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Data Quality Objectives Supporting the U.S. Transuranium and Uranium Registries Mission

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Preface

The progenitor of what is now the USTUR was formally established in August 1968 as the National Plutonium Registry by the Hanford Environmental Health Foundation (HEHF) under contract to the United States Atomic Energy Commission (AEC). The HEHF medical director, Philip A. Fuqua, invited a number of recognized scientists to serve as members of a blue ribbon Advisory Committee to help guide the Registry. The six initial committee members included three physicians: Clarence C. Lushbaugh, Thomas F. Mancuso, and J.H. Sterner; two physicists: Robley D. Evans and Herbert M. Parker; and a toxicologist Lloyd M. Joshel. Biophysicist, Wright Langham was added the following year. The National Plutonium Registry's name was changed to the United States Transuranium Registry (USTR) in its second year of operation, 1970. This change reflected the program's concern for other transuranic elements as well as plutonium.

Although uranium had been known to and used by man for more than two centuries, there was still a great deal to be uncovered regarding its behavior and effects in humans. Thus, the United States Uranium Registry (USUR) was established in 1978 by the Hanford Environmental Health Foundation under DOE funding. Although the USUR adopted a similar operational strategy to that of the USTR and utilized much of the same staff as its sister registry the USTR, the two organizations were administratively separate.

In 1981, the Advisory Committee recommended that the Transuranium and Uranium Registries be combined into a single entity. Although this recommendation was not immediately acted upon, a combined USTR and USUR Advisory Committee was created. The 1983 Advisory Committee discussed the Uranium Registry, suggesting that both the Uranium and the Transuranium registries should increase their understanding of uranium biokinetics and analysis. The USUR director, Robert Moore, retired in 1989 and USTR director, Margery Swint, assumed directorship of both registries for a short time before accepting a promotion at Hanford Environmental Health Foundation. Ronald Kathren then became the second USTR/USUR director. Though the two Registries remained administratively separate, the first combined Annual Report for the Registries was published in 1990.

The USTR and USUR were administratively joined in 1992 when the US DOE awarded a three year grant to Washington State University (WSU) for the management and operation of the Registries. The registries were renamed the U.S. Transuranium and Uranium Registries (USTUR) and continued to operate under the direction of Ronald L. Kathren. WSU's College of Pharmacy assumed responsibility for the registries at that time and radiochemistry operations moved from Los Alamos National Laboratory to WSU Pullman, WA in 1994.

Since moving to Washington State University in 1992, the USTUR has performed most of its own radiochemical analyses. This document forms a key part of the USTUR's evolving Quality Assurance Program Plan by documenting the computations performed using the results of radiochemical measurements, including mass and volume measurements, as well as counting results from alpha spectrometry. The USTUR uses the Multi-Agency Radiological Laboratory Analytical Protocols (MARLAP) Manual as its primary guidance, along with ANSI-ANS N41.5-2012 and other sources. Because results sometimes may be used for radiation dosimetry, USTUR uses radiological decay data from ICRP Publication 108 rather than other sources.

Acknowledgments

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Executive Summary

The “Data Quality Objectives” (DQO) process was originally developed by federal agencies to ensure that data of acceptable completeness and sufficient quality would be available to inform decisions about environmental cleanup. The USTUR generally does not make decisions based on its radiochemical measurement data, but rather uses the data it generates to quantitatively describe the biokinetics of uranium and transuranium elements in the human body. In this document, the USTUR presents its adaptation of the DQO process as described in MARLAP (2004) and ANSI/ANS-41.5-2012 as a key part of its Quality Assurance Project Plan (QAPP). Other parts of the QAPP can be found in other USTUR procedures and documents. The USTUR has chosen to adopt the methodology and terminology of MARLAP insofar as appropriate. The reader is directed to the Data Dictionary in Appendix B for definitions of symbols.

The DQO process includes specifying performance criteria for

- collecting, weighing, and packaging tissue samples at autopsy (completeness);
- radiochemical processing of partial or whole tissue samples to extract radionuclides of interest (most often isotopes of U, Pu, and Am, but with capabilities for isotopes of Ra, Th, Np, and Cm); and
- radioactivity measurements, uncertainty analysis, and quality assurance of results.

To meet the needs of the QAPP, the processes leading to measurement results are described in Chapter 4. Sample processing, collection, dissection, preservation, and storage, masses, and tracking are described. Sample radiochemical analysis includes a description of sequence of radiochemistry processing events, as well as considerations of aliquot size, analytes of interest, origins of radionuclides present at various stages of radiochemical analysis/times, activities, activity concentrations, and other quantities. Following a discussion of biokinetic modeling, dose assessment and a discussion of available resources and relevant timelines are presented.

Chapter 5 discusses performance indicators such as critical value, minimum detectable activity (*MDA*), and minimum quantifiable activity (*MQA*) for alpha spectroscopy measurements. The critical value is generally not used for autopsy samples because such samples are known to contain the radionuclides of interest from the Registrant’s work history and thus no detect/nondetect decisions are needed. Typical laboratory counting parameters are 15 counts observed during a 300,000-s background count time; a 150,000-s sample count time; a radiochemical recovery yield of 0.9; and a counter efficiency of 0.25. Using these parameters, the *MDA* for Pu or Am is 0.44 mBq (0.026 dpm) and the *MQA*(0.10) is 3.7 mBq (0.22 dpm).

Chapter 6 presents and summarizes an exhaustive listing of performance criteria for accuracy, precision, representativeness, completeness, and sensitivity. Two measurement quality objectives for Pu and Am are 1) to detect a known activity of

1/30 Bq (2 dpm) with a relative combined uncertainty u_R of 0.04 or less (this is the *MQA*(0.04)); and 2) to detect a known activity of 0.50 mBq (0.03 dpm) with u_R = 0.35(this is slightly above the *MDA* under optimal conditions). For perspective, note that the *MQA*(0.04) is roughly 76 × *MDA*, and the *MQA*(0.1) is roughly 8.4 × *MDA*. Laboratory detection capabilities are evaluated for 74 Bq (2 nCi) intakes of Pu and Am 50 years prior to death, and found to be adequate or barely adequate for the critical tissues: lung, liver, and skeleton. Technology shortfall is discussed, and its management presented, including resorting to commercial or university accelerator mass spectrometry services. Since intakes by many Registrants are much higher than 74 Bq, often samples have easily quantified activity. A comprehensive list of performance criteria and quality control (QC) samples, including those described above, appears in Table 15 at the end of Chapter 6.

Chapter 7 of this report discusses data validation, verification, and assessment. While these topics are not about DQOs, they form a key part of the QAPP and are included here for that purpose.

Appendix A presents the forward-calculation of the activity that is predicted to be on a planchet following a 74-Bq (2-nCi) intake of various chemical forms of 7 isotopes by inhalation or wound.

Appendix B is the data dictionary.

Among the innovations in this report are detailed equations, both in the text and in the Appendices, to calculate output quantities needed by USTUR researchers (typically mass, activity, and activity concentration) from the various radiochemistry measurement results. Appendix C contains the balance of equations not shown in the text, including the intricate mathematics of correcting raw activity results for interferences by both crosstalk during counting and presence and ingrowth of interfering radionuclides in tissue samples, solutions, and radioactivity standards. Uncertainties are calculated for all output quantities, and in many instances, derivations are given for uncertainties because they are unique to the USTUR. Additionally, the USTUR has chosen to use MARLAP's best-performing formulas for the statistics described in Chapter 5. These include the well-established decades-old Bayesian practice of using (N+1) counts in calculations when N counts were observed, as outlined in MARLAP.

Besides developing DQOs and measurement quality objectives (MQO)s, this report presents calculations of the critical values, MDAs, and MQAs for background counts from 0 to 30, underscoring the importance of keeping background as low as reasonably achievable. The relative uncertainty of any measurement is never less than about 3.3% due to the use of 1/30 Bq (2 dpm) of tracer activity, which produces about 1,000 net counts in routine measurements under normal conditions.

Short Forms: Abbreviations, Acronyms, Codes, Initialisms, and Symbols

Additional, specific notations are given in Tables 1, 2, 5, 6, 10, 14 (MARLAP flags), and most importantly, Appendix B (Data Dictionary).

Algebraic variables are in *italics*. Labels and constants are in a normal font.

Short Form	**Meaning**
$\%SD_{\mathrm{R}}$	relative standard deviation as a percent (same as u_{R} when expressed as a percent)
´	time derivative, e.g., S'; also used for “corrected volume”
°C	degree Celsius
A	*activity*; default unit is Bq but other units are used as needed, e.g., nCi
aliq	aliquot (subscript)
Am	chemical symbol for americium
ANS	American Nuclear Society
ANSI	American National Standards Institute
aut	autopsy (subscript)
avg	average
B	background (subscript)
b	*bias*
bkr	beaker (subscript)
Bq	becquerel
C	count (subscript); critical (subscript on L_{C})
Ci	curie
Cm	chemical symbol for curium
cpm	counts per minute
D	death (subscript on time t); detection (subscript on L_{D})
d	Stapleton’s d-factor
dis	disintegration
DL	ANSI N-13.30 (1996)’s *decision level*
dpm	disintegrations per minute
DQO	data quality objective
dry	dry (subscript)
DT	ISO’s *decision threshold*
EPA	Environmental Protection Agency
EST	Eastern Standard Time
fCi	femtocurie (10^{-15} Ci)
$f_{X,\mathrm{ROI}}$	*fraction of counts expected from X in the ROI*
g	gram
GUM	Guide to the Expression of Uncertainty in Measurement
HPS	Health Physics Society
hr	hour
ICP-MS	inductively-coupled mass spectrometry
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis

Short Form	**Meaning**
init	initial (subscript)
ISO	International Organization for Standardization
JCGM	Joint Committee for Guides in Metrology
k	*uncertainty expansion factor* (NIST)
keV	kiloelectronvolt, a unit of energy
L	liter
L_C	Currie's (1968) *critical level*
L_D	Currie's (1968) *detection level*
LOD	*limit of detection*
L_Q	Currie's (1968) *determination limit*
m	mass
MARLAP	Multi-Agency Radiological Laboratory Analytical Protocols Manual
mBq	millibecquerel (10^{-3} Bq)
MDA	minimum detectable amount
MDC	minimum detectable concentration
MeV	megaelectronvolt
mg	milligram
min	minute
mL	milliliter
MPBB	maximum permissible body burden
MQA	*minimum quantifiable activity*
MQO	measurement quality objective
N	*number of counts*
N	net (subscript)
N/A	not applicable
nCi	nanocurie (10^{-9} Ci)
NCRP	National Council on Radiation Protection and Measurements
NHRTR	National Human Radiological Tissue Repository
NIST	National Institute of Standards and Technology
Np	chemical symbol for neptunium
OIR	Occupational Intake of Radionuclides
p	probability
pCi	picocurie (10^{-12} Ci)
prep	prepared tissue sample (subscript)
Pu	chemical symbol for plutonium
QA	quality assurance
QAPP	Quality Assurance Project Plan
QC	quality control
r	*ratio*
R	*count rate*
Ra	chemical symbol for radium
RB	reagent blank
ref	reference
RESL	Radiological and Environmental Sciences Laboratory
ROI	region of interest (in an alpha particle energy spectrum)
s	second

Short Form	**Meaning**
S	sample (subscript)
S_C	*critical value of the net counts* (MARLAP notation)
S_D	*minimum detectable value of the net instrument signal*
samp	whole tissue sample (subscript)
sep	separated, separation (subscript)
soln	solution (subscript)
SOW	statement of work
SSPEP	Site Specific Performance Evaluation Program
t	*time* (s)
T	temperature
$T_{1/2}$	half-life (days)
T_A	ambient temperature
Th	chemical symbol for thorium
THEMIS	the USTUR's Management Information System
TL	tolerance level
tr	tracer (subscript)
U	chemical symbol for uranium
u	*uncertainty*
U-nat	natural uranium
u_R	relative uncertainty
USTUR	United States Transuranium and Uranium Registries
V	*volume* (L, ml, µL)
VIM	International Vocabulary of Measurement
wet	wet (subscript)
X	isotope of interest (subscript), e.g., ^{239}Pu
Y	tracer isotope (subscript), e.g., ^{242}Pu
y^*	ISO's "decision threshold"
y_{RR}	radiochemical recovery yield
Z	element (subscript), e.g., Pu or Am
z	standard normal deviate
α	alpha
β	beta
ε	counting efficiency
λ	decay constant
µ	prefix micro (10^{-6})
µBq	microbecquerel (10^{-6} Bq)
Π	pipette (subscript label)

1. INTRODUCTION

This Data Quality Objectives (DQOs) document addresses the sample collection and data analysis needs in support of the United States Transuranium and Uranium Registries' (USTUR) mission.

1.1. MISSION STATEMENT

The USTUR's mission is to:

- Follow up occupationally exposed individuals (volunteer Registrants) by studying the biokinetics (deposition, translocation, retention, and excretion) and tissue dosimetry of uranium and transuranium elements, such as plutonium, americium, curium, and neptunium;
- Obtain, analyze, preserve, and make available for future research, materials from individuals who had documented intakes of uranium and transuranium elements; and
- Apply USTUR data to refine dose assessment methods in support of reliable epidemiological studies, radiation risk assessment, and regulatory standards for radiological protection of workers and the general public.

1.2. REGISTRANTS

The current acceptance criterion to be a Registrant at the USTUR is to have a documented intake of 2 nCi (74 Bq) of actinides. Originally, it had been an intake of 4 nCi, which is 10% of the then-Maximum Permissible Body Burden (MPBB) of 40 nCi[1].

2. STATEMENT OF THE PROBLEM

Chapter 2.0 of this document describes the primary study question and discusses the preliminary data used to answer the study question. In addition, this chapter discusses the DQO team, available resources, and schedule for completion of the DQO document.

The goal of this document is to determine an optimal tissue sample collection, measurement, and data analysis techniques to meet the mission statement objectives.

2.1. DATA REQUIRED TO MEET THE MISSION STATEMENT OBJECTIVES

The USTUR studies the biokinetics and internal dosimetry of actinides, primarily uranium, plutonium, and americium. Systemic plutonium and americium concentrate in the liver and skeleton, while uranium primarily concentrates in the skeleton. Inhalation and wound intakes are the most common routes of intake. Thus, the following tissues are primarily collected and analyzed to test, validate, and improve the International Commission on Radiological Protection (ICRP) and National Council

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[1] The 2 nCi limit never applied to NHRTR ^{226}Ra cases in the USTUR archives. The radium cases generally had much higher intakes.

on Radiation Protection and Measurements (NCRP) biokinetic models: lungs, thoracic lymph nodes, liver, skeleton, and, for a wound intake, wound site and axillary lymph nodes. Even if a worker with a contaminated wound had no documented inhalation of plutonium, the lungs and thoracic lymph nodes are analyzed to confirm that no material was inhaled.

The above tissues are critical for biokinetic modeling; however, a broader set of tissues should still be collected and analyzed. Analysis of other soft tissues, such as muscle or spleen, reduces uncertainty during biokinetic modeling by allowing researchers to better estimate the amount of actinide in a donor's whole body. Additionally, analysis of internal organs provides data that will be readily available for new ideas and future research. Thus, most of the tissues collected during a partial-body donation must be analyzed and a survey analysis must be performed on tissues from whole-body donations. Survey analysis provides key initial information by analyzing the same organs and tissues from a whole-body donation that would be analyzed following a partial body donation.

Any remaining tissues are stored for future use**.** Most notably, this includes tissues from the left-hand side of the body, half of single organs such as the heart, bodily fluids, hair, and nails.

2.2. PLANNING TEAM MEMBERS

This step in the DQO process addresses development of a planning team that will define the problem and implement subsequent steps of the process. It also calls for the identification of a decision maker who will lead the planning team and make final resolutions during the process. The goal is to create a well-structured planning team that will work effectively and efficiently to develop a concise and complete description of the problem, which will provide the basis for the rest of the DQO development.

The DQO planning team includes:

- Lead Scientist with background in radiochemistry, radiation detection, and radiation dosimetry. This member is a final decision maker.
- Scientist with expertise in biokinetic modeling and internal dosimetry.
- Scientist with expertise in radiation measurements.
- Scientist with experience in tissue radiochemical analysis.
- Scientist with experience in Quality Assurance (QA) and background in the DQO process. This member is the DQO facilitator.

3. GOAL OF THE DQO PROCESS

The ultimate goal of the DQO process is to ensure that the USTUR collects and retains data of sufficient quality to support USTUR's research mission. The USTUR's mission requires

- the collection, mass measurement, packaging, labeling, and archiving of tissue samples at autopsy;
- the radiochemical analysis of all or parts of samples; and
- the analysis of measurement results to characterize their values and uncertainties, as well as providing metrics of the quality of the measurements.

The information required to optimize the process includes the data and bounding conditions to identify, as necessary, sample collection and analysis, equipment, and data use to demonstrate adequate assessment of radionuclide concentration in organs and tissues of USTUR donors.

3.1 TISSUE SAMPLES COLLECTED AT AUTOPSY

USTUR personnel identify the core tissue samples to be collected at autopsy consistent with the USTUR Tissue Sample Collection Procedure. The sample list is adjusted to maximize the usefulness of the available tissues for biokinetic modeling and dosimetry.

3.2 ACTIVITIES IN DOSIMETRICALLY IMPORTANT ORGANS AND TISSUES

Available exposure information from the USTUR health physics database and/or individual exposure file is used to identify the radionuclide(s) of concern, route(s) of intake, material solubility type, and worksite estimates of deposition or intake to guide the radiochemical analysis of tissue samples. Radiochemical analysis of tissue samples is performed.

3.3 UNCERTAINTY IN USTUR ACTIVITY MEASUREMENTS

Measurement uncertainties are calculated as a function of activity and number of background counts. Performance metrics, such as critical value of the net counts (S_C), p-value, minimum detectable activity (MDA), and minimum quantifiable activity (MQA), are calculated, recorded, and used as described below.

4. INPUTS

This chapter lists and describes the sources and methods used for addressing the needs identified in Section 3. All notation is identified in the Data Dictionary (Appendix B).

4.1. SAMPLE PROCESSING

4.1.1. SAMPLE COLLECTION

Organs from whole-body donations are collected during autopsy at the USTUR facility, and their weights are recorded. These samples are listed in Phase 1 of the Whole Body Specimen Worksheet (F402a). The remaining tissues are frozen, and the samples listed in Phase 2 of F402a are dissected in the following the autopsy.

Samples from partial-body donations are collected by a pathologist during the autopsy. The pathologist will weigh samples, record their weights on the Partial Body Specimen Worksheet (F402b), place them in pre-labelled specimen bags, and ship them to the USTUR. Form F402b contains the default list of samples that are collected during a partial-body autopsy; however, this list may be modified based upon factors such as the Registrant's exposure history.

The USTUR has a single pathologist who performs all whole-body and local partial-body autopsies with assistance of USTUR staff, resulting in greater sample collection consistency. Autopsies performed by external pathologists may introduce variability due to partial sample collection and/or omission of samples listed in F402b.

4.1.2. Sample Dissection

All samples are dissected in a consistent manner. For example, bones collected at autopsy are further dissected into predetermined subsamples as listed in F402a, Phase 2. Excess fat and/or tissue are removed from samples prior to analysis or storage.

4.1.3. Sample Preservation and Storage

Samples are routinely vacuum packaged, inventoried, and stored in a –30°C freezer. Occasionally, samples may be stored in neutral buffered formalin solution.

4.1.4. Sample Masses

We give unique names to each variable, including intermediate results that could be output in any specified format, so that calculations can easily be verified. Names used at the USTUR are often unique, with some generic names as well.

Tare, gross, and net mass measurements are made at several stages of radiochemical processing (Figure 1).

At autopsy, masses of various tissues are measured as a part of the typical autopsy process; these are denoted as m_{aut}. However, such masses are generally not used for analysis, as they are for tissues not dissected to the degree preferred for USTUR analysis, and frequently include mass lost in packaging, transport, and/or storage before tissues can be dissected to the degree required by the USTUR.

When packaged tissues are opened for dissection at the USTUR, the mass of the tissue is measured again after extraneous tissue has been removed. A sample such as an entire respiratory tract must be further dissected into separate physiologically and dosimetrically important tissues including but not limited to the larynx, trachea, bronchus, pulmonary lymph nodes, lung tissue, and possibly tumors. Each of these pieces will have its mass measured, and these masses are termed m_{init}. In the case of complete organs, such as the liver, that are further divided for analysis, sample masses are measured again. These are the "prepared samples" whose masses are measured and denoted as m_{prep}. If no further dissection is needed for a sample, m_{init} may equal m_{prep}.

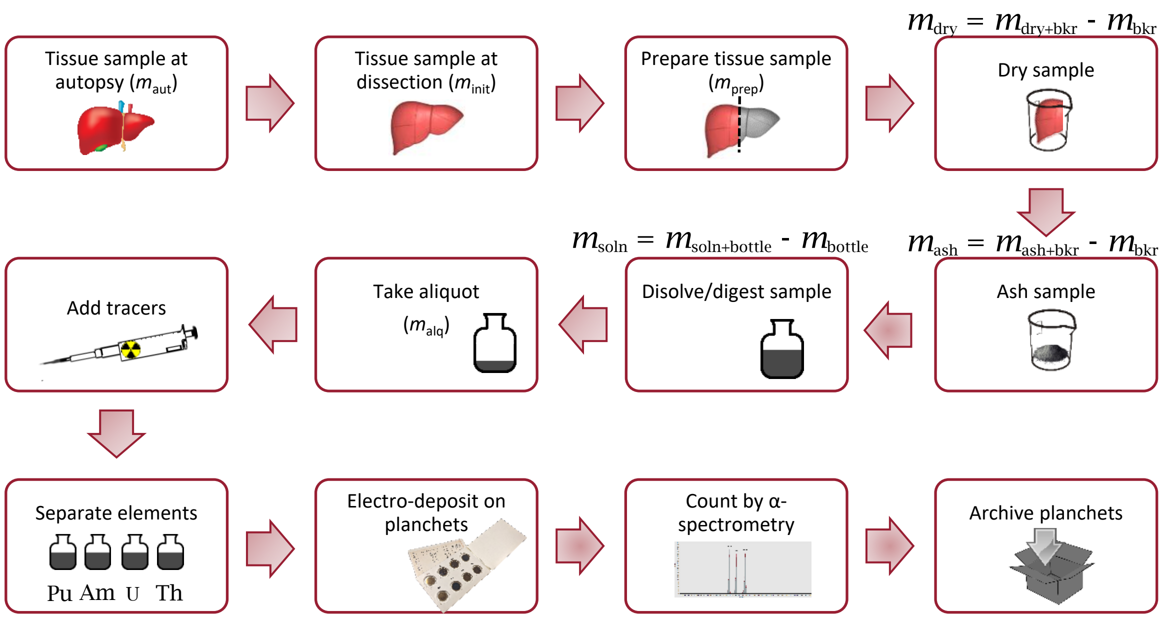

Figure 1. Mass measurements are made at several stages during radiochemical processing. Notation is described in Table 1.

Table 1 describes naming convention for stages of the radiochemistry process. All equations used during sample processing steps are given in Appendix C of this document.

Table 1. Naming convention for stages of the radiochemistry process at USTUR.

Name	Label
Tissue sample at autopsy	aut
Tissue at dissection	init
Prepared tissue sample	prep
Dry sample	dry
Ashed sample	ash
Acid solution	soln
Aliquot	aliq
Tracer	tr
Elemental fraction	*Z*

4.1.5 SAMPLE TRACKING

The Management Information System (THEMIS) inventory database used in the USTUR Radiochemistry laboratory automatically maintains each sample's location history. There is no distinction made in the history file between sample movement inside the laboratory and sample movement to locations outside the laboratory. Any time a sample is moved, the change in the location of the sample must be recorded by laboratory personnel. Any time a sample is moved outside of the USTUR laboratory facility, a chain of custody document (automatically generated when a sample or set of samples is moved in the inventory database) is printed out and shipped with the sample. A copy of the chain of custody is also saved in the electronic and hard case files.

4.2. SAMPLE RADIOCHEMICAL ANALYSIS

This document identifies an isotope of interest as “isotope *X*;” a tracer isotope as “isotope *Y*;” and an element as “element *Z*.” Table 2 shows shorthand notation for isotope names, intended to keep notation brief, yet specific. This notation is widely used in Appendix C.

Table 2. Isotope codes used in variable names.

Isotope	Code	Isotope	Code	Isotope	Code	Isotope	Code	Isotope	Code	Isotope	Code
^{228}Th	T8	^{232}U	U2	^{238}Pu	P8	^{241}Am	A1	^{237}Np	N7	^{244}Cm	C4
^{229}Th	T9	^{234}U	U4	^{239}Pu	P9	^{243}Am	A3				
^{230}Th	T0	^{235}U	U5	$^{239+240}$Pu	P90						
^{232}Th	T2	^{236}U	U6	^{240}Pu	P0						
		^{238}U	U8	^{241}Pu	P1						
				^{242}Pu	P2						

Additionally, standard chemical element symbols are used to refer to mixtures of isotopes of the same chemical element: U, Pu, or Am.

4.2.1. ALIQUOT SIZE

Table 3 shows the default aliquots that are used for different sample types.

Table 3. Aliquot sizes by tissue type.

Tissue type	Aliquot fraction	By mass
Lung	0.05	
Liver	0.1	
Wound	0.05	
Thoracic lymph nodes (LNTH)	0.2	
Skeletal	0.7	or 2 g ash, whichever is less solution
All other tissues	0.7	or 2 g ash, whichever is less solution
Formalin	0.9	or 2 g ash, whichever is less solution
Filters	0.9	or 2 g ash, whichever is less solution

The default aliquot fractions are smallest for the lung, liver, wound, and lymph nodes. These tissues typically contain relatively large activities of plutonium. Thus, the aliquots need to be small enough to not obscure the tracer signal during the analysis.

For all other tissues, the default aliquot size is either 70% or 2 g of ash, whichever results in a smaller aliquot. Generally, soft tissue samples are aliquoted at 70%, and bone samples are subject to the 2 g limitation. A 70% aliquot improves the likelihood of detecting plutonium in samples with low levels of plutonium. If, however, the activity is too low to be detected by the USTUR's in-house α-spectrometry, 30% of the solution will be saved, and an aliquot can be sent to another facility for inductively coupled plasma mass spectrometry (ICP-MS) analysis. Typically, 30% of a sample is at least 35 mL, which is more than the 10-15 mL needed for the highly sensitive ICP-MS analysis. If the intake was an inhalation of soluble material, the lungs will contain only low levels of plutonium and are aliquoted at up to 70% as with any other soft tissue.

4.2.2. ANALYTES OF INTEREST

Tissue sample analysis is a multi-step process. During the analysis, a tissue undergoes five different analytical steps: (i) drying and ashing, (ii) digestion and dissolution, (iii) radiochemical actinide separation, (iv) preparation of an α-counting source (planchet), and (iv) measurement of individual actinides – plutonium (^{238}Pu and $^{239+240}$Pu), americium (^{241}Am), uranium (^{234}U, ^{235}U, and ^{238}U), and thorium (^{232}Th) by α-spectrometry (in-house) or by ICP-MS (external). The ICP-MS is a method of choice for U and Th analysis.

