**EURADOS Annual Meeting**
**Porto, Portugal**
**June 12-15, 2023**

# EURADOS/REMPAN Wound Contamination Project

## Annex 1: Example of Plutonium Contaminated Wound

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# USTUR Whole-body Case 0262

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## USTUR WHOLE BODY CASE 0262: 33-Y FOLLOW-UP OF $PuO_2$ IN A SKIN WOUND AND ASSOCIATED AXILLARY NODE

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**This whole body donation case (USTUR Registrant) involved two suspected $PuO_2$ inhalation intakes, each indicated by a measurable Pu alpha activity in a single urine sample, followed about $1\frac{1}{2}$ y later by a puncture wound to the thumb while working in a Pu glovebox. The study is concerned with modelling simultaneously the biokinetics of deposition and retention in the respiratory tract and at the wound site; and the biokinetics of Pu subsequently transferred to other body organs, until the donor's death. Urine samples taken after the wound incident had readily measurable Pu alpha activity over the next 14 y, before dropping below the minimum detectable excretion rate (<0.4 mBq d$^{-1}$). The Registrant died about 33 y after the wound intake, at the age of 71, from hepatocellular carcinoma with extensive metastases. At autopsy, all major soft tissue organs were harvested for analysis of their $^{238}$Pu, $^{239+240}$Pu and $^{241}$Am content. The amount of $^{239+240}$Pu retained at the wound site was 68 ± 7 Bq (1 SD), measured by low-energy planar Ge spectrometry. A further 56.0 ± 1.2 Bq was retained in an associated axillary lymph node, measured by radiochemistry. Simultaneous mathematical analysis (modelling) of all *in vivo* urinary excretion data, together with the measured lung, thoracic lymph node, wound, axillary lymph node and systemic tissue contents at death, yielded estimated intake amounts of 757 and 1504 Bq, respectively, for the first and second inhalation incidents, and 204 Bq for the total wound intake. The inhaled Pu material was highly insoluble, with an estimated long-term absorption rate from the lungs of $2 \times 10^{-5}$ d$^{-1}$. The Pu material deposited at the wound site was mixed: ~14% was rapidly absorbed, ~49% was absorbed at the rate of about $6 \times 10^{-5}$ d$^{-1}$, and the remainder (~37%) was absorbed extremely slowly (at the rate of about $5 \times 10^{-6}$ d$^{-1}$). Thus, it was estimated that only ~40% of the Pu initially deposited in the wound had been absorbed systemically over the 33-y period until the donor's death. The biokinetic modelling also indicated that, in this individual case, some of the parameter values (rate constants) incorporated in the ICRP Publication 67 Pu model were up to a factor of 2 different from ICRP's recommended values (for reference man).**

### INTRODUCTION

The United States Transuranium and Uranium Registries (USTUR) maintains an extensive and growing database of health physics information and measured actinide content of tissues for former workers who were known to have had intakes of actinide elements during their employment in the nuclear industry. These registrants had volunteered to donate tissues after their death for radiochemical analyses and biokinetic modelling studies. Case 0262, the subject of this study, is of particular interest since his intakes were well documented, and they occurred as two discrete (acute) inhalation events, with a further intake via a cutaneous puncture wound. Case 0262 is a whole body donation, for whom USTUR has analysed radiochemically the contents of $^{239+240}$Pu in all major soft tissue organs and individual bones comprising about half the skeleton, plus $^{238}$Pu and $^{241}$Am. The $^{238}$Pu and $^{241}$Am data, and biokinetic modelling of $^{241}$Am in-growth in tissues from $^{241}$Pu decay of the Pu material taken into the body will be reported elsewhere. This paper is concerned with modelling the systemic uptakes of Pu from both inhalations and the wound, and the subsequent long-term tissue retention of Pu.

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This gentleman worked as an engineer at the Hanford site, from 1951 to 82. He died in 1990, at the age of 71, from hepatocellular carcinoma with metastases to the diaphragm, lungs and liver. Incidental autopsy findings included degenerative arthritis of hip and shoulder joints, and diffuse degenerative changes of the lumbar spine (indicative of osteoporosis). At autopsy, the skin was taken from both hands in order to examine both for

