since startup, *in-vitro* bioassay results were recorded on cards that were placed in the individual's personnel file. Starting in 1990, bioassay results were stored on a computer. At the same time, all plutonium results were converted from dpm per 1.5 liters to dpm per liter.

#### **Tritium Urinalysis**

From startup to 1958, tritium oxide in urine was analyzed by adding calcium metal to the urine and running the evolved hydrogen through an ionization chamber (Hursh 1958). The MDA for this method was 1  $\mu$ Ci/l. In 1958, an automatic sample changing liquid scintillation counter (LSC) was procured (Hursh 1958). This counter was much easier to use and, with a one-minute count, provided the same MDA as the ionization chamber method.

Through the years, the detection capability of the LSC method improved, but the 1  $\mu$ Ci per liter was retained as a reporting level. The reporting level was lowered, first to 0.5  $\mu$ Ci per liter and later to 0.1  $\mu$ Ci per liter, which is the level in use today.

#### **Uranium Urinalysis**

Analyzing uranium by measuring alpha radiation emitted is referred to as an analysis for "enriched uranium". The first method used for enriched uranium involved coprecipitation

followed by electrodeposition (depositing a substance at an electrode using a direct current) and Kodak film emulsion counting of alpha radiation (Boni 1960). This method had a reported sensitivity of 0.15 dpm per 1.5 liters of urine. In the mid 1960s, the TIOA/gross alpha counting method was adopted for enriched uranium analyses (Butler 1968). This method had an MDA of about 1 dpm per 1.5 liters, which was considered adequate at the time.

The analysis of uranium based on its chemical properties is referred to as a "depleted uranium" analysis. Analyses for depleted uranium were performed with the Oak Ridge fluorophotometric method from 1954 to 1982. This method involved using a spectroscope to measure total uranium.

The delayed neutron analysis (DNA) method (Hurley 1982) was adopted for both enriched and depleted uranium analyses around 1982. This method involved coprecipitating the uranium with calcium fluoride, activating the sample in a reactor, and counting the delayed neutrons emitted by the U-235. This procedure had an MDA of 0.14 ng of U-235, which provided a 1 µg per liter MDA for natural uranium and a 1 dpm per liter MDA for enrichments typically encountered at SRS.

With the shutdown in 1986 of the reactor facility used for DNA of uranium, the TIOA method was again adopted for enriched uranium and the Jarrell-Ash method (x-ray fluoroscopy) for depleted uranium. In 1990, an ion-exchange method was adopted for enriched uranium. This method, which is still in use today, uses alpha spectrometry with an internal U-232 tracer.

Kinetic phosphorimetry analysis (KPA) for depleted uranium was used from 1986 through 1994. Since 1994, both enriched and depleted uranium have been analyzed by the ion exchange/alpha spectrometry method.

#### **Trivalent Actinides**

The trivalent actinides consist of americium, curium, and californium. There was increased production of these materials at SRS in the early 1970s. Analysis was accomplished by extracting the trivalent actinides from the plutonium-TIOA raffinate with di,2-ethylhexylophosphoric acid (HDEHP) followed by gross alpha counting. Later, the HDEHP extraction was replaced with a organic extraction that was less complex. The procedure required 500 ml of urine and had an MDA of 0.3 dpm per 1.5 liters.

In 1990, ion exchange resin was used to separate the trivalent actinides from other actinides. The sample was direct mounted (placed on a planchet with rimmed edges and heated to evaporation), and was "gross-alpha" counted by using a wide window on the alpha spectrometer. This method can achieve MDAs on the order of 0.1 dpm per liter. This is a gross-alpha technique and therefore does not use an internal tracer.

Since 1994, extraction chromatography resin has been used to separate the trivalent actinides from other actinides. With the implementation of improved software and use of an Am-243 tracer, chemical recovery and counting efficiency may be determined for each sample.

#### **Neptunium Urinalysis**

Starting in 1959, neptunium was coprecipitated, ion exchanged, electrodeposited, and counted on Kodak film emulsion. In the mid 1960s (Butler 1968), the TIOA/gross alpha method was adopted. This method was replaced in 1993 by anion (negatively changed ions) exchange followed by direct mounting and gross-alpha counting.

Since 1994, extraction chromatography resin has been used to separate neptunium from fission products and other actinides, and electrodeposi-

tion has been used to mount the sample. There are no suitable isotopes of neptunium available to use as tracers, so this is still a gross-alpha counting technique.

