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Curium in the human body – USTUR case study

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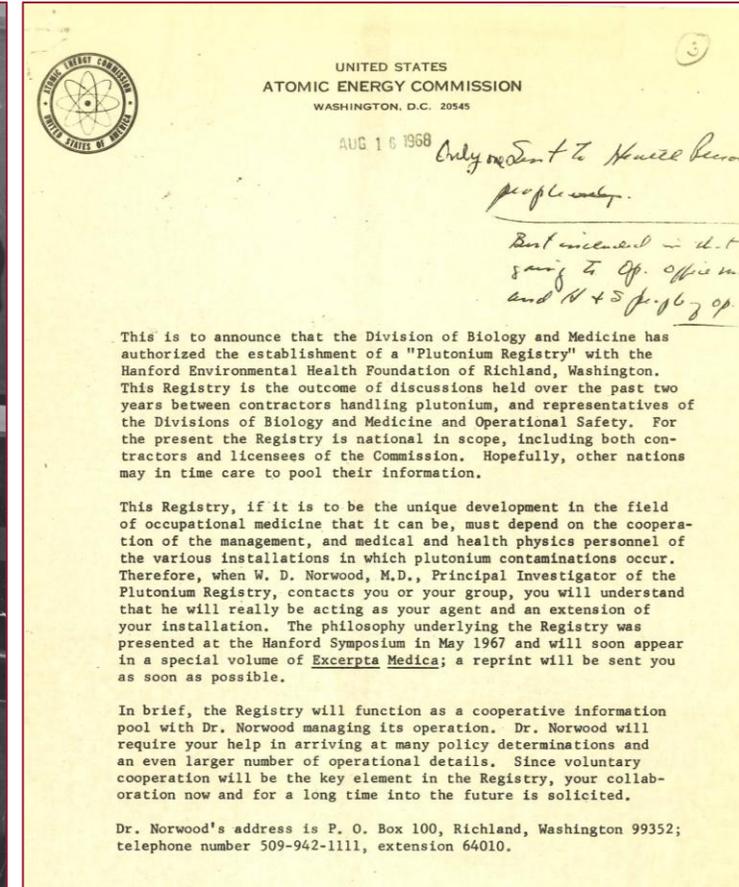
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College of Pharmacy and
Pharmaceutical Sciences



U.S. AEC Vision: 1966 Meeting on Plutonium Contamination in Man – Plutonium Registry (1968)



"We are proceeding deliberately with this project because it is one which should not be abandoned once it is taken up." (H.D. Bruner, A Plutonium Registry 1967)



The Registries



U.S. DEPARTMENT OF
ENERGY



REGISTRIES MANAGEMENT

ANALYTICAL SUPPORT

1968 Plutonium Registry
Hanford Environmental Health Foundation

Rocky Flats
Facility

Pacific Northwest
Laboratory

1970 United States Transuranium Registry (USTR)
Hanford Environmental Health Foundation

Los Alamos
Scientific Laboratory

1971

1978 United States Uranium Registry (USUR)
Hanford Environmental Health Foundation

1978

1992 United States Transuranium and Uranium Registries (USTUR)
College of Pharmacy, Washington State University

1989



Mission: Then and now

- Initial mission was “to protect the interests of workers, employees, and the public by the acquisition and provision of the latest and most precise information about **the effects of the transuranic elements** on man”

By the end of 1970s, the mission of the Registries moved **away** from **epidemiology**

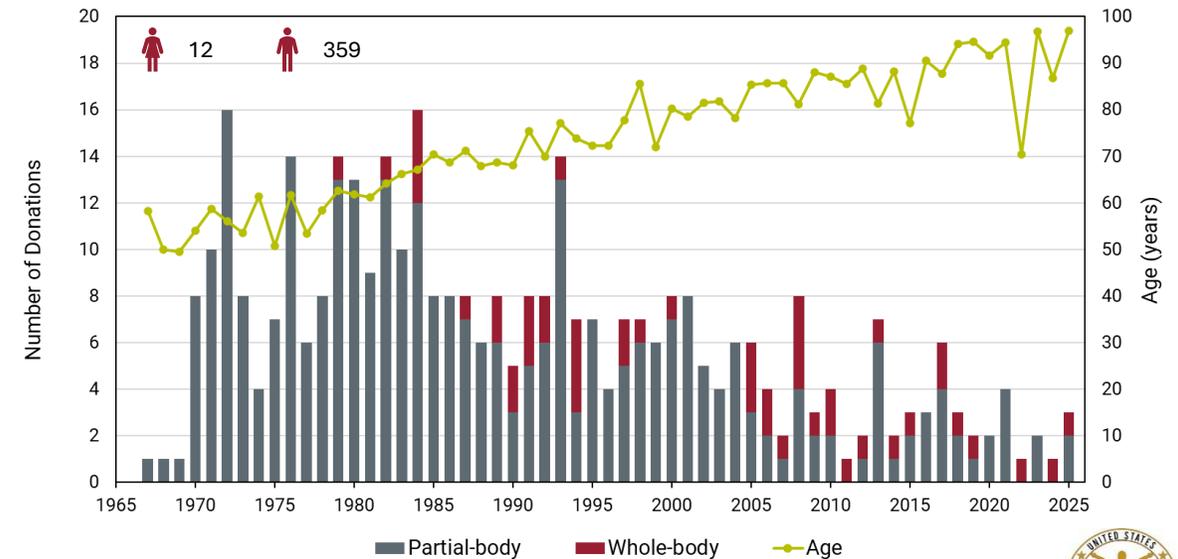
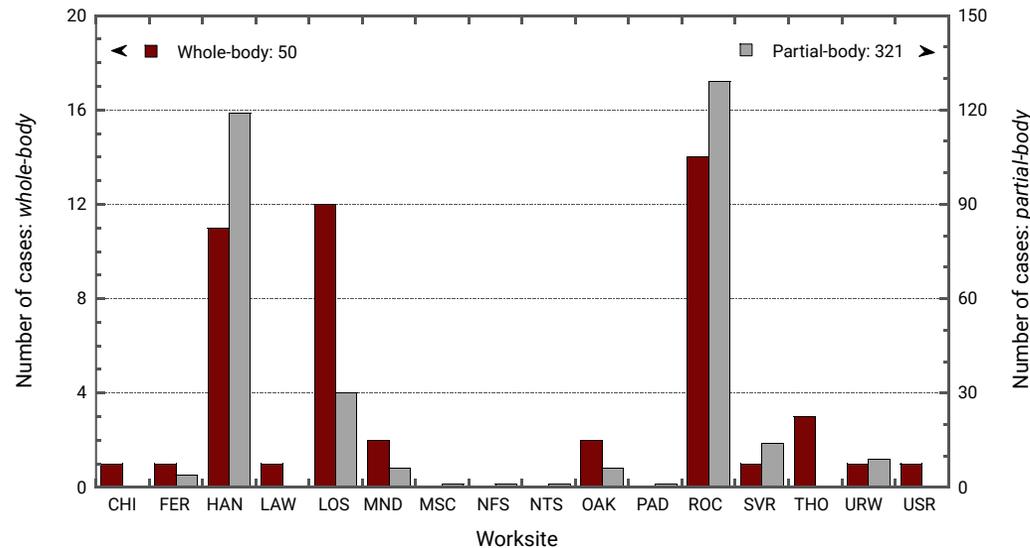
- Today’s mission is to follow up occupationally exposed workers by **studying the biokinetics and tissue dosimetry of the actinides** to refine dose assessment methods **in support of reliable epidemiological studies**, radiation risk assessment, and regulatory standards **for radiological protection** of workers and general public