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Paper

## REEVALUATION OF USTUR PLUTONIUM WOUND CASE 0262 USING BAYESIAN METHODOLOGY AND NEW DATA

Shane N. Weber,* Richard R. Brey,* and Anthony C. James†‡

***Abstract*—Skin penetration by radionuclide contaminants serves as a route of entry into the body and may pose a serious health risk to humans depending on the magnitude of intake. The United States Transuranium and Uranium Registry whole body Case 0262 was involved in a wound intake of plutonium at the Hanford Site. The registrant died about 33 years later. Results were initially reported in 2007 regarding the deposition and retention of plutonium in various tissues, including the wound site. However in 2009, an additional (previously unrecorded) sample of the wound tissue was located in the National Human Radiobiological Tissue Repository. The new sample was analyzed using inductively coupled plasma-mass spectrometry (ICP-MS), and the results were used to calibrate the measurement of emitted $^{239}$Pu x-rays from the original wound tissue sample made in 2007. In the present study, the analysis of $^{239}$Pu absorption rates from the wound and axillary lymph node from the initial study is repeated using the additional wound activity data and ICP-MS calibration. This new analysis is carried out using the Weighted Likelihood Monte Carlo Sampling (WeLMoS) method and code, which applies Bayesian inference to calculate the posterior probability distribution of intake and wound absorption parameters directly from the observed data and the assumed biokinetic model structure. The resulting central estimates of empirical wound absorption parameters and their associated uncertainties are here compared with the empirical values recommended in NCRP Report No. 156 for plutonium and with the maximum likelihood point estimates derived in the initial study from the Case 0262 data available at the time.
Health Phys. 103(3):286–300; 2012**

**Key words: biokinetics; dose, internal; dosimetry, internal; nuclear workers**

### INTRODUCTION

A breach of the skin poses a risk of internal deposition and translocation of radioactive material within the human body. Also, deposition of radioactive material at the wound site may be of concern because of the radiobiological effects on cells invoked to proliferate rapidly due to the physical injury. Tumor development at the wound site has been observed in several animal studies (Guilmette and Durbin 2003). There have been over 2,100 reported cases of skin injury involving radioactive contamination. Radionuclide-contaminated wound exposures were given a great deal of attention immediately after the Gulf War, where many soldiers were injured by depleted uranium shrapnel. More than 90% of radionuclide-contaminated wounds in the nuclear industry have involved injury to the hands (primarily the fingers) or arms. Most (almost 90%) of these wounds involved mechanical damage to the skin. The rest involved chemical or thermal burns (NCRP 2006). The NCRP Report 156 model focuses on the intake of actinides through skin punctures on the hands or arms because of the high proportion of intakes involving these scenarios.

An inadequate amount of human data unaffected by medical intervention (wound care, excision and/or chelation) is available to exhaustively validate the models of retention and translocation of radioactive material in contaminated wounds. Experimental animal data has been extrapolated to humans in the development of the currently recommended model for radionuclide-contaminated wounds (NCRP 2006). Intakes through wounds differ significantly from inhalation intakes because of the heightened pathophysiological response of nearby tissue. Inhalation and ingestion intakes are better understood than wound intakes because they are better described by normal biological processes. Wound intakes are usually harder to quantify than are inhalation and ingestion intakes because of the nature of the interruption of homeostasis often associated with wound intakes.

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# Case Description

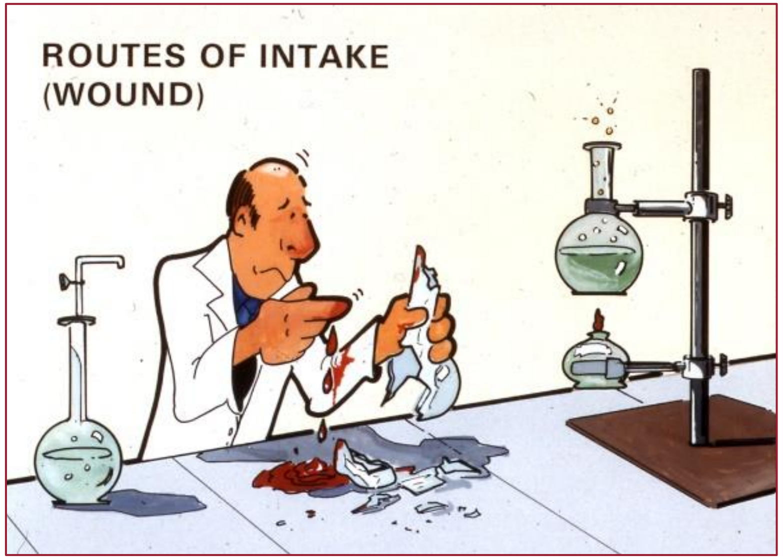
- Worked as an engineer at Nuclear Defense Facility for 31 years
- Routinely monitored for internal exposure to plutonium via bioassay measurement program: *urinalyses, in vivo chest counts*
- Several skin and workplace contamination incidents reported
- Two suspected plutonium inhalation intakes
- Major plutonium contaminated wound intake 1.5-y later
- Worksite estimate of $^{239}$Pu systemic deposition: 2.3 nCi (84 Bq)
- Died from hepatocellular carcinoma 33 y after wound intake

