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Measurement and uncertainty challenges in bringing USTUR's decades-old radiochemistry program into the 21st century: Part 1

George Tabatadze[†], Daniel J. Strom[†], Thomas L. Rucker[‡]

[†]United States Transuranium and Uranium Registries
1845 Terminal Drive, Suite 201, Richland, WA 99354
[‡]Leidos, Energy and Environmental Division, P.O. Box 2502
Oak Ridge, TN 37831

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65th Radiobioassay and Radiochemical Measurements Conference
Atlanta, Georgia, October 31 – November 4, 2022

U.S. AEC 1966 Meeting on Plutonium Contamination in Man (Rocky Flats Plant)

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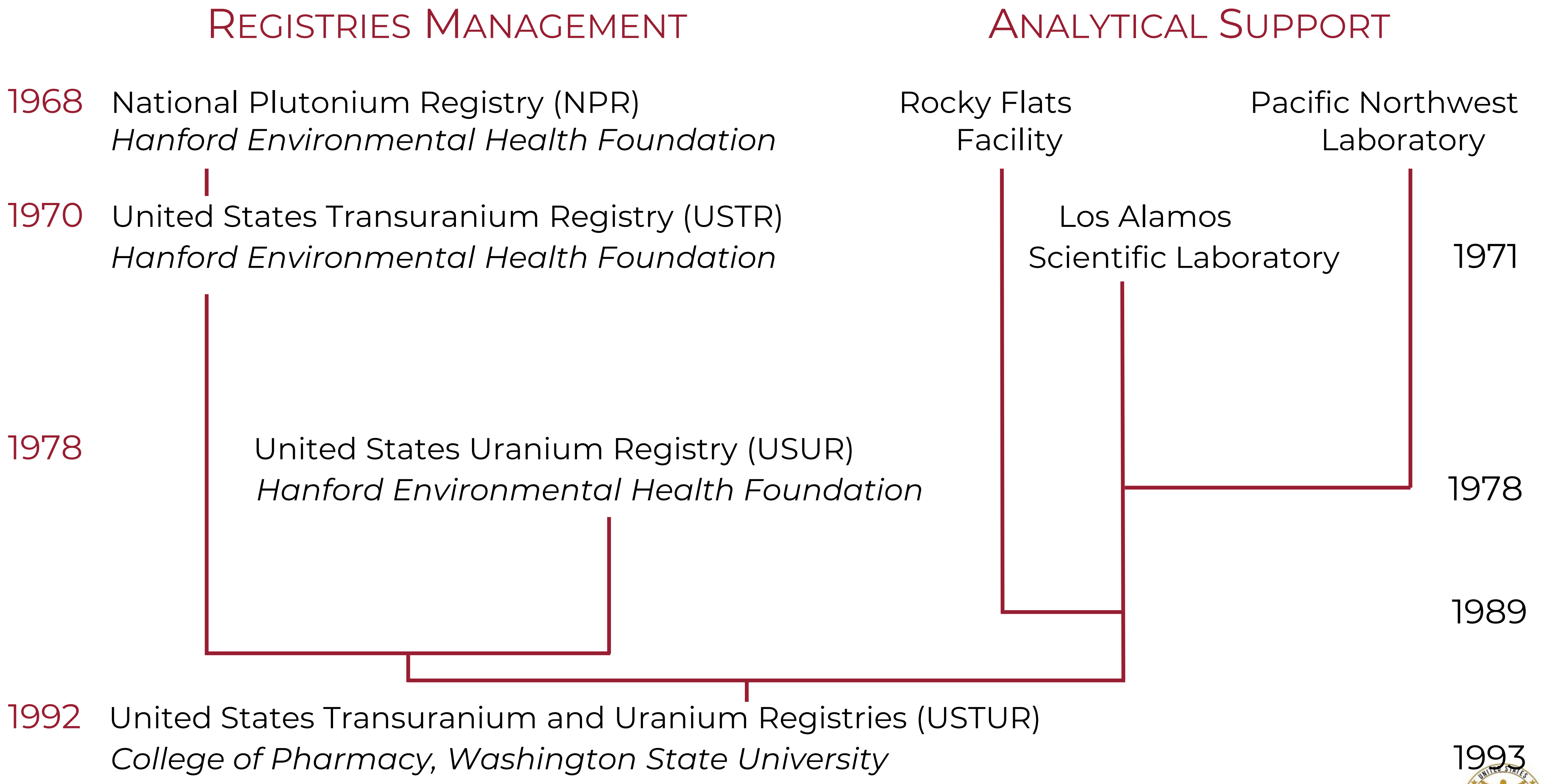
National Plutonium Registry: *Blue Ribbon Committee (1968)*