Microwave tissue digestion/dissolution and extraction chromatography separation methods are fully implemented by the USTUR Radiochemistry Laboratory.

4.2.3. ORIGINS OF RADIONUCLIDES PRESENT AT VARIOUS STAGES OF RADIOCHEMICAL ANALYSIS

Radionuclides in processed samples in the radiochemistry lab may come from more than one source. Table 4 shows the origins of radionuclides in the material (matrix) being processed for six different stages of radiochemical analysis. Two of the common tracer solutions contain contaminants. The ^{243}Am solution contains some ^{241}Am contaminant, and the ^{242}Pu tracer contains some ^{241}Pu, which decays into ^{241}Am.

Table 4. Origins of radionuclides in six locations during radiochemical processing.

Location of activity	Intended radionuclide in ^{242}Pu, ^{241}Am, ^{232}U, or ^{229}Th tracer	Contaminant in ^{242}Pu, ^{241}Am, ^{232}U, or ^{229}Th tracer	Ingrowth from radioactive parent in sample	Ingrowth from radioactive parent in tracer	Ingrowth from contaminant in the tracer
Prepared tissue sample(s)			yes		
Dissolved tissue (acid solution)			yes		
Aliquot of solution			yes		
Aliquot of solution to which tracer(s) has(have) been added	yes	yes	yes	yes	yes
Solution containing the element-*Z* fraction (for example, Am fraction, Pu fraction)	yes	yes	yes	yes	yes
Electrodeposited element-*Z* fraction on a planchet (for example, Am planchet, Pu planchet)	yes	yes	yes	yes	yes

In the plutonium-americium measurement and calculation processes, there are 11 distinct activity variables. These activities can be distinguished by values of three parameters: the isotope, the elemental fraction it ends up in, and its origin.

At the USTUR, there are 23 principal measurands that are activities for various radioelements, and each measurand is a function of time. Times of interest are given in Table 5. ^{239}Pu and ^{240}Pu are indistinguishable by α-spectrometry, so the measurand $A_{\mathrm{P90,Pu,prep}}(t)$ is the sum of the activities of these two isotopes of plutonium. When $^{239+240}$Pu is measured by mass spectrometry, measurement results are estimates of the measurands $A_{\mathrm{P9,Pu,prep}}(t)$ and $A_{\mathrm{P0,Pu,prep}}(t)$.

4.2.4. Dates/Times

Table 5 shows the nomenclature and notation for Date/Time variables. Variables are stored as Excel dates[2].

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[2] "Excel stores dates as sequential serial numbers so that they can be used in calculations. January 1, 1900 is serial number 1, and January 1, 2008 is serial number 39448 because it is 39,447 days after January 1, 1900." https://support.office.com/en-us/article/DATE-function-E36C0C8C-4104-49DA-AB83-82328B832349. Date values can include decimal fractions that signify the time of day and can be formatted to produce times in a variety of hour, minute, and second formats. Excel's dates assume that 1900 was a leap year (which it wasn't), so Excel dates are off by 1 day before March 1, 1900 (day 61 in the Excel system) and have incorrect weekday names before March 1, 1900. For USTUR purposes, this error is of no consequence, except for people born between January 1 and March 1, 1900, and even then, it's only 1 day. Negative numbers in an Excel date format simply generate errors. Excel dates are generated in python and validated with Excel. The correct Excel dates are generated by counting from December 30, 1899.

Table 5. Nomenclature for date/time variables.

Specific date/time	Symbol	Notes
Reference date/time for isotope Y tracer solution	$t_{\mathrm{ref},Y}$	may depend on sample batch
Date/time of death or of sample donation (e.g., wound debridement activity)	t_{D}	unique for each case
Date/time elements are separated into fractions	t_{sep}	
Date/time element Z fraction is counted, where $Z \in$ (Th, U, Pu, Am/Cm)	$t_{\mathrm{C},Z}$	may depend on sample batch

4.2.5. ACTIVITIES

Because there are so many isotopes involved in measurements, nomenclature becomes cumbersome. Table 6 lists brief, self-defining names for activity variables. Each isotope of interest is assigned a unique short code name consisting of the first letter of the element name and the last digit of the mass number, as shown in Table 2.

Each isotope of interest originates in one of the following: a prepared tissue sample, a tissue sample in an acid solution, an aliquot of the tissue sample in an acid solution, one of 4 tracer solutions, or the special case of ingrowth of ^{241}Am from ^{241}Pu.

Eventually, an isotope becomes a part of an elemental fraction[3], which is a solution that has been eluted from the various chromatography columns and, finally, is electrodeposited on a planchet. All equations used during sample analysis steps are given in Appendix C of this document.

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[3] Isotopes of interest depend on the intake(s) for each case. Not all cases are analyzed for all elements; most cases are not analyzed for all elements.

Table 6. Naming activity variables for the elements Th, U, Pu, and Am (Cm is not shown). Each activity can be evaluated at three different times, except for ID#s 3 and 19, which are zero before and at t_{sep}. Within a given line, the times are given from earliest to latest from left to right. t_{ref} may be before or after t_D for any given tracer and any given case. The date/time for a box containing a check mark is not used in calculations.

Time-dependent activity measurands identified by isotope, destination, origin	Unique symbol	Reference date/time, t_{ref}	Sample date/time	Solution containing an elemental fraction				Planchet with electrodeposited elemental fraction			
				Th	U	Pu	Am	Th	U	Pu	Am
^{228}Th in the thorium fraction from the prepared tissue sample	$A_{T8,Th,prep}(t)$		t_D	t_{sep}				$t_{C,Th}$			
^{228}Th in the thorium fraction from the ^{232}U tracer	$A_{T8,Th,U2tr}(t)$	$t_{ref,U2}$		t_{sep}				$t_{C,Th}$			
^{228}Th in the uranium fraction from the ^{232}U tracer	$A_{T8,U,U2tr}(t \geq t_{sep})$	$t_{ref,U2}$			t_{sep}				$t_{C,U}$		
^{229}Th in the thorium fraction from the ^{229}Th tracer	$A_{T9,Th,T9tr}(t)$	$t_{ref,T9}$		t_{sep}				$t_{C,Th}$			
^{230}Th in the thorium fraction from the prepared tissue sample	$A_{T0,Th,prep}(t)$		t_D	t_{sep}				$t_{C,Th}$			
^{232}Th in the thorium fraction from the prepared tissue sample	$A_{T2,Th,prep}(t)$		t_D	t_{sep}				$t_{C,Th}$			
^{232}U in the uranium fraction from the ^{232}U tracer	$A_{U2,U,U2tr}(t)$	$t_{ref,U2}$			t_{sep}				$t_{C,U}$		
^{234}U in the uranium fraction from the prepared tissue sample	$A_{U4,U,prep}(t)$		t_D		t_{sep}				$t_{C,U}$		
^{235}U in the uranium fraction from the prepared tissue sample	$A_{U5,U,prep}(t)$		t_D		t_{sep}				$t_{C,U}$		
^{238}U in the uranium fraction from the prepared tissue sample	$A_{U8,U,prep}(t)$		t_D		t_{sep}				$t_{C,U}$		
^{238}Pu in the plutonium fraction from the prepared tissue sample	$A_{P8,Pu,prep}(t)$		t_D			t_{sep}				$t_{C,Pu}$	
^{239}Pu in the plutonium fraction from the prepared tissue sample	$A_{P9,Pu,prep}(t)$		t_D			t_{sep}				$t_{C,Pu}$	
$^{239+240}$Pu in the plutonium fraction from the prepared tissue sample	$A_{P90,Pu,prep}(t)$		t_D			t_{sep}				$t_{C,Pu}$	
^{240}Pu in the plutonium fraction from the prepared tissue sample	$A_{P0,Pu,prep}(t)$		t_D			t_{sep}				$t_{C,Pu}$	
^{241}Pu in the plutonium fraction from the prepared tissue sample	$A_{P1,Pu,prep}(t)$		t_D			t_{sep}				$t_{C,Pu}$	
^{241}Pu in the plutonium fraction from ^{241}Pu in the ^{242}Pu tracer	$A_{P1,Pu,P1inP2tr}(t)$	$t_{ref,P2}$				t_{sep}				$t_{C,Pu}$	
^{242}Pu in the plutonium fraction from the ^{242}Pu tracer	$A_{P2,Pu,P2tr}(t)$	$t_{ref,P2}$				t_{sep}				$t_{C,Pu}$	
^{241}Am in the americium fraction from the prepared tissue sample	$A_{A1,Am,prep}(t)$		t_D				t_{sep}				$t_{C,Am}$
^{241}Am in the plutonium fraction from ^{241}Pu in the prepared tissue sample	$A_{A1,Pu,P1inprep}(t \geq t_{sep})$		t_D			t_{sep}				$t_{C,Pu}$	
^{241}Am in the americium fraction from ^{241}Am in the ^{243}Am tracer	$A_{A1,Am,A1inA3tr}(t)$	$t_{ref,A3}$					t_{sep}				$t_{C,Am}$
^{241}Am in the plutonium fraction from ^{241}Pu in the ^{242}Pu tracer	$A_{A1,Pu,P1inP2tr}(t \geq t_{sep})$	$t_{ref,P2}$				t_{sep}				$t_{C,Pu}$	
^{241}Am in the americium fraction from ^{241}Pu in the ^{242}Pu tracer	$A_{A1,Am,P1inP2tr}(t)$	$t_{ref,P2}$					t_{sep}				$t_{C,Am}$
^{243}Am in the americium fraction from the ^{243}Am tracer	$A_{A3,Am,A3tr}(t)$	$t_{ref,A3}$					t_{sep}				$t_{C,Am}$

Other activities not provided in Table 6:

- The gross "apparent activity of ^{238}Pu in the ^{238}Pu+^{241}Am ROI," $A^*_{\mathrm{P8+A1\text{-}ROI,Pu,3sources}}$, as of count date $t_{\mathrm{C,Pu}}$
- The gross "apparent activity of ^{241}Am in the Am-ROI," $A^*_{\mathrm{Am\text{-}ROI,Am,3sources}}$, as of count date $t_{\mathrm{C,Am}}$
- The "activity of isotope X or Y on the Z-fraction planchet," $A_{X,Z,\mathrm{aliq}}$ or $A_{Y,Z,\mathrm{aliq}}$ as of count date $t_{\mathrm{C},Z}$

4.2.6. ACTIVITY CONCENTRATIONS

There are two broad categories of *activity concentration* variables (Bq kg^{-1}).

1. The first category of activity concentration is the concentration of a radionuclide X in a tracer solution or diluted tracer solution of radionuclide Y, at reference time $t_{\mathrm{ref},X}$, which requires subscripts for unique identification: the isotope, its origin, and its reference date: $C_{X,Y\mathrm{tr}}(t_{\mathrm{ref},X})$
2. The second category of activity concentration is the concentration of radionuclide X in wet samples $C_{X,\mathrm{wet}}(t)$ and ash samples $C_{X,\mathrm{ash}}(t)$.

4.2.7. OTHER QUANTITIES

Variable names other than those in Table 6 are given in Appendix B, Table 20.

4.2.8. SEQUENCE OF RADIOCHEMISTRY PROCESSING EVENTS

Figure 2 shows variables of concern over time in USTUR radiochemistry processes for samples from cases who had intakes of Pu and Am. Here, C denotes concentration (Bq/kg) and A denotes activity (Bq); all notation is identified in the Data Dictionary (Appendix B).

Known volumes of one or more tracer solutions for radioelements of interest (Th, U, Pu, and Am) are added to aliquots of acid solutions following drying, ashing, digesting, and dissolving of samples. The tracer accounts only for losses in separation, and electroplating (electrodeposition), but not for other potential losses in processes such as ashing and digesting.

The ^{242}Pu tracer is contaminated with a small amount of ^{241}Pu, which decays into ^{241}Am. The ^{243}Am tracer is contaminated with ^{241}Am. The ^{232}U tracer is significantly contaminated with ^{228}Th – one of its progeny), requiring separate analyses for uranium and thorium.

After the tracers are added, aliquots are loaded on chromatography columns for separation into fractions by element. Separation occurs at date/time t_{sep}. Soon after separation, each fraction containing a single element is electroplated onto a separate planchet. If only Pu and Am are of interest, for example, then the end result of the radiochemical processing is two planchets, one containing isotopes of Pu (238, 239, 240, 241, and tracer 242) and one containing isotopes of Am (241 and tracer 243). However, as time passes, ^{241}Am grows on the Pu planchet from decay of ^{241}Pu on the planchet after separation and before counting.

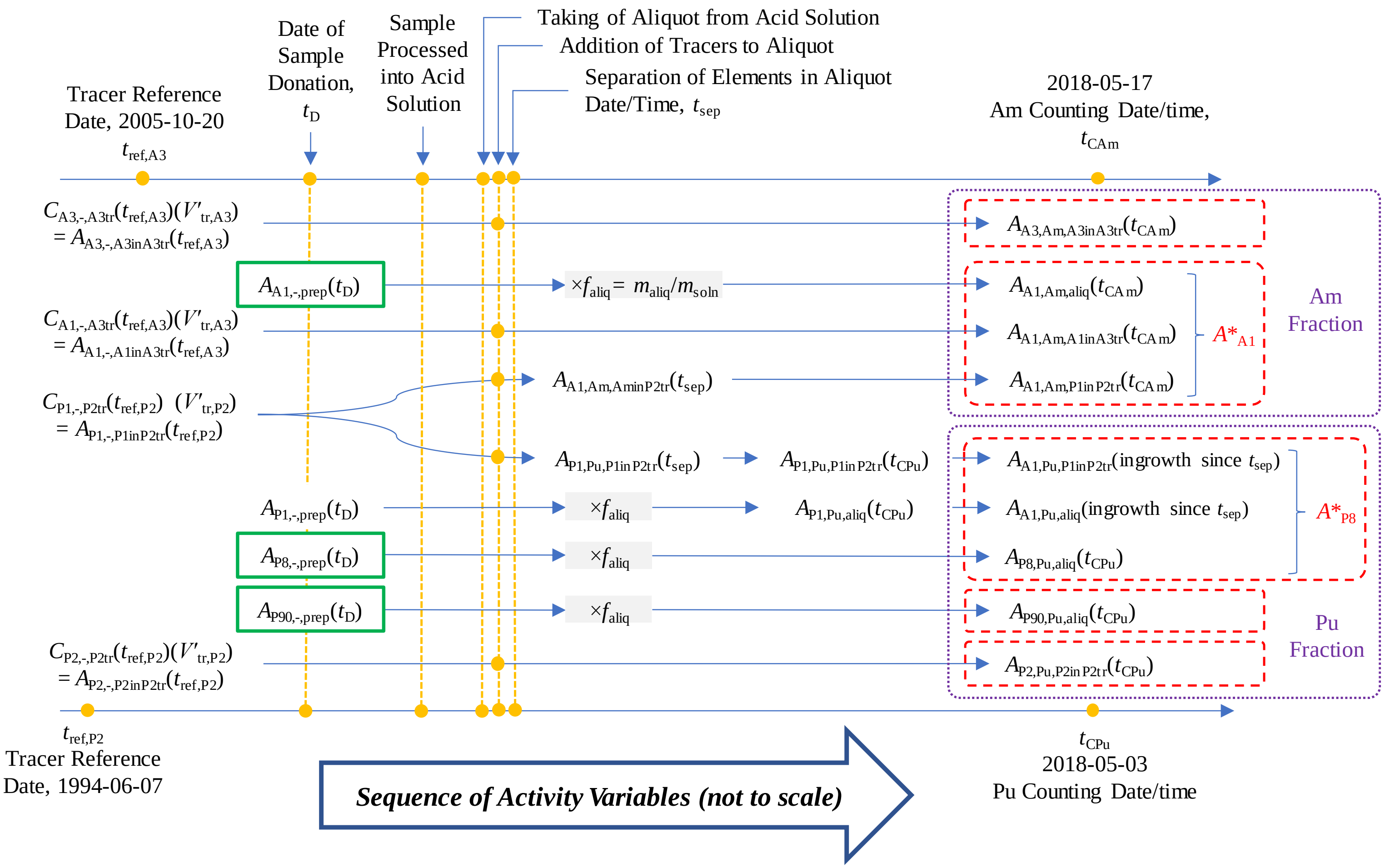

Figure 2. Sequence of processes over time in USTUR radiochemistry processing for Pu+Am cases. Beginning with the donation of Registrant samples through processing, including tracer issues and ^{241}Am ingrowth issues. Activity notation is $A_{\text{isotope,destination,origin}}(t)$. Green boxes are the desired measurands. Dashed red boxes are measurement results in a region of interest (ROI). Dotted purple boxes indicate element fractions (element planchets). A* indicates an "apparent activity" inferred from counts in an ROI in α-spectrometry. Corrections to A* must be made to infer values of desired measurands. Date/Time values are from a typical example of samples and tracers. Corrections for ingrowth and decay are applied at every stage. It is assumed in this document that no activity is lost, and no contamination is added during chemical processing of "prepared samples" before tracers are added.

4.3. BIOKINETIC MODELING AND DOSE ASSESSMENT

The results of radiochemical analysis of tissues from USTUR donors are used to test, validate, and improve the currently recommended biokinetic models. These are mathematical models which describe the intake, translocation and retention of a radionuclide in various organs or tissues of the body, as well as its excretion from the body by various pathways. The quantities needed for biokinetic modeling are activities of whole organs represented in the model as separate compartments or sets of compartments; e.g. lung, liver, skeleton, kidney etc. The radionuclide of concern and route of intake for a particular case determine the set of organ activities that can be used in modeling. Activities in the liver and skeleton are required for all actinides and all routes of intake. For uranium exposure, activity in the kidneys may also be used. If an individual's internal contamination resulted from inhalation intake(s) only, knowledge of the lung activity is critical to determine the case-specific respiratory tract parameters. Similarly, for wound intakes, the estimate of activity retained in the wound site is necessary to validate the model prediction.

Biokinetic modeling of individual cases at the USTUR is currently performed using internal dosimetry software IMBA Professional Plus® and Taurus. To estimate the intake using these software packages, the organ activities at time of death are used as bioassay quantities in addition to common bioassay data such as urinary excretion rates, lung count results, etc. The 'best' estimate of intake is obtained by simultaneously fitting all available data. Accounting for uncertainties in bioassay quantities is very important to obtain a reasonably accurate estimate of intake. IDEAS guidelines provide the reference variabilities (scattering factors) on bioassay monitoring data (Table 7).

Table 7. Typical values for the scattering factor SF for various types of in-vivo and in-vitro measurements (Castellani et al. 2013)

Quantity	Scattering factor (range)
In-vivo measurements[a]	
Low photon energy, E < 20 keV	2.3
Intermediate photon energy, 20 keV < E < 100 keV	1.4
High photon energy, E > 100 keV	1.2
In-vitro bioassay[b]	
True 24-hr urine	1.1
Simulated 24-hr urine, volume or specific gravity normalized	1.6 (1.3 – 1.8)
Spot urine sample	2.0
Fecal 24-hr sample	3 (2 – 4)
Fecal 72-hr sample	2 (1.5 – 2.2)

[a] Total type A (due to counting statistics) and B (all other) lognormal uncertainty
[b] Type B lognormal uncertainty

Since the software gives more weight to quantities with lower uncertainties, it is important to maintain low measurement uncertainties on organ activities. The lowest uncertainty recommended by IDEAS guidelines for in-vivo measurements of actinides in specific organs (lungs, liver, skeleton, or wound) is 40%. Therefore, in IMBA calculations, the uncertainties in organ activities estimated from tissue radiochemical analysis results should be lower than 40%. An uncertainty of 10% is commonly used for activities of soft tissue organs such as the lungs and liver while up to 30% is more appropriate for skeleton activity since it is mostly estimated based on a limited number of bone measurements except for whole-body cases. Inter-subject biological variability is one of the major contributors to the uncertainty of organ activities. Another important source of uncertainty is the uncertainty in the weights of the organs since only a part of the organ is radiochemically analyzed (e.g. right half of the liver, three lobes comprising the right lung, etc.). Therefore, the uncertainty in total organ activity is determined not only by uncertainty in the measured actinide concentrations, but, more significantly, by uncertainty in weight. Tissue and fluid loss during the dissection process may significantly contribute to the uncertainties in weights. Hence, it is important to have reasonably low measurement uncertainties on actinide concentrations in tissue samples (no more than 10%).

4.4. AVAILABLE RESOURCES AND RELEVANT TIMELINES

This section discusses the logistics of implementing the objectives. The purpose of this part is to define available resources and relevant timelines.

4.4.1. TISSUE SAMPLE BACKLOG

Generally, to complete the analysis of a whole-body donation, 150 tissue samples are analyzed. Typically, 35 samples are analyzed for a partial-body donation. To avoid accumulation of unanalyzed (intact) whole-body cases and to obtain key initial scientific information for biokinetic modeling, approximately 35 selected tissue samples are analyzed from each whole-body donation. This is called a survey analysis.

Based on the donation rate during the past five years, the USTUR expects to receive one whole- and three partial-body donations each year. The USTUR Radiochemistry Laboratory expects a throughput of 400 – 450 samples each year for Pu/Am analyses only. In the case of a U/Th exposure, tissues are acid-digested/dissolved in-house and sent to an external laboratory for U/Th ICP-MS measurements.

4.4.2. TISSUE ANALYSIS PRIORITY

Analysis priority

- Complete survey analyses of new whole-body donations as received
- Complete new partial-body donations as received
- Complete previous partial-body donations
- Complete previous whole-body donations.

4.4.3. EXPEDITIOUS ANALYSIS OF TISSUES FROM NEW DONATIONS

All tissues from new partial-body donations and selected tissues from whole-body donations (survey analysis) are analyzed in the year they are received. This will require analyses of an estimated 150 tissue samples from four expected donations. Given that the in-house radiochemistry laboratory has a throughput of 400 – 450 samples per year, 250 – 300 analyses will remain available for the analysis of previously donated tissues.

5. LABORATORY PERFORMANCE INDICATORS

In this document, radiometry means measuring the size of electrical impulses from α-particle detectors as they arrive over a fixed period of time and accumulating counts of those impulses in energy-specific channels. Energies of interest cover the range of α-particle energies from uranium and transuranium elements.

5.1. DETECT/NONDETECT DECISIONS ARE GENERALLY IRRELEVANT FOR AUTOPSY SAMPLES

For Registrants who meet the 2-nCi acceptance criterion, the presence of the primary analytes, for example, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am, is generally certain. For 2-nCi criterion Registrants, decisions about whether activity is present are not, in general, made on the basis of radiochemical measurement results of autopsy samples.

The exception to this is the Registrants with no reported intake. The highest activities are typically present in the lung (for oxide inhalation intakes), the liver (for nitrate inhalation or wound intakes), or the wound site. For such individuals, activities in those organs (or their net count rates) can be compared to a critical level, and, if none exceeds that criterion, there is little to be learned from α-spectrometric measurements of other samples.

When considered in an ensemble of data, it is important to record and report each of the radiochemical analysis results in a measurement result and its combined standard uncertainty, but there is no need to do a comparison with statistical criteria like a critical level. Measurement results that are below the critical level are still usable in dose reconstruction and model development and validation; they are simply more uncertain than measurement results that are at or above the threshold.

5.2. CRITICAL VALUE

Some radiochemical measurements need “an appropriate detection threshold,” which in MARLAP is called a critical value. The *critical value of a variable* is the smallest value of that variable that “can be reliably distinguished from zero” (MARLAP p. 20.1). When it is not known whether an analyte is present, the critical value represents the demarcation line between the decision “analyte is probably present” and “analyte is probably not present.” While critical values can be calculated for net counts, net count rate, or even activity, the most useful critical value is that of the net count rate, because if the net count rate does not exceed the critical value, there is no point in calculating the activity.

The critical value in counting experiments received many names, including Currie's (1968) "critical level" L_C; ANSI N-13.30's "decision level" *DL*; ISO's "decision threshold" y^*; and others.

MARLAP Eq. 20.11 offers this classic formula for the critical value of the net counts[4]. This is known in MARLAP as "critical value of the net signal" for isotope *X*, $S_{C,X}$:

$$S_{C,X}(N_B, z_{1\text{-}\alpha}, t_S, t_B) = z_{1\text{-}\alpha}\sqrt{N_B \frac{t_S}{t_B}\left(1+\frac{t_S}{t_B}\right)} = z_{1\text{-}\alpha}\sqrt{R_B t_S\left(1+\frac{t_S}{t_B}\right)}. \qquad (1)$$

where the term inside the radical is the variance of the net counts when no analyte is present. This formula does not perform well at low numbers of counts, giving far too many false positives ((Strom and MacLellan 2001); MARLAP (2004) Chapter 20).

However, the Stapleton Approximation, given in MARLAP as Eq. 20.54, is a much better-performing formula for the critical value of the net counts, S_C:

$$S_{C,X}(N_B, d, t_S, t_B, z_{1\text{-}\alpha}) = d\left(\frac{t_S}{t_B}-1\right) + \frac{z_{1\text{-}\alpha}^2}{4}\left(1+\frac{t_S}{t_B}\right) + z_{1\text{-}\alpha}\sqrt{(N_B+d)\frac{t_S}{t_B}\left(1+\frac{t_S}{t_B}\right)}. \qquad (2)$$

When $\alpha = 0.05$, the value $d = 0.4$ is the best choice (MARLAP p. 20–47). Strom and MacLellan (2001) have shown that the Stapleton Approximation is the best-performing decision rule, especially when background and sample count times are not equal.

Beginning in 2020, the USTUR uses the critical value of the net count rate, termed $S'_{C,X}$, computed by dividing $S_{C,X}$ by the sample count time t_S:

$$S'_{C,X} = \frac{S_{C,X}}{t_S}. \qquad (3)$$

If the net count rate exceeds S'_C, then one concludes that activity has been detected with a 5% or lower false positive probability when Eq. (2) is used for S_C. This means that there is less than a 5% chance that the observed result could be due to random fluctuations in background with no analyte (isotope *X*) present, that is, there is less than a 5% chance of making an incorrect decision that activity has been detected (a "false positive" mistake or "Type I error").

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[4] The concept of "net counts," when count times are not equal, is difficult to grasp. When $t_S \neq t_B$, "net counts" must be computed as the net count rate multiplied by the sample count time: $R_N t_S = (N_S/t_S - N_B/t_B)t_S = N_S - N_B(t_S/t_B)$. Because this calculated quantity will not be an integer unless t_S/t_B, is an integer, it is not "counts" in those cases and, thus, is not intuitive. Instead of "net counts," the USTUR uses the "net count *rate*" in its critical value.

5.3. THE *P*-VALUE OF THE NET COUNT RATE

In the context of low-level radioactivity measurements, a *p*-value is the probability that a measurand of 0 would produce the observed measurement result. This is also known as the “false positive probability.” A measurement result equal to S'_C has $p = 0.05$, meaning that 5% of measurements of a blank will have a net count rate greater than or equal to *DT*.