James AC, et al. (2007). Radiat. Prot. Dosim. 127(1-4): 114-119

Weber SN *et al*. (2012). Health Phys. 103(3): 286-300

# Contaminated Wound

- Injured finger on a broken drill while working in a glovebox
- Hood glove was contaminated to >40000 dpm (667 Bq)
- Laceration on left thumb: 0.6-cm long and 0.13-cm deep
- No apparent bleeding noted
- External measurement of the wound site: 500 dpm (8.3 Bq)
  - ✓ Decontaminated using pHisohex
- No chelation therapy and/or surgical excision attempted

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# Available Bioassay Data

- Five in vivo chest counts during last three years of employment: *all negative*
- Total of 86 Urine measurements over 30 years: *mostly positive after wound*

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# Autopsy Tissue Analyses

- Two samples of skin and muscle from left thumb containing the wound analyzed separately by γ-spectrometry and ICP-MS
- 83 soft tissue samples and 145 bones analyzed using α-spectrometry
- Analysis results published by James et al. (2007) and Weber et al. (2012)

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**Table 1. Measured $^{239+240}$Pu tissue contents.**

| Organ/tissue | Activity at death (Bq) |
|---|---|
| Wound site (left thumb) | 68 ± 7 (1 σ) |
| Left axillary lymph node | 56.0 ± 1.2 (1 σ) |
| Lungs, larynx, trachea | 2.6 |
| LNTH | 1.1 |
| Skeleton | 29.1 |
| Liver | 20.7 |
| Kidneys | 0.053 |
| Testes | 0.018 |
| All other soft tissues (total) | 8.6 |
| Spleen | 3.1 |
| Brain | 0.067 |
| Stomach wall | 0.44 |
| Small intestine wall | 0.056 |
| Large intestine wall | 0.008 |
| Total 'systemic' | 52.2 |

**REEVALUATION OF USTUR PLUTONIUM WOUND CASE 0262 USING BAYESIAN METHODOLOGY AND NEW DATA**

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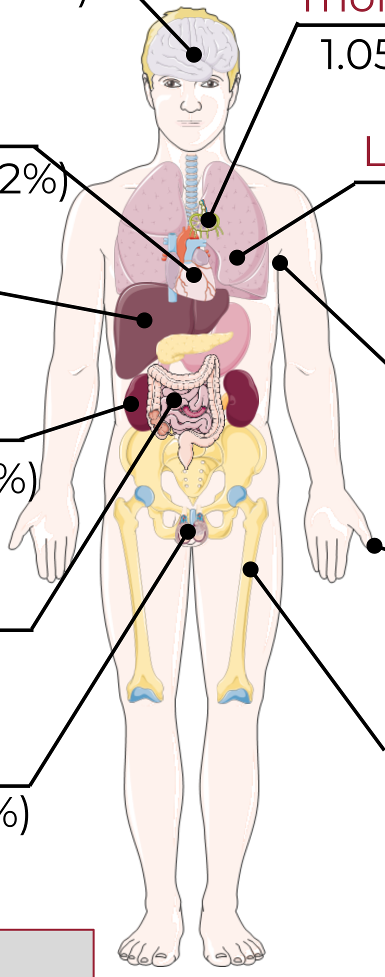
**Table 3.** Measured $^{239+240}$Pu tissue contents (adapted from James et al. 2007).

| Organ/tissue | Activity at death, Bq |
|---|---|
| Wound site (left thumb) | 103 ± 7 (1 σ) |
| Left axillary lymph node | 56 ± 1.2 (1 σ) |
| Lungs, larynx, trachea | 2.6 |
| Thoracic lymph nodes (LNTH) | 1.1 |
| Skeleton | 29.1 |
| Liver | 20.7 |
| Kidneys | 0.053 |
| Testes | 0.018 |
| All other soft tissues (total) | 8.6 |
| Spleen | 3.1 |
| Brain | 0.067 |
| Stomach wall | 0.44 |
| Small intestine wall | 0.056 |
| Large intestine wall | 0.008 |
| Total | 52.2 |