#### **Fission Product Urinalysis**

Strontium urinalysis was performed in the 1950s and 1960s by coprecipitation followed by beta counting on a Geiger Mueller counter. Since 1969, strontium has been analyzed by a liquid ion-exchange method that extracts the Y-90, which is counted on a beta proportional counter. In 1998, the strontium method was modified by using an ion exchange column to extract Sr-90, which is also counted on proportional counter. Gamma-emitting fission products have historically been analyzed by gamma-spectrometry with sodium iodide detectors.

### Fecal Bioassay

The history of fecal analysis is somewhat fragmented. It appears that a modified TIOA procedure (DPSOL 1972) was used in the 1970s for fecal analysis of actinides, but it is uncertain when the procedure was adopted or abandoned. The primary method for fecal bioassay used in the early and mid 1980s was gamma-spectrometry with a phoswhich detector. Some of the samples analyzed by this method were subsequently dried and sent to offsite laboratories for analysis. Since 1994, fecal samples have been analyzed by a modified urinalysis procedure.

# In-Vivo Bioassay

The history of *in-vivo* bioassay at SRS may be broken down into three periods that are separated by the introduction of new detector technologies. The first period of *in-vivo* bioassay began with the development of large solid scintillation detectors made of sodium iodide in the 1950s that were used to measure high-energy photon emitters in the body. The development of low-energy phoswich detectors, which made detection of low-energy photon emitters feasible, began the second period in 1970. Finally, the modern period began at SRS in

the late 1980s with the introduction of high-resolution germanium detectors.

#### Whole Body Counting

*In-vitro* bioassay was the only method available to assess occupational intakes of radionuclides in defense facilities before the mid 1950s. During this period, in-vivo bioassay became feasible with the introduction of large sodium iodide detectors at Argonne National Laboratory and 4p liquid scintillator detectors, which made it possible for radioactive emissions emerging from the top and bottom surfaces of a sample to be counted, at Los Alamos Scientific Laboratory. Before the development of these detectors, invivo bioassay was performed with ionization chambers or Geiger-Muller tubes that were insensitive and not practical for assessing occupational intakes of radionuclides. In 1957, Sanders (1965) proposed that a whole body counting facility similar to that at Argonne utilizing sodium iodide detectors be built at

This state-of-the-art whole body counting facility was completed in 1960. The room, known as the steel room, was built of 12-inchthick pre-WWII steel that was originally fabricated as armor for a battleship. Pre-WWII steel was selected because it did not contain manmade radionuclides (fallout Cs-137 and Co-60 from smelters) that can increase the background in the room. The steel room is 8-1/2 feet wide by 11 feet long by 8 feet high, weighs 155 tons, and is lined with 1/8-inch low-radioactivity lead sheet.

The first detector used in the steel room was an 8-inch diameter by 4-inch-high cylindrical sodium iodide detector. The detector was covered with 1/2-inch of lead (except for the bottom) and had three photo multiplier tubes, devices used to convert weak light output of a scintillation pulse into a corresponding electrical signal. This detector had an effective energy range of 100 keV to 2000 keV, which meant that it could not be used to detect or quantify radionuclides like plutonium and americium

that emit only low energy (<100 keV) photons.

The counting geometry, or source and detector position, used was the 40-cm arc. It was also known as the Argonne chair. The person sat in a reclining chair and the detector was suspended above the pelvic area. The chair geometry placed the body from chin to knees approximately 40 cm from the detector. Limited investigational counts of the lungs were available with the detector placed in contact with the chest.

The count data was collected in a 200-channel multichannel analyzer. Analysis of the count data with an IBM 1620 computer (Watts 1963) began in 1963. The 200 channels were divided into 15 energy regions that represented photons of various energies. The background in these regions was determined for a number of individuals who had only varying amounts of natural K-40 and Cs-137 fallout in their bodies. From these data, a "prediction equation" was calculated that was used to estimate the background in the corresponding energy regions for a worker whose body may have contained other radionuclides.

The whole body counter was calibrated with the REMCAL phantom, a butyrate plastic phantom (geometric model) designed to visually resemble man. An accepted practice at this time was to calibrate whole body counters with volunteers who were administered small quantities of radionuclides. This technique was never used at SRS, but intercomparisons were done with other defense facilities that did. SRS was within 2-40% of their calibrations for radionuclides like Cs-137.