A *p*-value greater than 0.05 does not mean that no activity was detected in the sample. For USTUR cases, there *is* activity in the sample, so no decision is needed. A *p*-value greater than 0.05 means that the result is highly uncertain but still usable.

To calculate a *p*-value, one first calculates the “*z*-score.” The *z*-score of the net count rate, whether it is positive, zero, or negative, is

$$z = R_N / u(R_N). \tag{4}$$

Under the assumption of a normal distribution, the probability p associated with the *z*-score of the net count rate is

$$p = 1 - P(z) = P(-z), \tag{5}$$

calculated in Microsoft Excel as

$$p = \mathrm{NORM.S.DIST}(-z,\mathrm{TRUE}) = \mathrm{NORM.S.DIST}(-R_N/u(R_N),\mathrm{TRUE}). \tag{6}$$

For $z \geq 7$, Excel returns $p = 1$, that is, if the net count rate is 7 or more times its uncertainty, there is no meaningful chance that it could be a false positive due to random count rate fluctuations.

5.4. LEGITIMATE BUT RARE USES OF THE CRITICAL VALUE AT THE USTUR

The critical value of the net count rate statistic may be needed for deciding whether activity is present for

- characterization of Registrants with no history of intake
- studies of specific tissues such as the brain
- measurements of radionuclides not known a priori to be present
- intercomparisons and DQOs.

5.5. MINIMUM DETECTABLE ACTIVITY

The minimum detectable activity (*MDA*) is the value of a theoretical measurand activity that, if present in a sample, will most likely produce a signal above a critical level. The *MDA* is often misunderstood and misused; see the discussion in MARLAP Section 20.2 (pp. 20-1 through 20-11).

MARLAP offers a variety of *MDA*-like quantities, but defaults to the minimum detectable concentration. “In radiochemistry the minimum detectable concentration

(*MDC*) is usually obtained from the *minimum detectable value of the net instrument signal*, S_{D}, which is the smallest mean value of the net signal at which the probability that the response variable will exceed its critical value is $1-\beta$" (MARLAP page 20-6).

The minimum detectable value of the *net counts* for isotope X is

$$S_{\mathrm{D},X}(S_{\mathrm{C},X}, R_B, t_S, t_B, z_{1-\beta}) = S_{\mathrm{C},X} + \frac{z_{1-\beta}^2}{2} + z_{1-\beta}\sqrt{\frac{z_{1-\beta}^2}{4} + S_{\mathrm{C},X} + R_B t_S\left(1 + \frac{t_S}{t_B}\right)}. \tag{7}$$

Of greater interest at the USTUR is the minimum detectable value of the *net count rate*,

$$S'_{\mathrm{D},X} = \frac{S_{\mathrm{D},X}}{t_S}. \tag{8}$$

The sample-specific *MDA* (Bq) for isotope X is

$$MDA_X = \frac{S'_{\mathrm{D},X}}{\varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}} = \frac{S_{\mathrm{D},X}}{t_S \varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}} = \frac{S_{\mathrm{C},X} + \frac{z_{1-\beta}^2}{2} + z_{1-\beta}\sqrt{\frac{z_{1-\beta}^2}{4} + S_{\mathrm{C},X} + R_B t_S\left(1 + \frac{t_S}{t_B}\right)}}{t_S \varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}}. \tag{9}$$

In Eq. (9), the ROI Intensity, $f_{X,\mathrm{ROI}}$, and the radiochemical recovery yield y_{RR} are both sample-specific, so an *MDA* calculated using this formula is sample-specific. Similarly, if the radiochemical recovery yield, y_{RR}, is included in the denominator, the *MDA* is sample-specific. MARLAP (pp. 20-8 and 20-9) explains that both generic and sample-specific *MDA*s are useful, and that *MDA* is a variable quantity with a well-defined uncertainty that can be determined by a Type A (statistical) uncertainty analysis. "MARLAP neither encourages nor discourages the reporting of sample-specific *MDC*s [minimum detectable concentrations] with measurement results" (MARLAP p. 20-12). A sample-specific *MDA* that is significantly higher than the USTUR's generic *MDA* for isotope X is a cause for an investigation.

A generic "worst case" value can be used to calculate a generic *MDA*. If $f_{X,\mathrm{ROI}}$ is set to the branching fraction only, ignoring the variable fraction of α particles that are expected to produce counts in the ROI, one will underestimate the *MDA*.

Table 8. Critical values and *MDA* values as a function of the number of background counts. Calculations assume $d = 0.4$; $t_B = 300{,}000$ s; $t_S = 150{,}000$ s; $\varepsilon = 0.25$; $f_{X,ROI} = 1$; $\alpha = \beta = 0.05$; and $y_{RR} = 0.9$

N_B	S_C	S'_C (s^{-1})	S_D	S'_D (s^{-1})	*MDA* (µBq)	*MDA* (fCi)
0	1.72	1.14E-05	5.61	3.74E-05	166	4.49
1	2.50	1.67E-05	7.11	4.74E-05	211	5.70
2	3.02	2.01E-05	8.12	5.42E-05	241	6.51
3	3.44	2.29E-05	8.94	5.96E-05	265	7.16
4	3.80	2.54E-05	9.65	6.44E-05	286	7.73
5	4.12	2.75E-05	10.3	6.86E-05	305	8.24
6	4.42	2.95E-05	10.9	7.24E-05	322	8.70
7	4.69	3.13E-05	11.4	7.60E-05	338	9.13
8	4.94	3.30E-05	11.9	7.94E-05	353	9.53
9	5.18	3.45E-05	12.4	8.25E-05	367	9.91
10	5.41	3.61E-05	12.8	8.55E-05	380	10.3
11	5.62	3.75E-05	13.3	8.83E-05	393	10.6
12	5.83	3.89E-05	13.7	9.11E-05	405	10.9
13	6.03	4.02E-05	14.1	9.37E-05	416	11.3
14	6.22	4.15E-05	14.4	9.62E-05	428	11.6
15	6.40	4.27E-05	14.8	9.87E-05	439	11.9
16	6.58	4.39E-05	15.2	1.01E-04	449	12.1
17	6.76	4.50E-05	15.5	1.03E-04	459	12.4
18	6.92	4.62E-05	15.8	1.06E-04	469	12.7
19	7.09	4.73E-05	16.2	1.08E-04	479	12.9
20	7.25	4.83E-05	16.5	1.10E-04	488	13.2
21	7.40	4.94E-05	16.8	1.12E-04	497	13.4
22	7.56	5.04E-05	17.1	1.14E-04	506	13.7
23	7.71	5.14E-05	17.4	1.16E-04	515	13.9
24	7.85	5.23E-05	17.7	1.18E-04	524	14.2
25	7.99	5.33E-05	18.0	1.20E-04	532	14.4
26	8.13	5.42E-05	18.2	1.22E-04	540	14.6
27	8.27	5.51E-05	18.5	1.23E-04	548	14.8
28	8.41	5.60E-05	18.8	1.25E-04	556	15.0
29	8.54	5.69E-05	19.0	1.27E-04	564	15.3
30	8.67	5.78E-05	19.3	0.000129	572	15.5

5.5.1. USES OF THE *MDA* AT THE USTUR

The *MDA* statistic can be used to

- characterize the detection capability of a system comprising both radiochemical processing and radioactivity measurements
- determine how long to count a sample to achieve a chosen degree of precision in measurement results

If the *MDA* is too high, that is, the measurements are not sensitive enough, potential Registrants may not provide usable data if their intake was so low that the range of measurands, that is, expected quantities in tissues, is less than the *MDA*. Larger samples or longer counting times may be needed if the *MDA* is too high.

5.5.2. SOME PRACTICAL CONSIDERATIONS

In recent years, the USTUR has counted routine samples for 150,000 s and backgrounds/blanks for 300,000 s. The sample counting time is 6 h 20 min less than 2 days, permitting an orderly changing of samples and vacuum pump-down time to be completed every two days.

For a 150,000 s (2500 minutes) sample count time, the addition of a nominal 2 dpm of tracer solution will yield about (2 dis min^{-1})(2500 min)(0.25) = 1250 counts, or fewer if the ROI fraction $f_{X,\mathrm{ROI}}$ is significantly less than 1. This leads to $u_\mathrm{R} \approx 3\%$ in y_RR.

5.6. MINIMUM QUANTIFIABLE ACTIVITY

The minimum quantifiable activity (*MQA*) is the value of a theoretical measurand activity that can be measured with a specified statistical precision (if the acceptable u_R is chosen to be 0.1, this is denoted *MQA*(0.1)). While the *MQA* at a precision of $u_\mathrm{R}(A^*) = 10\%$ would be desirable, it is not always attainable in practice. For illustration, Table 9 uses ±10%, t_B = 300,000 s and t_S = 150,000 s (count times that are routinely used at the USTUR). Each row in the tables is generated by choosing a value of N_B and calculating $u_\mathrm{R}(A^*)$ for increasing values of N_S to find the *MQA*(0.1) values. Table 9 used the $N + 1$ formulas in Eqs. (39) and (41) with and without the uncertainty in y_RR.

Table 9. *MQA*(0.1) values calculated using Eqs. (39) and (41) with $\varepsilon = 0.25$, $u_R(\varepsilon) = 0.01$, $f_{X,ROI} = 1$, $u_R(f_{X,ROI}) = 0.01$, and $y_{RR} = 0.9$, $u_R(y_{RR}) = 0.0316$.

				MQA(0.1) using ε, $f_{X,ROI}$ and y_{RR}	
N_B	N_S	$N_B + 1$	$N_S + 1$	(mBq)	(fCi)
0	114	1	115	3.39	91.7
1	116	2	117	3.44	92.9
2	117	3	118	3.45	93.3
3	118	4	119	3.47	93.7
4	119	5	120	3.48	94.1
5	120	6	121	3.50	94.5
6	122	7	123	3.54	95.7
7	123	8	124	3.56	96.1
8	124	9	125	3.57	96.5
9	125	10	126	3.59	96.9
10	126	11	127	3.60	97.3
11	128	12	129	3.64	98.5
12	129	13	130	3.66	98.9
13	130	14	131	3.67	99.3
14	131	15	132	3.69	99.7
15	132	16	133	3.70	100.1
16	133	17	134	3.72	100.5
17	134	18	135	3.73	100.9
18	135	19	136	3.75	101.3
19	137	20	138	3.79	102.5
20	138	21	139	3.81	102.9
21	139	22	140	3.82	103.3
22	140	23	141	3.84	103.7
23	141	24	142	3.85	104.1
24	142	25	143	3.87	104.5
25	143	26	144	3.88	104.9
26	144	27	145	3.90	105.3
27	145	28	146	3.91	105.7
28	146	29	147	3.93	106.1
29	147	30	148	3.94	106.5
30	148	31	149	3.96	106.9

It can be seen in Table 9 that for $N_B \leq 15$ counts, *MQA*(0.1) ≤ 3.7 mBq (≤ 100 fCi), regardless of which formula is used. Figure 3 shows graphs of *MDA* and *MQA* for u_R values between 0.05 and 0.50 as a function of N_B.

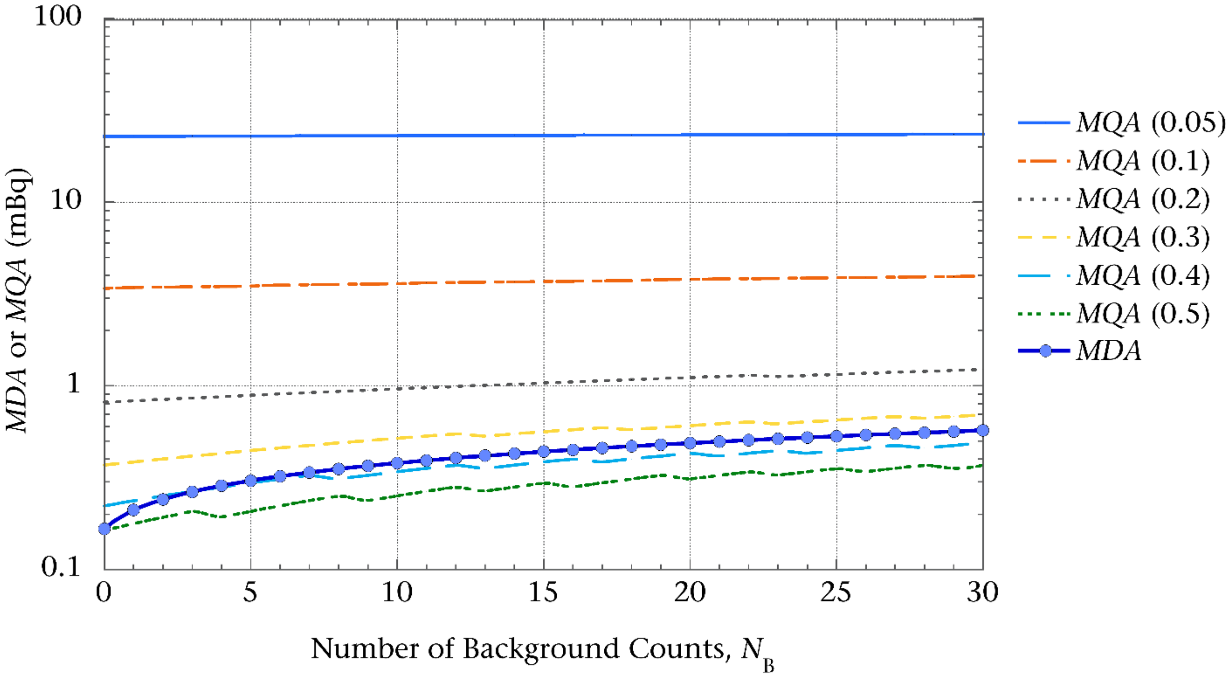

Figure 3. *MQA* and *MDA* as a function of N_B. The relative uncertainty for each *MQA* is given in parentheses in the legend.

5.7. ACTIVITY CONCENTRATIONS

5.7.1. ACTIVITY CONCENTRATION OF ISOTOPE X IN WET TISSUE AND ITS UNCERTAINTY

The activity in the prepared sample is assumed to be the same as the activity in the dissolved solution, that is, it is assumed that there are no losses in drying, ashing, and dissolving/digesting.

The activity concentration of isotope X in wet tissue, $C_{X,\text{wet}}(t_\text{D})$ (Bq kg^{-1} wet tissue), is the activity of isotope X in the dissolved sample back-corrected to the collection date, $A_{X,\text{-},\text{prep}}(t_\text{D})$ (Bq), divided by the “prepared mass of the sample,” m_prep (g):

$$C_{X,\text{wet}}(t_D) = \frac{A_{X,\text{-},\text{prep}}(t_D)}{m_\text{prep}}\left(\frac{1000\text{ g}}{1\text{ kg}}\right) = \frac{A_{X,\text{-},\text{soln}}(t_\text{C})e^{+\lambda_X(t_\text{C}-t_\text{D})}}{m_\text{prep}}\left(\frac{1000\text{ g}}{1\text{ kg}}\right) \quad (10)$$

The uncertainty in $C_{X,\text{wet}}(t_\text{D})$ is

$$u\left(C_{X,\text{wet}}(t_D)\right) = \left(\frac{1000\text{ g}}{1\text{ kg}}\right)\frac{1}{m_\text{prep}}\sqrt{\left(u^2\left(A_{X,\text{-},\text{prep}}(t_D)\right) + \frac{A^2_{X,\text{-},\text{prep}}(t_D)u^2\left(m_\text{prep}\right)}{m^2_\text{prep}}\right)} \quad (11)$$

5.7.2. Activity Concentration of Isotope *X* in Tissue Ash and Its Uncertainty

Currently, activity concentration in an ashed tissue is only calculated for bone samples. The activity concentration of isotope *X* in bone ash, $C_{X,\text{ash}}\ (t_\text{D})$ (Bq kg^{-1} ash), is the activity of isotope *X* in the dissolved prepared sample back-corrected to the collection date, $A_{X,\text{-},\text{prep}}(t_\text{D})$ (Bq), divided by the mass of the ashed bone sample, m_ash (g):

$$C_{X,\text{ash}}(t_D) = \frac{A_{X,\text{-},\text{prep}}(t_D)}{m_\text{ash}}\left(\frac{1000\text{ g}}{1\text{ kg}}\right) \tag{12}$$

The uncertainty in $C_{X,\text{ash}}(t_\text{D})$ is the uncertainty in $A_{X,\text{-},\text{prep}}(t_\text{D})$ divided by the mass of the ashed bone sample m_ash:

$$u\left(C_{X,\text{ash}}(t_D)\right) = \left(\frac{1000\text{ g}}{1\text{ kg}}\right)\frac{1}{m_\text{ash}}\sqrt{\left(u^2\left(A_{X,-,prep}(t_D)\right) + \frac{A^2_{X,-,prep}(t_D)u^2(m_\text{ash})}{m^2_\text{ash}}\right)} \tag{13}$$

5.8. Bias in the Measurement of the Activity of a Tracer Isotope

The bias in the measurement of the activity of (a tracer) isotope *Y*, b_Y, is the difference between the **known** activity of isotope *Y* placed in the aliquot referenced to count date for fraction *Z*, $A_{Y,\text{known-aliq},Y\text{tr}}(t_{\text{C},Z})$ (Bq) and the **measured** activity of isotope *Y* placed in aliquot referenced to count date for fraction *Z*, $A_{Y,\text{measured-aliq},Y\text{tr}}(t_{\text{C},Z})$ (Bq) divided by the **known** activity of isotope *Y* placed in sample referenced to count date:

$$b_Y = \frac{A_{Y,\text{known-aliq},Y\text{tr}}(t_{\text{C},Z}) - A_{Y,\text{measured-aliq},Y\text{tr}}(t_{\text{C},Z})}{A_{Y,\text{known-aliq},Y\text{tr}}(t_{\text{C},Z})} \tag{14}$$

5.9. Isotope Ratio

The ratio of the activity of isotope *X* to the activity of isotope *Y*, $r_{AX/AY}$, is the activity of isotope *X* divided by the activity of isotope *Y* at some time *t*:

$$r_{AX/AY} = \frac{A_{X,\text{-},\text{-}}(t)}{A_{Y,\text{-},\text{-}}(t)}. \tag{15}$$

The uncertainty in $r_{\text{AX/AY}}(t)$ is

$$u\left(r_{AX/AY}(t)\right) = \frac{1}{A_{Y,-,-}(t)}\sqrt{\left(u^2\left(A_{X,-,-}(t)\right) + \frac{A^2_{X,-,-}(t)u^2\left(A_{Y,-,-}(t)\right)}{A^2_{Y,-,-}(t)}\right)} \tag{16}$$

6. MEASUREMENT QUALITY OBJECTIVES

Measurement results and the combined standard uncertainty of the measurement results are the principal products of USTUR's radiochemistry laboratory. A discussion of statistical criteria is provided below to meet USTUR's DQOs.

This chapter describes the quality criteria for the data produced by the USTUR radiochemistry laboratory and the laboratory's MQOs, such as precision, accuracy (bias), representativeness, comparability, completeness, and sensitivity. Acceptance criteria and the control and tolerance limits of each MQO are covered in this section.

6.1. MEASURANDS AT USTUR

The measurand is "the quantity intended to be measured." From the standpoint of radiochemical measurements of tissue samples at the USTUR, one important measurand is the mass of a particular sample. A more difficult measurand is the true but unknown activity in a tissue, organ, or other sample. The distinction between measurands and measurement results is key to modern uncertainty analysis, as explained in the GUM (International Organization for Standardization (ISO) 2008) and the VIM (Joint Committee for Guides in Metrology (JCGM) 2012).

The goal of radiochemical measurements at the USTUR is to make probabilistic statements of the likely values the measurand could have given the measurement results and their combined standard uncertainty.

6.2. MEASUREMENT RESULTS THAT ARE RECORDED AT USTUR

For its measurements, the USTUR records

- the numbers of counts for sample, background, and tracer in the ROI
- the counting times, real and live, and dates of measurements
- mass of total wet samples, prepared samples, dry samples, ash, and aliquots
- QC and QA sample measurement results; and
- all additional data and metadata that the counting laboratory can supply.

These records enable future researchers to fully understand and use measurement results.

Prior to the implementation of this document, all calculations at the USTUR were performed using an Excel spreadsheet and a set of equations derived from ORTEC 2013, ANSI 2011, Gautier 1992, and Currie 1968.

6.3. STATISTICAL CRITERIA

Table 10 shows three statistical criteria commonly used in radiochemical measurements, and the names given to them by various authors over time (Currie 1968, Health Physics Society (HPS) 1996, U.S. Environmental Protection Agency (EPA) et al. 2004, International Organization for Standardization (ISO) 2010, American National

Standards Institute (ANSI) and Health Physics Society (HPS) 2011, Joint Committee for Guides in Metrology (JCGM) 2012). There is a good discussion of the history of the chaotic and uncoordinated development of these statistics in MARLAP §20.2.6.

Table 10. Three statistical criteria and the names used over the years by various authors

Author	Is anything there? Yes, if measurement result ≥ Criterion 1		What's the smallest measurand that will usually give a measurement result ≥ Criterion 1?		What's the smallest measurand that can be measured with fairly small uncertainty ($u_R \le 10\%$)?	
	Criterion 1	Symbol	Criterion 2	Symbol	Criterion 3	Symbol
Currie (1968)	Critical level	L_C	Detection limit	L_D	Determination limit	L_Q
ANSI N13.30-1996	Decision level	DL	Minimum detectable activity	MDA	–	–
ISO-11929-2010	Decision threshold	y^*	Detection limit	$y^\#$	–	–
VIM (2012) §4.16, 4.18	Discrimination threshold	-	Detection limit ≡ limit of detection[a]	LOD	–	–
MARLAP (2004)	Critical value of the net counts	S_C	Minimum detectable value of the net counts	S_D	Minimum quantifiable value	x_Q
This document	Critical value of the net count rate	S'_C	minimum Detectable activity	MDA	Minimum quantifiable activity	MQA

[a] According to MARLAP (2004; p. 20--10), in 1980, the term "limit of detection" was defined by some authors in the chemistry field as Criterion 1, rather than Criterion 2

6.4. USTUR'S MEASUREMENT CAPABILITIES

USTUR's ultimate goal is an acceptable relative uncertainty in its measurements.

A relative uncertainty of 10% or less in activity measurement results at specified values of the measurand would be an ideal outcome. However, achieving this level of uncertainty is difficult or impossible in some cases. For the USTUR's radiochemistry laboratory, uncertainty increases with decreasing activity on a planchet, as shown in Figure 4 for the example of $N_B = 15$ counts. Equations for S_C', MDA and MQA are given in Chapter 5.

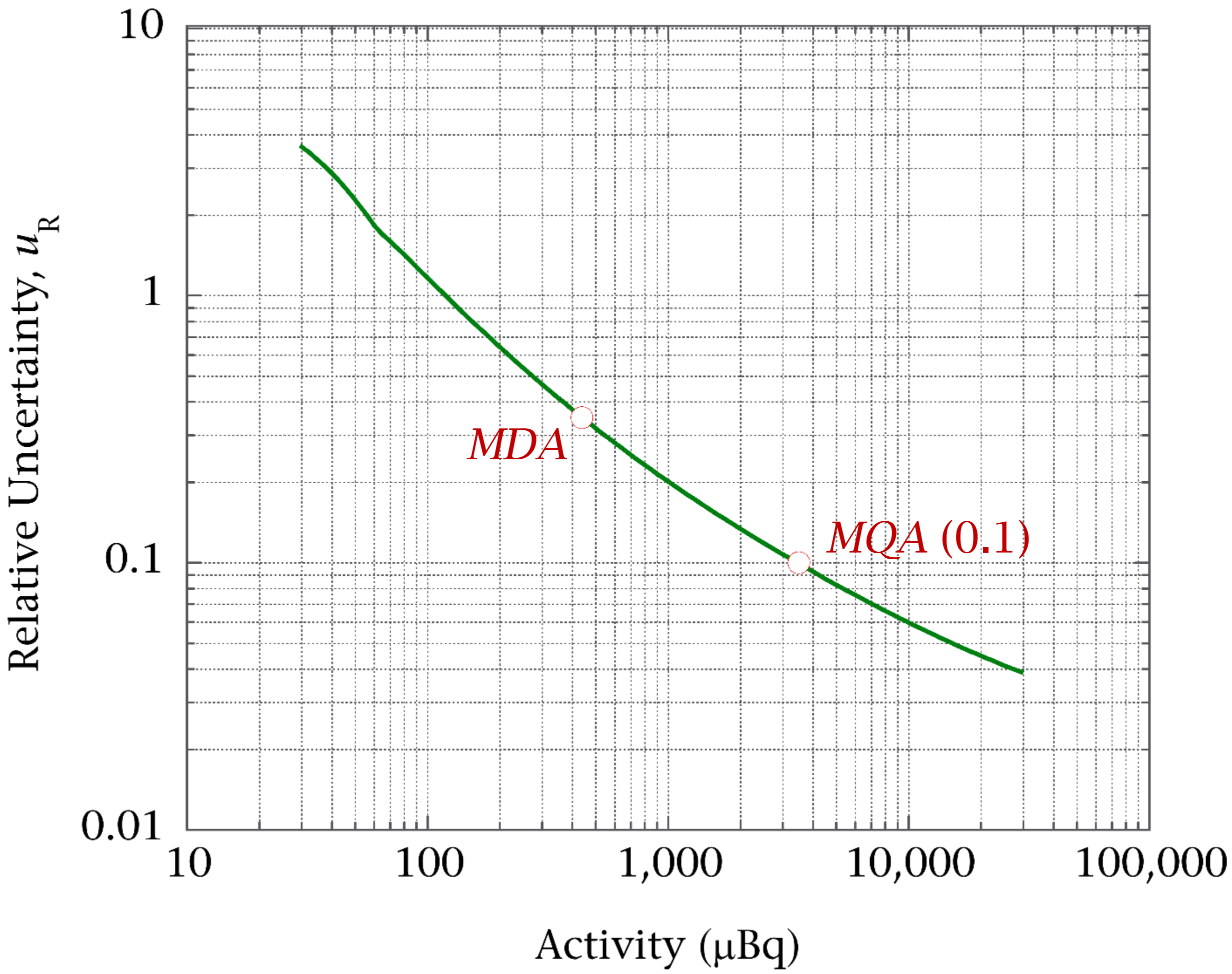

Figure 4. Relative uncertainty, u_R, as a function of activity on a planchet for $N_B = 15$ counts, $t_B = 300{,}000$s, $t_S = 150{,}000$s, $\varepsilon = 0.25$, $f_{X,ROI} = 1$, and $y_{RR} = 0.9$. For these parameters, the *MDA* = 439 μBq, while the MQA at 10% uncertainty is about 3,700 μBq.

