

**PNNL-SA-55009**

**“Where Did This Come From?”  
or  
Lessons Learned from High Routine  
Bioassay Investigations**

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# Where'd this come from?

- ▶ Fluor Hanford PHMC Lessons Learned Information Bulletin, 2006-RL-HNF-0052

“Awareness of Historical Information During Internal Dose Evaluations”

Robin Hill, December 6, 2006

# Bioassay

- ▶ Poor Man's Anti-Bioassay
  - Breathing air sample
  - Work place indicators
  
- ▶ Direct Measurements (In Vivo)
  - Whole Body Counts
  - Chest Counts
  
- ▶ Indirect Measurements (In Vitro, Excreta)
  - Urine sample analysis
  - Fecal sample analysis

# Some Typical Monitoring Programs

- ▶  $^{137}\text{Cs}$  – annual whole body exam
- ▶  $^{90}\text{Sr}$  – annual or biennial urinalysis
- ▶ Pu Mixtures – annual Pu urinalysis  
annual chest count for  $^{241}\text{Am}$
- ▶ Routine vs Special bioassay
- ▶ Investigate “High Routines”

# What's a “High Routine”

- ▶ A routine bioassay that exceeds a normal result screening level.

Screening levels are established for routine measurements based on one or more of the following criteria:

- Intake(s) may result in >10-mrem CEDE for a CY (e.g., tritium,  $^{90}\text{Sr}$ )
  - A result suggesting the abnormal presence of radioactivity (e.g., >background U, detection of Pu)
- 
- ▶ High routines suggest something may be amiss.

# What's Detection?

- ▶ A result greater than the decision level for determining a difference from background.
- ▶ The decision level (DL) is also known as the critical level ( $L_c$ ) for detection.
- ▶ The DL is NOT the Minimum Detectable Amount.
  - MDA is based on a 95% probability of seeing something that is there at the given level.
  - The DL defines the false-positive rate for a blank sample. 98% of the time, a true blank sample should show a result  $<DL$ ; 2% of the time a blank will show a false-positive result. Setting the rate (DL) is a business decision – what is acceptable?

# Reasons for High Routines

- ▶ Normal statistics of the program design
- ▶ Unusual non-occupational conditions
  - Uranium in groundwater
  - $^{137}\text{Cs}$  from wild game or foreign travel
  - Glow-in-the-dark devices ( $^3\text{H}$  in watches)
  - Medical administrations ( $^{131}\text{I}$ ,  $^{207}\text{Tl}$ ,  $^{131,134,137}\text{Cs}$ )
- ▶ Previous known intake
- ▶ Surprise
  - New intake
  - Lab issue

# High-Routine Case Studies from Hanford (CY 2005-2007)

- ▶ Case 1 – Simple False Positive
- ▶ Case 2 – The Classic High Routine
- ▶ Case 3 – Looks Suspicious - Urine
- ▶ Case 4 - We Forgot We Were Warned
- ▶ Case 5 – Looks Suspicious – Chest Count
- ▶ Case 6 – Guilty
- ▶ Case 7 – A Unique (?) Puzzle

# High Routine Investigation Protocol

- ▶ Verify initial result
  - by 1 recount with similar result
  - not verified if 2 recounts show no detection
- ▶ Investigate verified results
  - Special follow-up bioassay (typically 2 measurements)
  - Check recent work history
- ▶ General philosophy: It takes 2 independent measurements to rule out a positive one.

# CDLs, MDA, and DLs for $^{239}\text{Pu}$

Lab	Contractual Level (dpm)	Observed MDA (dpm)	Typical QC Blank DL (dpm)
Former Lab (thru 3/31/05)	0.02	0.014	0.0042
Current Lab (since 4/1/05)	0.02	0.0102	0.003

# Define “Positive”

- ▶ “Positive” means the analyte is detected by a given measurement
- ▶ Hanford defines “Positive” for most low-level measurements as a result greater than the decision level for the measurement, or

Result > 2 × Total Propagated Uncertainty

$$(DL = 2 \times TPU)$$

# False-Positives

- ▶ A fact of life based on your program statistical design
- ▶ No false positives suggests you may be running a relatively insensitive program
- ▶ Excessive false positives raise questions about validity of results and decreases everybody's confidence in the program
- ▶ Hanford program is designed for a false-positive rate of about 2%.

