WASHINGTON STATE UNIVERSITY
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Measurement and uncertainty challenges in bringing USTUR's decades-old radiochemistry program into the 21st century: Part 2

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Expected Sample Activity: *Objective*

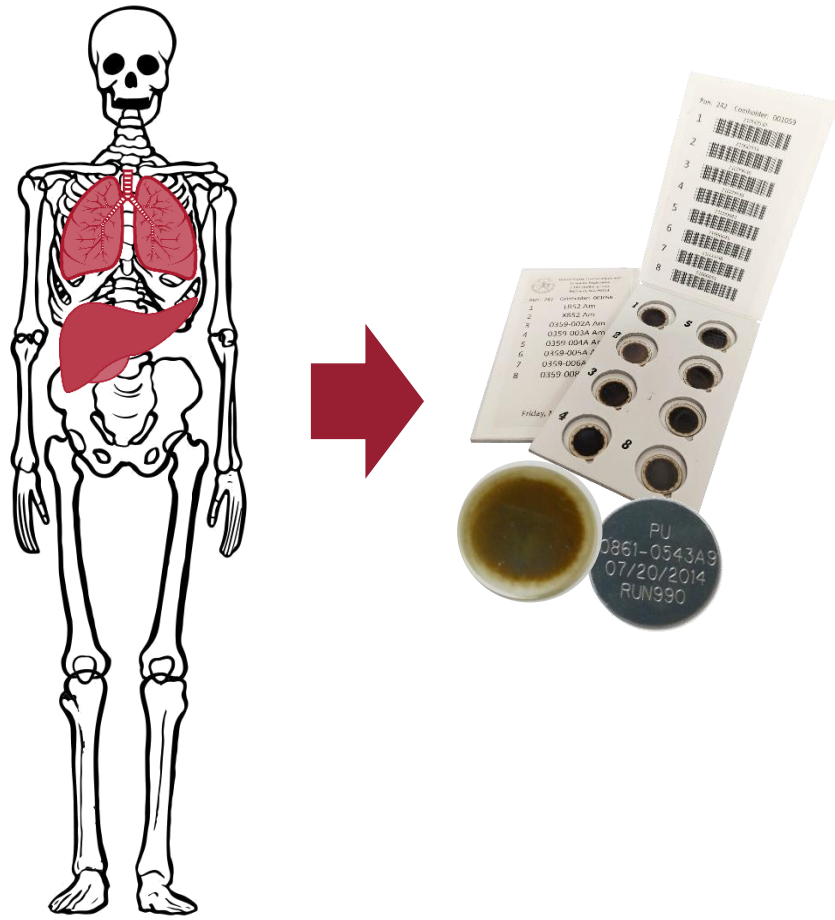
Using 74 Bq activity threshold for becoming a USTUR Registrant as a starting point

- through inhalation and/or wounds
- various chemical forms of the principal radionuclides

Objective is to predict

- activity on a planchet for an intake/uptake of ≥74 Bq, 50 years post intake
- relative uncertainties under realistic conditions

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



Expected Sample Activity: *Assumptions*

Initial Conditions

- m_{prep}/m_{init} range from 1 down to 0.1;
- m_{aliq}/m_{soln} ranges from 1 to 0.2;
- y_{RR} is typically 0.9 with ranges from 0.5 to 1.1
  - ✓ the worst (least sensitive) case value (the lower Tolerance Limit for y_{RR}) of 0.5 is used in calculations.