In this case, the measurand is the activity *A* of a principal radionuclide such as ^{238}Pu, $^{239+240}$Pu, and/or ^{241}Am. The expected value of the measurand *A* is determined by a forward calculation using IMBA. An intake of 2 nCi (74 Bq) or more[5] is assumed; this is the activity threshold for becoming a USTUR Registrant. From postulated intakes through inhalation and/or wounds, of various chemical forms of the principal radionuclides, we calculate the activity that would remain 50 years after intake in the fraction of an organ that is eventually counted. This is the activity on a planchet, $A_{planchet}$, which is treated in detail in Appendix A.

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[5] Originally, it had not been an intake, but a "body burden" of 4 nCi, which is 10% of the then-MPBB of 40 nCi. Also, the 2 nCi limit never applied to ^{226}Ra – those cases generally had much higher intakes.

The ideal objective then would be to ensure that the predicted activity on a planchet for an intake of ≥74 Bq (2 or more nCi) 50 years earlier can be detected with 10% relative uncertainty under realistic conditions.

The value of A_{planchet} is always less than the activity in the initial tissue or organ sample, A_{init}:

$$A_{\text{planchet}} = A_{\text{init}} \frac{m_{\text{prep}}}{m_{\text{init}}} \frac{m_{\text{aliq}}}{m_{\text{soln}}} y_{\text{RR}} \tag{17}$$

As detailed in Appendix A, typical values of $m_{\text{prep}}/m_{\text{init}}$ range from 1 down to 0.1; $m_{\text{aliq}}/m_{\text{soln}}$ ranges from 1 to 0.2; y_{RR} is typically 0.9 with ranges from 0.5 to 1.1, but the worst (least sensitive) case value (the lower Tolerance Limit for y_{RR}) of 0.5 is used in the following evaluations.

Ideally, the *MQA* should be less than the expected activity on a planchet:

$$MQA(\text{radionuclide}) \leq A_{\text{planchet}}(\text{radionuclide, chemical form, organ, route of intake}). \tag{18}$$

For Pu and Am, inhalation intakes of oxides and nitrates are most common. Many other chemical forms are encountered for other radionuclides, as listed in Table 11. Table 11 and Table 12 show the performance capabilities of the USTUR radiochemical laboratory expressed as fractions or multiples of the *MDA* and the *MQA*, respectively, for samples obtained 50 years after an intake of 74 Bq (2 nCi).

While the vast majority of USTUR Registrants have had intakes of ^{239}Pu or ^{241}Am, the USTUR also routinely analyzes for ^{238}Pu. Measurements of isotopes of Ra, Th, U, Np, and Cm are also made, but far less frequently.

6.5. TECHNOLOGY SHORTFALL

There is "technology shortfall" (as expressed in the DOE Technical Standard *Internal Dosimetry*) in many cases for this low of an intake. Fortunately, most Registrants have intakes that are sufficiently high so that material can be detected in many tissues, with some notable exceptions. For example, 50 years after an inhalation intake of Pu nitrate, less than 1/1000th of the initial activity in the lung remains (see Figure 5), so detecting Pu in lung tissue under these circumstances is very difficult for a 74 Bq intake.

Table 11. Predicted fraction of 1 *MDA* that will appear on a planchet after radiochemical preparations for 7 radionuclides in 4 tissues/organs for commonly encountered chemical forms. These are based on an intake of 74 Bq (2 nCi), which is lower, often far lower, than intakes experienced by most Registrants. For N_B = 15 counts in 300,000s, the worst-case background (the tolerance limit for background), the *MDA* = 0.44 mBq. Entries with a green background are greater than 1 *MDA*; entries with a yellow background are between 0.5 and 1 *MDA*; and entries with a pink background are less than 0.5 *MDA*. Note that if y_{RR} had been chosen as 0.9 instead of 0.5, all of the yellow boxes below would become green, but none of the pink boxes would become yellow.

Radionuclide	Tissue or organ	Systemic uptake	Inhalation								Ingestion
			Soluble		Moderately soluble			Insoluble			
			Hexafluoride	Citrate	Nitrate	Chloride, citrate, nitrate, sulphate, fluoride	Nitrate, oxide, chloride	Dioxide	Octoxide	Type S	
Pu-239	Lungs	--			1.59E-01			3.11E+01			
Pu-239	Liver	9.34E+02			3.41E+01			3.88E+00			
Pu-239	Skeleton	1.73E+01			6.34E-01			6.43E-02			
Pu-239	Other Soft Tissues	6.70E+00			2.42E-01			2.60E-02			
Am-241	Lungs	--			2.06E-01			5.58E+00			
Am-241	Liver	1.10E+02			6.11E+00			3.64E+00			
Am-241	Skeleton	3.33E+01			1.85E+00			5.71E-01			
Am-241	Other Soft Tissues	1.24E+01			6.89E-01			1.97E-01			
U-nat	Lungs	--	0.00E+00						3.93E-03		
U-nat	Liver	1.99E-01	2.79E-02						7.09E-03		
U-nat	Skeleton	2.17E-01	3.03E-02						6.26E-03		
U-nat	Other Soft Tissues	3.97E-01	5.55E-02						1.03E-02		
Th-232	Lungs	--				7.42E-39		5.97E+00			
Th-232	Liver	1.20E+01				7.03E-01		2.56E-01			
Th-232	Skeleton	3.08E+01				1.84E+00		5.47E-01			
Th-232	Other Soft Tissues	9.96E+00				5.98E-01		1.57E-01			
Ra-226	Lungs	--			0.00E+00						--
Ra-226	Liver	5.28E-04			1.26E-04						1.06E-04
Ra-226	Skeleton	1.50E-01			3.59E-02						8.10E-04
Ra-226	Other Soft Tissues	7.42E-04			1.78E-04						1.00E-06
Cm-244	Lungs	--		2.39E-01			2.09E-01			--	
Cm-244	Liver	1.76E+01		7.16E+00			6.15E+00			3.64E+00	
Cm-244	Skeleton	1.44E-01		2.16E+00			1.86E+00			5.71E-01	
Cm-244	Other Soft Tissues	1.34E-02		8.08E-01			6.89E-01			1.97E-01	
Np-237	Lungs	--			2.47E-39			5.97E+00			
Np-237	Liver	1.10E+01			1.75E+00			4.84E-01			
Np-237	Skeleton	8.77E+00			1.42E+00			2.12E-01			
Np-237	Other Soft Tissues	5.01E+00			8.13E-01			8.51E-02			
🟩 $f \geq 1.0$	🟨 $0.5 < f < 1.0$	🟥 $f \leq 0.5$						MDA = 0.000439 Bq		MQA = 0.0037 Bq	

Table 12. Predicted fraction of 1 *MQA* that will appear on a planchet after radiochemical preparations for 7 radionuclides in 4 tissues/organs for commonly encountered chemical forms. These are based on an *intake* of 74 Bq (2 nCi), which is lower, often far lower, than intakes experienced by most Registrants. For N_B = 15 counts in 300,000s, the worst-case background (the Tolerance Limit for background), the *MDA* = 0.44 mBq and the *MQA*(0.1) = 3.7 mBq. Entries with a green background are greater than 1 *MQA*; entries with a yellow background are between 0.5 and 1 *MQA*; and entries with a pink background are less than 0.5 *MQA*. Note that if y_{RR} had been chosen as 0.9 instead of 0.5, all of the yellow boxes below would become green, but none of the pink boxes would become yellow.

Radionuclide	Tissue or organ	Systemic uptake	Inhalation								Ingestion
			Soluble		Moderately soluble			Insoluble			
			Hexafluoride	Citrate	Nitrate	Chloride, citrate, nitrate, sulphate, fluoride	Nitrate, oxide, chloride	Dioxide	Octoxide	Type S	
Pu-239	Lungs				1.89E-02			3.69E+00			
Pu-239	Liver	1.11E+02			4.05E+00			4.60E-01			
Pu-239	Skeleton	2.05E+00			7.52E-02			7.63E-03			
Pu-239	Other Soft Tissues	7.95E-01			2.88E-02			3.09E-03			
Am-241	Lungs				2.45E-02			6.62E-01			
Am-241	Liver	1.31E+01			7.25E-01			4.32E-01			
Am-241	Skeleton	3.95E+00			2.19E-01			6.77E-02			
Am-241	Other Soft Tissues	1.47E+00			8.17E-02			2.34E-02			
U-nat	Lungs								4.67E-04		
U-nat	Liver	2.36E-02	3.31E-03						8.41E-04		
U-nat	Skeleton	2.57E-02	3.60E-03						7.43E-04		
U-nat	Other Soft Tissues	4.71E-02	6.59E-03						1.22E-03		
Th-232	Lungs					8.80E-40		7.09E-01			
Th-232	Liver	1.42E+00				8.34E-02		3.04E-02			
Th-232	Skeleton	3.66E+00				2.18E-01		6.49E-02			
Th-232	Other Soft Tissues	1.18E+00				7.10E-02		1.87E-02			
Ra-226	Lungs										
Ra-226	Liver	6.26E-05			1.50E-05						1.25E-05
Ra-226	Skeleton	1.78E-02			4.26E-03						9.60E-05
Ra-226	Other Soft Tissues	8.80E-05			2.11E-05						1.19E-07
Cm-244	Lungs			2.83E-02			2.48E-02				
Cm-244	Liver	2.09E+00		8.50E-01			7.30E-01			4.32E-01	
Cm-244	Skeleton	1.70E-02		2.57E-01			2.20E-01			6.77E-02	
Cm-244	Other Soft Tissues	1.59E-03		9.59E-02			8.17E-02			2.34E-02	
Np-237	Lungs				2.94E-40			7.09E-01			
Np-237	Liver	1.30E+00			2.08E-01			5.74E-02			
Np-237	Skeleton	1.04E+00			1.68E-01			2.51E-02			
Np-237	Other Soft Tissues	5.94E-01			9.64E-02			1.01E-02			
$f \geq 1.0$	$0.5 < f < 1.0$	$f \leq 0.5$									

The three most highly irradiated tissues or organs for transuranics are the lungs, liver, and skeleton. ICRP Occupational Intakes of Radionuclides (OIR) Part 4 (ICRP 2019) predictions of organ or tissue contents, expressed as Bq in the organ per Bq of intake, are shown in Figure 5 and Figure 6 for moderately transportable Pu nitrate and slowly transportable Pu dioxide, respectively. In both cases, the activity in the respiratory tract decreases with time, so a shorter interval between intake and death means relatively more activity in the lungs.

Often, concentrations in other tissues and organs are very similar; these are lumped together as “other soft tissues” for MQO purposes.

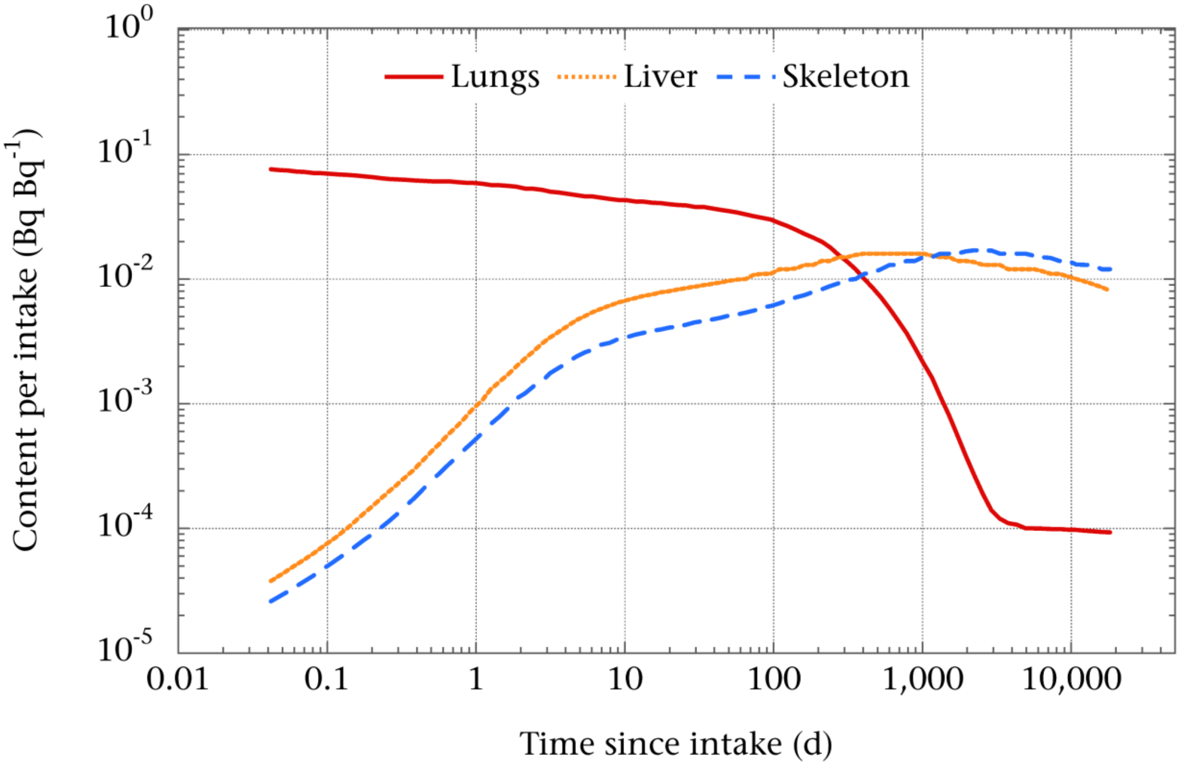

Figure 5. Tissue or organ content over time following a 1 Bq inhalation intake of 5-μm ^{239}Pu nitrate aerosol with f_A = 1E-4. Liver content changes only over a 2-fold range after 100 days, and skeleton content over a 3-fold range after 100 days. In contrast, lung content decreases by a factor of about 250 between 100 days and 4000 days. From ICRP (2019), OIR-4.

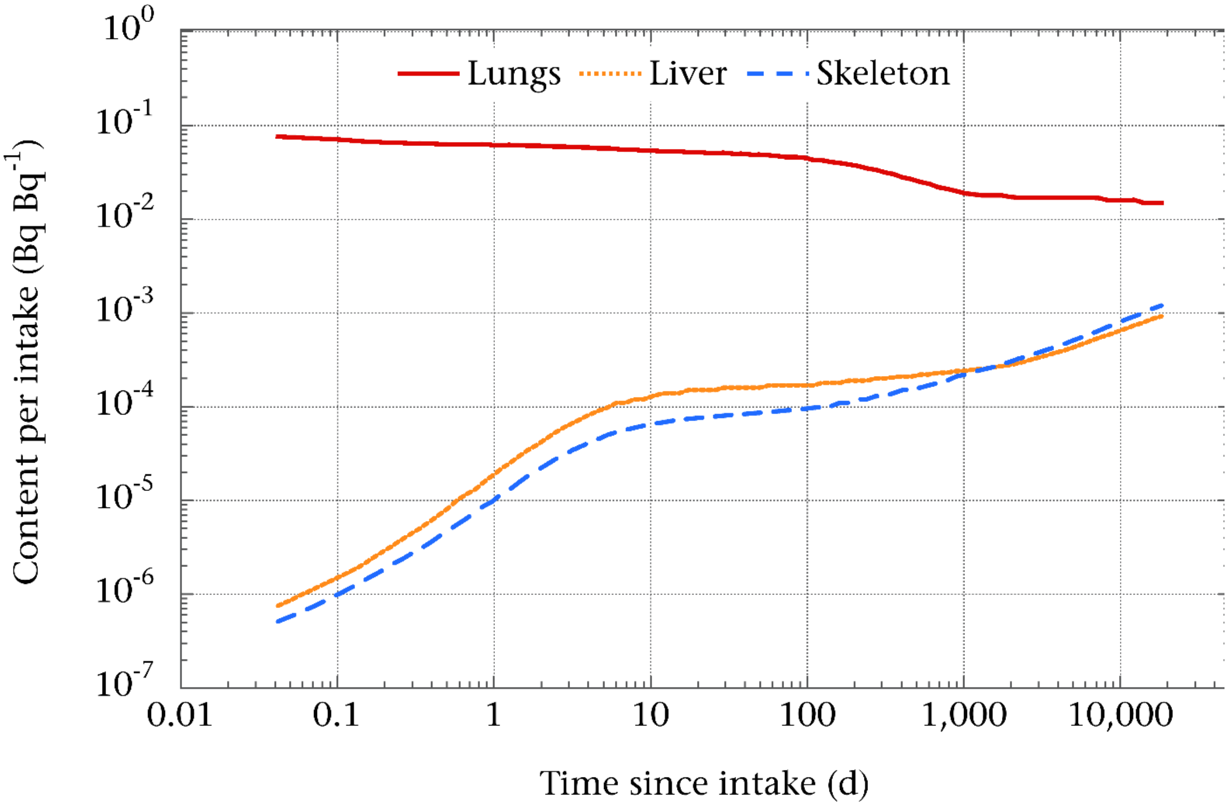

Figure 6. Tissue or organ content over time following a 1 Bq inhalation intake of 5-μm ^{239}Pu dioxide aerosol with f_A = 2E-6. Liver content shows a life-long slow increase by a factor of about 7 after 100 days, and skeleton shows a life-long slow increase by a factor of over 10 after 100 days. In contrast, lung content, which is much greater than that in the liver or skeleton, decreases by a factor of about 3 between 100 days and 4000 days. From ICRP (2019), OIR-4.

6.6. MANAGING TECHNOLOGY SHORTFALL

At least four approaches can be taken to manage technology shortfall.

6.6.1. INCREASING COUNTING TIME

In cases where technology shortfall does occur, selected samples can be counted for up to 600,000s, lowering the *MDA* by a factor of 2.

6.6.2. USING MASS SPECTROMETRY TECHNIQUE

Samples can be sent to an outside laboratory for measurement by ICP-MS.

6.6.3. COMBINING ELUATES FROM SEVERAL COLUMNS

Most of the time, sample sizes cannot be increased, because of the 2-g limit for the chemical separation columns. For instance, we already assume that 2 g of bone ash is analyzed, which is really the maximum that can be separated on a column. However, in rare circumstances, it may be possible to combine the eluates from several columns, running 2 g of ash through each, and combining the eluate from the columns to electroplate.

6.6.4. COMBINING COUNTS FROM SEVERAL SUB-SAMPLES OF A TISSUE OR ORGAN

The USTUR typically analyzes the right lung in 3 different lobes. The counting data collected from these 3 lobes (or from *n* different samples) can be combined to arrive at a pooled result for the samples, as shown in the following thought experiment. For t_B = 300,000 s and t_S = 150,000 s, Table 13 shows how counting results of different portions of the same tissue or organ can be combined to reduce uncertainty. If there are differences in y_{RR} between the samples, these can be accounted for by using weighted averages.

Table 13. Results of combining counts for 3 sub-samples of an organ to reduces the relative uncertainty of the net count rate, $u_R(R_N)$, in this case, by a factor of about 1.67 (≠ 5/3). Data are for ^{241}Am from Case 0340 with roughly 32-44% relative uncertainties.

Tissue sample	Mass (g)	N_B	N_S	Count rate ×10^6 (s^{-1})			$u(R_N)$	$u_R(R_N)$
				R_B	R_S	R_N		
Lung: superior lobe	96.03	5	9	20.0	66.7	46.7	20.5	44.0%
Lung: middle lobe	66.84	5	11	20.0	80.0	60.0	22.6	37.7%
Lung: inferior lobe	104.56	3	12	13.3	86.7	73.3	23.1	31.5%
All three lobes	267.43	13	32	46.7	220.0	173.3	39.2	22.6%
Approximate factor by which uncertainty in $u_R(R_N)$ is reduced:								**1.6703**

6.7. ACCURACY

"The term 'accuracy' describes the closeness of the result of a measurement to the true value of the quantity being measured" (the measurand; MARLAP p. 9–14)." "Since MARLAP uses 'accuracy' only as a qualitative concept, … the agreement between measured results and true values is evaluated quantitatively in terms of the 'precision' and 'bias' of the measurement process. 'Precision' usually is expressed as a standard deviation, which measures the dispersion of results about their mean. 'Bias' is a persistent deviation of results from the true value" (measurand or conventionally true value).

Accuracy of radiochemical analysis of the USTUR laboratory is determined through establishing acceptance criteria for bias and recovery on Laboratory Control Samples (spikes). Various blank samples are used to assess contamination of samples that may cause upward bias of results. Proper calibration of analytical and sampling equipment is determined on a monthly basis.

Additionally, blind testing of reference materials from Site Specific Performance Evaluation Program (SSPEP) is used to assess the accuracy of USTUR measurement techniques.

Acceptance criteria for the quality control sample measurements expressed in terms of bias are specified in Table 15.

6.8. PRECISION

Precision is the measure of agreement among repeated measurements. "'Precision' usually is expressed as a standard deviation, which measures the dispersion of results about their mean" (MARLAP p. 9-–14).

One or more of the following criteria are used to determine the precision of the data:

- analyses of the same sample using the same analytical methods (laboratory or analytical duplicates),
- analysis of samples with the standard method using external laboratory (inter-laboratory precision evaluation using analysis results of Site-Specific Performance Evaluation Program (SSPEP) samples[6]).
- Monitoring proper calibration of analytical and sampling equipment on a monthly basis.

Analysis of a split sample (subsample duplicate) to evaluate precision from sample collection to sample handling, preservation and storage is not done at USTUR because it would require homogenizing a tissue before splitting.

Precision is expressed measured as the relative standard deviation ($\%SD_R$) between two duplicate determinations. Acceptance criteria for duplicate sample measurements are specified in Table 15.

6.9. REPRESENTATIVENESS

Representativeness describes how the collected data accurately represents the whole-body radionuclide content in the USTUR donors. Samples from whole- and partial-body donations are collected during autopsy according to USTUR SOP F402a and F402b, respectively. Selection of samples for biokinetic modeling and assessment of the intake is described section 4.3 of this document. Acceptance criteria for representativeness of sample measurements are specified in Table 15.

6.10. COMPLETENESS

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[6] Radiological and Environmental Sciences Laboratory (RESL), Idaho Falls, ID

Completeness, or the amount of usable data collected compared to the amount of data expected to be obtained, is defined in terms of sample, analytical, and overall completeness.

Sample completeness, or the number of valid samples collected relative to the number of samples planned for collection depends on the type of donation. The complete set of tissues and organs from whole-body donations is collected during the autopsy. These samples are listed in Phase 1 of the Whole Body Specimen Worksheet (F402a). Samples from partial-body donations are collected by a pathologist during the autopsy (Partial Body Specimen Worksheet F402b). This list may be modified based upon factors such as the Registrant's exposure history. The USTUR has a single pathologist who performs all whole-body and local partial-body autopsies with assistance of USTUR staff, resulting in greater sample collection consistency. Autopsies performed by external pathologists may introduce variability due to partial sample collection and/or omission of samples listed in F402b.

Analytical completeness, or number of valid sample measurements relative to the number of valid samples collected typically varies for each project. For biokinetic modeling, a set of samples necessary for an accurate prediction of an individual intake is outlined in section 4.3.

Per the USTUR mission, overall completeness of data collected, which is defined as the number of valid sample measurements relative to the number of samples planned for collection, is achieved when analysis of all tissue samples is complete.

USTUR Cases are categorized as 'Intact,' 'Incomplete,' 'Surveyed,' or 'Complete'. 'Intact' means that no tissue samples have been analyzed. 'Incomplete' typically denotes that a selected sub-set of tissue analyses is in progress. The 'Surveyed' category is exclusive to whole-body cases. 'Surveyed' denotes that only analysis of selected tissue samples that provide key scientific information to determine the level of exposure has been completed, and can be used for biokinetic modeling. More tissue samples are available for 'Surveyed' cases. 'Complete' denotes that a full selection of tissue samples was analyzed and results were reported.

Data completeness is reported as the percent of tissue samples for which analysis is complete. Data completeness reaches 100% when all samples designated for analysis from a case have been analyzed. A lower level of data completeness may be adequate during the initial survey analysis of a whole-body donation to determine the level of exposure for use in biokinetic modeling.

6.11. SENSITIVITY

Sensitivity refers to the ability of an analytical procedure to quantify an analyte at a given concentration.

The following sensitivity requirements are established for USTUR tissue sample analysis

- radiochemical recovery yield (fractional tracer recovery) of the laboratory blank samples (reagent and method blanks),
- the *MDA* characterizing the detection sensitivity of a system, and
- the minimum quantifiable activity *MQA*.

"A method blank is a sample of a matrix as similar as practical to the associated samples that is free from the analytes (radionuclides) of interest to the extent possible. The method blank is processed simultaneously with, and under the same conditions as, samples through all steps of the analytical procedures. A reagent blank consists of the analytical reagent(s) in the procedure without the target analyte or sample matrix, introduced into the analytical procedure at the appropriate point and carried through all subsequent steps to determine the contribution of the reagents and of the involved analytical steps" (MARLAP p. 18–9).

The calculation of chemical recovery, the *MDA*, and the *MQA* is discussed in the following sections, and the equations used by the USTUR, adopted from MARLAP, appear in Chapter 5 and Appendix C. Acceptance criteria for sensitivity of sample measurements based on the *MQA* and other performance indicators are specified in Table 15.

6.12. MEASUREMENT PERFORMANCE CRITERIA

The measurement performance criteria described below permit USTUR analysts to flag data with verification and validation codes. The USTUR has adopted the MARLAP codes as shown in Table 14.

Data qualifiers are codes placed on an analytical result that alert data users to the validator's or verifier's concern about the result, and the basis for rejection or qualification of the data.

The validation process uses the qualifiers listed below to identify data points that do not meet the project MQOs or other analytical process requirements listed in the statement of work (SOW) or appropriate project plan document.

Table 14. Verification and validation (V&V) data qualifiers from MARLAP.

V&V Data Qualifier	Meaning
E	A notice to the validator that something was noncompliant. The verification process uses a qualifier ("E") to alert the validator to noncompliance, including missing documentation, contract compliance, etc. This qualifier may be removed or replaced during validation, based on the validator's interpretation of the effect of the noncompliance on the data's integrity.

V&V Data Qualifier	Meaning
U	A normal, not detected (< critical value) result.
Q	A reported combined standard uncertainty, which exceeds the project's required method uncertainty.
J	An unusually uncertain or estimated result. The assignment of the "J" qualifier relies heavily on the judgement and expertise of the reviewer and therefore, these qualifiers should be assigned as appropriate at the end of data validation.
R	A rejected result: the problems (quantitative or qualitative) are so severe that the data cannot be used. The assignment of the "R" qualifier relies heavily on the judgement and expertise of the reviewer and, therefore, these qualifiers should be assigned as appropriate at the end of data validation.
S	A result with a related spike result (laboratory control sample [LCS], matrix spike [MS] or matrix spike duplicate [MSD]) that is outside the control limit for recovery (%R); "S+" or "S-" used to indicate high or low recovery.
P	A result with an associated replicate result that exceeds the control limit.
B	A result with associated blank result, which is outside the control limit, "B+" or "B" used to indicate high or low results.

A data qualifier or a set of qualifiers does not apply to all similar data. The data validator incorporates USTUR's MQOs into the testing and qualifying decision-making process. The final validation report includes a summary of QC sample performance for use by the data assessor.

USTUR's measurement performance criteria are shown in Table 15. Additional discussion of two of these is provided in this section. Many of these criteria can also be found in ANSI/ANS-41.5-2012.