Standing left to right: Carlos E. Newton, Jr., W. Daggett Norwood, H.D. Bruner, Philip A. Fuqua
Seated left to right: Thomas F. Mancuso, J.H. Sterner, Robley D. Evans, Herbert M. Parker
Not photographed: Clarence C. Lushbaugh, Lloyd M. Joshel

Genealogy of the USTUR



USTUR Mission Statement

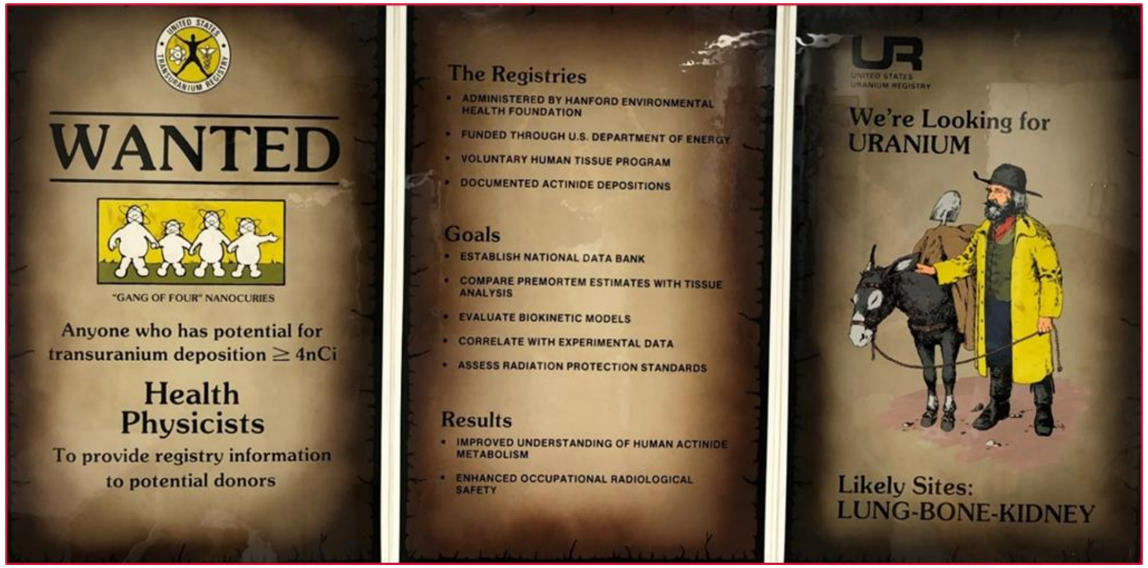
- Follow up occupationally-exposed individuals (volunteer Registrants) by studying the biokinetics (deposition, translocation, retention, and excretion) and tissue dosimetry of uranium and transuranium elements, such as plutonium, americium, curium, and neptunium
- Obtain, analyze, preserve, and make available for future research, materials from individuals who had documented intakes of uranium and transuranium elements
- 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