Registrants

Individuals with documented history of exposure to the actinides

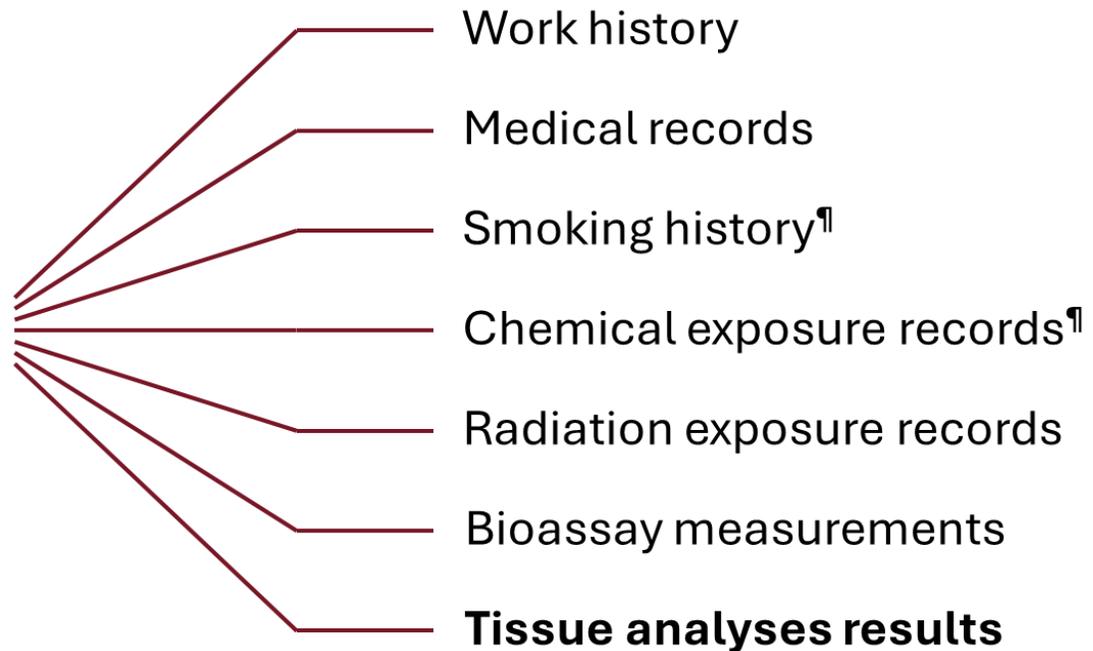
- Primary exposure: ^{239}Pu , ^{238}Pu , ^{241}Am , U_{nat} , HEU, DU, ^{244}Cm , ^{237}Np
- Criteria: 4 nCi (internal deposition) or 10 rem (external)
- Mainly former nuclear workers from DOE sites
- Voluntary tissue donors (posthumous)
- Statistics and trends:



Unique data resource



¶ - self-reported



National Human Radiobiology Tissue Repository (NHRTR)



USTUR research: *frozen and acid-digested tissues (acid solutions)*

Radium Dial Workers study: *frozen, plastic-embedded, and dry tissues*

Plutonium Injection study: *plastic-embedded bones*

(†)

Los Alamos Environmental study: *acid solutions*

(‡)

† - from Argonne National Laboratory

‡ - from Los Alamos National Laboratory



This study: Case description

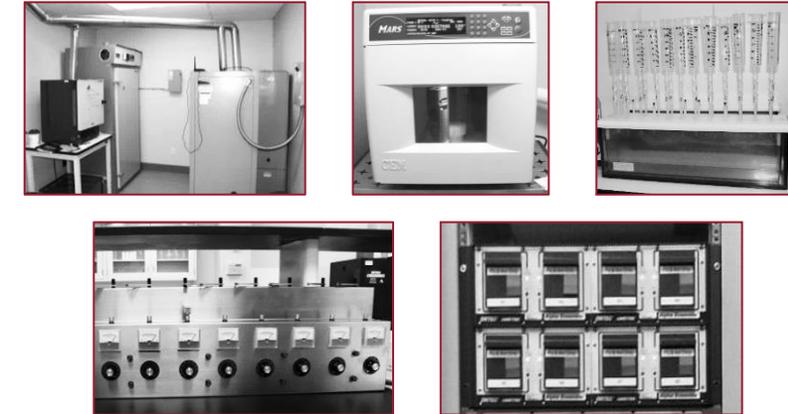
Individual with documented history of exposure to ^{239}Pu and ^{244}Cm

- Partial-body tissue donor worked with actinides for 33 years
- 44-year bioassay monitoring (Pu/Am/U): urine, feces, chest/wound counts
- Major inhalation and wound intake of soluble ^{239}Pu (*nitrite-chloride-acid mist*)
- Treated with 10 i.v. injections of 1.0-g Ca-DTPA
- **Acute inhalation of ^{244}Cm** (3 years after DTPA treatment)
- A **single** urine measurement **below detection limit** for ^{244}Cm
- Worksite estimated deposition, nCi: 8 (^{239}Pu), 4 (^{241}Am), **no estimate** (^{244}Cm)
- Died 52 years after ^{244}Cm intake from heart disease at age 86
- Ten bones and 18 soft tissue organs were collected at autopsy

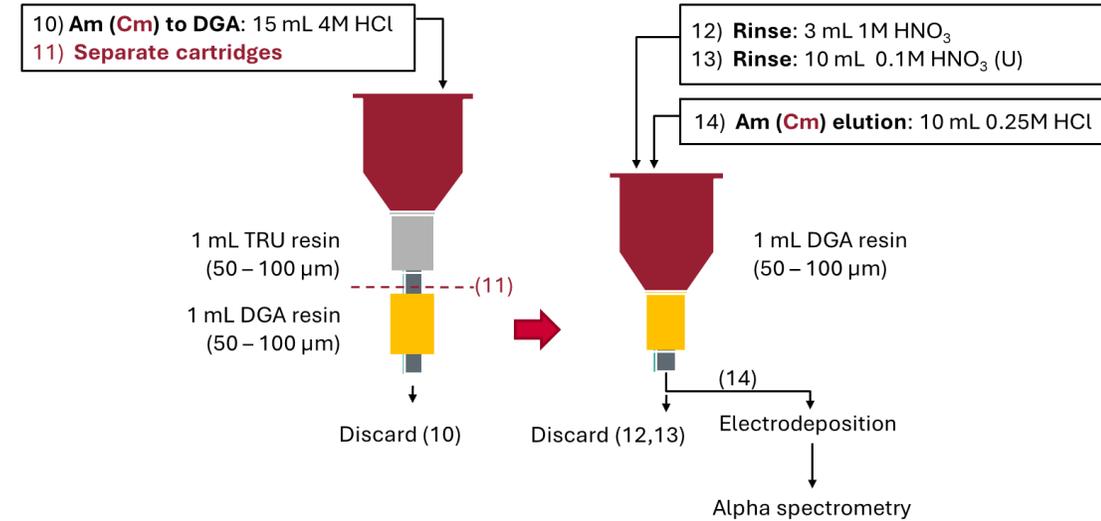
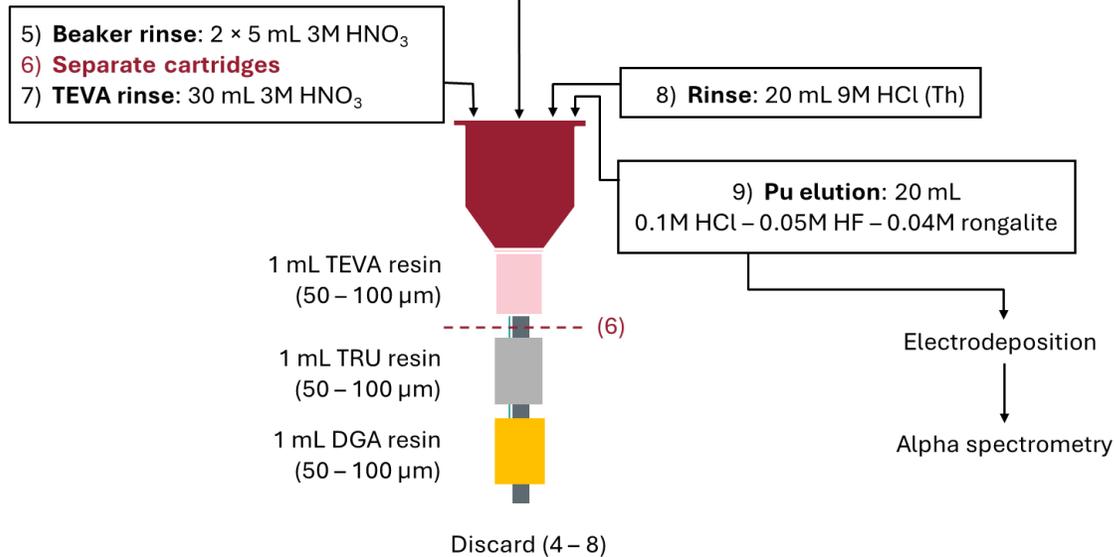