6.12.1. ACCURACY AND PRECISION MQO CRITERION

For accuracy and precision, the USTUR chooses to measure 1/30 Bq (2 dpm) with a *relative* combined standard uncertainty $u_R \leq 4\%$ as its primary MQO. This activity corresponds to the current spike level for tracers such as ^{243}Am and ^{242}Pu. For ^{239}Pu, this is about 76 × *MDA* and 8.4 × *MQA*(0.1). Effectively, we evaluate accuracy and precision at *MQA*(0.04). This choice was made so that historical measurements could be reviewed for their accuracy and precision. Also, since the *MDA* varies in a nonlinear way with the number of background counts that were observed, it is not specified as some multiple of the *MDA*.

6.12.2. SENSITIVITY MQO CRITERION

For sensitivity, the USTUR chooses to measure 0.5 mBq with a relative combined standard uncertainty $u_R \leq 35\%$ as its primary MQO for sensitivity. This activity is just slightly higher than the *MDA* as shown it Figure 3.

Table 15. Measurement performance criteria for use in data verification and validation.

QC sample or purpose	Associated MQO	Parameter type	Frequency or number	Measurement performance criteria (MPC)	Corrective action (CA) if MPC not met
Aliquot Split; only valid when each Sample contains > *MQA*(0.05)	Precision	Sample specific	1 per case (liver, lung, or large bone sample[7])	Tolerance Level (TL) for Precision: relative standard deviation, $s_R \le 0.15$	Investigate; Flag data as *J* or *U*
Aliquot Split	Representativeness	Sample specific	1 per case (liver, lung, or large bone sample)	TL: test of $H_0{:}C_1=C_2$ given u_{C1}, u_{C2} at $p = 0.05$	Investigate; Flag data as *J* or *U*
Sample-specific chemical yield	Sensitivity	Sample specific	Every sample	Sensitivity TLs: $0.5 \le y_{RR} \le 1.1$	Recount; Reanalyze (rerun); Flag data as *J* or *U*
(LCS) QC Samples	Accuracy & precision	Batch control	Every batch	Measure 1/30 Bq (2 dpm) with $u_R \le 4\%$	Investigate; Flag as *S*; Flag data as *J* or *U*
Reagent Blank (RB)	Accuracy: bias, sensitivity	Batch control	1 of every 20 samples	TL: $N_{RB,ROI} \le 15$. Note 1: any $N_{RB,ROI} \le 21$ counts gives $u_R \le 0.35$ and an *MDA* ≤ 0.5 mBq, USTUR's Sensitivity MQO. Note 2: some contamination may be expected in U and Th measurements	Investigate; consider using $N_{RB,ROI}$ in place of $N_{B,ROI}$ Flag as *B*; Flag data as *J* or *U*

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[7] Bone sample resulting in more than 2 g of ash.

QC sample or purpose	Associated MQO	Parameter type	Frequency or number	Measurement performance criteria (MPC)	Corrective action (CA) if MPC not met
Method blank (MB)	Accuracy: bias	Batch control	1 per ~12 samples (drying oven shelf)	TL: $N_{\mathrm{MB,ROI}} \le 15$. Some contamination may be expected in U and Th measurements.	Investigate; Flag as *B*; Flag data as *J* or *U*
QC samples	Accuracy: bias	Batch control	1 of every 20 samples	TL: $b_Y \le 0.1$	Check for detector contamination; check laboratory methods; Flag as *S;* Flag data as *J* or *U*;
Matrix spike (MS) (post digestion spike)	Accuracy	Batch control	Every sample	TL: Analyte u_{R} <10%; Professional judgment if Sample concentration > 4× spike level; possible tracer-analyte interference	Recount; Reanalyze (rerun); Flag as *S;* Flag data as *J* or *U*
QC samples	Sensitivity	Batch control	For each method blank, reagent blank, and spike solution	Sensitivity TL: $0.9 \le y_{\mathrm{RR}} \le 1.1$	Investigate; Flag data as *J* or *U*
Efficiency calibration	Accuracy	Instrument	Monthly	Measure standard. Control limits: $\bar{\varepsilon} - zu \le \varepsilon \le \bar{\varepsilon} + zu$ TL: $\frac{\lvert\varepsilon - \bar{\varepsilon}\rvert}{\bar{\varepsilon}} < 0.1$	Warning limits for *z* are ±2 (recount) and control limits are ±3 (Re-optimize, re-calibrate); OR 1% TL, whichever is less. Flag data as *E*

QC sample or purpose	Associated MQO	Parameter type	Frequency or number	Measurement performance criteria (MPC)	Corrective action (CA) if MPC not met
					previously counted on that detector
Background measurement (unused planchet)	Accuracy & sensitivity	Instrument	Monthly	Total Background $N_{\mathrm{B,total}} = \sum_{i} N_{\mathrm{B},i}$ (i = channel number) is compared to historical data. TLs: $\overline{N}_{\mathrm{B,total}} - zu \leq N_{\mathrm{B,total}} \leq \overline{N}_{\mathrm{B,total}} + zu$	Warning limits for *z* are ±2 (recount) and control limits are ±3 (Re-optimize, re-calibrate); Flag data as *E* previously counted on that detector
Background measurement (unused planchet)	Sensitivity	Instrument	Monthly	$N_{\mathrm{B,ROI}} \leq N_{\mathrm{B},MDA}$: for USTUR, any $N_{\mathrm{B}} \leq 21$ counts gives $u_{\mathrm{R}} \leq 0.35$ and an $MDA \leq 0.5$ mBq, USTUR's MQO. TL: $N_{\mathrm{B}} \leq 15$	Warning if >15 then recount; if > 21, decontaminate and repeat; Flag data as *E* previously counted on that detector;
Site-specific performance evaluation program (SSPEP) samples	Sensitivity	Instrument	Annually	Measure 0.5 mBq (0.03 dpm) with $u_{\mathrm{R}} \leq 35\%$	Investigate; Flag as *E*
SSPEP	Accuracy: bias	Instrument	Annually	TL for bias: $b_Y \leq 0.1$; nuclide identification	Investigate; Flag as *E*

QC sample or purpose	Associated MQO	Parameter type	Frequency or number	Measurement performance criteria (MPC)	Corrective action (CA) if MPC not met
SSPEP	Precision	Instrument	Annually	TL for precision: measure 1/30 Bq (2 dpm) with a *relative* combined standard uncertainty $u_\mathrm{R} \leq 4\%$	Investigate; Flag as *E*
Pipette check	Accuracy: bias	Instrument	1 per set of ICP-MS aliquots, or per set of tracer spikes	TL for bias: $b_\Pi < 5\%$	Use different pipette; Flag as *E*
Pipette calibration	Accuracy: bias, precision	Instrument	1 per year	TLs: Bias and u_R to manufacturers specifications	Replace pipette; Flag as *E*
Balance calibration	Accuracy: bias, precision	Instrument	1 per year	TLs: Bias and u_R to manufacturers specifications	Repair or replace balance; Flag as *E*
Furnace calibration	Accuracy	Instrument	1 per year	TLs: Temperature to manufacturers specifications	Repair or replace furnace and/or temperature controller; Flag as *E*
Based on valid/usable data collected	Data completeness	N/A	N/A	TL: Minimum > 90% Overall	Potential data usability / data gap issue; Flag as *E*

7. ASSESSMENT: VERIFICATION, VALIDATION, AND QUALITY ASSESSMENT

Figure 7 shows MARLAP's view of data assessment. These steps are described below for the USTUR.

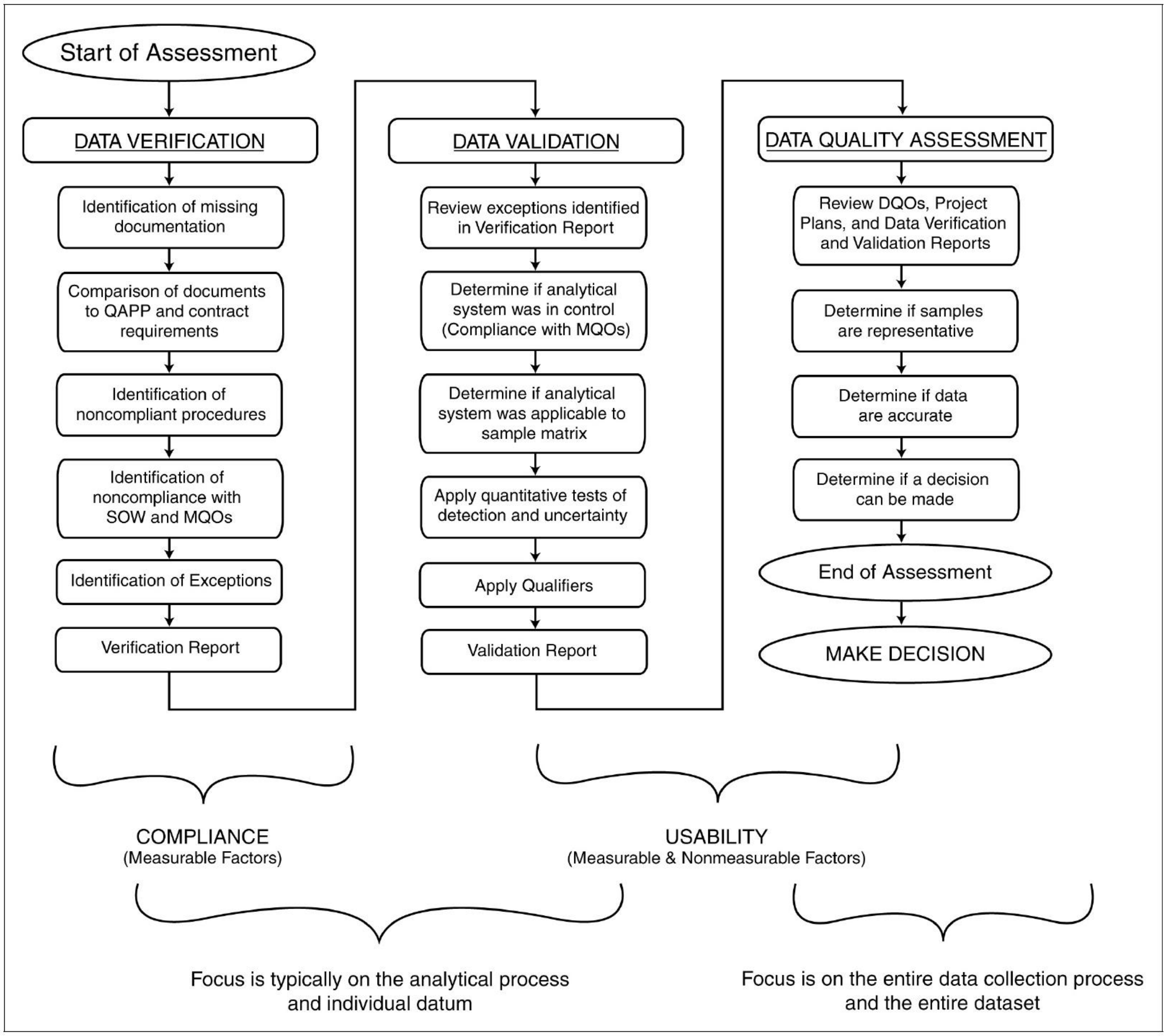

Figure 7. Overview of MARLAP's assessment process (MARLAP Fig. 8-1).

Both MARLAP and ANSI/ANS 41.5-2012 enumerate topics for data verification and validation. Table 16 shows where in the body of USTUR documents each of these topics (if applicable to the USTUR) can be found.

Table 16. Summary of V&V topics from MARLAP and ANSI/ANS 41.5-2012

Source	MARLAP / ANSI 41.5	Category	Variable, process, or task	Reference
MARLAP	8.5.1.1	Sample Handling & Analysis System	Sample Descriptors	DQO Doc, USTUR Procedures
MARLAP	8.5.1.2	Sample Handling & Analysis System	Aliquant Size	DQO Doc, USTUR Procedures
MARLAP	8.5.1.3	Sample Handling & Analysis System	Dates of Sample Collection, Preparation, and Analysis	DQO Doc, USTUR Procedures
MARLAP	8.5.1.4	Sample Handling & Analysis System	Preservation	USTUR procedures
MARLAP	8.5.1.5	Sample Handling & Analysis System	Tracking	USTUR procedures
MARLAP	8.5.1.6	Sample Handling & Analysis System	Traceability	DQO Doc
MARLAP	8.5.1.7	Sample Handling & Analysis System	QC Types and Linkages	Table 15
MARLAP	8.5.1.8	Sample Handling & Analysis System	Chemical Separation (Yield)	Table 15
MARLAP	8.5.1.9	Sample Handling & Analysis System	Self-Absorption	USTUR procedures
MARLAP	8.5.1.10	Sample Handling & Analysis System	Efficiency, Calibration Curves, and Instrument Background	Table 15, DQO Doc
MARLAP	8.5.1.11	Sample Handling & Analysis System	Spectrometry Resolution	USTUR procedures
MARLAP	8.5.1.12	Sample Handling & Analysis System	Dilution and Correction Factors	DQO Doc, USTUR Procedures
MARLAP	8.5.1.13	Sample Handling & Analysis System	Counts and Count Time (Duration)	DQO Doc
MARLAP	8.5.1.14	Sample Handling & Analysis System	Result of Measurement, Uncertainty, MDA, and Units	DQO Doc
MARLAP	8.5.2.1	Quality Control Samples	Method Blank	DQO Doc
MARLAP	8.5.2.2	Quality Control Samples	Laboratory Control Samples (LCS)	DQO Doc

Source	MARLAP / ANSI 41.5	Category	Variable, process, or task	Reference
MARLAP	8.5.2.3	Quality Control Samples	Laboratory Replicates	DQO Doc
MARLAP	8.5.2.4	Quality Control Samples	Matrix Spikes and Matrix Spike Duplicates	N/A
MARLAP	8.5.3.1	Tests of Detection and Unusual Uncertainty	Detection	DQO Doc
MARLAP	8.5.3.2	Tests of Detection and Unusual Uncertainty	Detection Capability	DQO Doc
MARLAP	8.5.3.3	Tests of Detection and Unusual Uncertainty	Large or Unusual Uncertainty	DQO Doc
ANSI	4.1	Sample-specific Parameters	Sample preservation	DQO Doc
ANSI	4.2	Sample-specific Parameters	Holding times	DQO Doc, USTUR Procedures
ANSI	4.3	Sample-specific Parameters	Sample-specific chemical yield	DQO Doc
ANSI	4.4	Sample-specific Parameters	Required detection level	DQO Doc
ANSI	4.5	Sample-specific Parameters	Nuclide identification	DQO Doc
ANSI	4.6	Sample-specific Parameters	Quantification and CSU	DQO Doc
ANSI	4.7	Sample-specific Parameters	Detectability	DQO Doc
ANSI	4.8	Sample-specific Parameters	Sample aliquot representativeness	N/A[a]
ANSI	5.1	Batch Control Parameters	LCS analysis	DQO Doc
ANSI	5.2	Batch Control Parameters	Matrix spike analysis	N/A
ANSI	5.3	Batch Control Parameters	Duplicate and MSD sample analysis	N/A
ANSI	5.4	Batch Control Parameters	Batch method blank analysis	DQO Doc
ANSI	6.1	Instrument Parameters	Counting efficiency calibration	DQO Doc
ANSI	6.2	Instrument Parameters	Energy calibration	DQO Doc
ANSI	6.3	Instrument Parameters	Background determination	DQO Doc

[a] not applicable, the USTUR does not implement this.

7.1. DATA VERIFICATION

Multiple data verification steps are described below.

7.1.1. IDENTIFICATION OF MISSING DOCUMENTATION

A complete list of USTUR Standard Operating Procedures (SOPs) is provided in Appendix XYZ, along with their revision dates and current status. This list is reviewed periodically for missing documentation.

7.1.2. COMPARISON OF DOCUMENTS TO QAPP AND CONTRACT REQUIREMENTS

This document forms the core of the USTUR Quality Assurance Project Plan (QAPP). As defined in MARLAP, a QAPP is a "formal document describing in detail the necessary quality assurance, quality control, and other technical activities that must be implemented to ensure that the results of the work performed will satisfy the stated performance criteria."

7.1.3. QAPP

Currently, the USTUR's QAPP consists of this document and the Quality Assurance Procedure (QAP), USTUR 800 (rev. 2000).

7.1.3.1. CONTRACT REQUIREMENTS

The USTUR is primarily funded by grant DE-HS0000073 from the US Department of Energy's Office of Health, Safety, and Security. The DOE grant proposal does not explicitly state what measurement capabilities must be. Consequently, the USTUR has developed its own requirements as summarized in this document and the QAPP.

Contract requirements for external ICP-MS measurements include the requirements of this document, including analysis of blanks, duplicates, splits, and blind spikes.

7.1.4. IDENTIFICATION OF NONCOMPLIANT PROCEDURES

Procedures at the USTUR are routinely reviewed and revised as necessary and as time and resources permit. Currently, there are plans to integrate the MQOs and Performance Indicators described here into the USTUR QAPP.

7.1.5. IDENTIFICATION OF NONCOMPLIANCE WITH THE SOW AND MQOS

USTUR's grant application serves as its SOW. Noncompliance with MQOs identified by the criteria in Table 15 for measurements made by the USTUR and by external contractors performing ICP-MS is documented and reported to management.

7.1.6. IDENTIFICATION OF EXCEPTIONS

Exceptions are identified by comparing performance indicators to control limits and tolerance limits as described in Table 15. Any exception from compliance is justified and documented, for example, an instance of technology shortfall in a specific case.

7.1.7. VERIFICATION REPORT

Verification of data is done at the same time as reporting and entry into the databases, so all verification is recorded in the database. It includes all the elements above.

7.2. DATA VALIDATION

"Validation addresses the reliability of the data. The validation process begins with a review of the verification report and laboratory data package to identify its areas of strength and weakness. This process involves the application of qualifiers that reflect the impact of not meeting the MQOs and any other analytical process requirements. Validation then evaluates the data to determine the presence or absence of an analyte, and the uncertainty of the measurement process. During validation, the technical reliability and the degree of confidence in reported analytical data are considered. The data validator should be a scientist with radiochemistry experience. Validation flags (i.e., qualifiers) are applied to data that do not meet the performance acceptance criteria established in the project plan documents. The products of the validation process are validated data and a validation report stating which data are acceptable, which data are sufficiently inconsistent with the validation acceptance criteria in the expert opinion of the validator, and a summary of the QC sample performance. The appropriate data validation tests should be established during the project planning phase. The point of validation is to perform a systematic check on a set of data being used to meet the project MQOs and any other analytical process requirements. Documenting that such a check cannot be done is an appropriate and essential validation activity" (MARLAP, Chapter 8).

At the USTUR, the data validator is a scientist with radiochemistry experience.

7.2.1. VALIDATION OF MASS DIFFERENCES

In addition to the quality measures listed in Table 15, the USTUR checks for consistency of mass measurements calculated as the difference of two masses. For the reader's convenience, definitions of mass quantities are repeated here. If a variable is outside of its valid value range and investigation is conducted, the variable is flagged if the discrepancy cannot be resolved.

Table 17. Summary of Measurement Performance Criteria. Valid value ranges for mass differences expressed as tolerance limits

Mass variable	Name of mass variable	Formula or measurement result	Valid value range[a]
m_{aliq}	mass of sample aliquot	measurement result	
m_{ash}	mass of ashed prepared sample	$m_{\mathrm{ash}} = m_{\mathrm{ash+bkr}} - m_{\mathrm{bkr}}$	$m_{\mathrm{ash}} \geq 0 \pm 2u$
$m_{\mathrm{ash+bkr}}$	mass of beaker and ashed prepared sample	measurement result	
m_{bkr}	tare mass of beaker	measurement result	
m_{bottle}	tare mass of empty solution bottle	measurement result	

Mass variable	Name of mass variable	Formula or measurement result	Valid value range[a]
m_{dry}	mass of dry sample	$m_{\mathrm{dry}} = m_{\mathrm{dry+bkr}} - m_{\mathrm{bkr}}$	$0 \pm 2u < m_{\mathrm{dry}} \le m_{\mathrm{prep}}$
$m_{\mathrm{dry+bkr}}$	mass of beaker and dry sample	measurement result	
m_{prep}	prepared mass of sample	measurement result	
m_{soln}	mass of dissolved prepared sample solution	$m_{\mathrm{soln}} = m_{\mathrm{soln+bottle}} - m_{\mathrm{bottle}}$	$m_{\mathrm{soln}} \ge 0 \pm 2u$
$m_{\mathrm{soln+bottle}}$	mass of filled solution bottle	measurement result	
$m_{\Pi\mathrm{avg}}$	average mass of deionized water pipetted ("π-petted")	$m_{\Pi\mathrm{avg}} = \frac{m_{\Pi 1} + m_{\Pi 2} + m_{\Pi 3} + m_{\Pi 4}}{4}$	
$m_{\Pi j}$	j^{th} mass of deionized water pipetted	measurement result	
f_{ash}	fraction of prepared sample mass that is ash, $= m_{\mathrm{ash}}/m_{\mathrm{prep}}$	$f_{\mathrm{ash}} = m_{\mathrm{ash}}/m_{\mathrm{prep}}$	$f_{\mathrm{ash}} \le 1$
f_{aliq}	fraction of mass of prepared sample that is sample aliquot	$f_{\mathrm{aliq}} = m_{\mathrm{aliq}}/m_{\mathrm{soln}}$	$f_{\mathrm{aliq}} \le 1$
m_{samp}	mass of whole tissue sample at autopsy	measurement result	
$\Delta m_{\mathrm{samp\text{-}prep}}$	mass removed in preparing sample (e.g., blood, fat, connective tissue, …)	$\Delta m_{\mathrm{samp\text{-}prep}} = m_{\mathrm{samp}} - m_{\mathrm{prep}}$	$m_{\mathrm{samp}} - m_{\mathrm{prep}} \ge 0 \pm 2u$

[a]depending on which balance is used, $u = 0.01$ g or $u = 0.0001$ g.

7.2.2. Validation of Volumetric Measurements

Volumetric measurements also have uncertainties. Since the bias-corrected tracer volume has an associated Type A uncertainty evaluation, it is appropriate to set its tolerance limit at a reasonable level for laboratory analysis.

Table 18. Tolerance limit associated with a bias-corrected volume measurement.

Quantity	Description	Unit	Value origin	Uncertainty type	Uncertainty, u_{R}
$V'_{Y\mathrm{tr}}$	bias-corrected tracer volume for tracer isotope Y, with unit change	mL	Eq. (34)	A	$u_{\mathrm{R}} \equiv s(m_{\Pi\mathrm{avg}})/m_{\Pi\mathrm{avg}} \le 0.01$

7.2.3. Review Exceptions Identified in Verification Report

Management reviews all reports submitted by the data analyst. Exceptions are approved as needed.

7.2.4. Determine Whether Analytical System Was in Control (Compliance with MQOs)

QC activities, tolerance limits, tolerance charts, and control charts are reviewed for compliance with MQOs using the criteria outlined in Table 15.

7.2.5. Determine Whether Analytical System Was Applicable to Sample Matrix

The sample matrix that the USTUR analyzes consists of human organ and tissue samples. Procedure USTUR 115 (Microwave Tissue Digestion for Isolation of

Radionuclides) describes applicability of the analytical system to various tissue types. For example, the radiochemistry technician examines the results of each digestion and dissolution by matrix type for undissolved solids, cloudiness, etc., to ensure optimal recovery.

7.2.6. APPLY QUANTITATIVE TESTS OF DETECTION AND UNCERTAINTY

Radiochemistry measurement results are automatically compared to the critical value of the net count rate, with total propagated uncertainty specified. Data not in compliance with the criteria in this document are automatically flagged during the data reduction process.

With the exception of Registrants with no history of intake, critical levels are not used as a basis for decisions since the presence of radionuclides is not in question. When a radionuclide is known to be present in a Registrant, the activity and its uncertainty are measured and reported, regardless of their values, but they are not compared to a critical level.

7.2.7. APPLY QUALIFIERS

Validation flags (i.e., qualifiers) are applied to data that do not meet the performance acceptance criteria outlined above. Validation flags or data qualifiers denote results not meeting the MQOs and any other analytical process requirements. An example of a validation flag would be one indicating the y_{RR} was too low or too high. A qualifier could indicate the presence or absence of an analyte, or excessive uncertainty of the measurement process. The data validator considers the technical reliability and the degree of confidence in reported analytical data.

7.2.8. VALIDATION REPORT

The validation report summarizes results of each data validation step.

7.3. DATA QUALITY ASSESSMENT

“Data Quality Assessment is the last phase of the data collection process, and consists of a scientific and statistical evaluation of project-wide knowledge to assess the usability of data sets. To assess and document overall data quality and usability, the data quality assessor integrates the data validation report, field information, assessment reports, and historical project data, and compares the findings to the original project DQOs. The DQA process uses the combined findings of these multi-disciplinary assessments to determine data usability for the intended decisions, and to generate a report documenting that usability and the causes of any deficiencies. It may be useful for a validator to work with the assessor to assure the value of the validation process (e.g., appropriateness of rejection decision) and to make the process more efficient” (MARLAP Chapter 8).

7.3.1. REVIEW DQOS, PROJECT PLANS, AND DATA VERIFICATION AND VALIDATION REPORTS

Management reviews DQOs and Project Plans on an annual basis. Verification and Validation Reports are reviewed as they are produced and data becomes available for addition to the databases.

7.3.2. DETERMINE WHETHER SAMPLES ARE REPRESENTATIVE

Representativeness of samples is rarely an issue at USTUR except for tissues such as the skeleton where only a limited number of bones (2–9) are analyzed for partial-body donations. In many cases, an entire organ is analyzed (e.g., the kidneys, spleen, esophagus, etc.), so representativeness is not an issue.

7.3.3. DETERMINE WHETHER DATA ARE ACCURATE

Accuracy, as characterized by measures of bias and precision, is assessed regularly by examining the indicators outlined in Table 15.

7.3.4. DETERMINE WHETHER DATA ARE USABLE

Data that meet all the DQOs are deemed to be usable.

However, the usability of data for a specific research purpose that was not considered when developing DQOs may depend on the data’s uncertainty.

7.4. SUMMARY OF ASSESSMENT

The Assessment process determines that the data are in compliance using measurable factors; and that the data are usable using both measurable and nonmeasurable factors. These actions take place for the data collection and the analytical process, for individual measurements and the entire dataset.

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Appendix A. PREDICTED ACTIVITY ON PLANCHETS

Plutonium concentrates in the liver and skeleton, but it is still present at lower concentrations in soft tissues, such as muscle or brain. Soft tissues with lower concentrations are not thought to be of dosimetric significance, and the ICRP systemic models combine most of them into a single pool called *Other soft tissues*. The concentration of plutonium in the individual soft tissues is assumed to be the same as the concentration of Other soft tissues.