Ideally:

$$MDA(\text{radionuclide}) \text{ OR } MQA(u_R) \leq A_{\text{planchet}}(\text{radionuclide, chemical form, organ, route of intake})$$

Expected Sample Activity: *Example*

Example: *Other* soft tissue

- Intake: 74 Bq
- Analyte: ^{239}Pu
- Chemical form: nitrate
- Route of intake: inhalation
- Time post intake: 50 y
- Ref. weight (ICRP 89): 59.5 kg
- Chemical recovery (TL): 50%

Activity estimated with IMBA Professional Plus ®

- Activity in soft tissues: 0.13 Bq

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 2g ash†
All other tissues	0.7	or 2 g ash†
Formalin	0.9	or 2 g ash†
Filters	0.9	or 2 g ash†

† whichever is less solution

$$\frac{0.13\ \text{Bq}}{59.5\ \text{kg}} = 2.1\times10^{-3}\ \text{Bq kg}^{-1}\ \text{of tissue.} \qquad 2.1\times10^{-3}\ \text{Bq kg}^{-1}\ \text{tissue} \times \frac{1\ \text{kg tissue}}{20\ \text{g ash}} = 1.1\times10^{-4}\ \text{Bq g}^{-1}\ \text{ash.}$$

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

Expected Sample Activity: *Results*

- Predicted activity on a planchet for key tissues and organs
- *50 years after inhaling 74 Bq*
- isotopes of Pu, Am, U, Th, Ra, Cm, and Np

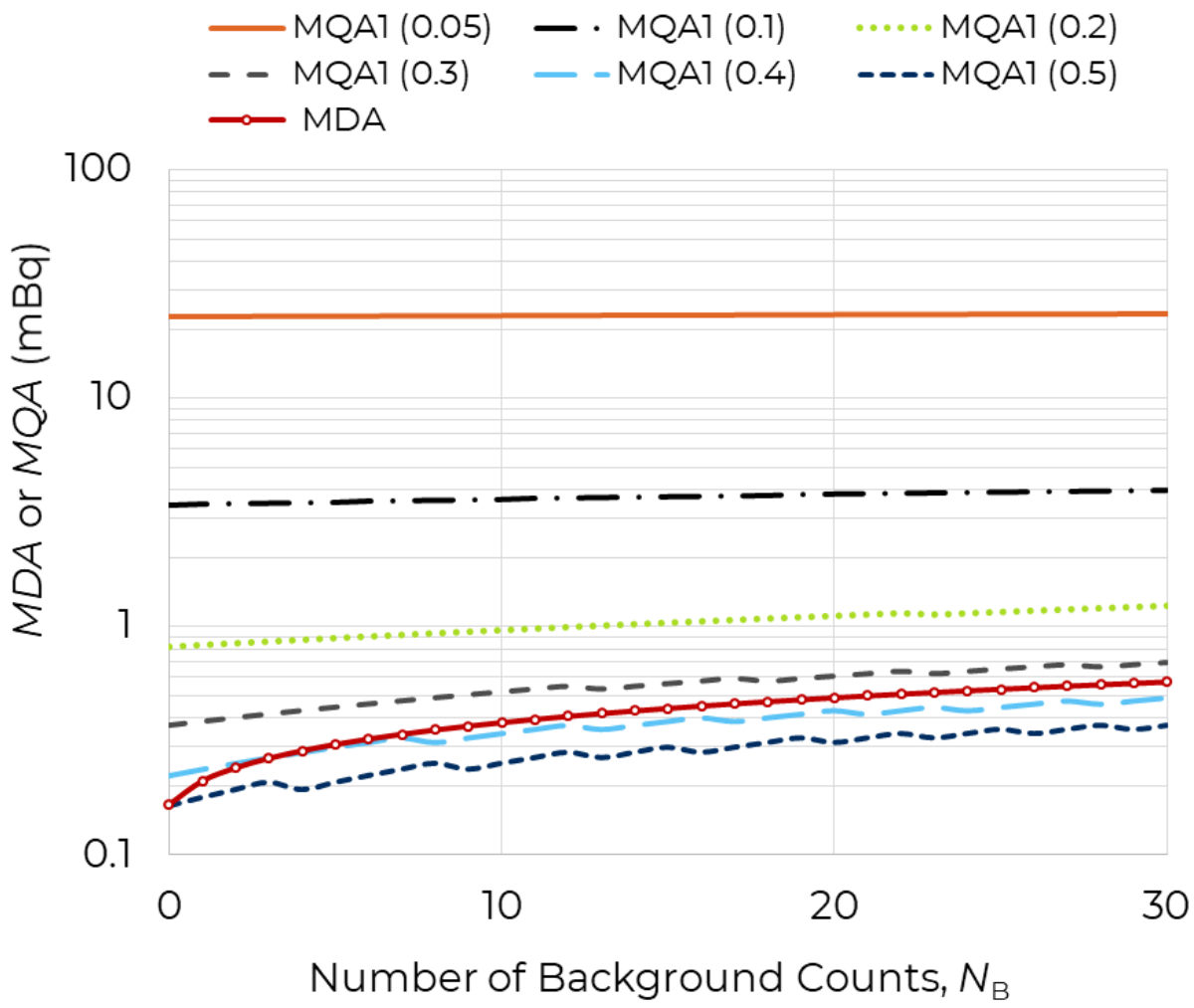
Radio-nuclide	Tissue or Organ	Systemic Uptake	Inhalation									Ingestion
			Soluble		Moderately Soluble			Insoluble				
			Hexafluoride	Citrate	Nitrate	Chloride/ Citrate/Nitrate/ Sulphate/ Fluoride	Nitrate/ Oxide/ Chloride	Dioxide	Octoxide	Type S	
Pu-239	Lungs				7.00E-05			1.36E-02			
Pu-239	Liver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