^{244}Cm analysis in tissue samples

- 50 tissue samples analyzed for Pu/Am/Cm



- 1) Sample in 40 mL 3M HNO_3 – 1M $\text{Al}(\text{NO}_3)_3$
- 2) Add 0.75 mL 1.5M sulfamic acid + 6 mL 0.75M ascorbic acid
- 3) Add 2.5 mL 3.5 M sodium nitrite
- 4) **Sample loading** (1 mL min^{-1})

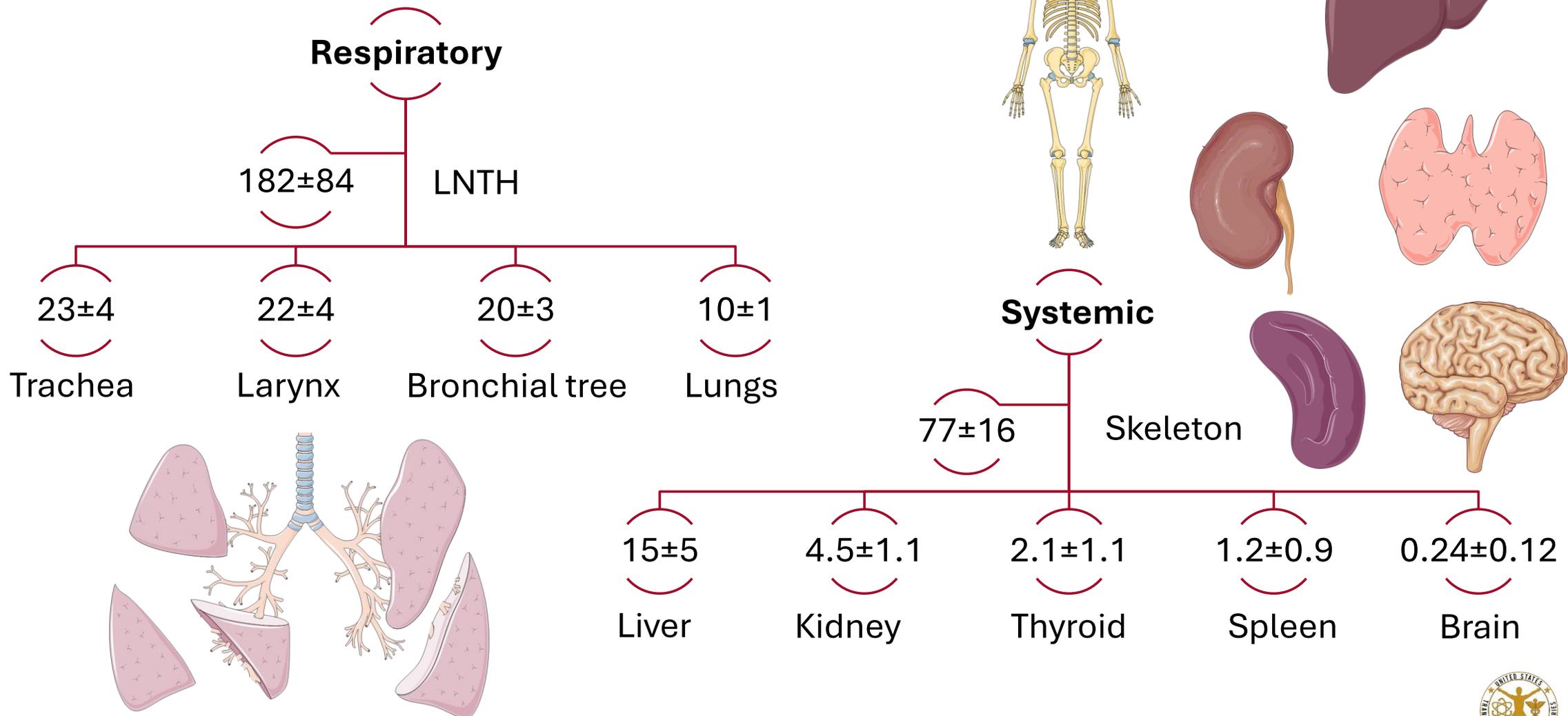


- ^{244}Cm measured by α -spectrometry for 600,000 sec ($s'_c = 6.8 \times 10^{-5}$ Bq)



