



Savannah River
Nuclear Solutions, LLC
A Fluor Daniel Partnership

Tritium In Human Urine by Liquid Scintillation Counting

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**Principal Scientist
Savannah River Nuclear Solutions
Savannah River Site
Aiken, SC, USA
April 17, 2009**

Health Physics Society Meeting

Aiken, SC

WHO AM I?

- **B.S. Chemistry, University of Cincinnati, 1981**
99mTc Radiopharmaceutical Development w/ UCCM, MU.
Undergraduate research director: Dr. Edward A. Deutsch.
- **M.S. Nuclear Chemistry, Florida State University, 1984**
Stability Constants of Lanthanides and Actinides with Salicylic Acid by Solvent Extraction; Graduate research director: Dr. Gregory R. Choppin.
- **Completed course work for Ph.D., University of South Carolina, 1992**
Analytical Chemistry
DOE sponsored program with SCUREF*
- **25 years at SRS:**
Naval Fuel Laboratory: (1984 – 1989).
SRNL: ADS & Environmental Compliance (~ 1993).
Analytical Lab: F-Area Count Room CTF (~ 1998).
E-Area Headspace Gas Analysis Laboratory (~ 2002).
Environmental / Bioassay Laboratory (- to present).
- **Collect Postage Stamps and Chemistry Books (oldest 1872).**