# Distribution $^{239}Pu$ in organs/tissues

**Whole-body activity:**
**221.8 ± 7.2 Bq**



Brain
0.067 ± 0.003 Bq (0.03%)

Heart
0.053 ± 0.003 Bq (0.02%)

Liver
20.7 ± 0.7 Bq (9.3%)

Kidneys
0.053 ± 0.003 Bq (0.02%)

Alimentary tract
0.57 ± 0.01 Bq (0.003%)

Testes
0.018 ± 0.002 Bq (0.0001%)

Thoracic lymph nodes
1.05± 0.04 Bq (0.5%)

Lungs+trachea+larynx
2.59 ± 0.06 Bq (1.2%)

Left axillary lymph nodes
56.0 ± 1.2 Bq (25.2%)

Wound (left thumb)
102.9 ± 7.1 Bq (46.4%)

Skeleton
29.6 ± 0.2 Bq (13.3%)

Remaining: 8.3 ± 0.1 Bq

Unpublished data revised from James et al. (2007) and Weber et al. (2012)

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# USTUR Case Evaluations

# James *et al.* 2007

- Multiple intake regimes: two inhalations and wound
- *Ad hoc* wound model, ICRP 66 HRTM & ICRP 67 Pu systemic model
- Birchall and James (1989) 'rate matrix' to solve compartmental models
  - ✓ Selected model parameters optimized
- Simultaneous fit to all urine and tissue retention data
  - ✓ Measured wound activity: 68 Bq – 51% lower than actual retention
- Estimated intakes:
  - ✓ Inhalations – insoluble Pu: (1) 757 Bq and (2) 1504 Bq
  - ✓ Wound – 86% insoluble & 14% soluble Pu: 204 Bq
- Total committed effective dose: 35 mSv

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# Weber *et al.* 2012

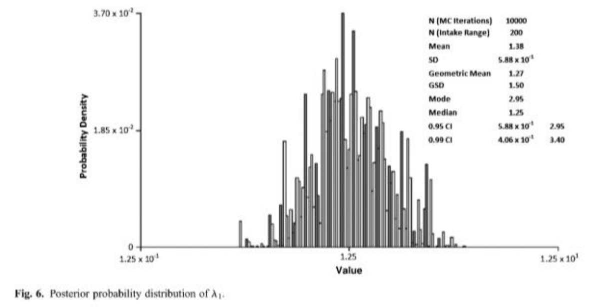


**Fig. 6.** Posterior probability distribution of $\lambda_1$.

- Single wound intake assumed
- NCRP 156 wound model & ICRP 67 Pu systemic model
- IMBA Professional Plus®: Maximum likelihood fit to urinary excretion
  - ✓ Deposition category 'Fragment' best described data
  - ✓ Model underestimated long-term wound and whole-body retention
- IMBA Uncertainty Analyzer: <u>We</u>ighted <u>L</u>ikelihood <u>Mo</u>nte Carlo <u>S</u>ampling
  - ✓ Posterior mean wound retention function:

    $R(t) = 0.0342e^{-1.38t}+0.117e^{-0.0112t}+0.849e^{-0.00000269t}$
  - ✓ Estimated posterior mean values (95% CI):

    Intake: 222 ± 6 Bq (210 Bq – 234 Bq)

    CED: 19.0 mSv (18.8 mSv – 52.3 mSv)