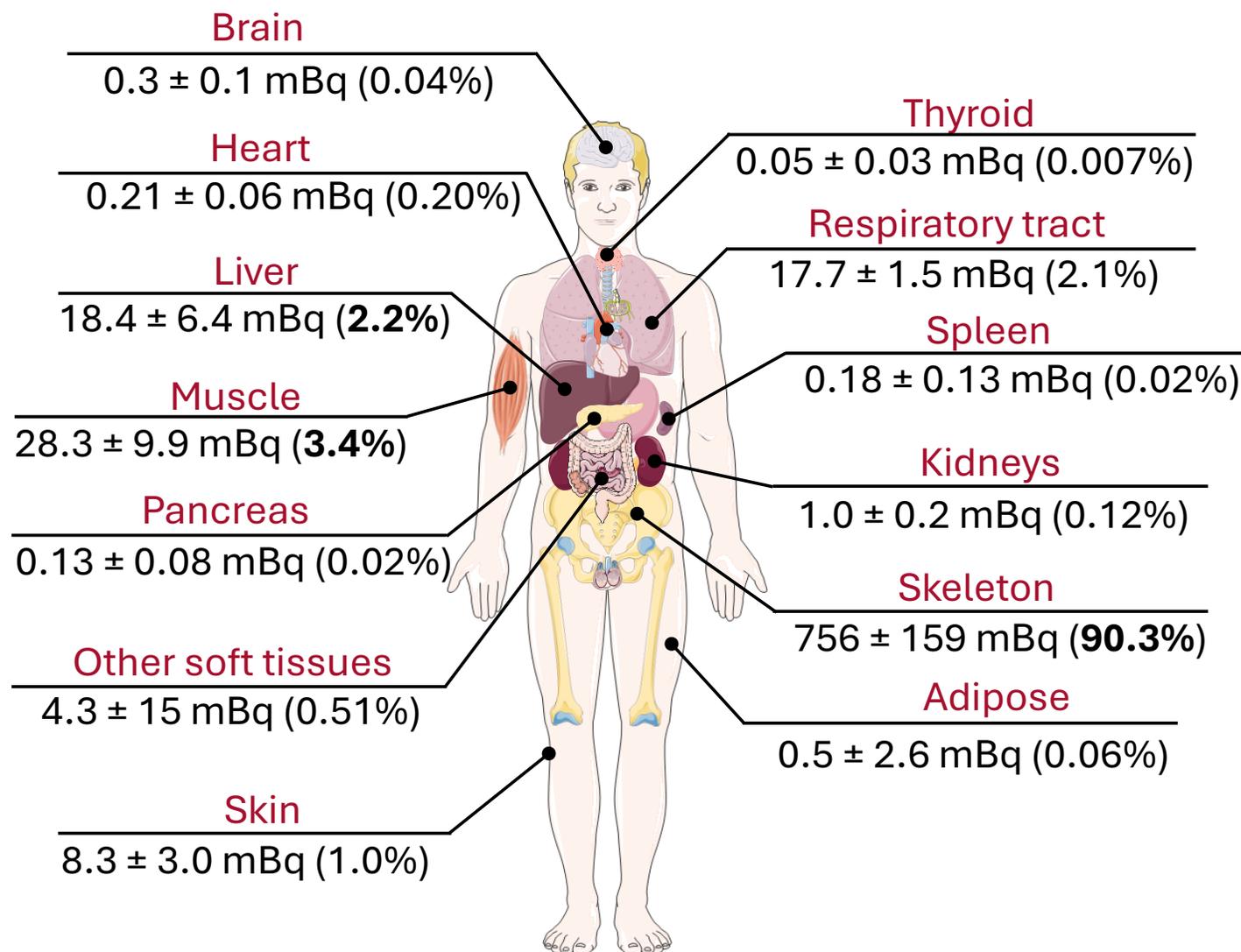
^{244}Cm concentrations in human tissues

- Total body concentration: $10.6 \pm 2.0 \text{ mBq kg}^{-1}$

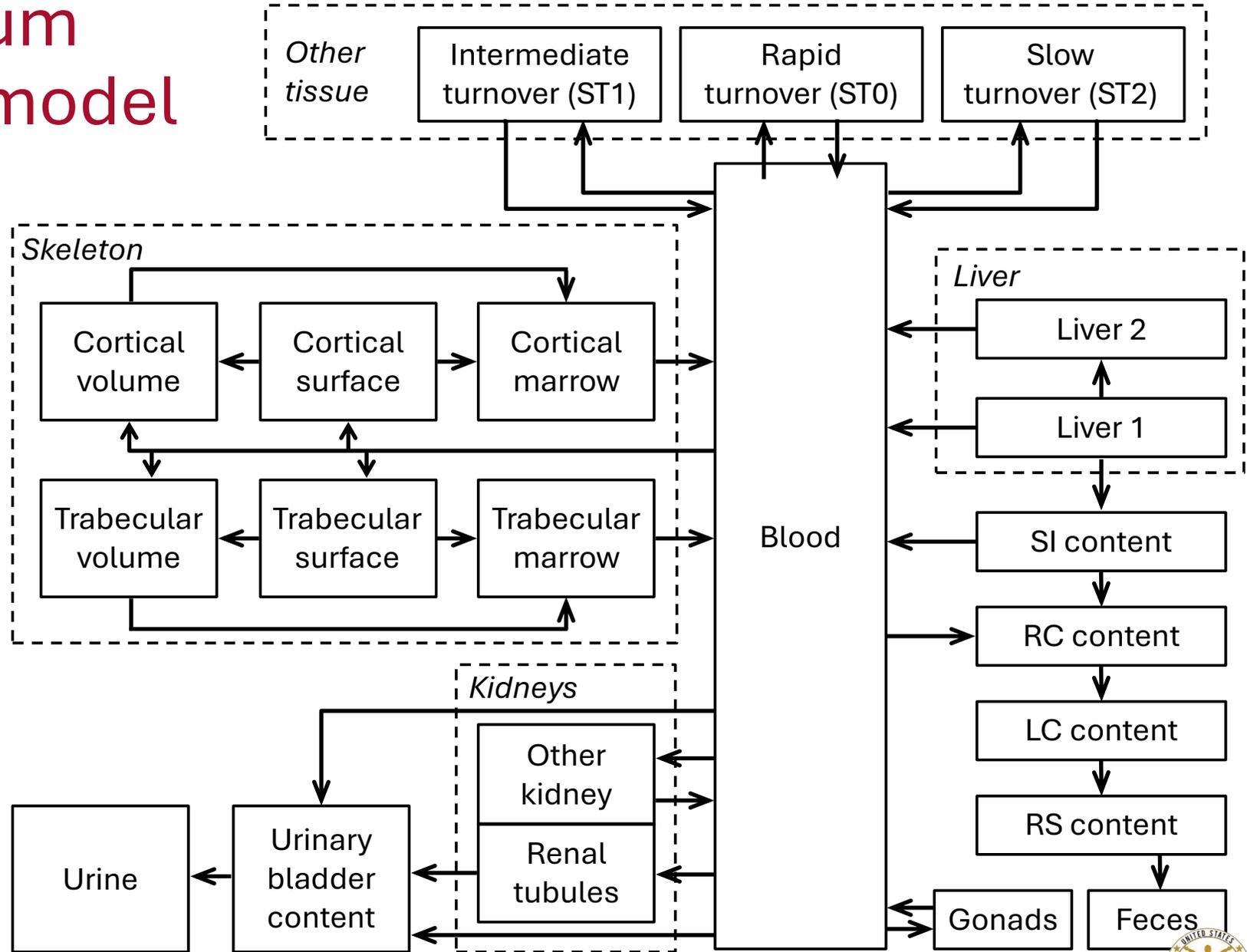
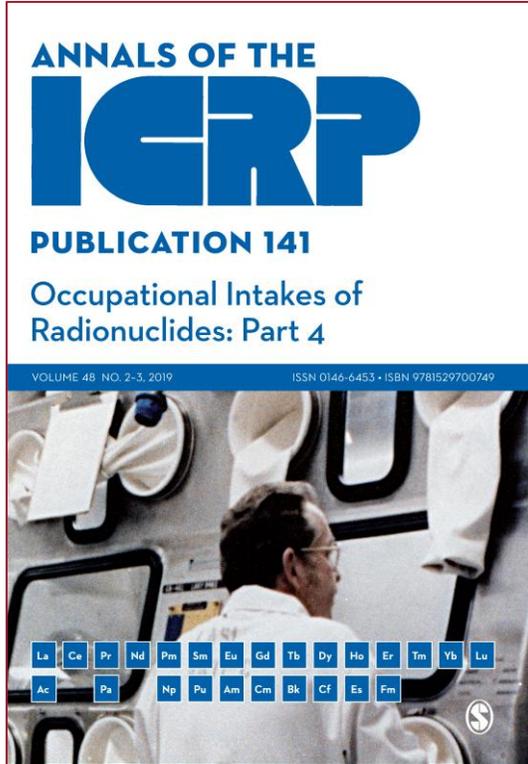