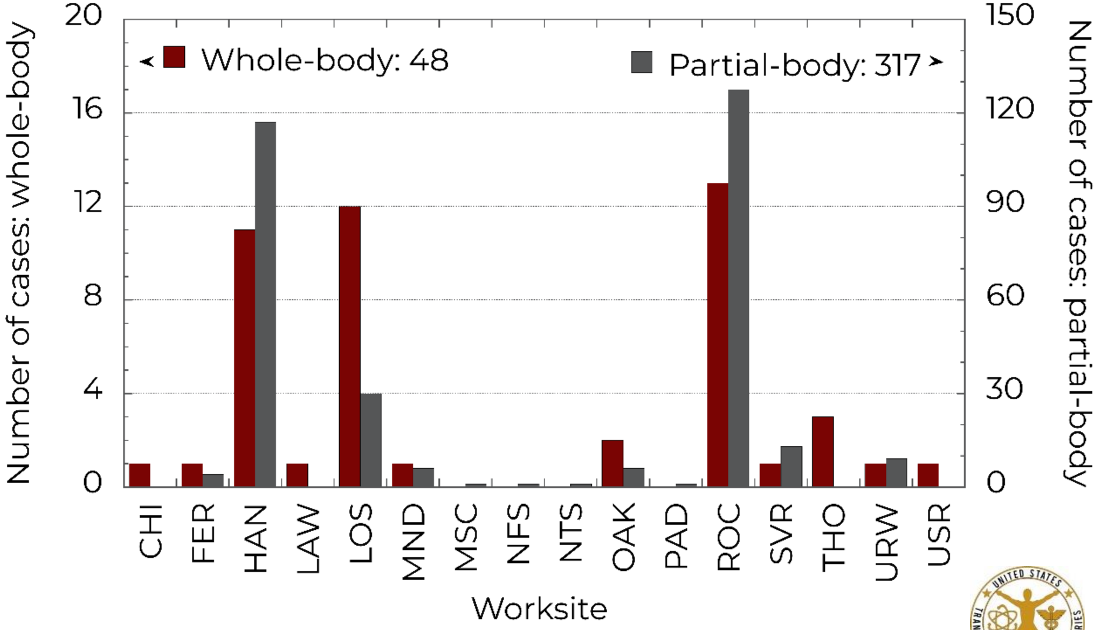
USTUR Registrants

Individuals with documented history of exposure to the actinides

- Selection criteria: ≥2 nCi (internal deposition) or ≥10 rem (external)
- Mainly former nuclear workers from DOE sites
- Voluntary tissue donors (posthumous):

whole- (48) and/or partial-body (317) donations





Unique Data Resource

12 353

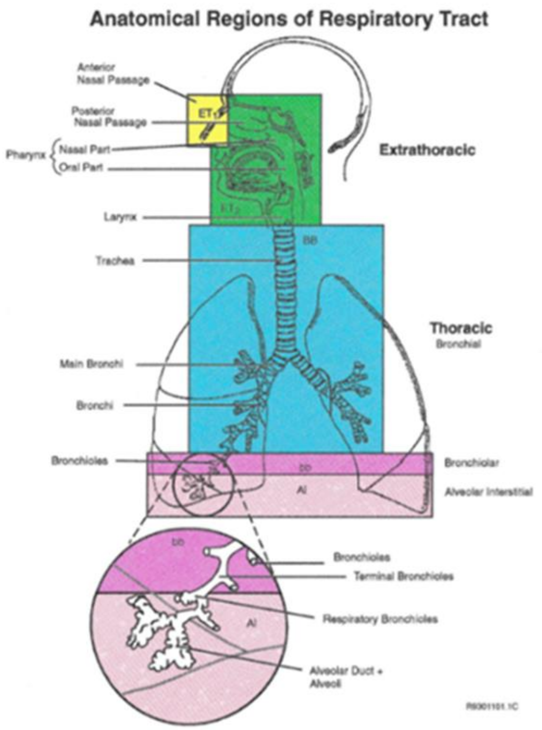
USTUR data

- Work history
- Medical records
- Smoking history†
- Chemical exposure records†
- Radiation exposure records
- Bioassay measurements
- Tissue analysis results (Pu, Am, U)

† - self-reported

Primary Research: *Biokinetic Modeling and Internal Dosimetry of Actinides*

- Testing, improving and parameterizing biokinetic models for radiological protection
  - ✓ Human Respiratory Tract Model (ICRP 130)
  - ✓ Wound Model (NCRP 156)
  - ✓ Systemic models for U, Pu, Am (ICRP 137 & 141)
- Modeling actinide decorporation
- Evaluating uncertainties in internal radiation dose assessment