The MDA was defined to be a count that exceeded three times the standard deviation (3s) of the expected count rate in an energy region. The MDAs calculated for various radionuclides during this time are shown in Table 1 along with the corresponding maximum permissible body burdens. As indicated in Table 1, even the very first whole body counter was capable of detecting small intakes of high-energy photon emitting radionuclides.

Sometime during the early 1970s, the chair geometry was replaced with a bed geometry. Shortly thereafter, the single 8-inch by 4-inch sodium iodide detector was replaced with five 4-inch by 4-inch cylindrical sodium iodide detectors placed under the bed. The sodium iodide detectors were positioned in an arc under the bed to provide constant efficiency for all detectors. In essence, the detectors rather than the person were placed in an arc. Count data continued to be analyzed by the 200-

**Table 1.** Minimum Detectable Levels for SRS *In-Vivo* Counting Whole Body Count with 4 x 8 Crystal

Isotope	Minimum Detectable Level (20 min count) (nCi)	1% MPBB Values (nCi)
<sup>144</sup> Ce	29	50
NatU	62	.05
$^{131}I$	1.4	7
<sup>106</sup> Ru	6.1	30
<sup>137</sup> Cs	1.0	300
<sup>95</sup> Zr- <sup>95</sup> Nb	2.2	200
$^{65}$ Zn	5.1	600
<sup>140</sup> Ba- <sup>140</sup> La	9.3	40

All 4 x 8 crystal "whole body" count MDAs are based on +3 deviation from average clean person

channel MCA.

In the mid 1980s, commercially available standup counters using large sodium iodide detectors and a shallow shield arrangement (FASTSCAN) were obtained to provide 2-to-4-minute counts for high-energy photon emitters. Two of the counters were mounted in trucks so that they could be moved from place to place as needed.

#### **Chest Counting**

The whole body counter, useful for monitoring workers for intake of gamma-emitting fission and activation products, was of no use to detect or quantify intakes of many actinides such as plutonium and americium. Efforts had begun at SRS in the mid 1960s to develop low-energy photon detectors that would detect actinides in the chest. Most efforts involved either thin sodium iodide detectors or xenon-filled proportional counters. The xenon proportional counters, with their high resolution, showed great promise but were eventually abandoned because of their high background and low efficiency.

Development of the thin sodium iodide detector continued. In 1966, a 1-mm thick by 5-inch diameter detector with a 0.001-inch window was first used to count workers involved in a contamination incident. Thin sodium iodide detectors were used with varying degrees of success from 1966 until 1971, when they were replaced with phoswich "phosphor sandwich" detectors, composed of a layer of sodium iodide on top of a layer of cesium iodide.

This was a major breakthrough in *in-vivo* bioassay for low-energy photon. This detector's thin construction and inherent anti-coincidence counting (the elimination of undesired radiation) capabilities greatly reduced the background in the low-energy region of the energy spectrum. In 1972, a dual phoswich system composed of two 5-inch detectors was placed in service. Although improvements were made in

data analysis and calibration over the years, this basic system was used for chest counting for nearly 15 years until the adoption of planar germanium detectors in the late 1980s.

The detection capability of the phoswich detectors for plutonium is somewhat uncertain. In 1969, researchers at Los Alamos Scientific Laboratory (Dean 1969) claimed an MDA of approximately 1 nCi for Pu-238. During the 1970s, measurable Pu-238 chest burdens on the order of 4 to 10 nCi were reported at SRS. Currently, however, estimates of the MDA for Pu-238 using state-of-the-art germanium detectors is approximately 60 to 70 nCi. The cause of this discrepancy is not known, but it may be related to differences in the way the MDAs were calculated in the 1970s versus today.

In the late 1970s, small coaxial germanium detectors were installed in the steel room. These detectors were pointed at the sides of the chest to detect the 186 keV photon from the U-235 in enriched uranium, which was not readily detected by the phoswich or sodium iodide detectors. These detectors were also used to identify high-energy photon emitters, taking advantage of the superior resolution of the germanium detectors. In the mid 1980s, thick phoswich detectors specifically designed for enriched uranium detection were purchased. This turnkey system was only used for a few years before the germanium chest counter replaced it.

Routine use of a six-detector germanium chest counter began at SRS in December 1989, replacing the phoswich system. The detectors were 2000 mm² surface area, housed separately in "organ-pipe" dewars (Canberra ACT-I design). A reclining chair counting geometry was used. In August 1995, a pressure transient damaged the detector array. Because of this, a planned move to a new facility was accelerated, and new 2800 mm² detectors, two each housed in a 7-liter dewar (Canberra ACT-II design) were placed into service in September 1995.