Currently, a potential tissue donor must have received at least a 74 Bq (2 nCi) intake to become a USTUR Registrant. If the individual inhaled ^{239}Pu nitrate, 0.13 Bq would remain in the Other soft tissues at 50 years post-intake[8]. The mass of Other soft tissues in the reference worker is 59.5 kg (ICRP 89)[9]. Therefore, 50 years after the reference worker inhaled 74 Bq, the concentration in Other soft tissues would be

$$\frac{0.13\ \text{Bq}}{59.5\ \text{kg}} = 2.1\times10^{-3}\ \text{Bq kg}^{-1}\ \text{of tissue.}$$

The ash fraction for soft tissues is 2%. Thus, 1 kg of soft tissue results in 20 g of ash, and a 74 Bq intake would result in

$$2.1\times10^{-3}\ \text{Bq kg}^{-1}\ \text{tissue} \times \frac{1\ \text{kg tissue}}{20\ \text{g ash}} = 1.1\times10^{-4}\ \text{Bq g}^{-1}\ \text{ash.}$$

This ash is dissolved into acid, and an aliquot of the resulting solution is loaded onto a separation column. The aliquot size is limited to 2 g of ash. If more than 2 g of ash is loaded onto the columns, the actinide separation chemistry is compromised due to the matrix effect. Therefore, if the lowest acceptable chemical recovery of 50% is assumed, there would be

$$1.1\times10^{-4}\ \text{Bq g}^{-1}\ \text{ash} \times 2\ \text{g ash} \times 0.5 = 1.1\times10^{-4}\ \text{Bq on the planchet.}$$

The above value was calculated for plutonium nitrate. If the reference worker inhaled $^{239}PuO_2$ dioxide and passed away 50 years later, there would be an estimated 1.1×10^{-5} Bq on the planchet following analysis of Other soft tissue. The above calculations were repeated for the skeleton for both plutonium nitrate (2.8×10^{-4} Bq) and oxide (2.8×10^{-5} Bq).

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[8] The retention in the *Other soft tissues* was calculated with the USTUR's research edition of IMBA Professional Plus®, ver. 4.1.66. The calculation used the plutonium systemic model and absorption and lung clearance parameters from ICRP's Occupational Intakes of Radionuclides series (ICRP 130 and ICRP 141).

[9] This value was calculated based upon the values in Table 2, where Other soft tissues = Total body - Skeleton - Liver - Lungs

Predictions of plutonium activity on a planchet following analysis of the liver or lungs are simpler because the aliquot size for the liver and lungs is based upon a set fraction of the entire acid solution, rather than the ash fraction. For example, if the reference worker inhaled 74 Bq of plutonium nitrate, there would be 0.599 Bq in his liver 50 years later. Assuming half of the liver is analyzed, an aliquot fraction of 0.1, and 50% recovery, there would be

0.599 Bq $\times 0.5 \times 0.1 \times 0.5 = 1.5 \times 10^{-2}$ Bq on the planchet.

A similar calculation can be carried out to predict the activity that would be on a planchet if the liver were analyzed 50 years after an intake of $^{239}PuO_2$, and for the lungs following a nitrate or oxide inhalation.

Table 19 summarizes the predicted activity that would be on a planchet following analysis of the key tissues, if the reference worker passed away 50 years after inhaling 74 Bq for isotopes of Pu, Am, U, Th, Ra, Cm, and Np.

Table 19. Predicted activity on planchets (Bq) 50 years after a 74 Bq (2 nCi) intake of Pu, Am, U-nat, Th, Ra, Cm, and Np.

			Inhalation								
			Soluble		Moderately soluble			Insoluble			
Radionuclide	Tissue or organ	Systemic uptake	Hexafluoride	Citrate	Nitrate	Chloride, citrate, nitrate, sulphate, fluoride	Nitrate, oxide, chloride	Dioxide	Octoxide	Type S	Ingestion
^{239}Pu	Lungs				7.00E-05			1.36E-02			
^{239}Pu	Liver	4.10E-01			1.50E-02			1.70E-03			
^{239}Pu	Skeleton	7.59E-03			2.78E-04			2.82E-05			
^{239}Pu	Soft Tissue	2.94E-03			1.06E-04			1.14E-05			
^{241}Am	Lungs				9.05E-05			2.45E-03			
^{241}Am	Liver	4.83E-02			2.68E-03			1.60E-03			
^{241}Am	Skeleton	1.46E-02			8.10E-04			2.51E-04			
^{241}Am	Soft Tissue	5.46E-03			3.02E-04			8.64E-05			
^{235}U	Lungs								1.73E-06		
^{235}U	Liver	8.75E-05	1.22E-05						3.11E-06		
^{235}U	Skeleton	9.51E-05	1.33E-05						2.75E-06		
^{235}U	Soft Tissue	1.74E-04	2.44E-05						4.53E-06		
^{232}Th	Lungs					3.26E-42		2.62E-03			
^{232}Th	Liver	5.27E-03				3.09E-04		1.12E-04			
^{232}Th	Skeleton	1.35E-02				8.08E-04		2.40E-04			
^{232}Th	Soft Tissue	4.37E-03				2.63E-04		6.91E-05			
^{226}Ra	Lungs										
^{226}Ra	Liver	2.32E-07			5.55E-08						4.63E-08
^{226}Ra	Skeleton	6.57E-05			1.58E-05						3.55E-07
^{226}Ra	Soft Tissue	3.26E-07			7.80E-08						4.40E-10
^{244}Cm	Lungs			1.05E-04			9.16E-05				
^{244}Cm	Liver	7.72E-03		3.14E-03			2.70E-03			1.60E-03	
^{244}Cm	Skeleton	6.30E-05		9.50E-04			8.16E-04			2.51E-04	
^{244}Cm	Soft Tissue	5.88E-06		3.55E-04			3.02E-04			8.64E-05	
^{237}Np	Lungs				1.09E-42			2.62E-03			
^{237}Np	Liver	4.81E-03			7.69E-04			2.12E-04			
^{237}Np	Skeleton	3.85E-03			6.23E-04			9.30E-05			
^{237}Np	Soft Tissue	2.20E-03			3.57E-04			3.74E-05			

Appendix B. DATA DICTIONARY

Table 20. Data dictionary for other variables, in alphabetical order by variable name

Symbol	Quantity	Unit	Radiochemistry database	Radiochemistry summary spreadsheet	Sample analysis spreadsheets
$A_{\mathrm{isotope,destination,origin}}(t)$	activity variable, identified by isotope, the chemical fraction Z it is ultimately found in, and its origin (samp = whole tissue sample; prep = prepared tissue sample, dry, ash, soln = solution, aliq = aliquot, tr = tracer).				
$A_{\mathrm{A1,Am,A1inA3tr}}(t_{\mathrm{C,Am}})$	activity of ^{241}Am in Am fraction from ^{243}Am tracer as of count date	Bq			1
$A_{\underline{\mathrm{A1,Am,aliq}}}(t_{\mathrm{C,Am}})$	activity of ^{241}Am on Am-fraction planchet at count date, corrected for ^{241}Am contamination	Bq			1
$A_{\mathrm{A1,Am,P1inP2tr}}(t_{\mathrm{C,Am}})$	activity of ^{241}Am in Am fraction that has grown in from decay of ^{241}Pu in Pu tracer as of count date	Bq			1
$A_{\underline{\mathrm{P8,Pu,aliq}}}(t_{\mathrm{C,Pu}})$	activity of ^{238}Pu on Pu-planchet at count date, corrected for ^{241}Am contamination that produces counts in the same ROI as ^{238}Pu				
$A_{\mathrm{A1,Pu,P1inP2tr}}(t_{\mathrm{C,Pu}})$	activity of ^{241}Am in Pu fraction that has grown in from decay of ^{241}Pu in Pu tracer as of count date	Bq			1
$A_{X,\text{-},\mathrm{prep}}(t_{\mathrm{D}})$	activity of isotope X in dissolved prepared sample at collection date	Bq	1	1	
$A_{X,\text{-},\mathrm{prep}}(t_{\mathrm{D}})$	activity of isotope X in solution, which is assumed to be the same as in the prepared sample, corrected back to date/time of collection t_{D}	Bq			1
$A_{X,Z,\text{-}}(t_{\mathrm{C},Z})$	activity of isotope X on planchet at counting date/time for element fraction Z	Bq			
$A_{X,Z,\mathrm{soln}}(t_{\mathrm{C},Z})$	activity of isotope X in solution at count date for Z-planchet	Bq			1
$A_{Y,\mathrm{known\text{-}aliq},Y\mathrm{tr}}(t_{\mathrm{C},Z})$	known activity of tracer isotope Y placed in aliquot referenced to count date for Z-fraction planchet	Bq		1	
$A_{Y,\mathrm{measured\text{-}aliq},Y\mathrm{tr}}(t_{\mathrm{C},Z})$	measured activity of tracer isotope Y placed in aliquot referenced to count date for Z-fraction planchet	-		1	

Symbol	Quantity	Unit	Radiochemistry database	Radiochemistry summary spreadsheet	Sample analysis spreadsheets
b_X	bias in measurement of isotope *X*	-		1	
b_Π	bias of pipette volume		1		
$C_{X,\text{ash}}(t_{\text{C},Z})$	activity concentration of isotope *X* referenced to count date for *Z*-fraction planchet	Bq kg^{-1} tissue ash	1	1	
$C_{X,\text{wet}}(t_\text{D})$	activity concentration of isotope *X* in wet tissue at collection date	Bq kg^{-1} wet tissue	1	1	
$C_{X,Y\text{tr}}(t_{\text{ref},Y})$	activity concentration of isotope *X* in *Y* tracer as of tracer reference date as specified on calibration certificate supplied with tracer	Bq mL^{-1}			1
d	Stapleton's *d*-factor, used in calculating S_C and *MDA*; $d = 0.4$ for $\alpha = 0.05$	-			
f_aliq	fraction of mass of prepared sample that is sample aliquot, $= m_\text{aliq}/m_\text{soln}$	-		1	
f_ash	fraction of prepared sample mass that is ash, $= m_\text{ash}/m_\text{prep}$	-		1	
$f_{X,\text{ROI}}$	these can be called "ROI intensity," and is the "fraction of all α-particles emitted by isotope *X* expected to produce counts in the ROI" for each ROI choice and each radionuclide, $f_{X,\text{ROI}}$. They are NOT branching fractions or branching ratios. (Provided by AlphaVision, where it is incorrectly called "Branching Ratio"). $f_{X,\text{ROI}}$ may be expressed in % but is used in calculations as a fraction				1
m_aliq	mass of sample aliquot	g		1	1
m_ash	mass of ashed prepared sample	g	1	1	
$m_\text{ash+bkr}$	mass of beaker and ashed prepared sample	g	1	1	
m_bkr	tare mass of beaker	g	1	1	
m_bottle	tare mass of empty solution bottle	g	1	1	
MDA_X	minimum detectable activity of isotope *X*	Bq			1
m_dry	mass of dry sample	g	1	1	
$m_\text{dry+bkr}$	mass of beaker and dry sample	g	1	1	

Symbol	Quantity	Unit	Radiochemistry database	Radiochemistry summary spreadsheet	Sample analysis spreadsheets
m_{prep}	prepared mass of sample	g	1	1	
m_{soln}	mass of dissolved prepared sample solution	g	1	1	1
$m_{\text{soln+bottle}}$	mass of filled solution bottle	g	1	1	
$m_{\Pi\text{avg}}$	average mass of deionized water pipetted (“π-petted”)	g	1		
$m_{\Pi j}$	j^{th} mass of deionized water pipetted	g	1		
$N_{\text{B}}(X)$	blank (background) counts within the ROI for isotope X over the live time as determined by the AlphaVision Program	-			1
$N_{\text{N}}(X)$	number of net counts for isotope X. Note that N_{N} may not be an integer if $t_{\text{B}} \neq t_{\text{S}}$, so N_{N} is an unphysical quantity.				
$N_{\text{S}}(X)$	sample (gross) counts within the ROI for isotope X over the live time as determined by the AlphaVision Program	-			1
$r_{\text{A}}(X,Y)$	ratio of the activity of isotope X to the activity of isotope Y	-		1	
$R_{\text{B}}(X)$	background count rate for isotope X	s^{-1}			
$R_{\text{N}}(X)$	net count rate for isotope X	s^{-1}			
$R_{\text{S}}(X)$	sample (gross) count rate for isotope X	s^{-1}			
S'_{C}	critical value of the net count rate R_{N} (the decision threshold for the net count rate)	s^{-1}			
$S'_{\text{D},X}$	minimum detectable value of net count rate	s^{-1}			
S_{C}	critical value of the net counts N_{N}	-			
$S_{\text{D},X}$	minimum detectable value of net counts	-			
$\text{T}_{1/2,X}$	half-life of isotope X (may be treated as a constant or an uncertain variable)	day			1
T_{A}	ambient temperature	˚C	1	1	
t_{B}	background count live time duration	s			1

Symbol	Quantity	Unit	Radiochemistry database	Radiochemistry summary spreadsheet	Sample analysis spreadsheets
$t_{\mathrm{C},Z}$	count date/time for element fraction Z , where $Z \in$ (Th, U, Pu, Am, Cm). For maximum accuracy, $t_{\mathrm{C},Z}$ should be the midpoint of the counting interval	days[10]			1
t_D	collection date/time for whole tissue sample (date of death)	days[10]			1
$t_{\mathrm{ref},Y}$	reference date/time for applicable standard for tracer isotope Y; for example, $t_{\mathrm{ref,P2}}$ $t_{\mathrm{ref,A3}}$	days[10]			1
t_S	sample (gross) count live time duration	s			1
t_sep	date/time of separation of elements into, for example, Pu fraction and Am fraction	days[10]			
$u(A_{X,\text{-},\mathrm{prep}}(t_\mathrm{D}))$	uncertainty of activity of isotope X in dissolved prepared sample at collection date	Bq	1	1	
$u(C_\mathrm{ash}(X))$	uncertainty in activity concentration of isotope X for tissue ash	Bq kg^{-1} tissue ash	1	1	
$u(C_\mathrm{wet}(X))$	uncertainty in activity concentration of isotope X for wet tissue	Bq kg^{-1} wet tissue	1	1	
$u_\mathrm{R}(A_{X,\mathrm{soln},\text{-}}(t_{\mathrm{C},Z}))$	relative uncertainty of activity of isotope X in solution at count date	-			1
$u_\mathrm{R}(A_{X,\mathrm{soln},\text{-}}(t_\mathrm{D}))$	relative uncertainty of activity of isotope X in solution at collection date	-			1
$u_\mathrm{R}(A_{\underline{X,\mathrm{Z,aliq}}}(t_{\mathrm{C},Z}))$	relative uncertainty of activity of isotope X on Z-fraction planchet at count date, corrected for ^{241}Am contamination	-			1
$u_\mathrm{R}(A_{X,Z,\mathrm{aliq}}(t_{\mathrm{C},Z}))$	relative uncertainty of activity of isotope X on planchet at count date	-		1	1
$u_\mathrm{R}(V_\Pi)$	relative uncertainty of pipette volume	-	1	1	
$u_\mathrm{R}(y_\mathrm{RR}(Y))$	relative uncertainty for fractional radiochemical recovery yield of tracer Y	-			1

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[10] Excel date

Symbol	Quantity	Unit	Radiochemistry database	Radiochemistry summary spreadsheet	Sample analysis spreadsheets
$u_R(\varepsilon)$	relative uncertainty in the efficiency of the detector (provided by AlphaVision)	-			1
V_{Ytr}	volume of isotope *Y* tracer added	µL			1
V'_{Ytr}	bias-corrected tracer volume for tracer isotope *Y*, with unit change	mL			
V_{Π}	volume pipette is set to	µL	1	1	
X	isotope *X*, an isotope of interest (note 1)				
Y	isotope *Y*, usually a tracer isotope				
$y_{RR}(Y)$	radiochemical recovery yield (fractional tracer recovery) for tracer *Y*	-			1
Z	element *Z*, the elemental fraction after radiochemical separation				
$z_{1-\alpha}$	standard normal deviate for Type I error, set at 1.645 corresponding to a false positive probability $\alpha = 0.05$	-			1
$z_{1-\beta}$	standard normal deviate for Type II error, set at 1.645 corresponding to a false negative probability $\beta = 0.05$	-			1
ε	efficiency of the detector in any ROI (provided by AlphaVision); differs for each detector	-			1
λ_X	decay constant of isotope *X*	day^{-1}			1

Appendix C. EQUATIONS COMPENDIUM

C.1. MASSES

For each directly measured mass (as opposed to a calculated mass), the uncertainty is assigned by the radiochemistry technician based on the manufacturer's uncertainty associated with each digital balance.

C.1.1. MASS OF THE DRY PREPARED SAMPLE

The mass of the dry prepared sample, m_{dry}, is the difference between the mass of the beaker plus the dry prepared sample $m_{\text{dry+bkr}}$ and the tare mass of the beaker m_{bkr}:

$$m_{\text{dry}} = m_{\text{dry+bkr}} - m_{\text{bkr}} \tag{19}$$

The uncertainty in m_{dry} is

$$u(m_{\text{dry}}) = \sqrt{u^2(m_{\text{dry+bkr}}) + u^2(m_{\text{bkr}})} \tag{20}$$

where the uncertainties in $m_{\text{dry+bkr}}$ and m_{bkr} are typically ±0.01 g.

C.1.2. MASS OF THE ASHED SAMPLE

The mass of the ashed prepared sample, m_{ash}, is the difference between the mass of the beaker and the ashed prepared sample $m_{\text{ash+bkr}}$ and the tare mass of the beaker m_{bkr}:

$$m_{\text{ash}} = m_{\text{ash+bkr}} - m_{\text{bkr}} \tag{21}$$

The uncertainty in m_{ash} is

$$u(m_{\text{ash}}) = \sqrt{u^2(m_{\text{ash+bkr}}) + u^2(m_{\text{bkr}})} \tag{22}$$

where the uncertainties in $m_{\text{ash+bkr}}$ and m_{bkr} are typically ±0.01 g.

C.1.3. MASS OF THE DISSOLVED SAMPLE

The mass of the dissolved prepared sample solution, m_{soln}, is the difference between the mass of the filled solution bottle $m_{\text{soln+bottle}}$ and the tare mass of the empty solution bottle m_{bottle}:

$$m_{\text{soln}} = m_{\text{soln+bottle}} - m_{\text{bottle}} \tag{23}$$

The uncertainty in m_{soln} is

$$u(m_{\text{soln}}) = \sqrt{u^2(m_{\text{soln+bottle}}) + u^2(m_{\text{bottle}})} \tag{24}$$

where the uncertainties in $m_{\text{soln+bottle}}$ and m_{bottle} are typcially ±0.01 g.

C.2. MASS FRACTIONS

Two mass fractions are of interest: the fraction of a sample that is ash, and the fraction of a sample that is represented by an aliquot.

C.2.1. ASH FRACTION

The fraction of prepared sample mass that is ash, f_{ash}, is the ratio of the mass of the ashed sample, m_{ash} (g) to the mass of the prepared sample m_{prep} (g):

$$f_{\mathrm{ash}} = \frac{m_{\mathrm{ash}}}{m_{\mathrm{prep}}} \tag{25}$$

The uncertainty in f_{ash} is a function of the mass measurements and their uncertainties:

$$u(f_{\mathrm{ash}}) = \frac{1}{m_{\mathrm{prep}}}\sqrt{u^2(m_{\mathrm{ash}}) + \frac{m_{\mathrm{ash}}^2}{m_{\mathrm{prep}}^2}u^2\left(m_{\mathrm{prep}}\right)}. \tag{26}$$

C.2.2. ALIQUOT FRACTION

The fraction of mass of sample that is in the sample aliquot, f_{aliq}, is the ratio of the mass of sample aliquot, m_{aliq} (g) to the mass of the dissolved sample solution m_{soln} (g):

$$f_{\mathrm{aliq}} = \frac{m_{\mathrm{aliq}}}{m_{\mathrm{soln}}} \tag{27}$$

The uncertainty in f_{aliq} is a function of the mass measurements and their uncertainties:

$$u\left(f_{\mathrm{aliq}}\right) = \frac{1}{m_{\mathrm{soln}}}\sqrt{u^2\left(m_{\mathrm{aliq}}\right) + \frac{m_{\mathrm{aliq}}^2}{m_{\mathrm{soln}}^2}u^2(m_{\mathrm{soln}})}. \tag{28}$$

C.3. PIPETTING

C.3.1. AVERAGE MASS OF FOUR PIPETTINGS

The average of 4 pipettings, $m_{\Pi\mathrm{avg}}$ (g), is the arithmetic mean of the masses of deionized water pipetted in 4 repetitions, $m_{\Pi j}$:

$$m_{\Pi\mathrm{avg}} = \frac{m_{\Pi 1} + m_{\Pi 2} + m_{\Pi 3} + m_{\Pi 4}}{4} \tag{29}$$

The Type B uncertainty in $m_{\Pi\mathrm{avg}}$ is

$$u\left(\, m_{\Pi avg}\right) = \frac{\sqrt{u^2(m_{\Pi 1}) + u^2(m_{\Pi 2}) + u^2(m_{\Pi 3}) + u^2(m_{\Pi 4})}}{4} \tag{30}$$

where the uncertainties in $m_{\Pi i}$ are typically ±0.0001 g.

The Type A relative uncertainty of the 4 pipettings due to variability only is

$$u_R(V_\Pi) = \frac{\sqrt{\frac{\sum_{i=1}^{4}(m_{\Pi i} - m_{\Pi\text{avg}})^2}{3}}}{m_{\Pi\text{avg}}} \tag{31}$$

C.3.2. BIAS OF PIPETTE VOLUME

The bias of pipette volume, b_Π (dimensionless), is determined by temperature-correcting the density of water and using the average of 4 pipettings set at V_Π (μL):

$$b_\Pi = \frac{m_{\Pi\text{avg}}\left(1000\,\frac{\mu\text{L}}{\text{mL}}\right)}{V_\Pi\left(1 - \frac{T_A + 288.9414}{508929.2(T_A + 68.12963)}(T_A - 3.9863)^2\right)} - 1 \tag{32}$$

where T_A is the ambient temperature (°C). The denominator term in large parentheses is an expression for the density of water (g mL^{-1}) as a function of temperature (Koech 2015).

C.3.3. RADIOACTIVE DECAY CONSTANT

The radioactive decay constant of isotope *X*, λ_X, is related to the half-life of isotope *X*, $T_{1/2X}$, as a reciprocal with proportionality of the natural log of two:

$$\lambda_X = \frac{\ln 2}{T_{1/2X}} \tag{33}$$

The USTUR uses half-lives in days, and radioactive decay constants in inverse days (day^{-1}).

C.3.4. BIAS-CORRECTED TRACER VOLUME

To simplify equations, the bias-corrected tracer volume for tracer isotope *Y*, $V'_{Y\text{tr}}$ (mL) is introduced:

$$V'_{Y\text{tr}} = \frac{V_{Y\text{tr}}(1 + b_\Pi)}{1000\ \mu\text{L mL}^{-1}}, \text{ and} \qquad u_R(V'_{Y\text{tr}}) = u_R(V_{Y\text{tr}}) \tag{34}$$

C.4. MEASURAND: ACTIVITY OF ISOTOPE *X* ON THE ELEMENT *Z*-FRACTION PLANCHET

For low numbers of counts, MARLAP recommends (p. 19-89) estimating the variance of an observed number of counts *N* as *N*+1, and its standard deviation as $(N+1)^{1/2}$. This recommendation can be traced back to Dodson (1945) at Los Alamos, and cited by numerous authors ever since (Dodson 1945, Friedlander and Kennedy 1949, Thomas

1963, Stevenson 1966, Friedlander et al. 1981). The N+1 recommendation also applies wherever N is used unless other steps have been taken to account for the underestimate of the variance at low numbers of counts (e.g., the Stapleton approximation that appears in Eq. (**2**) and in the *MDA* discussion).

The calculation of activity begins with background and sample counts and count times. These are used to compute count rates,

$$R_\mathrm{B} = \frac{N_\mathrm{B} + 1}{t_\mathrm{B}}, \tag{35}$$

$$R_\mathrm{S} = \frac{N_\mathrm{S} + 1}{t_\mathrm{S}}, \tag{36}$$

$$R_\mathrm{N} = R_\mathrm{S} - R_\mathrm{B} \tag{37}$$

where

R_B	s^{-1}	denotes	the blank or background count rate
N_B	–	denotes	the number of background counts
t_B	s	denotes	the blank (background) count time
R_S	s^{-1}	denotes	the sample (gross) count rate
N_S	–	denotes	the number of sample counts
t_S	s	denotes	the sample (gross) count time in MARLAP notation. For t_S, USTUR uses what Ortec's AlphaVision software reports as "live time," t_L
R_N	s^{-1}	denotes	the net count rate.

The uncertainty and relative uncertainty in R_N are

$$u(R_\mathrm{N}) = \sqrt{\frac{N_S + 1}{t_S^2} + \frac{N_B + 1}{t_B^2}}$$

$$u_\mathrm{R}(R_\mathrm{N}) = \frac{u(R_\mathrm{N})}{R_N} = \frac{\sqrt{\frac{N_S + 1}{t_S^2} + \frac{N_B + 1}{t_B^2}}}{\frac{N_S + 1}{t_S} - \frac{N_B + 1}{t_B}} \tag{38}$$

The "apparent activity" of isotope X (or tracer Y) on the element Z-fraction planchet as of the count date, $A^*_{X,Z,\mathrm{aliq}}(t_{\mathrm{C},Z})$ in Bq is

$$A^{*}_{X,Z,\mathrm{alq}}(t_{\mathrm{C},z}) = \frac{R_N}{\varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}} \tag{39}$$

where

$t_{\mathrm{C,Z}}$	d	denotes	the date/time that the *Z*-fraction planchet is counted (Excel date)
ε	–	denotes	the counting efficiency within a given ROI for a given detector
$f_{X,\mathrm{ROI}}$	–	denotes	the "ROI intensity," which is the sum of intensities of all α-particles emitted by isotope *X* expected to produce counts the ROI
$y_{\mathrm{RR}}(Y)$	–	denotes	the radiochemical recovery yield for the tracer *Y* for isotope *X*. For some calculations, y_{RR} is set to 1 and its uncertainty is set to zero.

If there are no contaminant isotopes present, then the "apparent activity" is simply the activity: $A = A^*$. If there are contaminant isotopes present, as in the case of Am and Pu fractions, then the contributions of these contaminants must be subtracted from A^*.

C.4.1. UNCERTAINTIES OF THE ACTIVITY OF ISOTOPE *X* ON THE ELEMENT *Z*-FRACTION PLANCHET

For large numbers of counts, the relative uncertainty of the activity of isotope *X* on the *Z*-fraction planchet as of the count date, $u_{\mathrm{R}}(A_{X,Z,\mathrm{aliq}})$, is estimated using a Type B uncertainty analysis as

$$u_R(A_{X,Z,\mathrm{alq}}) = \sqrt{u_R^2(R_{\mathrm{N}}) + u_R^2(\varepsilon) + u_R^2(f_{X,\mathrm{ROI}}) + u_R^2(y_{\mathrm{RR}})}$$

so that (40)

$$u(A_{X,Z,\mathrm{alq}}) = A_{X,Z,\mathrm{alq}} u_R(A_{X,Z,\mathrm{alq}})$$

This result is derived in Derivation of the Uncertainty of the Activity of Isotope X on the Element Z-Fraction Planchet.