Typical MDAs & MQAs Depend on Background

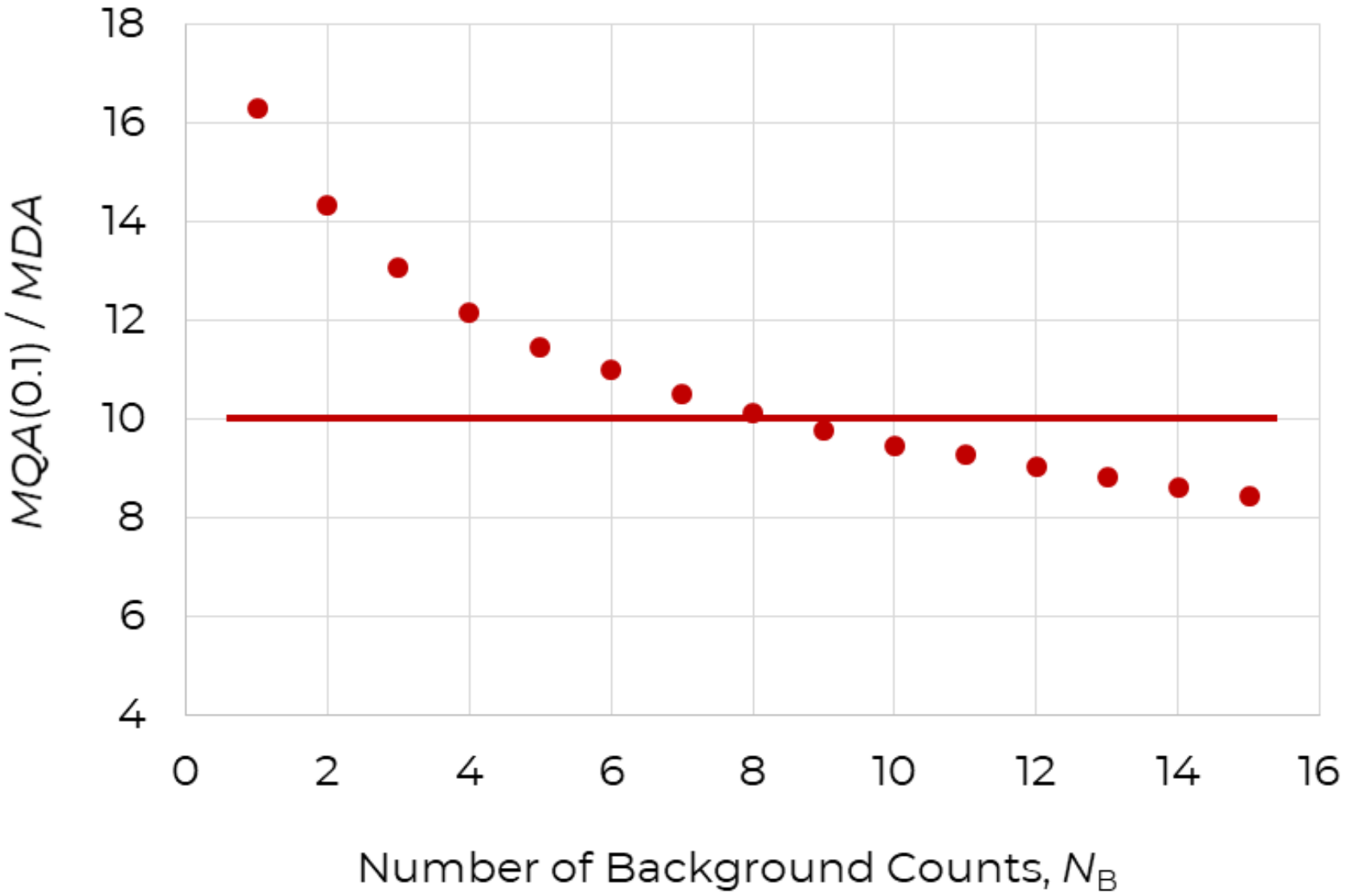
Calculating *MDA* and *MQA*

A generic approach

- N_B: 0 – 30
- t_B = 300,000 s
- t_S = 150,000 s
- ε = 0.25
- $f_{X,ROI}$ = 1
- $\alpha = \beta$ = 0.05
- d = 0.4
- y_{RR} = 0.9



Typical MQA / MDA Ratios



Chosen MDA & MQA: *Uncertainty & Measurand*

Uncertainty increases with decreasing activity

- $u_R(Activity) \approx 35\%$ at the *MDA*

For

N_B = 15 counts (TL)