# Case 1 – Simple False-Positive

- ▶ Annual Pu urinalysis, March 2006, 1176 ml
- ▶ Indication of detection of  $^{239}\text{Pu}$  (Result  $> 2\sigma$  DL)
- ▶ Action (procedural): Recount sample twice

Nuclide	Result $\pm 1\sigma$ (dpm)	DL (dpm)	Detection?
$^{238}\text{Pu}$	$-0.00108 \pm 0.00285$ dpm	0.0057	No
$^{239}\text{Pu}$	$0.00628 \pm 0.00298$ dpm	0.006	Maybe

# Case 1 – Simple False-Positive

- ▶ Now have 3 counts for each of 2 nuclides for one sample

Nuclide	Result $\pm 1\sigma$ (dpm)	DL (dpm)	Detection
$^{238}\text{Pu}$ initial	$-0.0011 \pm 0.0028$	0.0057	No
$^{239}\text{Pu}$ -initial	$0.0063 \pm 0.0030$	0.0060	Maybe
$^{238}\text{Pu}$ -recount1	$-0.0014 \pm 0.0019$	0.0038	No
$^{239}\text{Pu}$ -recount1	$0.0031 \pm 0.0023$	0.0046	No
$^{238}\text{Pu}$ -recount2	$0.0044 \pm 0.0018$	0.0037	No
$^{239}\text{Pu}$ -recount2	$0.0030 \pm 0.0023$	0.0045	No

# Case 1 – Simple False-Positive

- ▶ Initial indication not verified by 2 recounts.
- ▶ Conclusion of statistical false-positive
- ▶ All results recorded
- ▶ Documentation is in procedure, no investigation report is written.
- ▶ Transparent to the worker. A normal result letter is provided.

# Expected Statistical High Routines

Hanford Pu urinalyses: 3000 per year

Pu Results: 6000 per year

Expected statistical false positives at 2% rate if no activity is present: 120 per year

## Case 2: Classic High Routine

- ▶ Annual Pu urinalysis, Aug 2005, 1745 ml
- ▶ Indication of detection of  $^{238}\text{Pu}$
- ▶ Action: Recount sample twice

Nuclide	Result $\pm 1\sigma$ (dpm)	DL (dpm)	Detection
$^{238}\text{Pu}$	0.0083 $\pm$ 0.0033	0.0066	Maybe
$^{239}\text{Pu}$	0.0051 $\pm$ 0.0027	0.0054	No

## Case 2: Classic High Routine

► Now have 3 counts of same sample

Nuclide	Result $\pm 1\sigma$ (dpm)	DL (dpm)	Detection
$^{238}\text{Pu}$ -initial	$0.0083 \pm 0.0033$	0.0066	Maybe
$^{238}\text{Pu}$ -recount1	$0.0079 \pm 0.0038$	0.0075	Yes
$^{238}\text{Pu}$ -recount2	$0.0098 \pm 0.0042$	0.0085	Yes

## Case 2: Classic High Routine

- ▶ Presence of  $^{238}\text{Pu}$  is verified for the sample
- ▶ Where might it have come from?
  - Intake?
  - External sample contamination?
  - Lab interference problem?
- ▶ Request work history review
- ▶ Request 2 special follow-up urine samples

## Case 2: Classic High Routine

▶ Work review report:

“...works as a nuclear waste operator at the xyz facility. Has had multiple RWP entries into xyz work sites, but none have required Pu bioassay. Therefore the potential for exposure to Pu is extremely low.”

## Case 2: Classic High Routine

- ▶ Two follow-ups in Oct 2005; 1416 ml & 1866 ml
- ▶ Now have 5  $^{238}\text{Pu}$  results (original, 2 recounts, 2 specials)

Nuclide	Result $\pm 1\sigma$ (dpm)	DL (dpm)	Detection
$^{238}\text{Pu}$ -initial	$0.0083 \pm 0.0033$	0.0066	Maybe
$^{238}\text{Pu}$ -recount1	$0.0079 \pm 0.0038$	0.0075	Yes
$^{238}\text{Pu}$ -recount2	$0.0098 \pm 0.0042$	0.0085	Yes
$^{238}\text{Pu}$ -special1	$0.00 \pm 0.000062$	0.00012	No
$^{238}\text{Pu}$ -special2	$-0.0015 \pm 0.0019$	0.0039	No

## Case 2: Classic High Routine

- ▶ Initial result was verified showing activity
- ▶ 2 special follow-up samples did not confirm initial indication. No detectable excretion.
- ▶ Work place reviewed showed no real potential
- ▶ Intake was not confirmed and no dose assigned
- ▶ Reason for high routine is unknown. Possibilities include:
  - External contamination of sample
  - intake below capability of program for confirming (i.e., below the minimum detectable intake and any dose associated is below the minimum detectable dose)
  - Normal statistical fluctuation of the measurement process
- ▶ Evaluation report with summary letter to worker.