Curium distribution in the human body

- Estimated ^{244}Cm total body activity: 837 ± 160 mBq



ICRP: Curium biokinetic model



Autopsy data: Estimation of ²⁴⁴Cm intake and dose

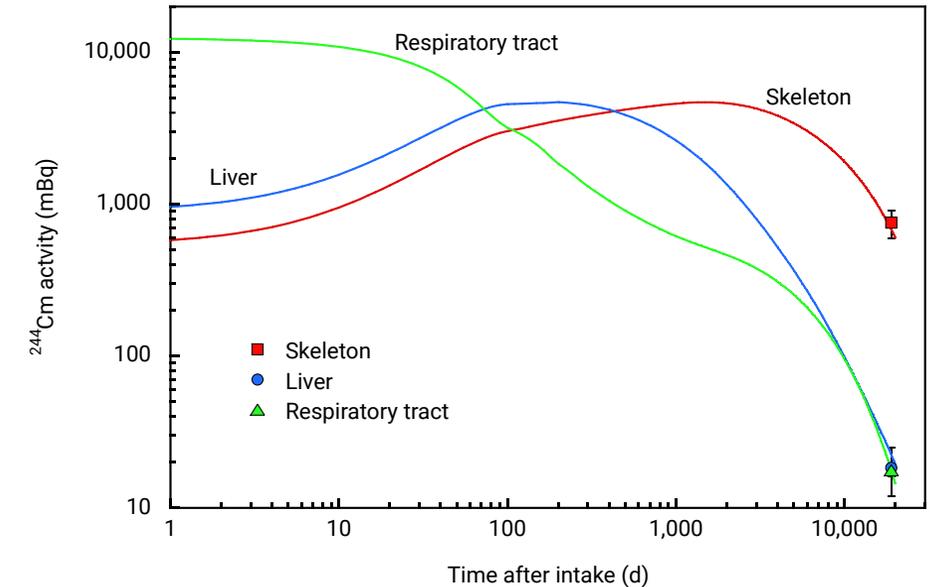
Input

Reference: Case_0641_Cm-244 R1
 Nuclide: Cm-244 18.10y
 Deposition parameters: Light work, 5.0 microns AMAD
 Absorption parameters: ICRP OIR series defaults

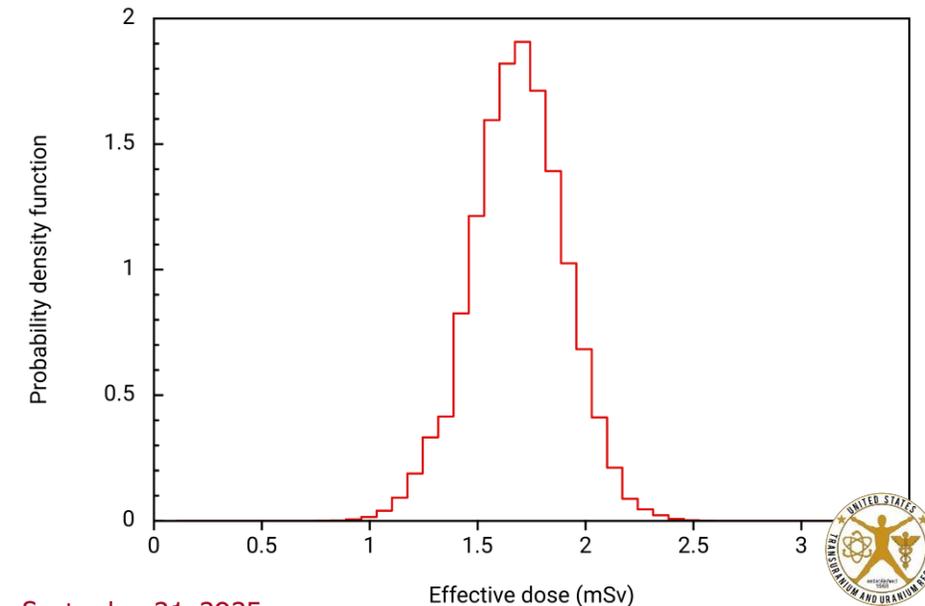
Systemic biokinetics: ICRP OIR series defaults
 Alimentary tract: ICRP OIR series defaults
 Respiratory tract: ICRP OIR series defaults
 Wound model: NCRP defaults

Intake regimes: Number of intake regimes (max. 20): 2
 Reference year: 1901, End year: 2022

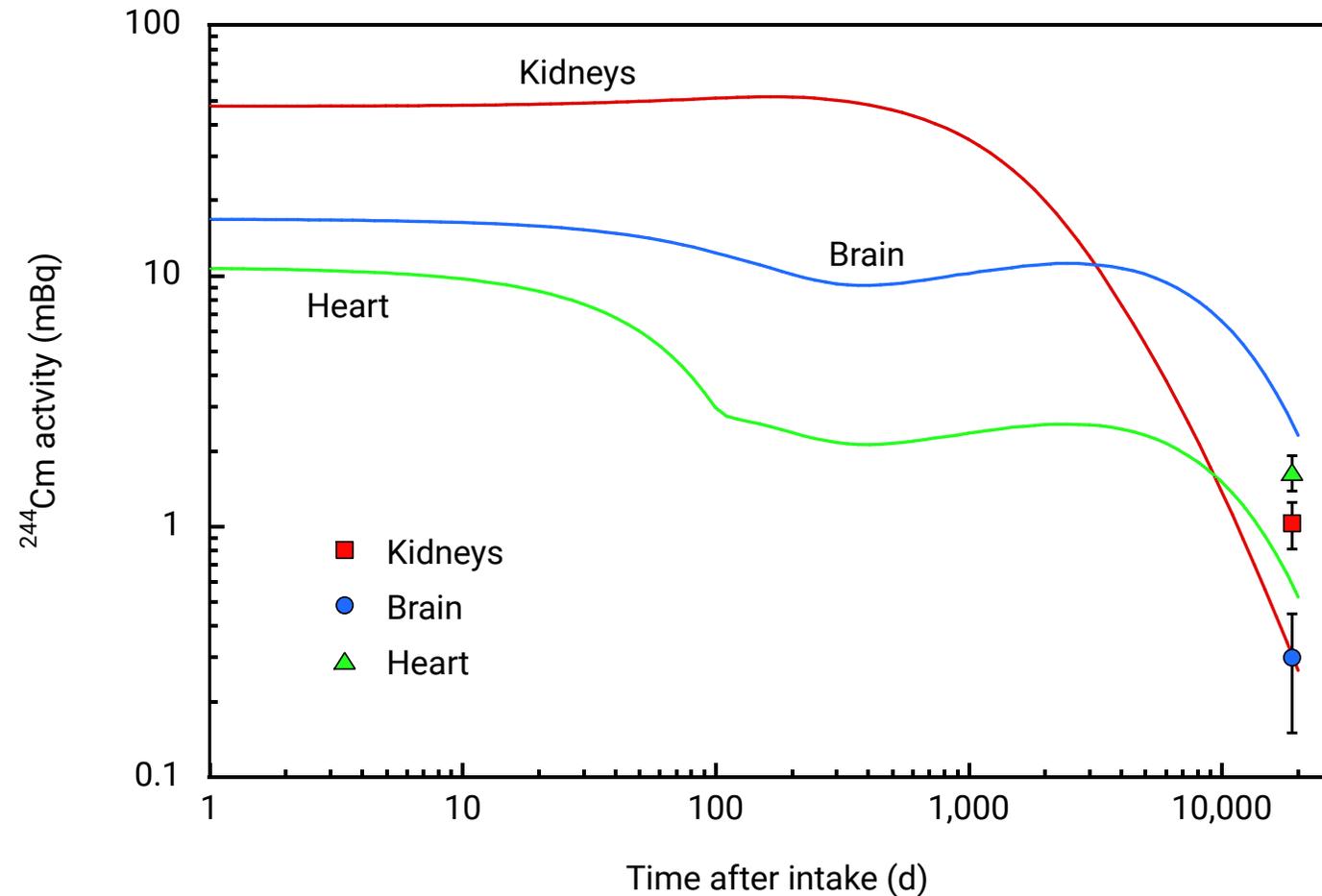
	LR	LRfrac	Dep	Form	Route	Mode	Start	End	Intake	fA	fr	sr	ss	fb
1	A	1.0000	5.0 um AMAD	CMO2	INH	Acute	0		1.9223E+02	2.50E-04	0.50000	0.40000	1.00000E-02	2.00000E-03
2	B	1.0000	5.0 um AMAD	S	INH	Acute	0		3.7625E+01	5.00E-06	1.00000E-02	0.40000	1.00000E-04	2.00000E-03