t_B = 300,000 s

t_S = 150,000 s

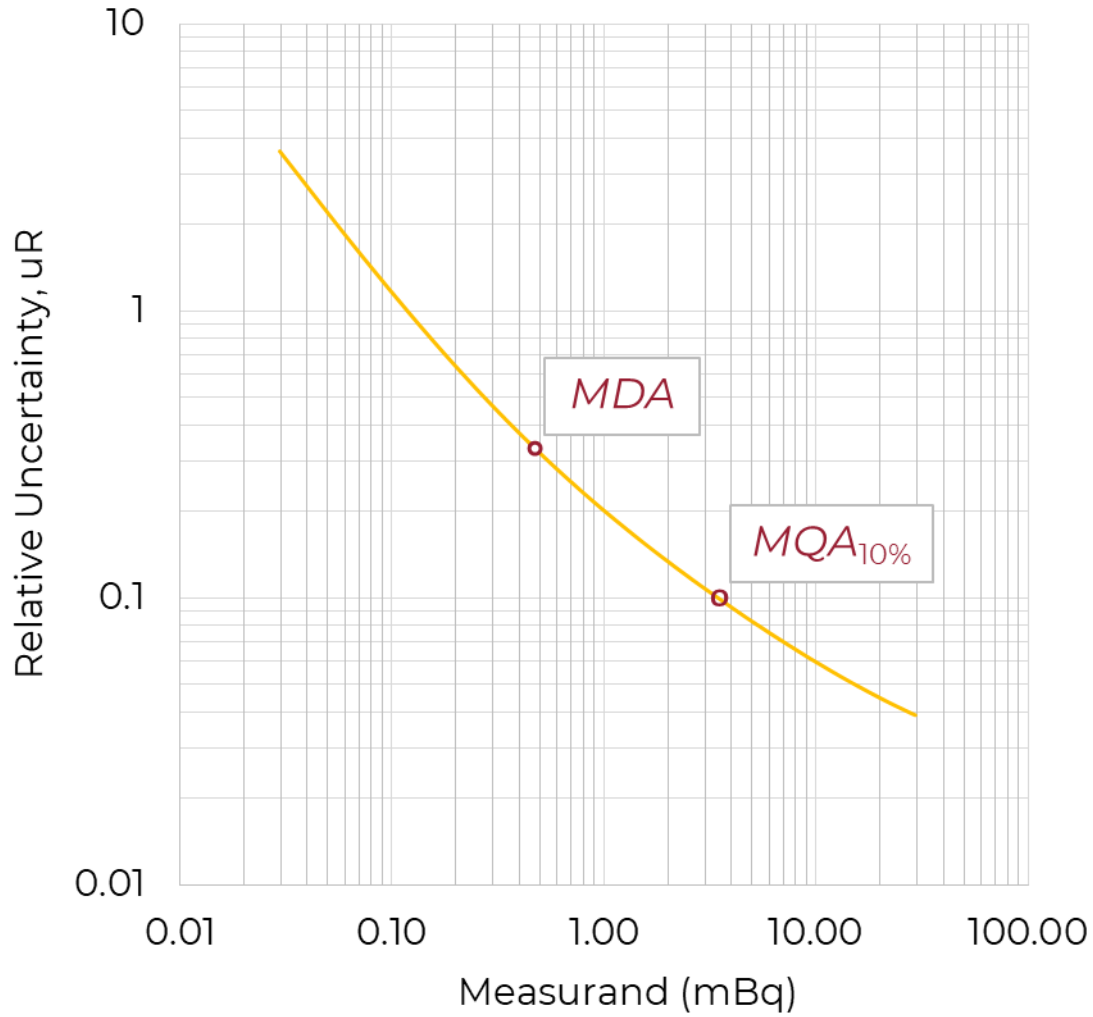
ε = 0.25

$f_{X,ROI}$ = 1

y_{RR} = 0.9

the *MDA* = 0.44 mBq

the $MQA_{10\%}$ ~ 3.7 mBq



Expected Sample Activity: *MDA & MQA*

- N_B = 15 counts in 300,000s assumed (background tolerance limit)

MDA = 0.44 mBq

Radio-nuclide	Tissue or Organ	Uptake	Inhalation	
			Nitrate	Dioxide
Pu-239	Lungs	N/A	X	√
Pu-239	Liver	√	√	√
Pu-239	Skeleton	√	O	X
Pu-239	Other Soft Tissues	√	X	X
Am-241	Lungs	N/A	X	√
Am-241	Liver	√	√	√
Am-241	Skeleton	√	√	O
Am-241	Other Soft Tissues	√	O	X

MQA = 3.7 mBq

Radio-nuclide	Tissue or Organ	Uptake	Inhalation	
			Nitrate	Dioxide
Pu-239	Lungs	N/A	X	√
Pu-239	Liver	√	√	X
Pu-239	Skeleton	√	X	X
Pu-239	Other Soft Tissues	O	X	X
Am-241	Lungs	N/A	X	O
Am-241	Liver	√	O	X
Am-241	Skeleton	√	X	X
Am-241	Other Soft Tissues	√	X	X

$f \geq 1.0$
$0.5 < f < 1.0$
$f \leq 0.5$

- f is $A_{50,predicted}$ / *MDA* or $A_{50,predicted}$ / $MQA_{10\%}$
- Green: meets *MDA* or $MQA_{10\%}$ criterion
- Yellow: within a factor of 2 below *MDA* or $MQA_{10\%}$ criterion
- Pink: "technology shortfall," more than 2× below *MDA* or $MQA_{10\%}$ criterion