## Case 3 – Looks Suspicious

- ▶ Pu baseline, May 2005, detected  $^{239}\text{Pu}$
- ▶ Verified and confirmed by follow-ups

Nuclide	Result $\pm 1\sigma$ (dpm)	DL (dpm)	Detection
$^{239}\text{Pu}$ -initial	$0.0132 \pm 0.0050$	0.010	Maybe
$^{239}\text{Pu}$ -recount1	$0.0145 \pm 0.0049$	0.0098	Yes
$^{239}\text{Pu}$ -recount2	$0.0100 \pm 0.0043$	0.0086	Yes
$^{239}\text{Pu}$ -special1	$0.0078 \pm 0.0037$	0.0075	Yes
$^{239}\text{Pu}$ -special2	$0.0047 \pm 0.0029$	0.0058	No

# Case 3 – Looks Suspicious

## ▶ Work History Review

- Recent years a lab supervisor with low risk work
- No recent Pu work without fission products
- Purex work in the early-mid 1980s
- Worker thought some internal exposure may have occurred in the 1980s
- Surgery in 2004
- Medications for blood pressure and cholesterol
- Drinks normal amounts of cranapple juice, drank a lot of tea after surgery, has since returned to coffee

# Mild Chelating Drugs – “The Short List” (courtesy Patrick Lowry, MD, REAC/TS)

- Anti-Inflammatory Drugs
- Salicylates
- Indocin
- Aminopyrine
- Tylenol
- Butazolidin Group
- Steroids
- Cortisone, Hydrocortisone, etc.
- Neuropsych Drugs
- Chlorpromazine
- Dilantin
- Antimicrobial Drugs that Chelate
- p-Aminosalicylic Acid Fe, Cu
- Bacitracin Zn
- Isoniazid Fe, Cu, Mn, Co
- Kanamycin Ca
- Neomycin Fe, Al
- Novobiocin Mg
- Penicillin Co
- Polymyxin Mg, Mn, Ca, Fe
- Streptomycin Mn
- Tetracycline Fe, Mg, Mn, Mo, Al, Ca

## Case 3 – Looks Suspicious

- ▶ Reviewed worker's history file back to start of employment
- ▶ Routine Pu urinalyses and chest counts from 1983 to 1992 showed no detection
- ▶ Pu urinalyses in 1999 and 2000 also showed no detection
- ▶ Purex was in prime operation in the mid-1980s
- ▶ 6 personal contamination events at Purex (1981 to 1987)
- ▶ Purex job tasks involved highly contaminated work

## Case 3 – Looks Suspicious

- ▶ A 1985 special urine sample, in hindsight, looked like detection

$$0.0175 \pm 0.0083$$

In 1985, “detection” was defined as  $>0.05$  dpm

By today’s  $>2\sigma$  method, “detection”  $>0.0166$  dpm

- ▶ Conclusion:

Confirmed 2.5 nCi  $^{239}\text{Pu}$  inhalation in 1985

CEDE = 2.3 rem; (Bone Surface = 43 rem)

# Case 3 – Looks Suspicious

- ▶ Why not detected from 1985 to 2005?
  - Change in definition of detection (late 1980s)
  - April 1, 2005 change in analytical labs. New lab appears to be showing slightly better detection capability
  - Only a slight change in sensitivity could move from non-detection to detection
  - New lab with lower background
  - Difference in how background = “0” is treated
- ▶ Medical conditions (surgery and medication) could have resulted in a mild Pu chelation effect
- ▶ Excretion near DL can be expected to show hit-and-miss detection

# Case 4 - Forgot We Were Warned

- ▶ Termination Pu, April 2005, detected  $^{239}\text{Pu}$
- ▶ Verified and confirmed by follow-ups (Barely!)