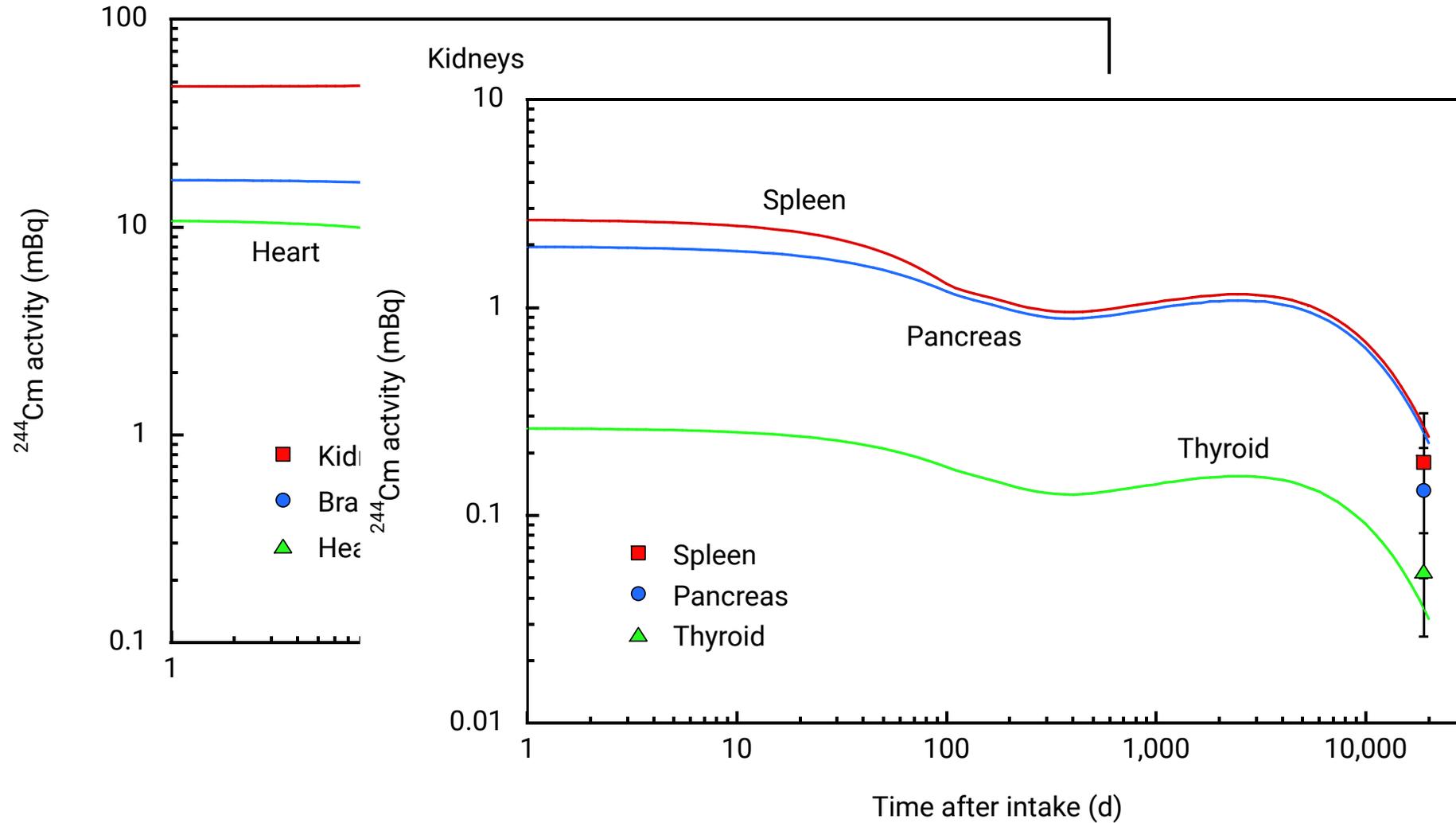
- Inhaled material: Curium oxide, nitrate, and chloride ('soluble') + type S (insoluble)
- Data: post-mortem organ activities
- Goodness-of-fit, $\chi^2 = 0.615$ ($NDF = 1$)
- Estimated intake: 266 ± 39 Bq (84% soluble fraction)
- Committed effective dose (mean): 1.62 ± 0.24 mSv



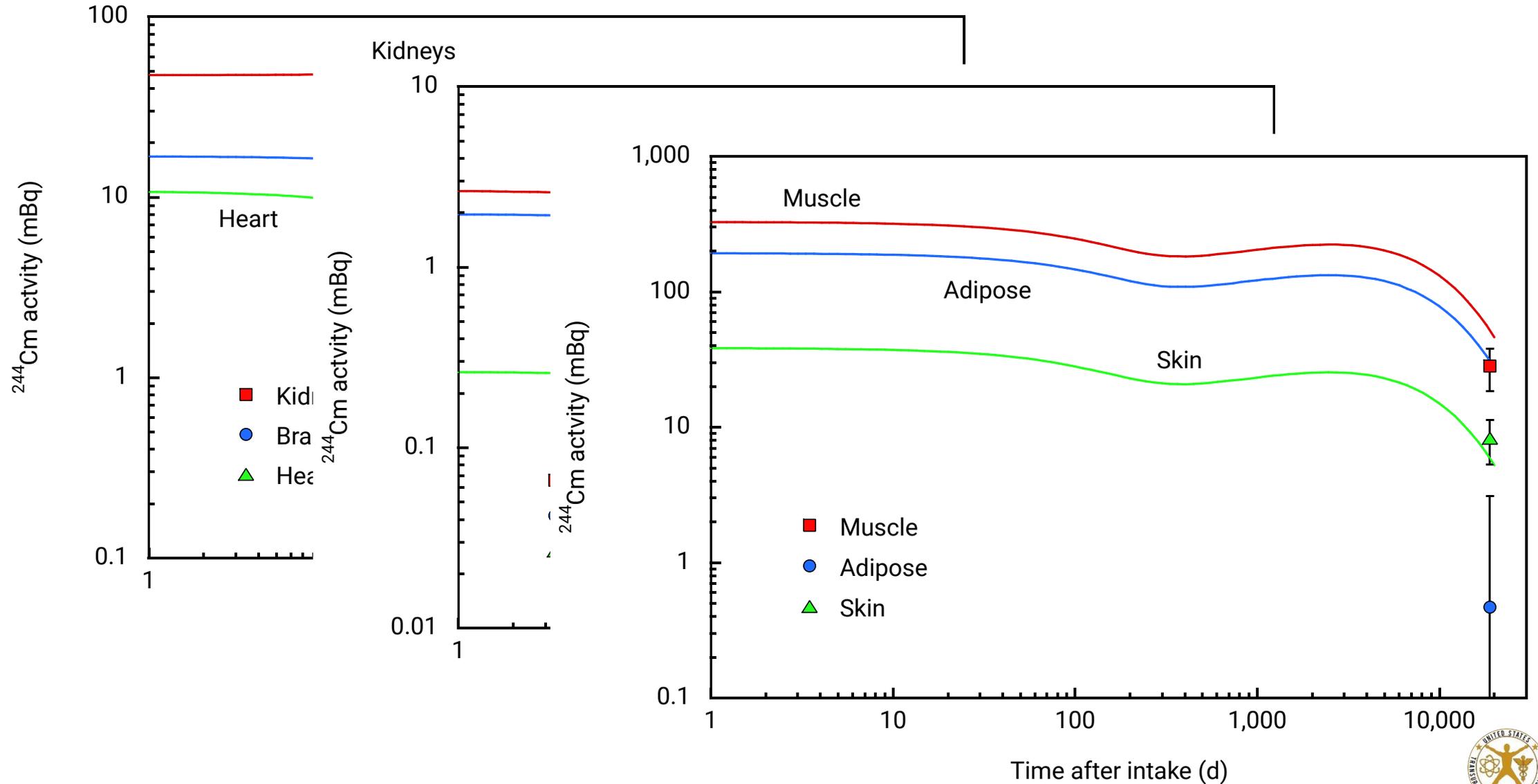
Model validation: Organ activity predictions