An Achievable MQO: *Sensitivity*

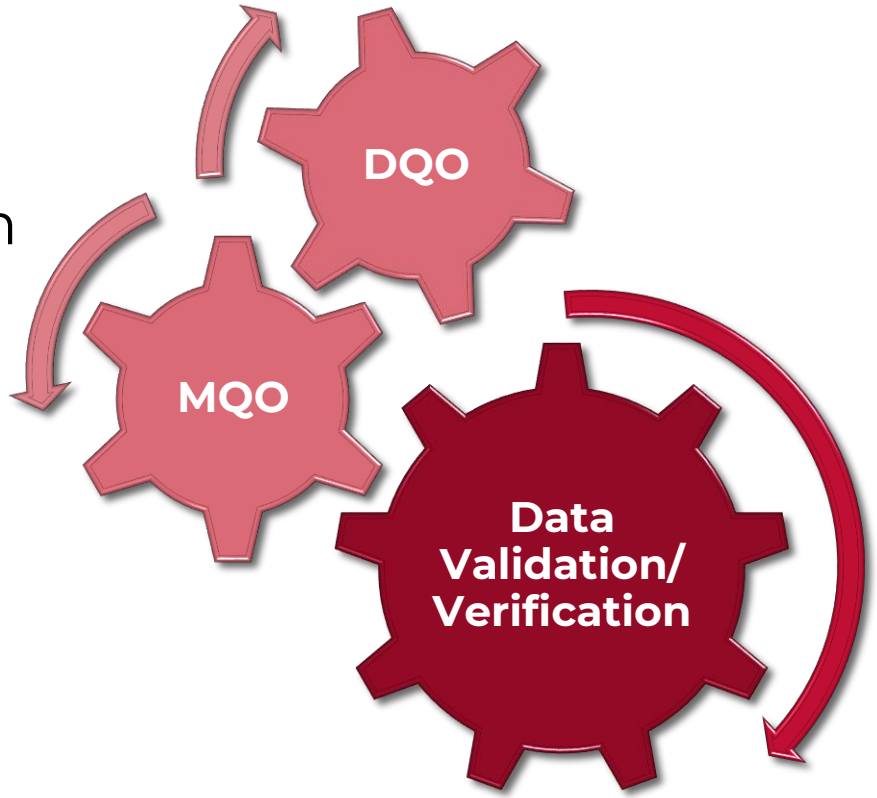
- MQO for sensitivity is that the expected activity on a planchet which should be greater than the *MDA*

$$A_{\text{planchet}}(\text{radionuclide, chemical form, organ, route of intake}) \geq MDA(\text{radionuclide})$$

- There will be about 35% combined standard relative uncertainty with *MDA* rather than 10% using the $MQA_{10\%}$

An Achievable MQO: *Accuracy and Precision*

- USTUR chooses 1/30 Bq (2 dpm) with $u_R \leq 4\%$
  - ✓ ~ 10 × $MQA_{10\%}$
  - ✓ ~ 100 × $MDA(^{239}Pu)$
  - ✓ Current (historical) spike level for ^{243}Am and ^{242}Pu tracers
  - ✓ historical measurements can be reviewed for their accuracy and precision
- Basis for over 20 measurement performance criteria for accuracy, precision, sensitivity, representativeness, and completeness



An Achievable MQO: *MPI*

Measurement performance criteria (MPI) for use in data verification and validation