Nuclide	Result $\pm 1\sigma$ (dpm)	DL (dpm)	Detection
$^{239}\text{Pu}$ -initial	$0.00565 \pm 0.00268$	0.00536	Maybe
$^{239}\text{Pu}$ -recount1	$0.0043 \pm 0.0026$	0.0052	No
$^{239}\text{Pu}$ -recount2	$0.0064 \pm 0.0028$	0.0055	Yes
$^{239}\text{Pu}$ -special1	$0.0101 \pm 0.0041$	0.0082	Yes
$^{239}\text{Pu}$ -special2	$0.0031 \pm 0.0023$	0.0046	No

# Case 4 - Forgot We Were Warned

## ▶ Recent Work History

- Multiple recent entries into TRU waste sites
- No indication of likely intake

## ▶ Personal History File Review

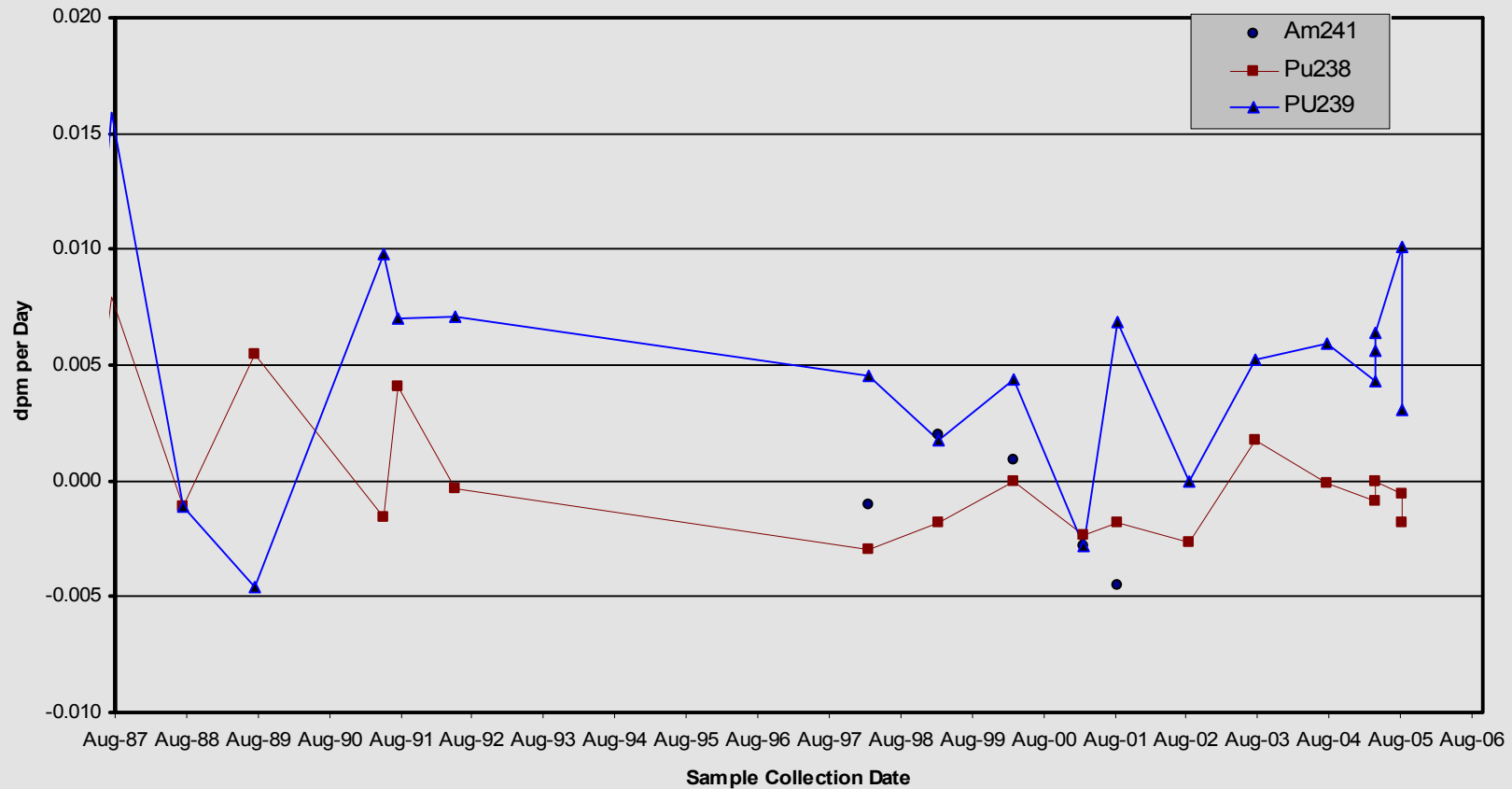
- 1987 to 2004, 15 Pu urinalyses with no detection
- 1987 employment history form noted Rocky Flats
- 1987 Rocky Flats Deposition Report