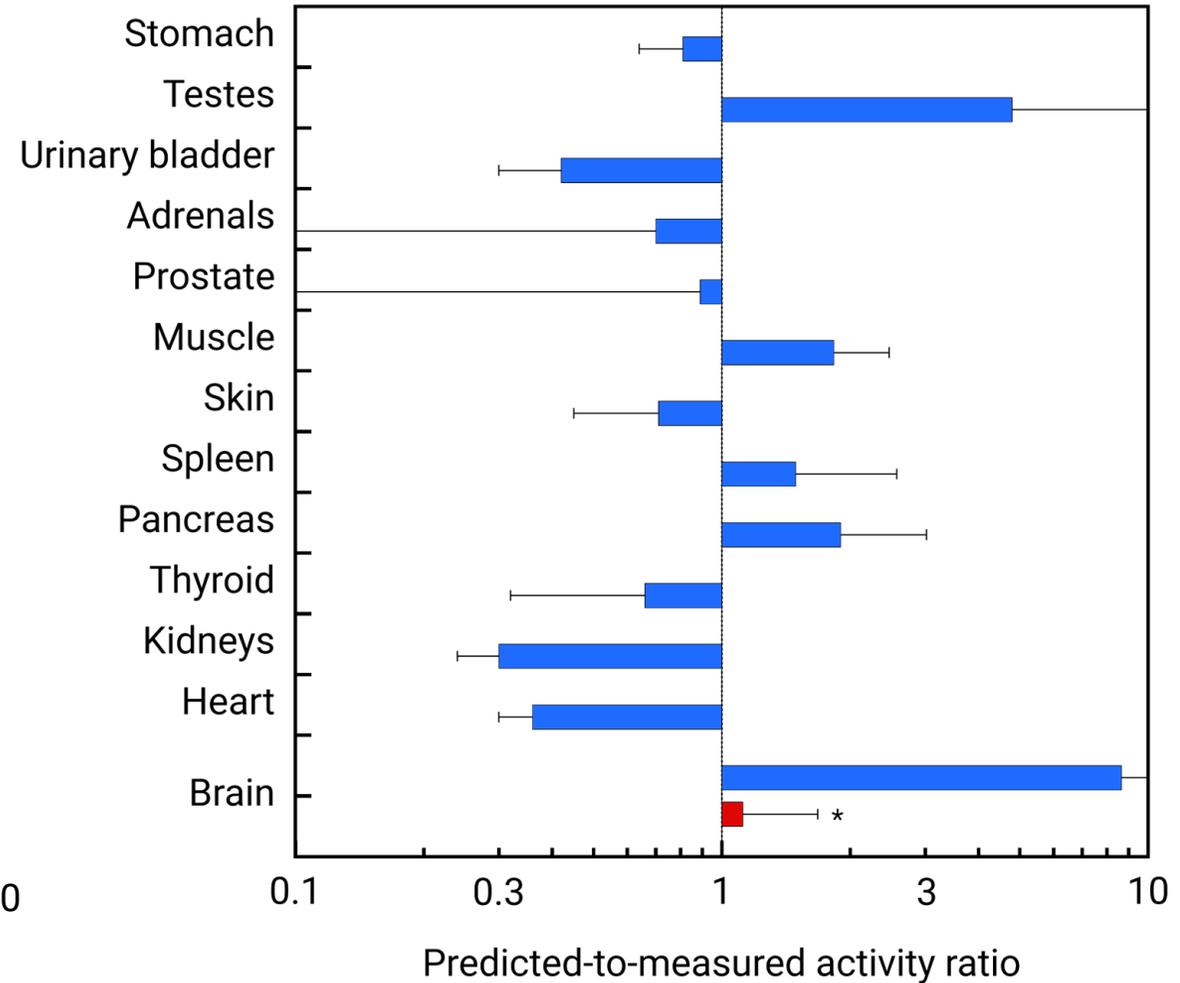
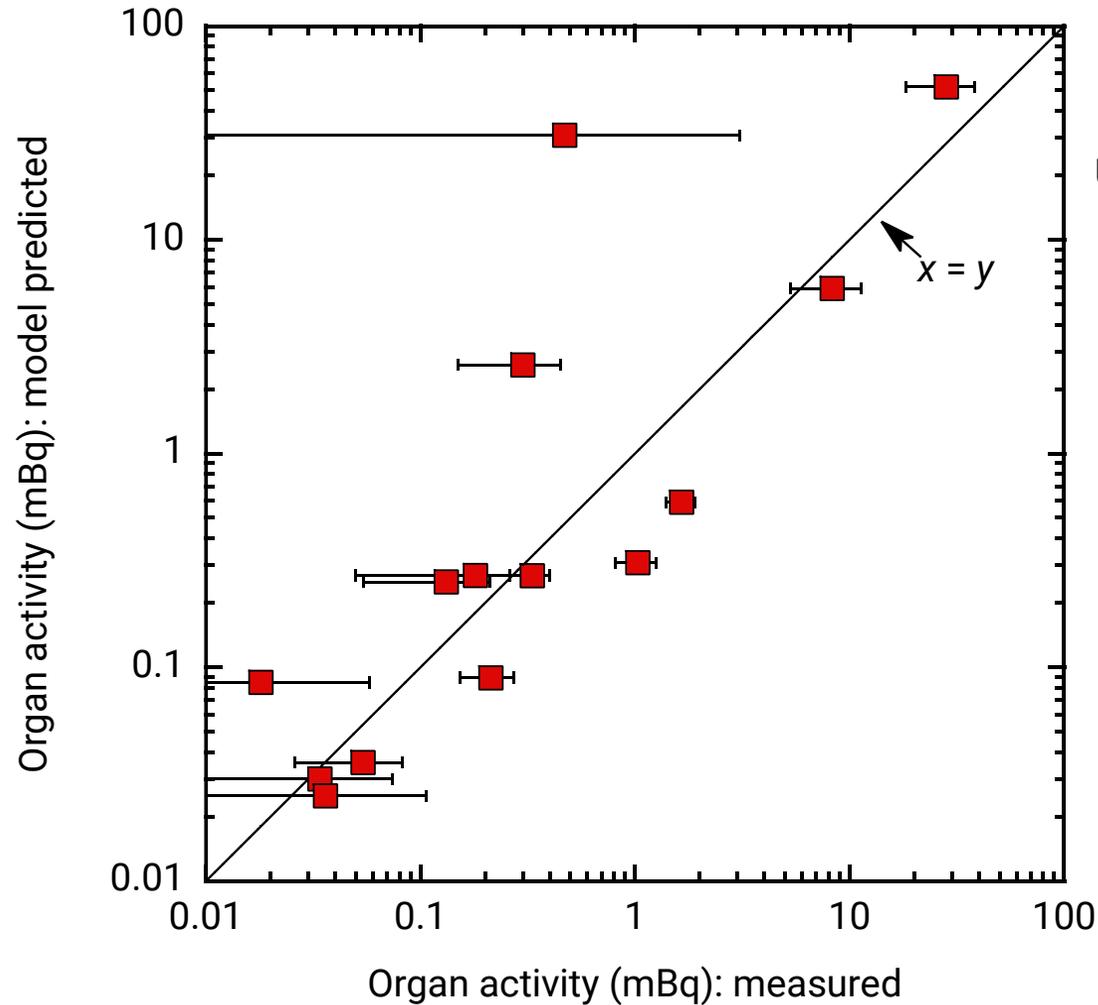
Model validation: Organ activity predictions