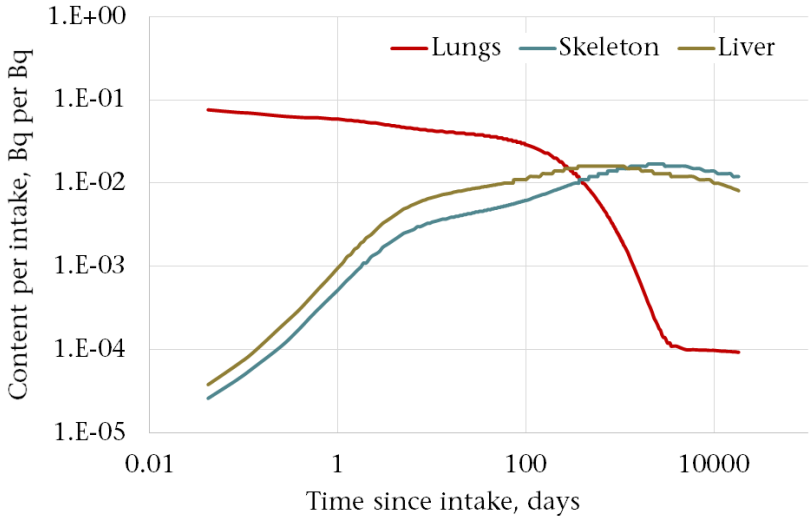
QC Sample or Purpose	Associated MQO	Parameter Type	Frequency or Number	Measurement Performance Criteria (MPC)	Corrective Action (CA) if MPC Not Met
Aliquot Split; only valid when each Sample contains > MQA(0.05)	Precision	Sample specific	1 per case (liver, lung, or large bone)	Tolerance Level (TL) for Precision: relative standard deviation, $s_R \leq 0.15$	Investigate; Flag data as J or U
Aliquot Split	Represent-ativeness	Sample specific	1 per case (liver, lung, or large bone)	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: $0.5 \leq y_{RR} \leq 1.1$	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 $u_R \leq 4\%$	Investigate; Flag as S; Flag data as J or U
Reagent Blank (RB)	Accuracy: Bias, Sensitivity	Batch control	1 of every 20 Samples	TL: $N_{RB,ROI} \leq 15$. Note 1: any $N_{RB,ROI} \leq 21$ counts gives $u_R \leq 0.35$ 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

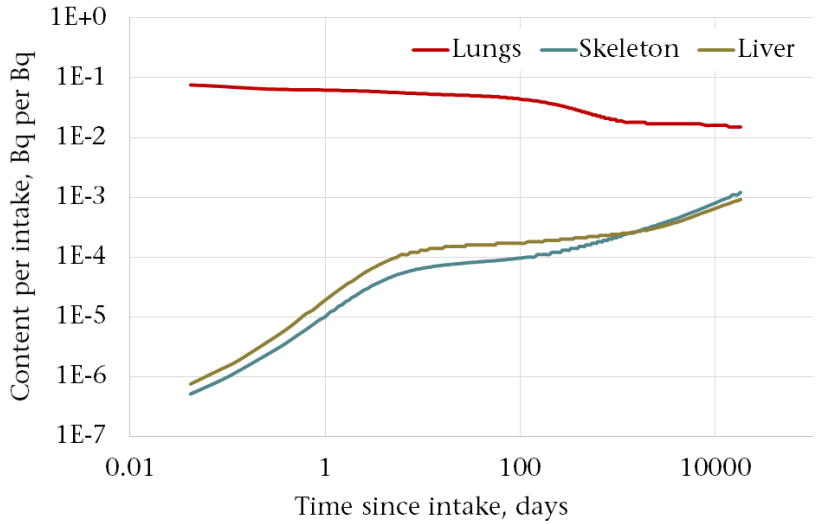
Technology Shortfall

Most registrants have intakes that are sufficiently high that material can be detected in many tissues, with some notable exceptions

- 50 years after an inhalation intake of plutonium nitrate, less than 1/1000th of the initial activity in the lung remains
- Technology shortfall is not often encountered because most Registrants had intakes that exceeded 74 Bq

Tissue or organ content over time following 1 Bq inhalation intake of 5-µm ^{239}Pu nitrate (a) and oxide (b) aerosols





Managing Technology Shortfall

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

- Increasing α-spectrometry counting time
- Using mass spectrometry technique
- Combining eluates from several sub-samples
- Combining counts from several sub-samples of a tissue or organ

  Lung lobes: superior, middle, inferior

  Combining counts reduces uncertainty by 40% in a typical case

	Mass (g)	N_B	N_S	R_B (Ms^{-1})	R_S (Ms^{-1})	R_N (Ms^{-1})	$u(R_N)$	$u_R(R_N)$
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 3 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

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Thank you for your attention