*Paper*

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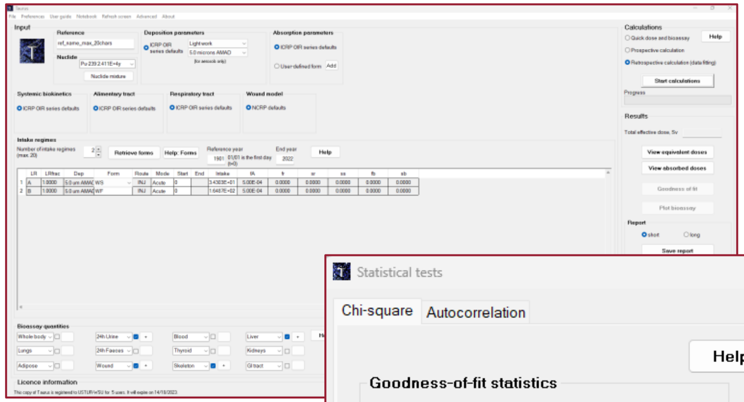
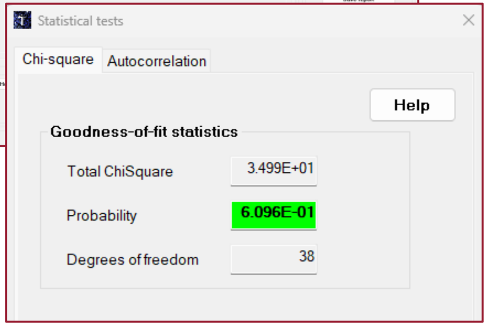
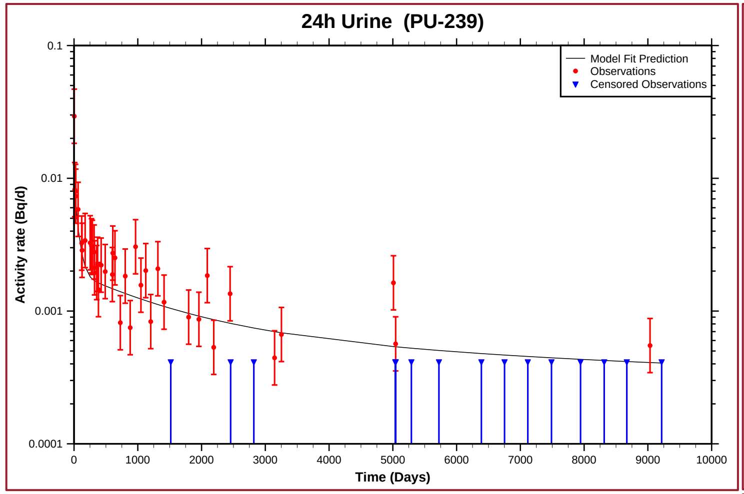
# Current Work (I)

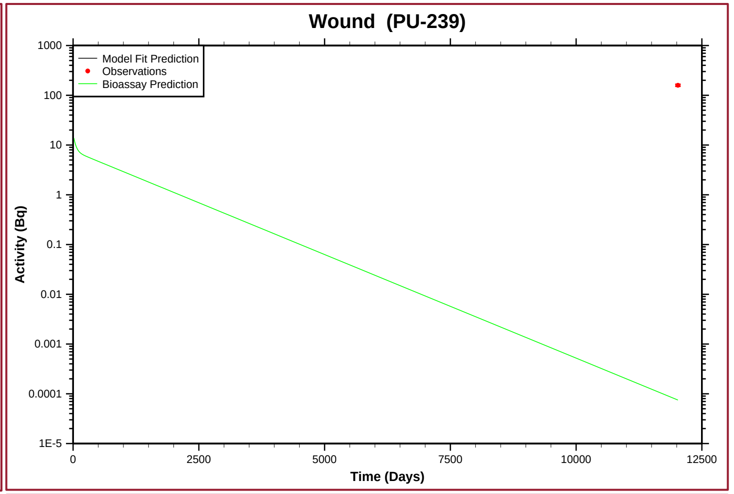
TAURUS internal dosimetry software, Version A_0.1

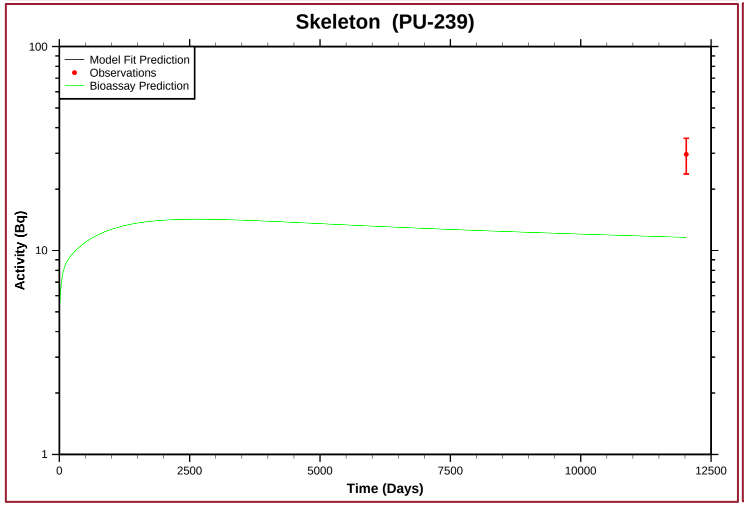
- Strongly retained soluble Pu assumed
- Urine bioassay only
- Intake: 34.4 Bq
- CED: 8.45 mSv