#### **Chest Wall Thickness**

The thickness of the chest wall is an important factor in quantifying actinides in the chest. In 1970, ultrasound equipment was purchased to measure the chest-wall thickness. Through the years, the equipment was abandoned and upgraded several times. Eventually, an empirical height-weight to chest wall thickness algorithm was adopted to estimate the thickness of the chest wall. This algorithm, which is still in routine use today, is known to be inaccurate for females. Presently, in cases which intake evaluation is based on chest count bioassay data, direct measurement of chest wall thickness can be made by ultrasound techniques.

#### Calibration

To this day, a major problem with chest counting is the accurate calibration of the system. The early thin NaI detectors were calibrated with simple sources consisting of a suitable radioactive material plated on metal planchets. In 1969, a human skeleton with tissue-equivalent lungs was first used for calibrations. This phantom was used until the Livermore phantom was adopted in 1982. The Livermore phantom was designed and prototyped by in-vivo experts in the weapons complex. After a series of round-robin intercomparisons in the complex with the phantom in the late 1970s, the phantom was turned over to commercial firm for mass production. Up to that time, all sites in the complex were using different phantoms of their own design and construction. Thus, with the introduction of the Livermore phantom, for the first time a standard chest phantom was readily available to anyone with enough money to buy it (~\$20,000). Over the years, this *de facto* standard has greatly improved the consistency of chest counting throughout the complex. More significantly, the success of the Livermore phantom in the early 1980s started a move at SRS and in the complex to "turnkey" commercial phantoms, detectors, and software that have essentially eliminated in-house research and development in the area of *in-vivo* bioassay.

# Internal Dose Computational Methods

Assessment of internal dose for workers in the nuclear weapons complex has always been based on the evaluation of bioassay data rather than air monitoring data. The methods used to evaluate bioassay data throughout the history of SRS have changed to keep pace with improvements in the technology of internal dose assessment and the evolution of internal dose regulations. All radionuclides have not been impacted equally as things have changed. For example, the methods used to evaluate intakes of tritiated water have not changed significantly in over 40 years whereas the methods used to evaluate intakes of plutonium have changed significantly.

# **Calculating Body Burdens**

From plant startup until 1975, internal dose limits were expressed in terms of the MPBB or maximum permissible organ burden (MPOB). The body or organ burden for radionuclides that emit high-energy photon radiation and target specific locations in the body, like Cs-137 and I-131, are directly determined by *in-vivo* bioassay. No calculations are required. No significant intakes of these materials has ever occurred at SRS, with few measured burdens exceeding a percentage of the MPBB.

Intakes of radioactive materials that cannot be quantified by *in-vivo* bioassay are more difficult to evaluate because inference must be made to what is in the body from what is coming out. The standard practice for evaluating intakes of these materials (DPSOL 1968 and DPSOL 1968a) except for tritiated water during the 1954-1975 period was to:

- collect enough excretion data, usually urine, to define the excretion curve
- integrate the excretion curve from the time of uptake to infinity to calculate the total amount of material excreted through the urine

 adjust the integral for the fraction of an uptake excreted by pathways other than urine to determine the uptake

The excretion data were typically fit to simple power functions of time like

$$A = Et^{-b}$$

where A is the amount of material in the urine at time t, and E is the amount of material in the urine at t=1. E and b were determined in the least squares fit. The power function was popular at this time because it can describe multicomponent excretion curves and be readily transformed into a simple linear form that is easy to manipulate mathematically without the aid of sophisticated computers. If few data were available, default values were used for the exponent b such as -0.74 for plutonium, and the available data were used to determine E. Evaluations from this period that appear in personnel files are typically not well documented as to the exact methods used. However, it can be safely assumed that the power function method was used because no evidence supporting the use of another method has ever been found.

Notice that the power function method gives an uptake, which is the systemic body burden shortly after the time of the intake. Material in the lungs or in a wound was not accounted for and was usually ignored until it eventually became systemic (absorbed in the blood). Starting with the installation of the dual phoswich chest counter in 1971, it became possible to directly measure chest burdens as long as they were quite large. Measured chest burdens were compared with the maximum permissible lung burden (MPLB) to determine their significance.