The use of $N + 1$ in the above relationships gives significantly greater estimates of uncertainty and relative uncertainty at low numbers of counts. Using $N + 1$ corrects for the fact that, at low numbers of counts, the observed number of counts, *N*, is a very uncertain estimator of the mean of the underlying Poisson distribution and an even worse estimator of its variance (Strom and MacLellan 2001). A comparison of the *N* and *N* + 1 formulas is given in (Strom and MacLellan 2001).

The above results are used in

- reporting activity and its uncertainty
- computing two statistics that characterize the radiochemistry measurement system at the USTUR, the lowest usable *action level* of net count rate S_C' and the *MDA*.

Equation (40)) can only be used when *measurements* (that is, counting results) and not *calibration certificates* are the basis for calculating uncertainty. Different equations are needed for uncertainty in activities that are calculated rather than measured. In particular, $A_{\mathrm{A1,Am,A1inA3tr}}(t_{\mathrm{C,Am}})$ and $A_{\mathrm{A1,Am,P1inP2tr}}(t_{\mathrm{C,Am}})$ are calculated activities whose uncertainties need to be assessed separately.

C.5. COUNTING EFFICIENCY

The counting efficiency ε (counts per second per Bq, that is, dimensionless) for each radionuclide *X*'s region-of-interest (ROI) is needed to compute activity.

For tracers ^{243}Am and ^{242}Pu, AlphaVision software provides a download of counting efficiencies and their uncertainties based on archived detector-specific measurements of known standards and blanks, and it includes an estimate of relative uncertainty that is typically less than 1%.

Alternatively, ε also can be calculated from measurements made with each batch of samples of calibration standards containing known activities, decay corrected to the date and time of counting[11]. For a standard (std) source of known activity of isotope *X* for which no radiochemical recovery yield is needed,

$$\varepsilon_X = \frac{R_{\mathrm{N},X.\mathrm{std}}}{A_{X.\mathrm{std}}(t_{\mathrm{ref},X\mathrm{std}})e^{-\lambda_X\left(t_{C,X}-t_{\mathrm{ref},X.\mathrm{std}}\right)}f_{X,\mathrm{ROI}}} \tag{41}$$

where

$R_{\mathrm{N},X\mathrm{std}}$	denote	the source (subscript S) and background (subscript B) counts that are observed at ($t_{\mathrm{C,Z}}$) for isotope *X* in the standard	s^{-1}

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[11] For the next few sections, many extra identifying subscripts are added, because these variables have different values for different isotopes and sources (unknown, standard, blank), and must be distinguished in data reduction and calculations, such as programming in python.

$A_{X,X.\mathrm{std}}(t_{\mathrm{ref},X.\mathrm{std}})$	denotes	the activity of radioisotope X in the standard as of the reference date/time for the standard	Bq mL^{-1}
λ_X	denotes	the radioactive decay constant of radioisotope Y	d^{-1}
$t_{\mathrm{C},X}$	denotes	date/time of counting of isotope X (Excel date)	d
$t_{\mathrm{ref},X\mathrm{std}}$	denotes	reference date/time of standard X (Excel date)	d
$f_{X,\mathrm{ROI}}$	denotes	the fraction of counts from isotope X that will fall within the ROI	-

Note that blank counts must be made on the same detector in the same ROI as the source in every paired measurement of a source and a blank.

The result of Eq. (41) is important for QA.

C.5.1. RELATIVE UNCERTAINTY IN THE MEASURED COUNTING EFFICIENCY

The uncertainty in the measured counting efficiency ε (Eq. (41)) is complex, but full analytical modeling is not needed. Instead, sources of uncertainty that are obviously minor will be treated as precise constants. In Eq. (41), terms treated as constants include λ_X, $t_{\mathrm{C},X}$, $t_{\mathrm{ref},X\mathrm{std}}$, and $f_{X,\mathrm{ROI}}$.

Ignoring the uncertainties in λ_X, $t_{\mathrm{C},X}$, $t_{\mathrm{ref},X\mathrm{std}}$, and $f_{X\mathrm{ROI}}$, the relative uncertainty and uncertainty in ε are

$$u_{\mathrm{R}}(\varepsilon) = \sqrt{u_{\mathrm{R}}^2\left(R_{\mathrm{N},X.\mathrm{std}}\right) + u_{\mathrm{R}}^2\left(A_{X.\mathrm{std}}\left(t_{\mathrm{ref},X.\mathrm{std}}\right)\right)}$$

so that

$$u(\varepsilon) = u_{\mathrm{R}}(\varepsilon)\varepsilon \tag{42}$$

C.6. RADIOCHEMICAL RECOVERY YIELD (FRACTIONAL TRACER RECOVERY)

The radiochemical recovery yield (fractional tracer recovery) for tracer Y (of element Z), $y_{\mathrm{RR}}(Y)$, is calculated for each sample by

$$y_{\mathrm{RR}}(Y) = \frac{\text{Observed Net Count Rate of Tracer } Y}{\text{Decay Corrected Activity of Tracer } Y}$$

$$= \frac{R_{\mathrm{N},Y}}{C_{Y,Y\mathrm{tr}}\left(t_{\mathrm{ref},Y}\right)e^{-\lambda_Y\left(t_{C,Z} - t_{\mathrm{ref},Y}\right)}V'_{Y\mathrm{tr}}\varepsilon_Y f_{Y,\mathrm{ROI}}} \tag{43}$$

where

Z	denotes	the element of which radioisotope Y is a member	-
$C_{Y,Y\text{tr}}(t_{\text{ref},Y})$	denotes	the concentration of radioisotope Y in the Y tracer as of the reference date/time for tracer Y ($t_{\text{ref},Y}$), given by the manufacturer of the tracer solution and found in the Tracer Information tab of the Workbook	Bq mL^{-1}
λ_Y	denotes	the radioactive decay constant of radioisotope Y	d^{-1}
$t_{\text{C,Z}}$	denotes	date/time of counting of elemental fraction Z (Excel date)	d
$t_{\text{ref},Y}$	denotes	reference date/time of tracer Y (Excel date)	d
V'_{Ytr}	denotes	bias-corrected pipetted volume of Y tracer	mL
ε_Y	denotes	counting efficiency (counts per second per Bq, that is, dimensionless) for isotope Y	
$f_{Y,\text{ROI}}$	denotes	the fraction of counts from isotope Y in the ROI	-
$R_{\text{N},Y}$	denotes	the net count rate for isotope Y	

In practice, Eq.(46) is rearranged to give the product $\varepsilon_Y y_{\text{RR}}$ for each tracer Y in terms of observables and standard concentration data from NIST. The $\varepsilon_Y y_{\text{RR}}$ product is assumed to apply to all isotopes of an element, so, for example, the unknown activity of ^{241}Am can be calculated from its net count rate, its $f_{\text{A1,ROI}}$, and the $\varepsilon_Y y_{\text{RR}}$ value observed for ^{243}Am. Using the detector-specific ε_Y values stored by AlphaVision from calibration runs, y_{RR} values are calculated for use in quality assurance.

C.6.1. Relative Uncertainty and Uncertainty in the Radiochemical Recovery Yield

The relative uncertainty in the radiochemical recovery yield (Eq. (43)) is given by

$$u_R\big(y_{\text{RR}}(Y)\big) = \sqrt{u_R^2\big(R_{\text{N},Y}\big) + u_R^2\left(C_{Y,Y\text{tr}}\big(t_{\text{ref},Y}\big)\right)}$$

$$\text{so that} \tag{44}$$

$$u(y_{\text{RR}}(Y)) = y_{\text{RR}}(Y)\, u_R(y_{\text{RR}}(Y))$$

The second term under the radical is provided by the manufacturer of tracer solution Y (given in the Tracer Information tab of the Excel Workbook) combined with USTUR's uncertainties in pipetting and mass measurements.

C.7. Measurand: Activity of ^{241}Am in an Aliquot of Digested Tissue

Measurement of ^{241}Am is complicated by the fact that there are three potential sources of ^{241}Am on a given planchet.

C.7.1. Three Sources of Americium

The purpose of the next three calculations is to account for contributions to the americium fraction from two known sources of small activities of ^{241}Am and use them to correct the gross signal so that the net signal due to ^{241}Am in the sample alone can be calculated.

The “apparent activity” in the Am-ROI, $A^*_{\text{A1-ROI,Am,3sources}}$, on count date $t_{\text{C,Am}}$, is calculated from the observed sum of counts due to 3 sources. The sources are 1) ^{241}Am activity in the Am fraction from the aliquot of the dissolved tissue solution; 2) ^{241}Am activity in the Am fraction from ^{241}Am contaminant in the ^{243}Am tracer; and 3) ^{241}Am activity in the Am fraction from the ^{241}Am that has grown in from the ^{241}Pu contaminant in the ^{242}Pu tracer up until the time of element separation.

$$\begin{aligned} A^*_{\text{A1-ROI,Am,3sources}}(t_{\text{C,Am}}) \\ &= A_{\text{A1,Am,aliq}}(t_{\text{C,Am}}) + A_{\text{A1,Am,A1inA3tr}}(t_{\text{C,Am}}) \\ &+ A_{\text{A1,Am,A1inP2tr}}(t_{\text{sep,Am}}) \end{aligned} \tag{45}$$

C.7.2. ^{241}AM IN THE AM FRACTION FROM ^{241}AM CONTAMINATION IN THE ^{243}AM TRACER

The first known source is the contribution to the total ^{241}Am counts that is due to ^{241}Am contamination in the ^{243}Am tracer solution. The “activity of ^{241}Am in Am fraction from ^{243}Am tracer as of count date t_{C},” $A_{\text{A1,Am,A1inA3tr}}(t_{\text{C,Am}})$ (Bq), is the product of

- the “concentration of ^{241}Am in the tracer as of the tracer reference date,” $C_{\text{A1,A3tr}}(t_{\text{ref,A3}})$ (Bq)
- an exponential decay factor for ($t_{\text{C,Am}}$ - $t_{\text{ref,A3}}$), the time interval (days) between the midpoint of the americium fraction counting date/time $t_{\text{C,Am}}$ and reference date/time for the tracer $t_{\text{ref,A3}}$; and
- the volume of tracer pipetted corrected for pipetting bias and converted to mL, V'_{A3tr}.

$$A_{\text{A1,Am,A1inA3tr}}(t_{\text{C,Am}}) = C_{\text{A1,A3tr}}(t_{\text{ref,A3}})e^{-\lambda_{\text{A1}}(t_{\text{C,Am}}-t_{\text{ref,A3}})}V'_{\text{A3tr}} \tag{46}$$

The relative uncertainty of concentration of the ^{241}Am contaminant in the ^{243}Am tracer, $u_{\text{R}}(C_{\text{A1,A3tr}}(t_{\text{ref},x}))$, is known from its calibration certificate. The u_{R} of the tracer volume that was pipetted is the same as the u_{R} of the pipetting. The u_{R} of the decay correction factor depends on the time difference between counting and reference times, as well as the uncertainty in λ_{A1}, which can be evaluated using IAEA MassChains data (International Atomic Energy Agency (IAEA) 2019), $T_{1/2,\text{A1}}$ = 432.6±0.6 y, so u_{R} = 0.6/432.6 = 0.0014.

As derived in Derivation of the Uncertainty of the Concentration of the $^{241\text{Am}}$ Contaminant in the $^{243\text{Am}}$ Tracer, the uncertainty of $A_{\text{A1,Am,A1inA3tr}}$ is

$$u(A_{\text{A1,Am,A1inA3tr}}) = u_R(A_{\text{A1,Am,A1inA3tr}})A_{\text{A1,Am,A1inA3tr}} \tag{47}$$

C.7.3. ^{241}AM IN AM FRACTION FROM DECAY OF ^{241}PU IN ^{242}PU TRACER PRIOR TO ELEMENT SEPARATION

The activity of ^{241}Am in (Bq) in the Am fraction from decay of ^{241}Pu in the ^{242}Pu tracer as of count date/time for the Am fraction $t_{\mathrm{C,Am}}$,

$$A_{\mathrm{A1,Am,P1inP2tr}}(t_{\mathrm{C,Am}}) = C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}})V'_{\mathrm{P2tr}}\frac{\lambda_{\mathrm{A1}}}{\lambda_{\mathrm{A1}}-\lambda_{\mathrm{P1}}}\left(e^{-\lambda_{\mathrm{P1}}(t_{\mathrm{sep}}-t_{\mathrm{ref,P2}})} - e^{-\lambda_{\mathrm{A1}}(t_{\mathrm{sep}}-t_{\mathrm{ref,P2}})}\right)e^{-\lambda_{\mathrm{A1}}(t_{\mathrm{C,Am}}-t_{\mathrm{sep}})} \tag{48}$$

is the product of 5 terms:

- the concentration of ^{241}Pu in the ^{242}Pu tracer solution $C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}})$ on the reference date $t_{\mathrm{ref,P2}}$,
- the ^{242}Pu tracer volume corrected for bias and converted to mL, V'_{P2tr}
- the decay constant ratio from the Bateman equation for ^{241}Am and ^{241}Pu
- the difference of the fractional ingrowth of ^{241}Am from ^{241}Pu and the fractional decay of ^{241}Am during the interval between the separation date for the element fractions t_{sep} and the reference date for the ^{242}Pu standard, $t_{\mathrm{ref,P2}}$
- the decay of ^{241}Am between the separation date of the element fractions t_{sep} and the counting date for the Am fraction, $t_{\mathrm{C,Am}}$ (days); no ingrowth occurs during the interval between t_{sep} and $t_{\mathrm{C,Am}}$ because there is no ^{241}Pu present

The uncertainty in $A_{\mathrm{A1,Am,P1inPutr}}$ is dominated by the uncertainty in $C_{\mathrm{P1,P2tracer}}(t_{\mathrm{ref,P2}})$, and uncertainty contributions from the other terms are ignored.

$$u_R(A_{\mathrm{A1,Am,P1inP2tr}}(t_{\mathrm{C,Am}})) \approx u_R(C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}})) = \frac{u(C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}}))}{C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}})} \tag{49}$$

In the case of tracer UTR0039, $u_R(C_{\mathrm{P1,P2tr}}) = 0.098$ (see Example Calibration Certificates for Standards and Tracers).

C.7.4. ^{241}AM ACTIVITY IN THE AM FRACTION DUE TO ^{241}AM IN THE ALIQUOT OF THE DISSOLVED TISSUE SOLUTION

The ^{241}Am activity from the aliquot of the dissolved tissue solution is calculated by

$$A_{\mathrm{A1,Am,aliq}}(t_{\mathrm{C,Am}}) = A^{*}_{\mathrm{Am\text{-}ROI,Am,3sources}}(t_{\mathrm{C,Am}}) - A_{\mathrm{A1,Am,A1inA3tr}}(t_{\mathrm{C,Am}}) - A_{\mathrm{A1,Am,A1inP2tr}}(t_{\mathrm{sep,Am}}) \tag{50}$$

C.7.5. RELATIVE UNCERTAINTY OF ^{241}AM ACTIVITY

The uncertainty[12] in the activity of ^{241}Am on the planchet at the count date, corrected for ^{241}Am contamination, is

$$u(A_{\text{A1,Am,aliq}}) = \sqrt{u^2(A_{\text{Am-ROI,Am,3sources}}) + u^2(A_{\text{A1,Am,A1inA3tr}}) + u^2(A_{\text{A1,Am,A1inP2tr}})} \tag{51}$$

where

$$\begin{aligned} u^2(A_{\text{Am-ROI,Am,3sources}}) &= A^2_{\text{Am-ROI,Am,3sources}}(u_R^2(R_{\text{N}}) + u_R^2(\varepsilon)), \\ u^2(A_{\text{A1,Am,A1inA3tr}}) &= A^2_{\text{A1,Am,A1inA3tr}} u_R^2(A_{\text{A1,Am,A1inA3tr}}), \text{ and} \\ u^2(A_{\text{A1,Am,A1inP2tr}}) &= A^2_{\text{A1,Am,A1inP2tr}} u_R^2(A_{\text{A1,Am,A1inP2tr}}). \end{aligned} \tag{52}$$

In the case of tracer UTR0038, $u_{\text{R}}(A_{\text{A1,Am,A1inA3tr}}) = u_{\text{R}}(C_{\text{A1,A3tr}}) = 0.333$ (see Example Calibration Certificates for Standards and Tracers). In the case of tracer UTR0039, the relative uncertainty of ^{241}Am in the ^{242}Pu tracer is the same as the u_{R} of its parent, ^{241}Pu: $u_{\text{R}}(A_{\text{A1,Am,A1inP2tr}}) = u_{\text{R}}(C_{\text{P1,P2tr}}) = 0.098$ (see Example Calibration Certificates for Standards and Tracers).

C.8. MEASURAND: ACTIVITY OF ^{238}PU IN AN ALIQUOT OF DIGESTED TISSUE

As in the case of Am, there are three sources of counts in the ^{238}Pu + ^{241}Am region of interest (ROI) in the α-particle energy spectrum.

C.8.1. THREE SOURCES OF COUNTS IN THE ^{238}PU + ^{241}AM ROI

The apparent activity in the P8+A1-ROI, $A^*_{\text{P8+A1-ROI,Pu,3sources}}$, on count date $t_{\text{C,Pu}}$, is calculated from the observed sum of counts due to 3 sources. The sources are 1) ^{238}Pu activity in the Pu fraction from the aliquot of the dissolved tissue solution; 2) ^{241}Am activity in the Pu fraction that has grown in since separation of elements, arising from the ^{241}Pu contaminant in the ^{242}Pu tracer; and 3) ^{241}Am activity in the Pu fraction that has grown in since separation of elements, arising from ^{241}Pu in the aliquot of the dissolved tissue solution. The “apparent activity” (“uncorrected activity”) calculated from the counts in the α-spectrometry ROI associated with ^{238}Pu and ^{241}Am is the sum of these 3 contributions:

$$A^*_{\text{P8+A1-ROI,Pu,3sources}}(t_{\text{C,Pu}}) = A_{\text{P8,Pu,aliq}}(t_{\text{C,Pu}}) + A_{\text{A1,Pu,P1.in.aliq}}(t_{\text{C,Pu}}) + A_{\text{A1,Pu,P1inP2tr}}(t_{\text{C,Pu}}). \tag{53}$$

C.8.2. ^{241}AM IN THE PU FRACTION FROM DECAY OF ^{241}PU IN THE ^{242}PU TRACER

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[12] Note: When the uncertain quantity is a sum, one must add variances (as done correctly above) and not simple squares of u_{R}s.

The activity of ^{241}Am in the Pu fraction from the decay of ^{241}Pu in the Pu tracer as of count date $t_{\mathrm{C,Pu}}$, $A_{\mathrm{A1,Pu,P1inP2tr}}(t_{\mathrm{C,Pu}})$ (Bq),

$$\begin{aligned} A_{\mathrm{A1,Pu,P1inP2tr}}(t_{\mathrm{C,Pu}}) \\ &= C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}})V'_{\mathrm{P2tr}}\frac{\lambda_{\mathrm{A1}}}{\lambda_{\mathrm{A1}}-\lambda_{\mathrm{P1}}}\left(e^{-\lambda_{\mathrm{P1}}(t_{\mathrm{C,Pu}}-t_{\mathrm{sep}})}\right. \\ &\left.- e^{-\lambda_{\mathrm{A1}}(t_{\mathrm{C,Pu}}-t_{\mathrm{sep}})}\right)e^{-\lambda_{\mathrm{P1}}(t_{\mathrm{sep}}-t_{\mathrm{ref,P2}})} \end{aligned} \tag{54}$$

is the product of 5 terms:

- the concentration of ^{241}Pu in the ^{242}Pu tracer solution, $C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}})$, on the reference date, $t_{\mathrm{ref,P2}}$
- the ^{242}Pu tracer volume corrected for bias and converted to mL, V'_{P2tr}
- the decay constant ratio from the Bateman equation for ^{241}Am and ^{241}Pu
- the difference of the fractional ingrowth of ^{241}Am from ^{241}Pu and the fractional decay of ^{241}Am during the interval between the separation date of the element fractions, t_{sep}, and the counting date, $t_{\mathrm{C,Pu}}$
- the decay of ^{241}Pu between the separation date of the element fractions, t_{sep}, and the reference date for the ^{242}Pu standard, $t_{\mathrm{ref,P2}}$

Note that if ($t_{\mathrm{C,Pu}}$ – t_{sep}) is small, that is, counting is done shortly after separation of elements, the ^{241}Am activity due to this source is negligible.

The uncertainty in $A_{\mathrm{A1,Pu,P1inPutr}}(t_{\mathrm{ref,P2}})$ is dominated by the uncertainty in $C_{\mathrm{P1,P2tracer}}(t_{\mathrm{ref,P2}})$, as above.

$$u_R(A_{\mathrm{A1,Pu,P1inP2tr}}(t_{\mathrm{C,Pu}})) \approx u_R(C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}})) = \frac{u(C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}}))}{C_{\mathrm{P1,P2tr}}(t_{\mathrm{ref,P2}})} \tag{55}$$

In the case of tracer UTR0039, $u_{\mathrm{R}}(C_{\mathrm{P1,P2tr}})$ = 0.098 (see Example Calibration Certificates for Standards and Tracers).

C.8.3. ^{241}AM ACTIVITY IN THE PU FRACTION DUE TO INGROWTH FROM ^{241}PU IN THE ALIQUOT OF THE DISSOLVED TISSUE SOLUTION BETWEEN T_{SEP} AND $T_{\mathrm{C,PU}}$

In principle, the activity of ^{241}Am in the Pu fraction at t_{sep},

$$\begin{aligned} A_{\mathrm{A1,Pu,P1aliq}}(t_{\mathrm{C,Pu}}) = \ &\text{ingrowth of }^{241}\text{Am in Pu fraction from }^{241}\text{Pu} \\ &\text{in the sample between } t_{\mathrm{sep}} \text{ and } t_{\mathrm{C,Pu}}, \end{aligned} \tag{56}$$

could be estimated from measurement of $^{239+240}$Pu if ^{241}Pu if fraction in the material is known. Otherwise, the activity of ^{241}Am in the Pu fraction at t_{sep} is unknown. However, if ($t_{\mathrm{C,Pu}}$ – t_{sep}) is small, that is, counting is done shortly after separation of elements, the ^{241}Am activity due to this source is negligible.

C.8.4. ^{238}Pu Activity in the Pu Fraction due to ^{238}Pu in the Aliquot of the dissolved tissue solution

The quantity of interest for USTUR studies is the activity of ^{238}Pu from the aliquot of the dissolved tissue solution,

$$\begin{aligned} A_{\text{P8,Pu,aliq}}(t_{\text{C,Pu}} = A^{*}_{\text{P8+A1-ROI,Pu,3sources}}(t_{\text{C,Pu}}) - A_{\text{A1,Pu,P1.in.aliq}}(t_{\text{C,Pu}}) \\ - A_{\text{A1,Pu,P1inP2tr}}(t_{\text{C,Pu}}). \end{aligned} \tag{57}$$

The activity of ^{241}Am in the Pu fraction at t_{sep} is the decay product of the ^{241}Pu contaminant in the ^{242}Pu tracer. This activity is given in Eq. (54) and its relative uncertainty is given in Eq. (55).

If values for both of these quantities can be determined, then the activity of ^{238}Pu at $t_{\text{C,Pu}}$ can be calculated.

If the isotopic mix is not known, an excellent approximation for $A_{\text{P8,Pu,aliq}}(t_{\text{C,Pu}})$ can be made if the ingrowth of ^{241}Am on the Pu fraction planchet is minimized by counting as soon as possible after electroplating. This is the usual practice at the USTUR.

C.8.5. Relative Uncertainty of Apparent ^{238}Pu Activity

The relative uncertainty in the apparent activity in the P8+A1 ROI is given by Eq. (40), here with the Source and Blank counts and times used for Pu-238 counting:

$$u_R(A^{*}_{\text{P8+A1-ROI,Pu,3sources}}) = \sqrt{u_R^2(R_{\text{N,P8+A1−ROI}}) + u_R^2(\varepsilon)} \tag{58}$$

$$\begin{aligned} &u_R(A_{\text{P8,Pu,aliq}}(t_{\text{C,Pu}})) \\ &= \frac{\begin{pmatrix} u_R^2(A^{*}_{\text{P8+A1-ROI,Pu,3sources}}(t_{\text{C,Pu}}))A^{*2}_{\text{P8+A1-ROI,Pu,3sources}} \\ +u_R^2(A_{\text{A1,Pu,P1inaliq}}(t_{\text{C,Pu}}))A^{2}_{\text{A1,Pu,P1inaliq}} \\ +u_R^2(A_{\text{A1,Pu,P1inP2tr}}(t_{\text{C,Pu}}))A^{2}_{\text{A1,Pu,P1inP2tr}} \end{pmatrix}^{1/2}}{A_{\text{P8,Pu,aliq}}(t_{\text{C,Pu}})}, \text{so} \\ &u(A_{\text{P8,Pu,aliq}}(t_{\text{C,Pu}})) = \begin{pmatrix} u_R^2(A^{*}_{\text{P8+A1-ROI,Pu,3sources}}(t_{\text{C,Pu}}))A^{*2}_{\text{P8+A1-ROI,Pu,3sources}} \\ +u_R^2(A_{\text{A1,Pu,P1inaliq}}(t_{\text{C,Pu}}))A^{2}_{\text{A1,Pu,P1inaliq}} \\ +u_R^2(A_{\text{A1,Pu,P1inP2tr}}(t_{\text{C,Pu}}))A^{2}_{\text{A1,Pu,P1inP2tr}} \end{pmatrix}^{1/2}. \end{aligned} \tag{59}$$

C.9. Activity of Isotope *X* in the Dissolved Tissue Solution and Its Relative Uncertainty

The activity (Bq) of isotope *X* in the dissolved tissue solution at the count date for the element *Z* planchet is

$$A_{X,Z,\mathrm{soln}}(t_{\mathrm{C},Z}) = \frac{A_{X,\mathrm{Z,aliq}}(t_{\mathrm{C},Z})}{y_{\mathrm{RR}}(Y)}\frac{m_{\mathrm{soln}}}{m_{\mathrm{aliq}}}, \tag{60}$$

where

$A_{X,\mathrm{soln,prep}}(t_{\mathrm{C},Z})$	denotes	the activity of radioisotope X in the dissolved tissue solution on $t_{\mathrm{C},Z}$, the counting date/time for element Z	Bq
$A_{X,\mathrm{aliq,prep}}(t_{\mathrm{C},Z})$	denotes	the activity of radioisotope X in the aliquot of dissolved tissue solution on $t_{\mathrm{C},Z}$, the counting date/time for element Z	Bq
$y_{\mathrm{RR}}(Y)$	denotes	the radiochemical recovery yield for tracer Y	-
m_{soln}	denotes	the mass of the dissolved tissue solution	g
m_{aliq}	denotes	the mass of the aliquot	g

Note that $A_{X,Z,\mathrm{soln}} = A_{X,Z,\mathrm{prep}}$ under the assumption of no loss of isotope X between beginning of processing the prepared sample and its final form as an acid solution.