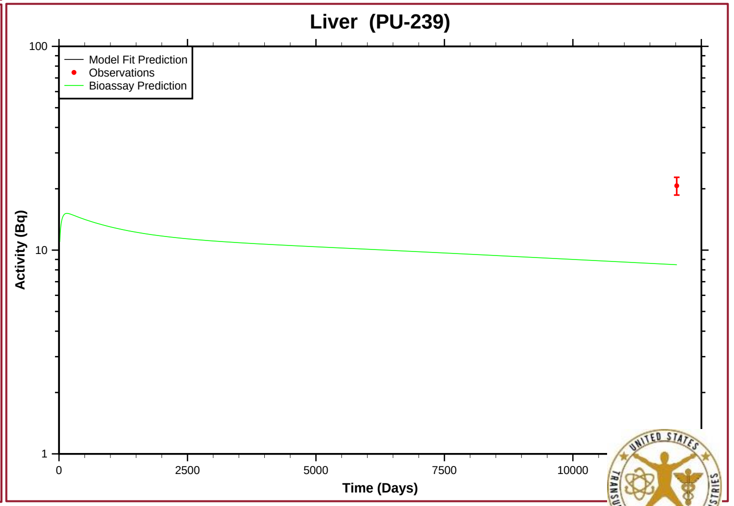




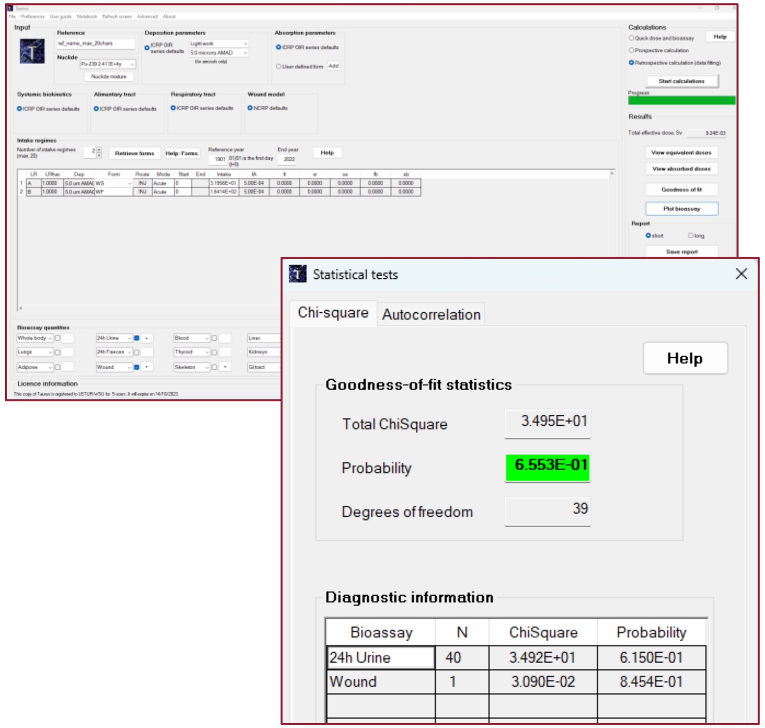



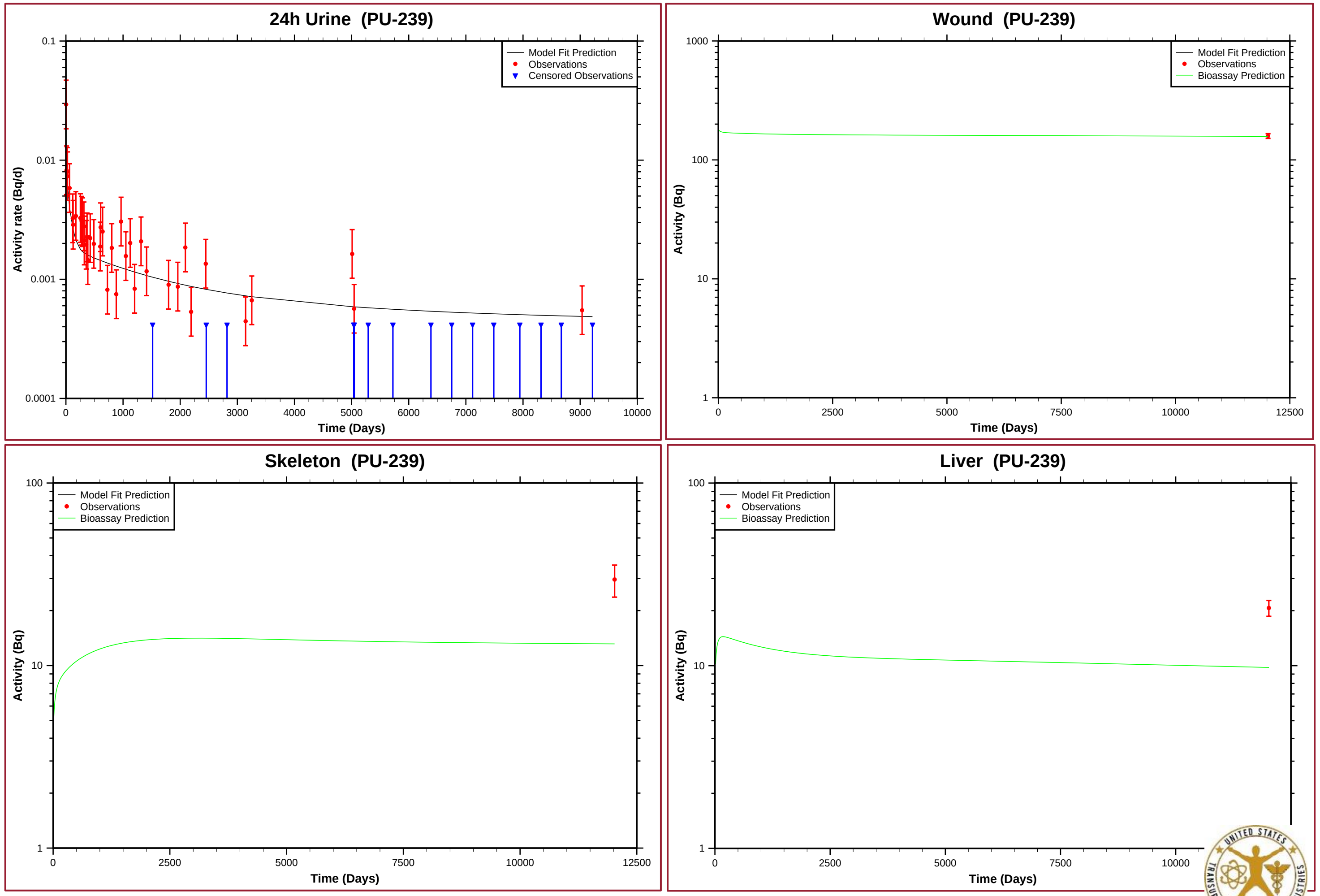



# Current Work (II)

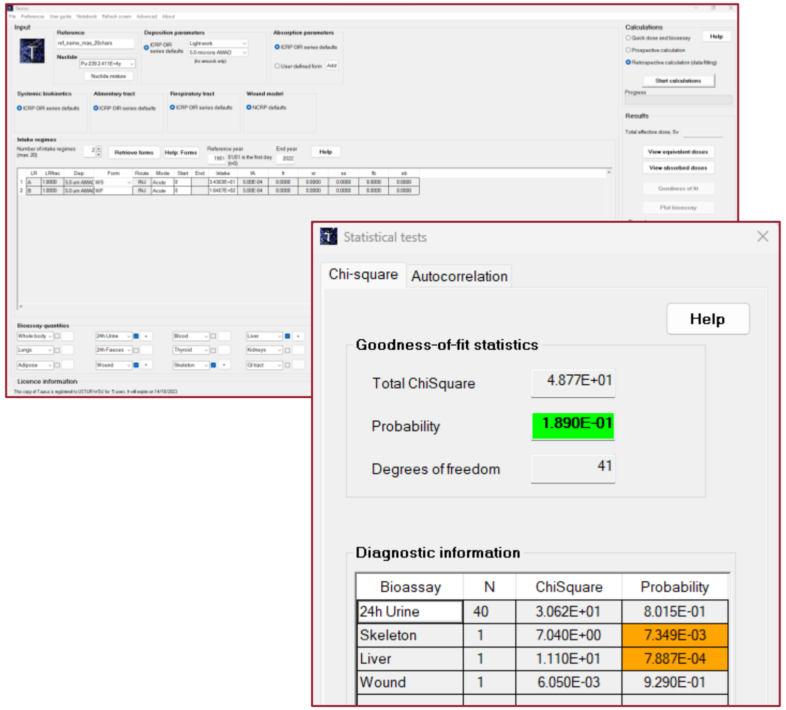
- Simultaneous fit to urine & post-mortem wound retention
- Mixture: 16.3% 'Soluble Strong' + 83.7% 'Fragment'
- Intake: 196.1 Bq
- CED: 9.2 mSv