#### Whole Body Dose from Tritiated Water

Tritiated water is unique among internal emitters at SRS because intakes of it have always been evaluated in terms of whole body dose equivalent that has been added to whole body dose equivalent from external sources. This was true even during the 1954-1975 period when limits were expressed in terms of the MPBB. The basic technique for calculating dose from tritiated water urinary excretion data has changed little over the years. For an acute intake of tritiated water it consists of

- fitting the urinary tritium concentration data to a single exponential
- integrating the urinary concentration curve
- multiplying by a constant to get dose.

The primary differences between the current method, which is based on Publication 30 of the International Commission on Radiological Protection (ICRP 30), and earlier methods, which were based on ICRP 2, are the

- default biological half-life (12 days then, 10 days now)
- target tissue (43 kg of body water then, 63 kg of soft tissue now)
- default mass of body water (43 kg then, 42 kg now)
- quality factor for tritium (1.7 then, 1.0 now)
- mean energy of tritium beta particle (6.1 keV then, 5.7 keV now).

Note that the default half-life is used only to interpolate between samples under a chronic, or recurrent, intake pattern. For acute intakes, the person's biological half-life, the time at which one half of a given substance is eliminated by biological processes was determined from his excretion curve. The ICRP 30 methodology for calculating tritium dose, especially the quality factor and target tissue, was adopted in 1986 (Reinig 1986). Changes in the default biological half-life and mean beta energy were probably made before 1981.

The exact methods used to calculate dose from chronic exposure to tritiated water prior to 1980 are not known for certain. In the days prior to computer evaluation of the data, only results greater than 5  $\mu$ Ci/l were evaluated (Reinig 1963), probably by a linear interpolation method (La Bone 1992). As computers became available,

all results were evaluated in terms of dose by an exponential interpolation method (Boone 1994).

## **Organ Doses**

From 1975 to 1987, limits for internal emitters were given in terms of organ doses in a year. Organ dose equivalent ( $H_T$ ) over the time period from  $t_1$  to  $t_2$  was calculated using the following formula (Hall 1980):

$$H_{T} = \frac{1.87 \times 10^{4} q \mathcal{E} f}{m} \int_{t_{1}}^{t_{2}} e^{-\lambda t} dt,$$

where

 $q = body burden at time t_1$ 

f = fraction of body burden in organ, and

e = effective energy for radionuclide in organ.

Although this is an ICRP 2 type calculation, the referenced document, which was issued to document the first Annual Exposure Reports, uses many ICRP 30 parameters. For example, the f for plutonium in bone is 0.45, which is the parameter given in ICRP 30. As new internal dosimetry methods evolve, it is quite common for parts of the new methods to be incorporated into the old methods. This is as true today as it was in 1960 and is a natural consequence of trying to use the most current technology.

#### **Intake Assessment**

As discussed earlier, ICRP 30 methods began to be incorporated into existing intake evaluation procedures in the late 1970s and early 1980s. In ICRP 30, the concept of calculating an intake from bioassay data and using this intake to calculate a 50-year committed effective dose equivalent (CEDE) was introduced. In 1987, the contamination cases of all active employees who had intakes of radioactive materials other than tritium were re-evaluated using ICRP 30 methods. All of these individuals were informed of the reassessment and were given the new dose estimates. DOE Order 5480.11, which required the calculation of annual and CEDE, was adopted in 1989. *The DOE Radiological Control* 

*Manual,* which required the exclusive use of CEDE, was adopted in 1993.

Work continued on the reevaluation of the cases of all the non-active (retired, terminated, or deceased) employees, and work was completed in 1992. These individuals were not informed of the reassessment unless they requested dosimetry information after 1987. Thus, as of 1992, all known intakes of radioactive materials other than tritium had been reassessed with ICRP 30 methods.

The methods used to evaluate bioassay data to calculate intakes and dose are documented in the SRS Internal Dosimetry Technical Basis Manual. The methods used to evaluate bioassay data have not changed since 1987. Internal doses from radionuclides other than tritium will be continually refined as new and improved biokinetic and dosimetric models are introduced and additional bioassay data are collected. Because intakes of tritiated water have always been addressed in terms of whole body dose, historic intakes of tritiated water are not reevaluated to incorporate the newest models. This is the same policy used for historic external doses, which are not reassessed.