Principal Study Questions

Maximizing measurement quality and utility considering limitations

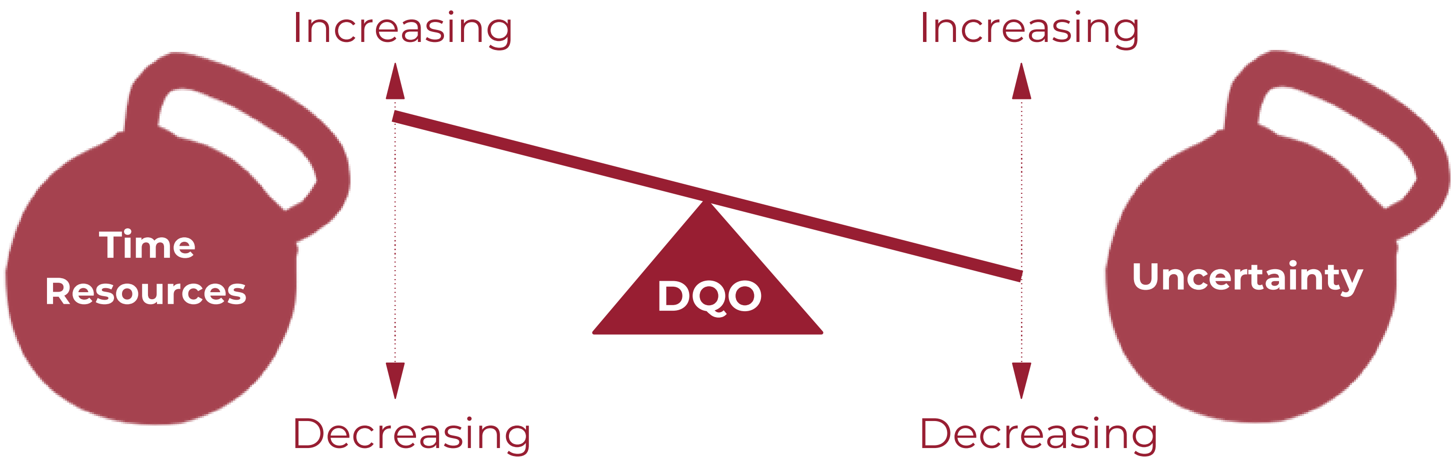
1. Activities
   For the radionuclide of concern, what are the activities in dosimetrically important organs and tissues?

2. Uncertainties
   What is an acceptable uncertainty in USTUR activity measurements?

The goal is to develop USTUR-Specific Data Quality Objectives (DQO)

Limitations

- Acceptance limit for registrants (74 Bq uptake)
- Limited samples from each registrant
- Historical spike of 1/30 Bq
- Use of 2 g of ash maximum in separation process
- Counting time
- Tissue sample backlog
- cost



Measurement Quality Objectives

Measurement results and the combined standard uncertainty of the measurement results are the principal products of USTUR's radiochemistry laboratory

Measurand – the quantity intended to be measured

- True but unknown activity in the USTUR tissue, organ or other sample

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

MQOs

- Accuracy
- Precision
- Sensitivity
- Representativeness
- Completeness

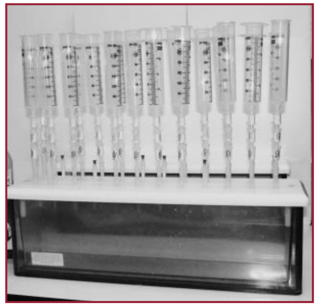
International Organization for Standardization (ISO). Uncertainty of measurement - part 3: Guide to the expression of uncertainty in measurement (GUM: 1995). Geneva, Switzerland; Guide 98-3 (2008); 2008.

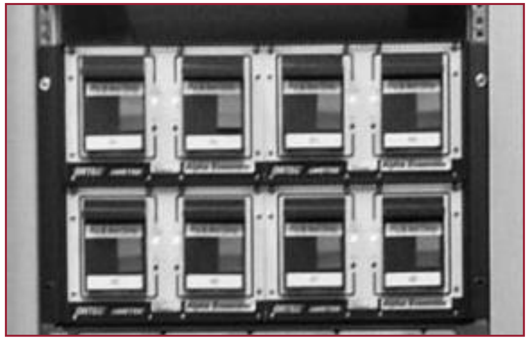
Joint Committee for Guides in Metrology (JCGM). International vocabulary of metrology – basic and general concepts and associated terms (VIM). JCGM 200:2012 (JCGM 200:2008 with minor corrections); 2012.

USTUR Analytical Methods

- USTUR tissue radiochemical analysis protocol

Drying & Ashing → Digestion & Dissolution → Actinide separation → α-source preparation → Actinide measurement