1984  $^{239}\text{Pu}$  deposition = 2.7% MPBB (1.1 nCi)

Hanford baseline had not confirmed this. Never assigned in the Hanford record.

# Case 4 - Forgot We Were Warned

*Long-term data is consistent*  
 *$^{239}\text{Pu}$  is detected,  $^{241}\text{Am}$  &  $^{238}\text{Pu}$  are not*

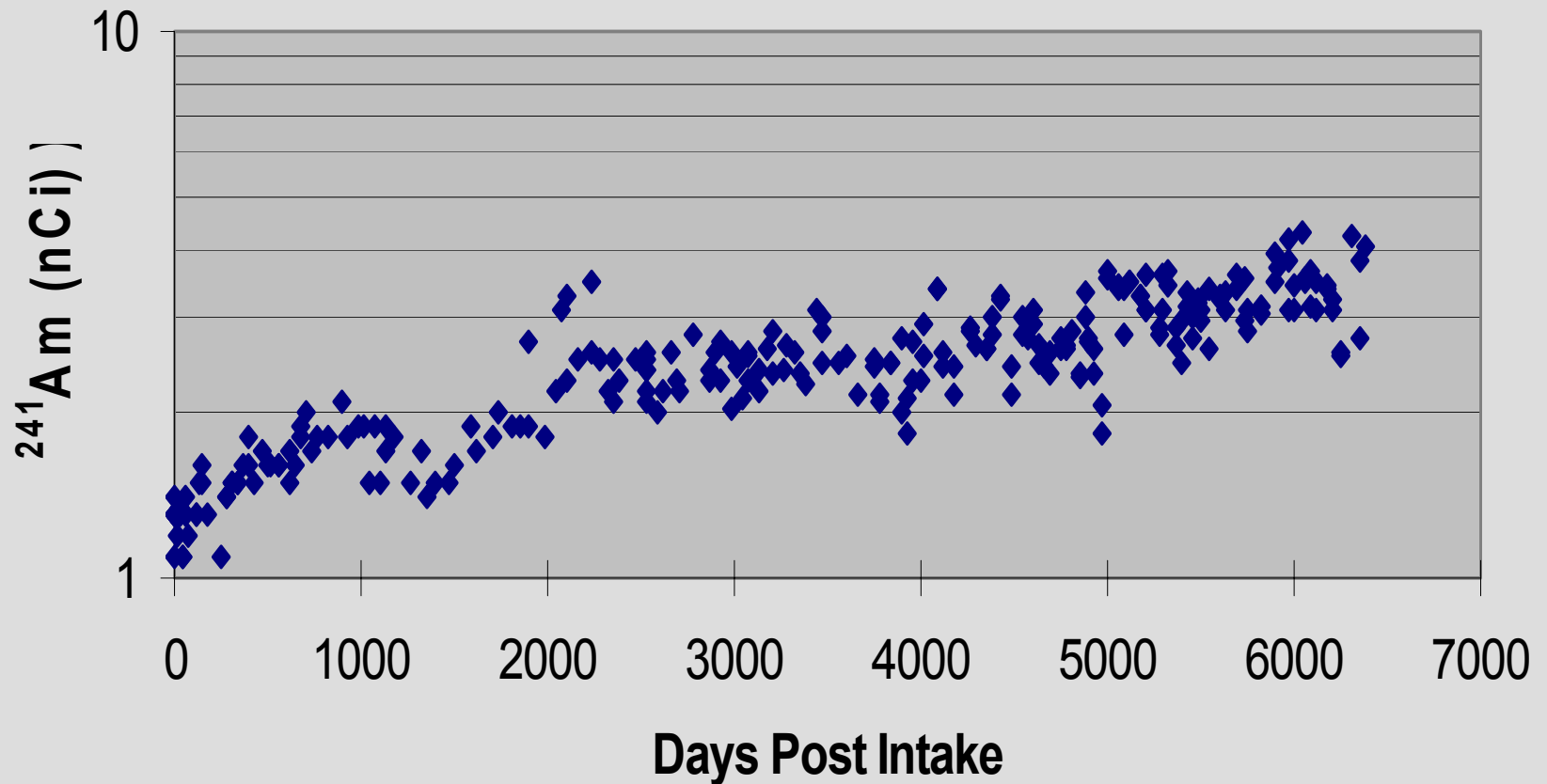