#### Chelation Therapy

Once an intake of a transuranic (plutonium, americium, and curium) occurs, there are only three therapeutic procedures available to reduce the intake and mitigate the dose: excision of material from wounds, removal of the material from the lungs with lavage, and chelation therapy. Chelation therapy enhances the urinary excretion of transuranics from the body by binding of a chelation agent to transuranic elements in the bloodstream. Hall et al. (Hall 1978) published a mathematical model for evaluating intakes of plutonium after chelation therapy with diethylene-triamine-pentaacetic acid, better known as DTPA. This model was derived from bioassay data of workers at SRS who had received DTPA treatments. Prior to the publication of this model, intake evaluation following chelation therapy was not possible using the conventional methods.

#### References

Boni, A. L., 1960, "Urinalysis Method for Enriched Uranium," *Health Physics* (2), 1960, pp. 288-290.

Boone, F. W., 1994, Description of Routine Tritium Dose Calculational Model, ESH-HPT-940226, July 25, 1994.

Butler, F. E., 1965, "Determination of Uranium and Americium-Curium in Urine by Liquid Ion Exchange," *Analytical Chemistry* (37), 1965, pg 340.

Butler, F. E., 1968, "Rapid Bioassay Methods for Plutonium, Neptunium, and Uranium," *Health Physics* (15), 1968, pp. 19-24.

Butler, H. L., and W. F. Splichal, Jr., 1966, "Solid State Alpha Counters: A Replacement for Nuclear Track Counting in Bioassay Procedures," *Health Physics* (12), 1966, pp. 1627-1630.

Dean, P. N., W. H. Langham, and H. M. Ide, 1969, External Measurement of Plutonium Lung Burdens 15th Annual Bioassay and Analytical Chemistry Conference, October 9-10, 1969.

DPSOL Manual 47, 1972, "Plutonium, Neptunium, Enriched Uranium, Americium-Curium-Califorium Sequential Determination in Feces," Procedure 330, May 1972.

DPSOL Manual 47, 1968, Enriched Uranium, Procedure 504, Rev. 0, June 1968.

DPSOL Manual 47, 1968a, Plutonium (Estimation of Body Burden and Radiation Dose), Rev. 0, May 1968.

Hall, R. M., G. A. Poda, R. Fleming, and J. A. Smith, 1978, "A Mathematical Model for Estimation of Plutonium in the Human Body from

Urine Data Influenced by DTPA Therapy", *Health Physics* (34), 1978, pp. 419-431.

Hurley, J. A., 1982, *Uranium Bioassay by Delayed Neutron Counting*, Proceedings of the 28th Annual Conference on Bioassay, Analytical and Environmental Chemistry, Boston, Massachusetts, October 1982.

Hursh, J. B., 1958, editor, *Chemical Methods for Routine Bioassay*, AECU-4024, November 1958.

Kressin, I. K., 1981, "Separation of Plutonium in Urine without Sample Ashing for Determination by a-Spectrometry, "Analytical Chemistry (53), 1981, pp. 1270-1274.

Reinig, W. C., 1986, "Tritium Dosimetry," memo to W. T. Thornton dated April 14, 1986.

Reinig, W. C. and E. L. Albenesius, 1963, "Control of Tritium Health Hazards at the Savannah River Plant," *American Industrial Hygiene Association Journal* (24), May-June 1963, pp. 276-283.

Sanders, S. M., 1956, *Determination of Plutonium in Urine*, DP-146, March 1956.

Sanders, S. M., 1965, *In Vivo Measurements of Radionuclides*, undated document circa 1965.

Sanders, S. M., and S. C. Leidt, 1961, "A New Procedure for Plutonium Urinalysis," *Health Physics* (6), 1961, pp. 189-197.

Watts, J. R., 1963, "Health Physics Utilization of an IBM 1620 Computer", *Health Physics* (9), 1963, pp. 1001-1003.

# **Biography**

George A. Taylor began his professional career at the Savannah River Site in 1989 after receiving a MS in Health Physics at Texas A&M University. From 1989 to 1991 he was assigned to Environmental Monitoring where he was responsible for issuing various environmental reports to DOE and State officials. He also authored the Site's annual Environmental Monitoring Report and managed the environmental TLD program. In 1992 he became a member of the Field Technical Support group in Health Physics Technology. He has developed several site-wide technical guidance documents that have been incorporated into the SRS radiological control program. In 1995 he captured historical dosimetry practices in "A History of Personnel Radiation Dosimetry at the Savannah River Site". Since joining Health Physics Technology, he also has held technical support roles in Internal Dosimetry Programs and Instrument Programs.

Intentionally left blank