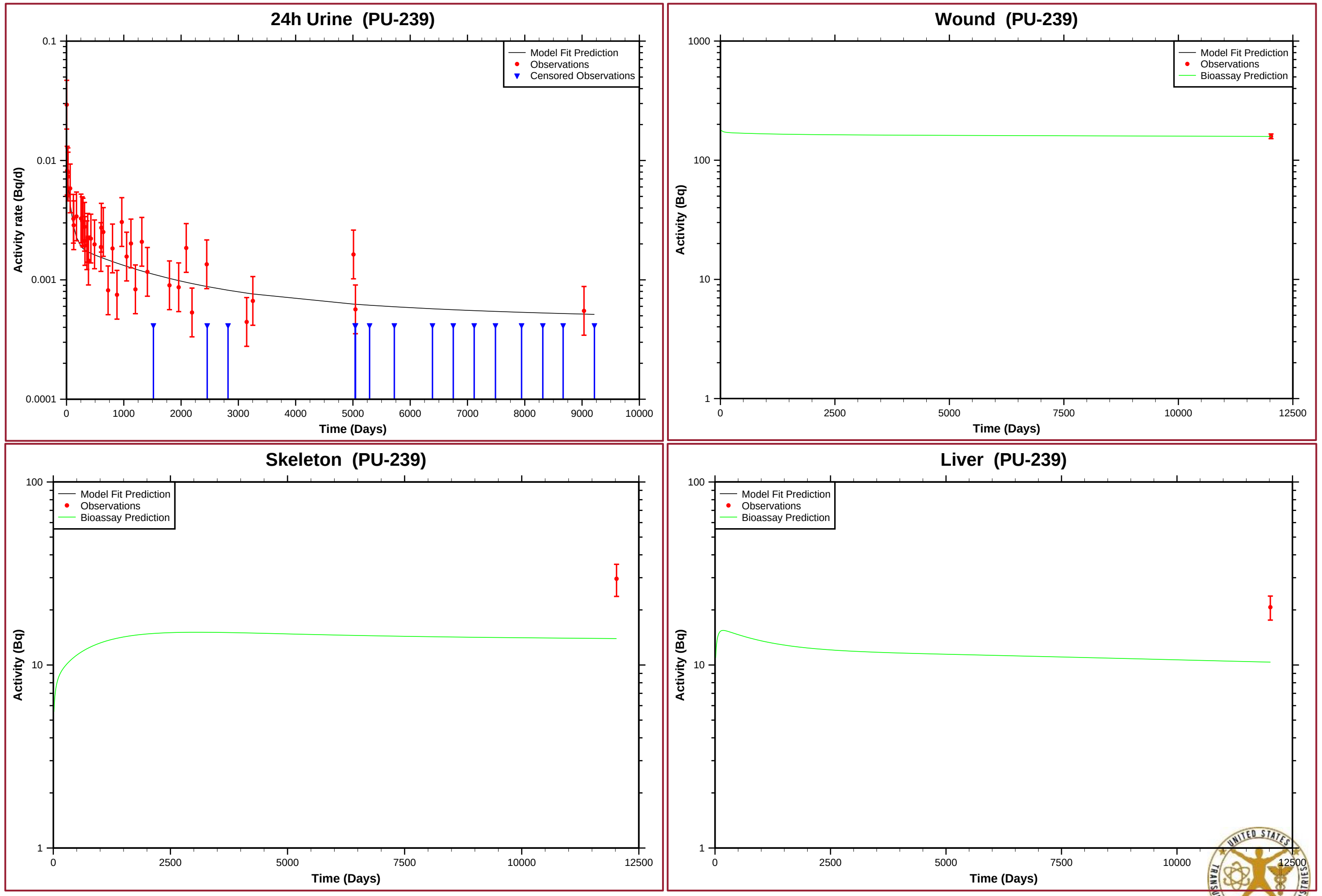


# Current Work (III)

- Simultaneous fit to urine & post-mortem wound, liver, skeleton
- Mixture: 17.2% ‘Soluble Strong’ + 82.8% ‘Fragment’
- Intake: 199.2 Bq
- CED: 9.8 mSv



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# Summary

EURADOS/REMPAN Wound Contamination Project, Annex 1

- USTUR whole body Case 0262
- Plutonium contaminated laceration
- No surgical excision or chelation
- 33 years of follow-up: urine bioassay, tissue retention data
- Two case evaluation studies published
- Final data analysis in progress

# QUESTIONS?

https://brand.wsu.edu/imagery/

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