Model validation: Organ activity predictions



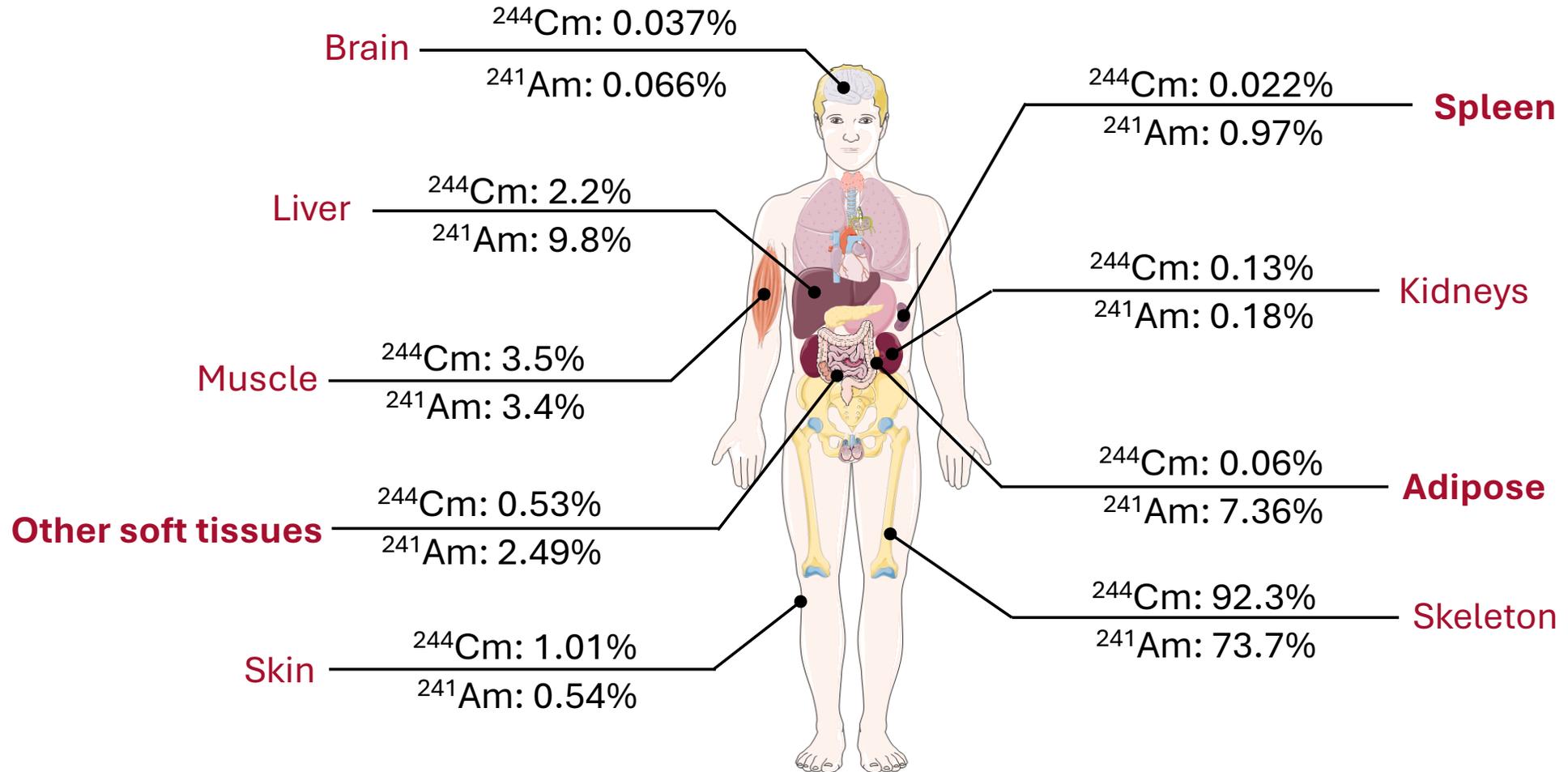
Oran activity: Model prediction vs measurement



* using NCRP 31 'explicit brain' model



Cm and Am 'systemic' distribution: Comparison



Skeleton-to-liver activity ratio (Bq/Bq)

- ²⁴⁴Cm: 41±17 (measured) – 30 (model predicted)
- ²⁴¹Am: 8±2 (measured) – 33 (model predicted)



Summary

- The most extensive **case study** on the biokinetics, tissue concentrations, and organ distribution of curium (^{244}Cm) in the human body
- The total body concentration of ^{244}Cm was estimated at **$10.6 \pm 2.0 \text{ mBq kg}^{-1}$** . Among **systemic tissues**, the highest concentration was in the **skeleton** followed by the **liver, kidney, spleen, and brain**
- **92.3%** of systemic ^{244}Cm is deposited in the **skeleton**, followed by **3.5%** in **muscle** and **2.2%** in the **liver**; for ^{241}Am , **73.7%** is retained in the **skeleton**, with **9.8%** in the **liver**, and **7.4%** in the **adipose tissue**
- Estimated **intake** of ^{244}Cm was **$268 \pm 39 \text{ Bq}$** with corresponding **committed effective dose** of **$1.62 \pm 0.24 \text{ mSv}$**
- Overall, current **biokinetic model for actinides** predicts the distribution of ^{244}Cm in the human body reasonably well. The observed **skeleton-to-liver** activity ratio is **41**, compared to a predicted ratio of **30** from the model





Thank you for your attention!



¹⁶ S Sulfur 32.07	⁶⁸ Er Erbium 167.26	³² Ge Germanium 72.63	³⁹ Y Yttrium 88.905
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