# Case 4 - Forgot We Were Warned

## ► Conclusion

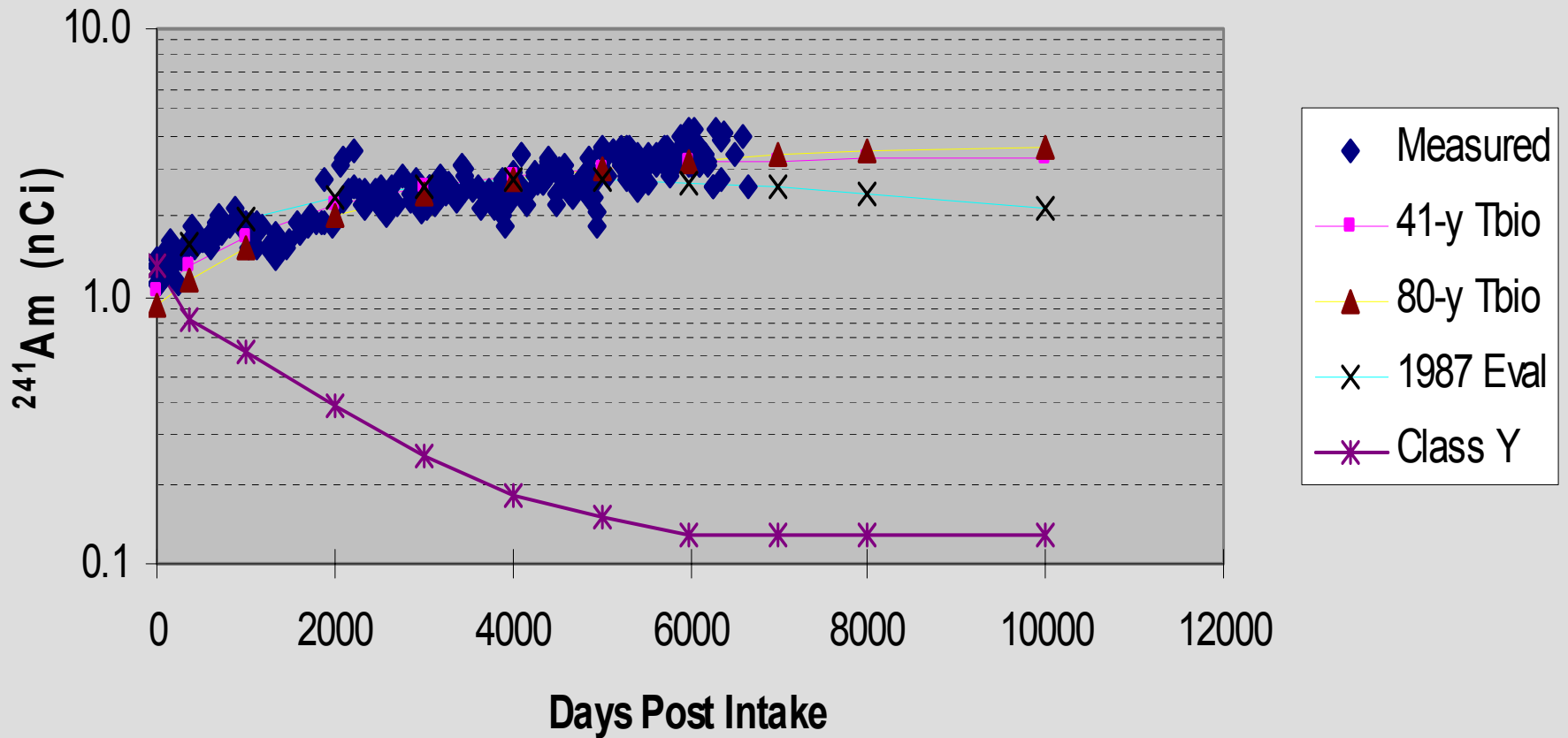
- Confirmed intake at Rocky Flats
- Worker was not surprised by this call
- MPBB to CEDE conversion at Rocky Flats in 1989
- Worker had never received CEDE dose report
- Requested updated dose report from Rocky Flats  
CEDE = 12 rem
- Dose added to Hanford records as prior-to-Hanford employment (now included in lifetime cumulative)