The relative uncertainty of the activity of isotope X in the dissolved tissue solution at count date, $u_{\mathrm{R}}(A_{X,Z,\mathrm{soln}}(t_{\mathrm{C}.Z}))$, is

$$u_{\mathrm{R}}(A_{X,Z,\mathrm{soln}}(t_{\mathrm{C},Z})) = \sqrt{u_{\mathrm{R}}^2\left(A_{X,Z,\mathrm{aliq}}(t_{\mathrm{C},Z})\right) + u_{\mathrm{R}}^2(y_{\mathrm{RR}}(Y)) + u_{\mathrm{R}}^2(m_{\mathrm{soln}}) + u_{\mathrm{R}}^2(m_{\mathrm{aliq}})}. \tag{61}$$

The activity of isotope X in the total volume of solution decay-corrected from the date of counting to the date of the tissue donation is

$$A_{X,Z,\mathrm{soln}}(t_{\mathrm{D}}) = A_{X,Z,\mathrm{soln}}(t_{\mathrm{C},Z})e^{-\lambda_X(t_{\mathrm{C},Z}-t_D)}. \tag{62}$$

The relative uncertainty of the activity of isotope X in the dissolved tissue solution at the donation date $u_{\mathrm{R}}(A_{X,Z.\mathrm{soln}}(t_{\mathrm{D}}))$ is the same as the relative uncertainty of the activity of isotope X in the dissolved tissue solution at the count date $u_{\mathrm{R}}(A_{X,Z,\mathrm{soln}}(t_{\mathrm{C},Z}))$,

$$u_{\mathrm{R}}(A_{X,Z,\mathrm{soln}}(t_{\mathrm{D}})) = u_{\mathrm{R}}(A_{X,Z,\mathrm{soln}}(t_{\mathrm{C},Z})). \tag{63}$$

C.10. Uncertainty of the Activity of Isotope *X* in the Initial Mass

In general, the activity in the initial mass is estimated by scaling the activity measured on a planchet, as shown in this rearrangement of Eq. (17):

$$A_{\mathrm{init}} = A_{\mathrm{planchet}}\,\frac{m_{\mathrm{init}}}{m_{\mathrm{prep}}}\,\frac{m_{\mathrm{soln}}}{m_{\mathrm{aliq}}}\,\frac{1}{y_{\mathrm{RR}}}. \tag{64}$$

The scaling factors are

- the ratio of initial mass to prepared mass

- the ratio of the mass of the entire acid solution to the mass of the aliquot that is used in isotope separation
- the reciprocal of the radiochemical recovery yield.

Uncertainty in each of these measured factors contributes to having greater uncertainty in A_{init} than in A_{planchet}.

If $m_{\mathrm{a}} \neq 0$ and $m_{\mathrm{b}} \neq 0$, and ignoring covariance terms that are included in MARLAP formulas (MARLAP Tables 19.1 and 19.2), the combined uncertainty in the ratio of two masses, $m_{\mathrm{a}}/m_{\mathrm{b}}$, is

$$u_{\square}^{2}(m_{\mathrm{a}}/m_{\mathrm{b}}) = \frac{m_{\mathrm{a}}^{2}}{m_{\mathrm{b}}^{2}}\left[\frac{u^{2}(m_{\mathrm{a}})}{m_{\mathrm{a}}^{2}} + \frac{u^{2}(m_{\mathrm{b}})}{m_{\mathrm{b}}^{2}}\right]. \tag{65}$$

The variance (that is, square of the uncertainty) of A_{init} is

$$\begin{gathered} u^{2}(A_{\mathrm{init}}) = \left[\frac{u^{2}(A_{\mathrm{planchet}})}{A_{\mathrm{planchet}}^{2}} + \frac{u^{2}(m_{\mathrm{init}}/m_{\mathrm{prep}})}{m_{\mathrm{init}}^{2}/m_{\mathrm{prep}}^{2}} + \frac{u^{2}(m_{\mathrm{soln}}/m_{\mathrm{aliq}})}{m_{\mathrm{soln}}^{2}/m_{\mathrm{aliq}}^{2}} + \right. \\ \left. \frac{u^{2}(y_{\mathrm{RR}})}{y_{\mathrm{RR}}^{2}}\right]\left(A_{\mathrm{planchet}}\ \frac{m_{\mathrm{init}}}{m_{\mathrm{prep}}}\ \frac{m_{\mathrm{soln}}}{m_{\mathrm{aliq}}}\ \frac{1}{y_{\mathrm{RR}}}\right)^{2}. \end{gathered} \tag{66}$$

Substituting Eq. (65) into Eq. (66) for the two mass ratios,

$$\begin{gathered} u^{2}(A_{\mathrm{init}}) = \left[\frac{u^{2}(A_{\mathrm{planchet}})}{A_{\mathrm{planchet}}^{2}} + \left\{\frac{u^{2}(m_{\mathrm{init}})}{m_{\mathrm{init}}^{2}} + \frac{u^{2}(m_{\mathrm{prep}})}{m_{\mathrm{prep}}^{2}}\right\} + \left\{\frac{u^{2}(m_{\mathrm{soln}})}{m_{\mathrm{soln}}^{2}} + \frac{u^{2}(m_{\mathrm{aliq}})}{m_{\mathrm{aliq}}^{2}}\right\} + \right. \\ \left. \frac{u^{2}(y_{\mathrm{RR}})}{y_{\mathrm{RR}}^{2}}\right]\left(A_{\mathrm{planchet}}\ \frac{m_{\mathrm{init}}}{m_{\mathrm{prep}}}\ \frac{m_{\mathrm{soln}}}{m_{\mathrm{aliq}}}\ \frac{1}{y_{\mathrm{RR}}}\right)^{2}. \end{gathered} \tag{67}$$

The relative variance is

$$u_{\mathrm{R}}^{2}(A_{\mathrm{init}}) = \frac{u^{2}(A_{\mathrm{planchet}})}{A_{\mathrm{planchet}}^{2}} + \frac{u^{2}(m_{\mathrm{init}})}{m_{\mathrm{init}}^{2}} + \frac{u^{2}(m_{\mathrm{prep}})}{m_{\mathrm{prep}}^{2}} + \frac{u^{2}(m_{\mathrm{soln}})}{m_{\mathrm{soln}}^{2}} + \frac{u^{2}(m_{\mathrm{aliq}})}{m_{\mathrm{aliq}}^{2}} + \frac{u^{2}(y_{\mathrm{RR}})}{y_{\mathrm{RR}}^{2}}.$$

In many but not all cases, the uncertainties in the mass measurements are negligible.

C.11. DERIVATION OF THE UNCERTAINTY OF THE ACTIVITY OF ISOTOPE *X* ON THE ELEMENT *Z*-FRACTION PLANCHET

If $y = f(x_1, x_2, \ldots, x_N)$, the combined standard uncertainty is calculated from the Taylor series expansion[13] (MARLAP Eq. 19.11; GUM 2008):

---

[13] Equation (68) is probably accurate enough for USTUR counting applications. This equation starts to significantly lose accuracy for u_{R}s > 0.2! It's just the first term of a

$$u_c^2(y) = \sum_{i=1}^{N} \left(\frac{\partial f}{\partial x_i}\right)^2 u^2(x_i) + 2\sum_{i=1}^{N-1} \sum_{j=i+1}^{N} \frac{\partial f}{\partial x_i}\frac{\partial f}{\partial x_j} u(x_i, x_j) \tag{68}$$
$$= \text{ Sum of variance terms and covariance terms}$$

We assume the covariance terms are negligible. In the following expression, t_S, t_B, and $f_{X,\mathrm{ROI}}$, are considered arbitrarily precise, so they are treated as constants.

Substituting Eq. (35) for f in the equation above,

$$f = \frac{\left(\frac{N_S+1}{t_S} - \frac{N_B+1}{t_B}\right)}{\varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}} = \frac{R_S - R_B}{\varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}} = \frac{R_N}{\varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}} \tag{69}$$

one can calculate the partial derivatives (using the methods of MARLAP Table 19.1) for the five variables that contribute to uncertainty:

$$\begin{gathered}
\frac{\partial f}{\partial N_S} = \frac{1}{t_S \varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}} \\
\frac{\partial f}{\partial N_B} = \frac{1}{t_B \varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}} \\
\frac{\partial f}{\partial \varepsilon} = \frac{-1}{\varepsilon^2} \frac{\left(\frac{N_S+1}{t_S} - \frac{N_B+1}{t_B}\right)}{f_{X,\mathrm{ROI}} y_{\mathrm{RR}}} \\
\frac{\partial f}{\partial f_{X,\mathrm{ROI}}} = \frac{-1}{{f_{X,\mathrm{ROI}}}^2} \frac{\left(\frac{N_S+1}{t_S} - \frac{N_B+1}{t_B}\right)}{\varepsilon y_{\mathrm{RR}}} \\
\frac{\partial f}{\partial y_{\mathrm{RR}}} = \frac{-1}{{y_{\mathrm{RR}}}^2} \frac{\left(\frac{N_S+1}{t_S} - \frac{N_B+1}{t_B}\right)}{\varepsilon f_{X,\mathrm{ROI}}}
\end{gathered} \tag{70}$$

the variances of the two Poisson variates, N_S and N_B, are simply the values themselves since the variance of a Poisson distribution is equal to its mean. For observations at low numbers of counts, a better estimate of the variance is the mean + 1:

$u^2(N_S) = N_S + 1$
$u^2(N_B) = N_B + 1$

The variance of the counting efficiency ε is

$u^2(\varepsilon) = (\varepsilon u_R(\varepsilon))^2$;

Taylor series expansion. More difficult uncertainty problems, including those with very large u_Rs, must be solved by Monte Carlo simulations.

the variance of $f_{\mathrm{X,ROI}}$ is

$u^2(f_{\mathrm{X,ROI}}) = (f_{\mathrm{X,ROI}}\ u_{\mathrm{R}}(f_{\mathrm{X,ROI}}))^2$;

and the variance of y_{RR} is

$u^2(y_{\mathrm{RR}}) = (y_{\mathrm{RR}}\ u_{\mathrm{R}}(y_{\mathrm{RR}}))^2$,

where $u_{\mathrm{R}}(*)$ is the relative uncertainty (coefficient of variation) of *.

Using the five partial derivatives and the five variances, the summation in Eq. (68) for the square of the combined standard uncertainty, $u_{\mathrm{C}}{}^2(A_{\mathrm{AX}})$, can be performed. The terms in the top line of the next equation are in the order of (partial derivative squared)(variance) + (partial derivative squared)(variance) + (partial derivative squared)(variance), from the formula above for $u_{\mathrm{C}}{}^2$:

$$
\begin{aligned}
u_{\mathrm{C}}^2(A_{X,Z}) &= \left(\frac{1}{t_{\mathrm{S}}\varepsilon f_{X,\mathrm{ROI}}y_{\mathrm{RR}}}\right)^2 N_{\mathrm{S}} + \left(\frac{1}{t_{\mathrm{B}}\varepsilon f_{X,\mathrm{ROI}}y_{\mathrm{RR}}}\right)^2 N_{\mathrm{B}} \\
&\quad + \left(\frac{-1}{\varepsilon^2}\frac{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}}-\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}}\right)}{f_{X,\mathrm{ROI}}y_{\mathrm{RR}}}\right)^2 (\varepsilon u_{\mathrm{R}}(\varepsilon))^2 \\
&\quad + \left(\frac{-1}{{f_{X,\mathrm{ROI}}}^2}\frac{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}}-\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}}\right)}{\varepsilon y_{\mathrm{RR}}}\right)^2 \left(f_{X,\mathrm{ROI}}u_{\mathrm{R}}(f_{X,\mathrm{ROI}})\right)^2 \\
&\quad + \left(\frac{-1}{{y_{\mathrm{RR}}}^2}\frac{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}}-\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}}\right)}{\varepsilon f_{X,\mathrm{ROI}}}\right)^2 (y_{\mathrm{RR}}u_{\mathrm{R}}(y_{\mathrm{RR}}))^2 \\
&= \left(\frac{1}{\varepsilon f_{X,\mathrm{ROI}}y_{\mathrm{RR}}}\right)^2 \left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}^2}+\frac{N_{\mathrm{B}}+1}{t_B^2}\right) \\
&\quad + \left(\frac{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}}-\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}}\right)}{\varepsilon f_{X,\mathrm{ROI}}y_{\mathrm{RR}}}\right)^2 \left(u_{\mathrm{R}}^2(\varepsilon)+u_{\mathrm{R}}^2(f_{X,\mathrm{ROI}})+u_{\mathrm{R}}^2(y_{\mathrm{RR}})\right)
\end{aligned}
\tag{71}
$$

The square of the relative uncertainty, $u_{\mathrm{R}}{}^{2}(A_{\mathrm{A}X})$ is

$$u_{\mathrm{R}}^{2}(A_{AX}) = \frac{u_{\mathrm{C}}^{2}(A_{X,Z})}{A_{X,Z}^{2}}$$

$$= \frac{\left(\frac{1}{\varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}}\right)^{2}\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}^{2}}+\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}^{2}}\right)+\left(\frac{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}}-\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}}\right)}{\varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}}\right)^{2}\left(u_{\mathrm{R}}^{2}(\varepsilon)+u_{\mathrm{R}}^{2}(f_{X,\mathrm{ROI}})+u_{\mathrm{R}}^{2}(y_{\mathrm{RR}})\right)}{\left[\frac{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}}-\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}}\right)}{\varepsilon f_{X,\mathrm{ROI}} y_{\mathrm{RR}}}\right]^{2}} \qquad (72)$$

$$= \frac{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}^{2}}+\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}^{2}}\right)}{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}}-\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}}\right)^{2}}+u_{\mathrm{R}}^{2}(\varepsilon)+u_{\mathrm{R}}^{2}(f_{X,\mathrm{ROI}})+u_{\mathrm{R}}^{2}(y_{\mathrm{RR}})$$

Taking the positive square root of the last group in Eq. (72)) above, the relative uncertainty of the activity of isotope X on the Z-fraction planchet as of the count date, $u_{\mathrm{R}}(A_{X,Z})$, is

$$u_{\mathrm{R}}(A_{X,Z}) = \frac{u(A_{X,Z})}{A_{X,Z}} = \sqrt{\frac{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}^{2}}+\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}^{2}}\right)}{\left(\frac{N_{\mathrm{S}}+1}{t_{\mathrm{S}}}-\frac{N_{\mathrm{B}}+1}{t_{\mathrm{B}}}\right)^{2}}+u_{\mathrm{R}}^{2}(\varepsilon)+u_{\mathrm{R}}^{2}(f_{X,\mathrm{ROI}})+u_{\mathrm{R}}^{2}(y_{\mathrm{RR}})} \qquad (73)$$

C.12. Derivation of the Uncertainty of the Concentration of the ^{241}Am Contaminant in the ^{243}Am Tracer

The relative uncertainty of concentration of the ^{241}Am contaminant in the ^{243}Am tracer, $u_{\mathrm{R}}(C_{\mathrm{A1,Ytr}}(t_{\mathrm{ref},X}))$, is known from its calibration certificate. The u_{R} of the tracer volume that was pipetted is the same as the u_{R} of the pipetting. The u_{R} of the decay correction factor depends on the time difference between counting and reference times, as well as the uncertainty in λ_{A1}, which can be evaluated using IAEA MassChains data, $T_{1/2,\mathrm{A1}}$ = 432.6±0.6 y, so u_{R} = 0.6/432.6 = 0.0014.

The u_{R} of the exponential term requires knowing its partial derivative with respect to $T_{1/2,\mathrm{A1}}$ (according to the formulas in MARLAP Table 19.1) and its uncertainty as a function of the uncertainty in $T_{1/2,\mathrm{A1}}$. Setting $k = \ln(2)\ (t_{\mathrm{C,Am}} - t_{\mathrm{ref,A3}})$,

$$\frac{\partial\left(e^{-k/T_{1/2,\mathrm{A1}}}\right)}{\partial T_{1/2,\mathrm{A1}}} = \frac{+k\left(e^{-k/T_{1/2,\mathrm{A1}}}\right)}{T_{1/2,\mathrm{A1}}^{2}}. \qquad (74)$$

The partial derivative of the exponential expression with respect to the half-life of ^{241}Am,

$$\frac{\partial(e^{-(ln(2)/T_{1/2,\mathrm{A1}})(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})})}{\partial T_{1/2,\mathrm{A1}}} = \frac{+\,ln(2)(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})(e^{-(ln(2)/T_{1/2,\mathrm{A1}})(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})})}{T^2_{1/2,\mathrm{A1}}},$$

so the square of the partial derivative is (75)

$$\left(\frac{\partial(e^{-(ln(2)/T_{1/2,\mathrm{A1}})(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})})}{\partial T_{1/2,\mathrm{A1}}}\right)^2 = \frac{+\,ln^2(2)(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})^2(e^{-(ln(2)/T_{1/2,\mathrm{A1}})(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})})^2}{T^4_{1/2,\mathrm{A1}}}.$$

Applying MARLAP Eq. 19.11,

$$\begin{aligned} u_C^2(e^{-(ln(2)/T_{1/2,\mathrm{A1}})(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})}) &= \left(\frac{\partial(e^{-(ln(2)/T_{1/2,\mathrm{A1}})(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})})}{\partial T_{1/2,\mathrm{A1}}}\right)^2 u^2(T_{1/2,\mathrm{A1}}) \\ &= \frac{+\,ln^2(2)(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})^2(e^{-(ln(2)/T_{1/2,\mathrm{A1}})(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})})^2}{T^4_{1/2,\mathrm{A1}}} u^2(T_{1/2,\mathrm{A1}}). \end{aligned} \tag{76}$$

Converting IAEA MassChains values to days, $T_{1/2,\mathrm{A1}}$ = 432.6 y = 158007 d; $u(T_{1/2,\mathrm{A1}})$ = 0.6 y = 219 d. As an example, consider $t_{\mathrm{C,Am}}$ = 2018 May 3 (2018-05-03) and $t_{\mathrm{ref,A3}}$ = 2005 October 20 (2005-10-20), and $\Delta t \equiv (t_{\mathrm{C,Am}} - t_{\mathrm{ref,A3}})$ = 4578 d.

$$\begin{aligned} &u_C^2(e^{-(ln(2)/T_{1/2,\mathrm{A1}})(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})}) \\ &\quad = (0.693)^2(4578)^2\left(e^{-(0.693/158007)(4578)}\right)^2\left(\frac{1}{158007^4}\right)(219)^2 \\ &= 7.45306\text{E-}10\text{, so} \\ &u_C(e^{-(ln(2)/T_{1/2,\mathrm{A1}})(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})}) = 2.730\text{E-}05 \end{aligned} \tag{77}$$

The value of the exponential term is 0.980117±0.000027, for a u_R of 2.8E-5. In the example above, this u_R of less than 3 parts in 100,000 is negligible and will not affect uncertainties. Finally, calculating the u_R for the activity of contaminant ^{241}Am from the ^{243}Am tracer solution at the time of counting,

$$\begin{aligned} u_R(A_{\mathrm{A1,Am,A1inA3tr}}(t_{\mathrm{C,Am}})) &= \sqrt{u_R^2(C_{\mathrm{A1,A3tr}}) + u_R^2(V'_{\mathrm{A3tr}}) + u_R^2(e^{-\lambda_{\mathrm{A1}}(t_{\mathrm{C,Am}}-t_{\mathrm{ref,A3}})})} \\ &= \sqrt{\frac{u^2(C_{\mathrm{A1,A3tr}})}{C^2_{\mathrm{A1,A3tr}}} + u_R^2(V'_{\mathrm{A3tr}}) + 0.000028^2} \\ &= \sqrt{(0.02/0.06)^2 + 0.0014^2 + 0.000028^2} = 0.333 \text{ for Case 0688} \end{aligned} \tag{78}$$

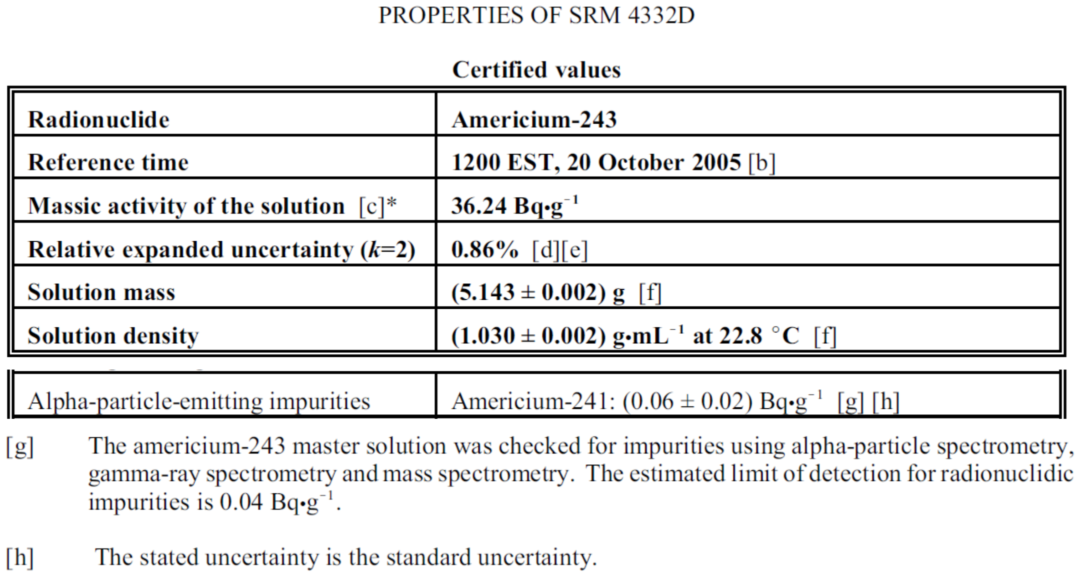
The u_R is dominated by a single term, the uncertainty supplied by the standards laboratory that produced the ^{243}Am tracer solution. This term accounts for 99.9965% of the u_R in this case.

The uncertainty of $A_{\mathrm{A1,Am,A1inA3tr}}$ is

$$u(A_{\mathrm{A1,Am,A1inA3tr}}) = u_R(A_{\mathrm{A1,Am,A1inA3tr}})A_{\mathrm{A1,Am,A1inA3tr}} \tag{79}$$

C.13. Example Calibration Certificates for Standards and Tracers

The ^{243}Am tracer solution UTR0038 derives from NIST Standard Reference Material SRM 4332D. Figure 8 shows the massic activity (activity concentration) of ^{243}Am is 36.24 Bq g^{-1} with u_R = 0.43% (k = 1). The massic activity (activity concentration) of ^{241}Am is 0.06 Bq g^{-1} with u_R = 33% (k = 1).



PROPERTIES OF SRM 4332D

Certified values

Radionuclide	**Americium-243**
Reference time	**1200 EST, 20 October 2005** [b]
Massic activity of the solution [c]*	**36.24 Bq·g^{-1}**
Relative expanded uncertainty (*k*=2)	**0.86%** [d][e]
Solution mass	**(5.143 ± 0.002) g** [f]
Solution density	**(1.030 ± 0.002) g·mL^{-1} at 22.8 °C** [f]

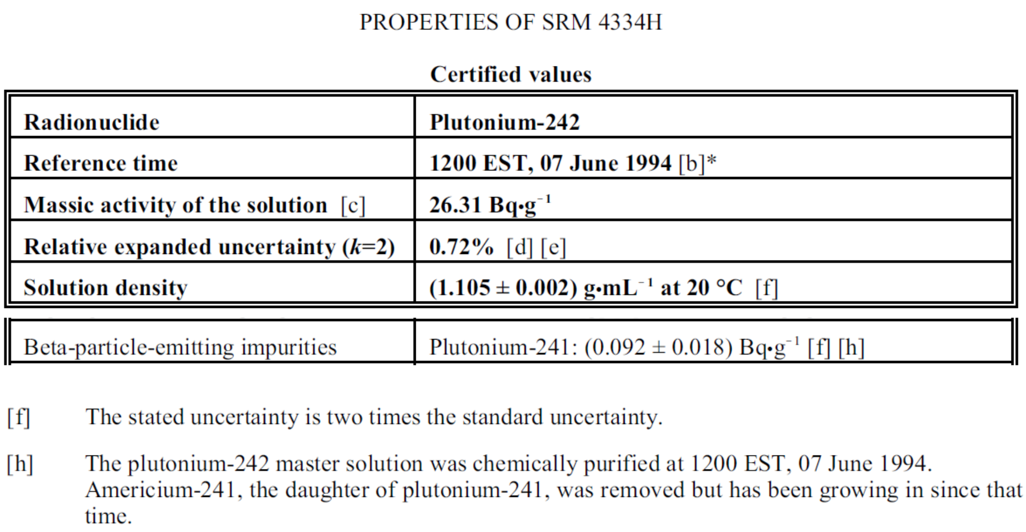
Alpha-particle-emitting impurities	Americium-241: (0.06 ± 0.02) Bq·g^{-1} [g] [h]

[g] The americium-243 master solution was checked for impurities using alpha-particle spectrometry, gamma-ray spectrometry and mass spectrometry. The estimated limit of detection for radionuclidic impurities is 0.04 Bq·g^{-1}.

[h] The stated uncertainty is the standard uncertainty.

Figure 8. Part of the NIST calibration documentation for the ^{243}Am standard solution

The ^{242}Pu tracer solution UTR0039 derives from NIST Standard Reference Material SRM 4334H. Figure 9 shows the massic activity (activity concentration) of ^{242}Pu is 26.31 Bq g^{-1} with u_R = 0.36% (k = 1). The massic activity (activity concentration) of ^{241}Pu is 0.092 Bq g^{-1} with u = 0.009 Bq g^{-1} (k = 1). Consequently, u_R = 9.8% (k = 1).



PROPERTIES OF SRM 4334H

Certified values

Radionuclide	**Plutonium-242**
Reference time	**1200 EST, 07 June 1994** [b]*
Massic activity of the solution [c]	**26.31 Bq·g^{-1}**
Relative expanded uncertainty (*k*=2)	**0.72%** [d] [e]
Solution density	**(1.105 ± 0.002) g·mL^{-1} at 20 °C** [f]

Beta-particle-emitting impurities	Plutonium-241: (0.092 ± 0.018) Bq·g^{-1} [f] [h]

[f] The stated uncertainty is two times the standard uncertainty.

[h] The plutonium-242 master solution was chemically purified at 1200 EST, 07 June 1994. Americium-241, the daughter of plutonium-241, was removed but has been growing in since that time.

Figure 9. Parts of the NIST calibration documentation for the ^{242}Pu standard solution.

In 1994, NIST quoted the uncertainty of the impurity was given as 2× the standard uncertainty, while in 2005, NIST quoted the uncertainty of the impurity as 1× the standard uncertainty.

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