Sample Processing & Analytes

- Sample processing

Collection → Dissection → Preservation & Storage → Tracking

- Analytes of interest

$^{238, 239, 239+240, 240, 241, 242}$Pu

$^{228, 229, 230, 232}$Th

$^{241, 243}$Am

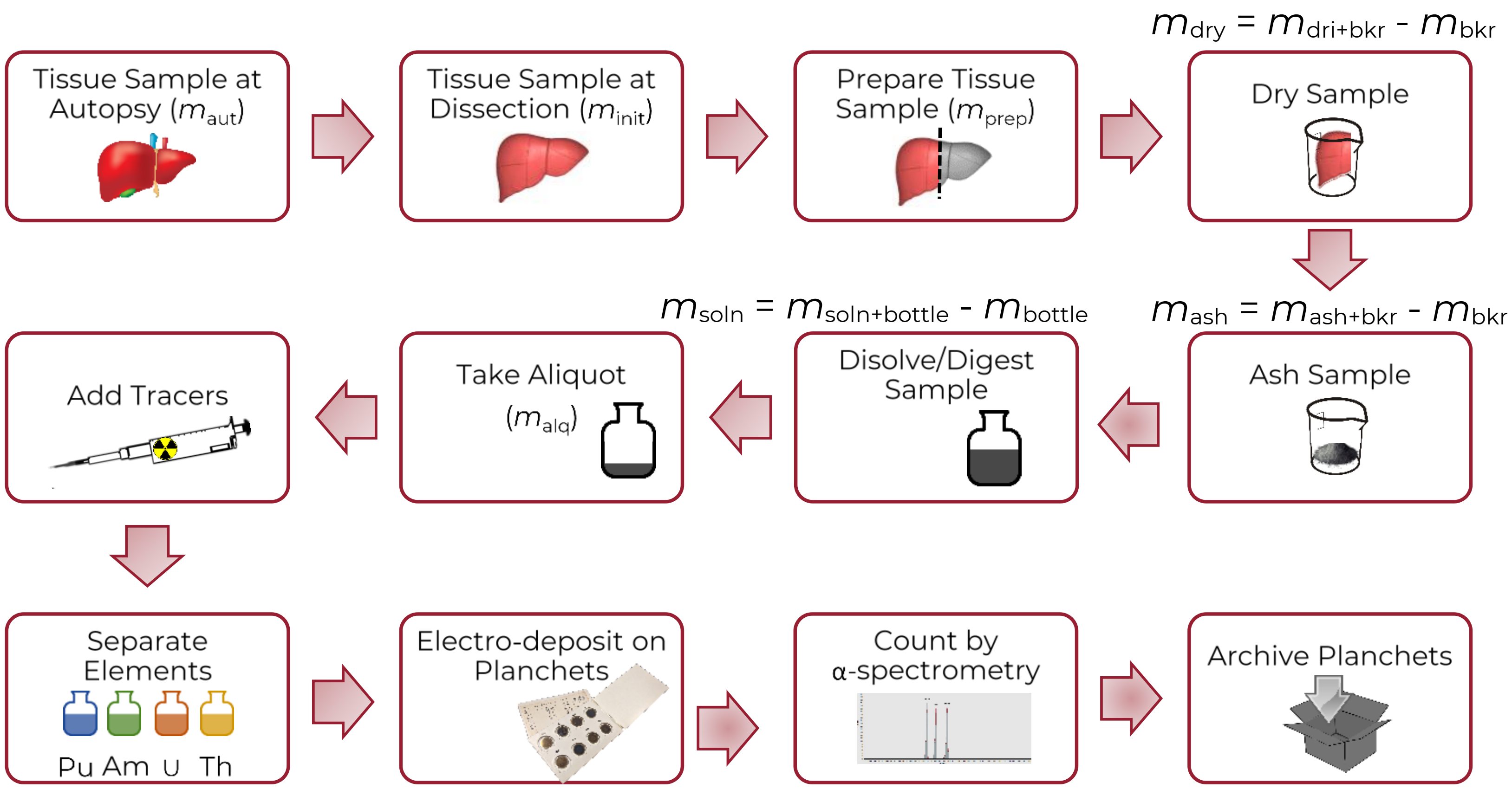
$^{232, 234, 235, 236, 238}$U

^{237}Np

^{244}Cm

Sample Processing: *Masses*

- Mass (tare, gross, and net) measurements are made at several stages of radiochemical processing