# HAN-1 Super Class Y Pu – Lung Count



# HAN-1 Super Class Y Pu – Data Fits (Carbaugh & La Bone, 2003)

*Radiation Protection Dosimetry* Vol. 105, Nos 1-4, pp 133-138



# Minimum detectable dose at 25 years for a HAN-1 type case

- ▶ If same lung retention pattern, but smaller intake with no initial detection and lung count now showing nominal detection (0.2 nCi  $^{241}\text{Am}$ )

CEDE: 8 rem

Lung CDE: 60 rem

Bone Surface CDE: 12 rem

## Case 5: Looks Suspicious – Chest Count (a newly discovered, old super class Y Pu case)

- ▶ High end-of-assignment chest count (Feb 2006)
  - Confirmed by recount & spectrum summing
  - Previous 2003 detection had not been confirmed
- ▶ Periodic urinalysis of Jan 2006 showed  $^{239}\text{Pu}$ 
  - Special follow-ups confirmed Pu but not  $^{241}\text{Am}$
- ▶ Recent work history showed very limited potential for intake. Worker recalled a 1985 glove breach.
- ▶ Two previous intakes (1980, 1985)
  - 2006 data not explained by 1985 mixture (glove breach), but was consistent with the 1980 mixture (Pu fire).

# Case 5: Looks Suspicious – Chest Count

(a newly discovered, old super class Y Pu case)

- ▶ Iterative trials resulted in a fit to a 3- $\mu$ m AMAD aerosol, 30% class Y and 70% super class Y.
  - Similar to the fit used for other workers in the 1980 incident.

CEDE: 6.8 rem

Lung CDE: 43 rem

Bone Surface CDE: 29 rem

- ▶ Similar bioassay effect for a wound.

## Case 6 – “Guilty”

- ▶ Minor wound (scraped knees in April, 2005)
- ▶ Special bioassay – no detection
  - Wound count, WBC, Pu urinalysis
- ▶ 3 months later: Pu detection in periodic analysis
- ▶ Verified by rcounts
- ▶ Confirmed by follow-up samples
- ▶ Conclusion: Minor wound resulted in an intake. Missed on the first special bioassays.
  - 0.02 nCi Pu, 86 mrem CEDE

# Case 7 – A Unique (?) Puzzle

- ▶ Confirmed Pu urinalysis
  - Baseline 1-y earlier did not show Pu
- ▶ Special chest count showed Am (no baseline)
- ▶ Hanford 2004-2006
- ▶ Rocky Flats 1995-97
- ▶ Sellafield 1964-1995
- ▶ Conclusion: Old intake at Sellafield based on worker's recollections (facility condition, nature of work). Dose not assigned but range noted in report.
  - Chronic Exposure: 200 to 500 mrem/year CEDE
  - Acute Exposure: 5 to 10 rem CEDE

# In Conclusion

- ▶ Hanford takes positive bioassay results very seriously and works hard to prove or disprove their implications.
- ▶ Archived records are important to history reviews
  - Readily retrievable means they'll be used!
- ▶ Measurement capability changes over time – yesterday's no detection could be today's detection
  - Even relatively small changes can move from nondetection to detection.
- ▶ It's possible to newly discover old intakes.
- ▶ Credibility and integrity of both measurements and the investigation leads to confidence in conclusions.

# Conclusion – A Guide to the Process

1. Verify the initial result (e.g., recount, data check)
2. Confirm the measurement by additional measurements
3. Review the recent work history (since last normal result)
4. Interview the worker
  1. Recent work potential
  2. Old work potential
  3. Possible interferences from life style
5. Follow the leads
6. Exercise good judgment
7. Communicate the conclusions