Sequence of Events: *Radiochemical Processing*

Activity notation is $A_{isotope,destination,origin}(t)$

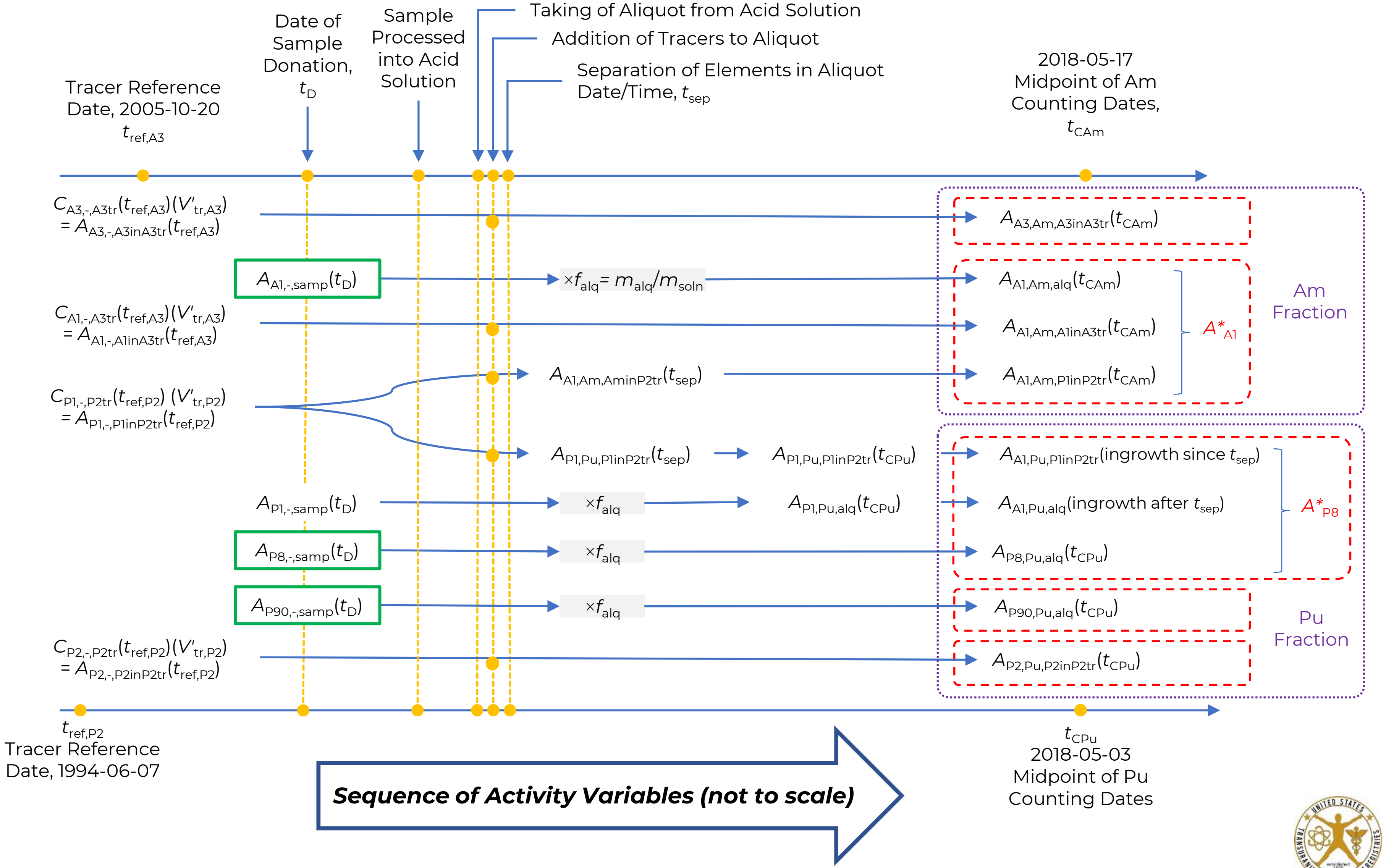
Green boxes are the desired measurands.

Dashed red boxes are measurement results in an ROI.

Dotted purple boxes indicate element fractions (element planchets).

A^* indicates an "apparent activity" inferred from counts in a region-of-interest (ROI) in alpha spectrometry.

Corrections to A^* must be made to infer values of desired measurands.



Calculations: *MARLAP's N+1 Counting Statistics*

USTUR α-spectrometry measurements

- Sample count time t_S = 150,000 s (~1.75 days)
- Background count time t_B = 300,000 s (~3.5 days)
- 5 to 15 background counts typically observed in an ROI
  - ✓ MARLAP: these are "low numbers of counts"
- For best accuracy and realistic statistics, use *N*+1 counting stats formulas (first published by Rainwater & Wu, Nucleonics 1:60-69 1947 and many others since)

$$R_N = \frac{N_S + 1}{t_S} - \frac{N_B + 1}{t_B} \qquad \mathrm{Var}(R_N) = \sqrt{\frac{N_S + 1}{t_S^2} + \frac{N_B + 1}{t_B^2}}$$

Calculations: *Activity*

- Activity of isotope *X* on the element *Z*-fraction planchet

$$A^{*}_{X,Z,alq}(t_{C,Z}) = \frac{\left(\frac{N_S}{t_S} - \frac{N_B}{t_B}\right)}{\varepsilon f_{X,ROI} y_{RR}} = \frac{R_S - R_B}{\varepsilon f_{X,ROI} y_{RR}} = \frac{R_N}{\varepsilon f_{X,ROI} y_{RR}}$$

$$A^{*}_{X,Z,alq}(t_{C,Z}) = \frac{\left(\frac{N_S + 1}{t_S} - \frac{N_B + 1}{t_B}\right)}{\varepsilon f_{X,ROI} y_{RR}}$$

- If there are no contaminant isotopes present, then the “apparent activity” is simply the activity: *A* = *A**.
- Relative Uncertainty of the activity of isotope *X*

$$u_R(A_{X,Z,alq}) = \frac{u(A_{X,Z,alq})}{A_{X,Z,alq}} = \sqrt{\frac{\left(\frac{N_S + 1}{t_S^2} + \frac{N_B + 1}{t_B^2}\right)}{\left(\frac{N_S + 1}{t_S} - \frac{N_B + 1}{t_B}\right)^2} + u_R^2(\varepsilon) + u_R^2(f_{X,ROI}) + u_R^2(y_{RR})}$$

Calculations: ^{241}*Am Activity*

- Contribution to the Am fraction
  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 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 the ^{242}Pu tracer up until the time of element separation.

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

- Relative uncertainty of ^{241}Am activity

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

Calculations: ^{238}Pu Activity

- Contribution to the Pu fraction

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
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.

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

- Relative uncertainty of ^{238}Pu activity

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

Calculations: *Summary*

- Equations Compendium: mass, mass fraction, volume (pipetting), isotope ratio, activity, activity concentration, radiochemical recovery yield and all associated uncertainties

$$A_{\text{init}} = A_{\text{planchet}} \frac{m_{\text{init}}}{m_{\text{prep}}} \frac{m_{\text{soln}}}{m_{\text{aliq}}} \frac{1}{y_{\text{RR}}}$$

$$u^2\left(m_{\text{a}}/m_{\text{b}}\right) = \frac{m_{\text{a}}^2}{m_{\text{b}}^2}\left[\frac{u^2(m_{\text{a}})}{m_{\text{a}}^2} + \frac{u^2(m_{\text{b}})}{m_{\text{b}}^2}\right]$$

$$u^2(A_{\text{init}}) = \left[\frac{u^2(A_{\text{planchet}})}{A_{\text{planchet}}^2} + \frac{u^2(m_{\text{init}}/m_{\text{prep}})}{m_{\text{init}}^2/m_{\text{prep}}^2} + \frac{u^2(m_{\text{soln}}/m_{\text{aliq}})}{m_{\text{soln}}^2/m_{\text{aliq}}^2} + \frac{u^2(y_{\text{RR}})}{y_{\text{RR}}^2}\right]\left(A_{\text{planchet}} \frac{m_{\text{init}}}{m_{\text{prep}}} \frac{m_{\text{soln}}}{m_{\text{aliq}}} \frac{1}{y_{\text{RR}}}\right)^2$$

$$u^2(A_{\text{init}}) = \left[\frac{u^2(A_{\text{planchet}})}{A_{\text{planchet}}^2} + \left\{\frac{u^2(m_{\text{init}})}{m_{\text{init}}^2} + \frac{u^2(m_{\text{prep}})}{m_{\text{prep}}^2}\right\} + \left\{\frac{u^2(m_{\text{soln}})}{m_{\text{soln}}^2} + \frac{u^2(m_{\text{aliq}})}{m_{\text{aliq}}^2}\right\} + \frac{u^2(y_{\text{RR}})}{y_{\text{RR}}^2}\right]\left(A_{\text{planchet}} \frac{m_{\text{init}}}{m_{\text{prep}}} \frac{m_{\text{soln}}}{m_{\text{aliq}}} \frac{1}{y_{\text{RR}}}\right)^2$$

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

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

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

$$y_{\text{RR}}(Y) = \frac{A_{Y,Z,Y\text{tr}}(t_{\text{C},Z})}{C_{Y,Y\text{tr}}(t_{\text{ref},Y})\,e^{-\lambda_Y(t_{\text{C},Z}-t_{\text{ref},Y})}\,V'_{Y\text{tr}}}$$

$$u_R(y_{\text{RR}}(Y)) = \sqrt{u_R^2(A_{Y,Z,Y\text{tr}}(t_{\text{C},Z})) + u_R^2(C_{Y,Y\text{tr}}(t_{\text{ref},Y}))}$$

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

$$u_R(A_{X,Z,\text{soln}}(t_{\text{C},Z})) = \sqrt{u_R^2(A_{X,Z,\text{aliq}}(t_{\text{C},Z})) + u_R^2(y_{\text{RR}}(Y))}$$

$$A_{X,Z,\text{soln}}(t_D) = A_{X,Z,\text{soln}}(t_{\text{C},Z})\,e^{-\lambda_X(t_{\text{C},Z}-t_D)} \qquad u_R\left(A_{X,Z,\text{soln}}(t_D)\right) = u_R\left(A_{X,Z,\text{soln}}(t_{\text{C},Z})\right)$$

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

$$u\left(A_{\text{A1,Am,A1inA3tr}}\right) = u_R\left(A_{\text{A1,Am,A1inA3tr}}\right)A_{\text{A1,Am,A1inA3tr}}$$

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

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

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

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

$$A_{\text{A1,Pu,P1aliq}}(t_{\text{C,Pu}}) = \text{ ingrowth of } {}^{241}\text{Am in Pu fraction from } {}^{241}\text{Pu}$$

Calculations: *Statistical Criteria*

- Critical Value (S'_C)

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

- Minimum Detectable Activity (*MDA*)

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

- Minimum Quantifiable Activity (*MQA*) - value of a theoretical measurand activity that can be measured with a specified statistical precision e.g. *MQA*(0.1) for $u_R = 10\%$

Routine Measurements at USTUR

The critical value of the net count rate, S'_C, is the smallest count rate that has only a 5% chance of being due to background noise

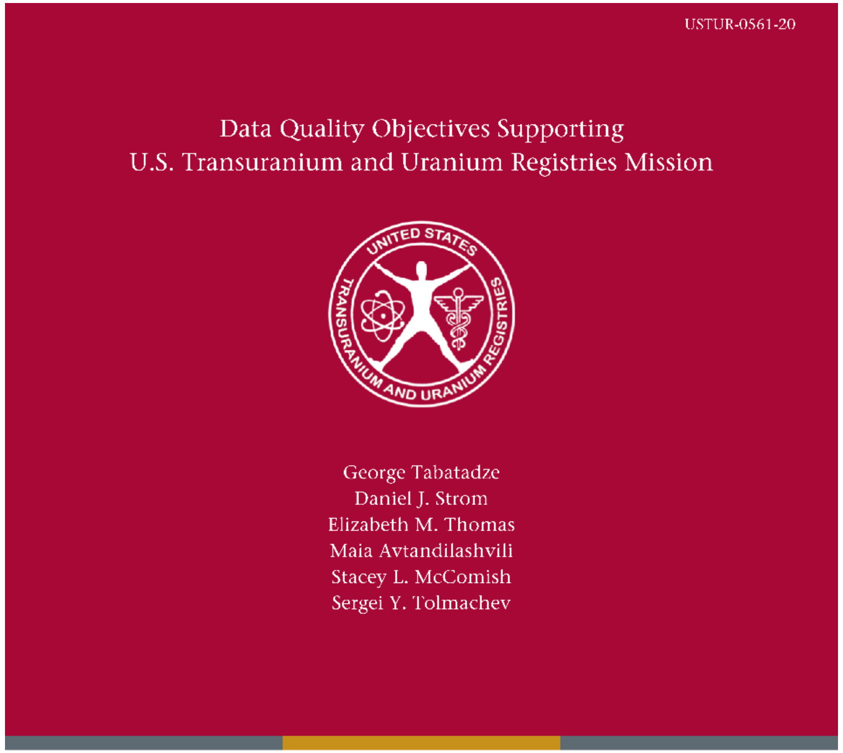
- For some applications, these are recorded as “nondetects” or “less-than” values
- USTUR does not censor its measurement results by changing numbers to nondetects

USTUR is not determining whether activity is present in autopsy samples – we know it’s there, so it’s only a question of how much

USTUR records and reports values less than S'_C along with their uncertainties

Highly uncertain measurements (rather than nondetects or less-thans) contain information that can be used for calculating intake and dose

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

George Tabatadze
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Elizabeth M. Thomas
Maia Avtandilashvili
Stacey L. McComish
Sergei Y. Tolmachev

March 25, 2022

College of Pharmacy and Pharmaceutical Sciences
WASHINGTON STATE UNIVERSITY

Daniel J. Strom

Thomas L. Rucker

USTUR Team

- Maia Avtandilashvili
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