*South Carolina Universities Research and Education Foundation

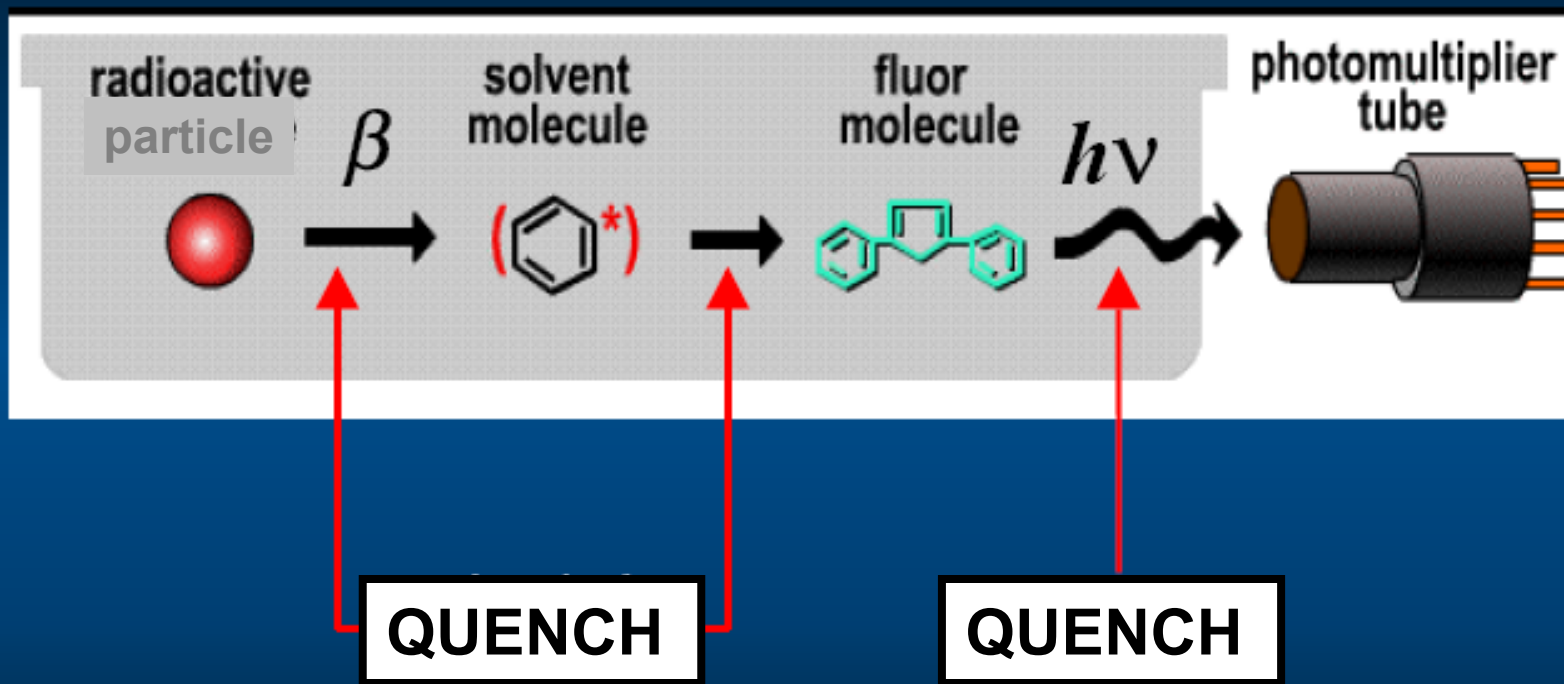
Tritium Bioassay By LSC

- **Basic Principals**
 - Theory
 - Quenching
 - Calibration
- **Interferences**
 - Chemiluminescence
 - Medical Isotopes
- **Solutions**
 - Quench Curve, Filtering and Dilutions
 - LUMEX® Correction
 - Extended hold times...
- **Current Practice**
 - Error Analysis & Quality Control
 - ORNL Intercomparison
 - DOELAP Accreditation
- **Tritium exposure at SRS**

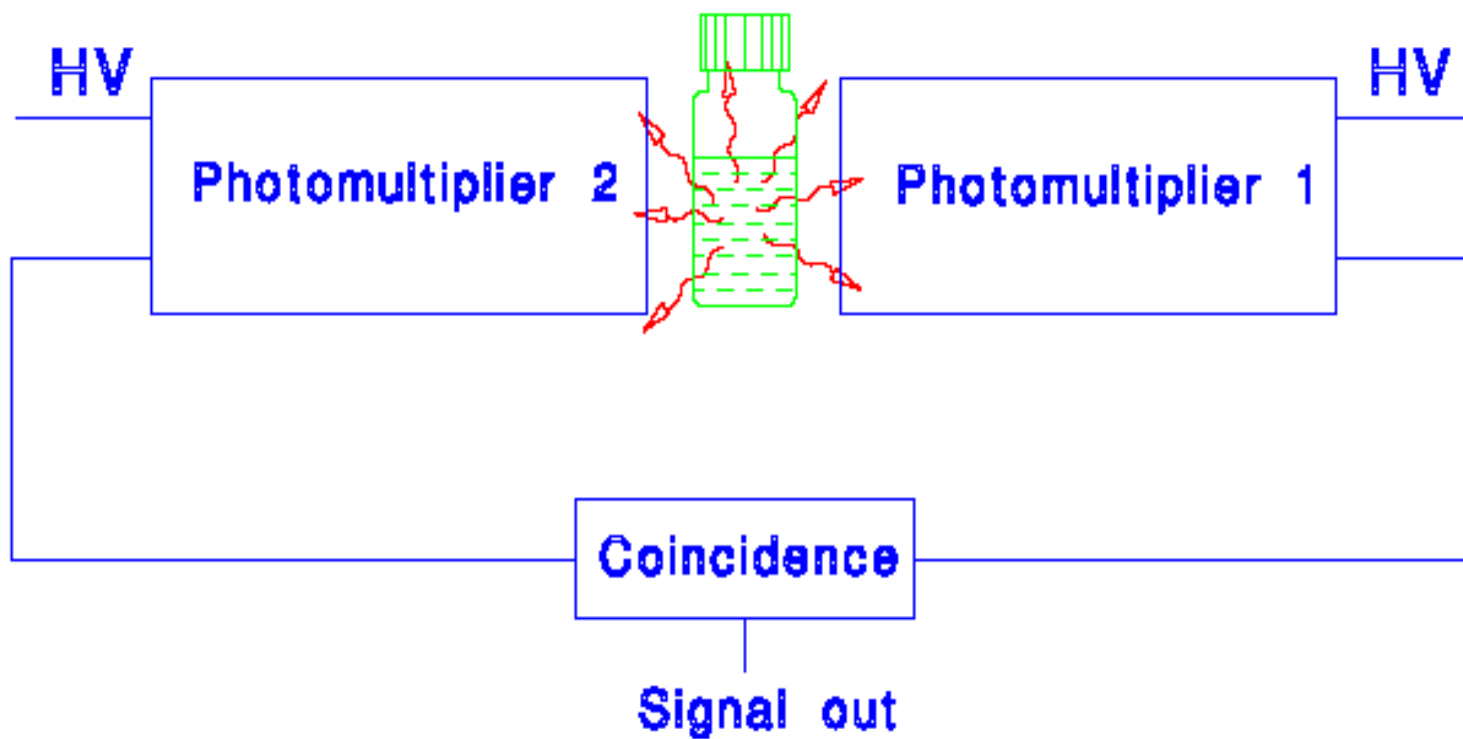
Liquid Scintillation Counting: Theory

- **Decay energy is converted to visible light in an organic liquid.**
- **Amount of light produced is proportional to the amount of radiation present.**
- **Energy of light produced is proportional to the energy of the radiation present.**

Liquid Scintillation Counting: Theory

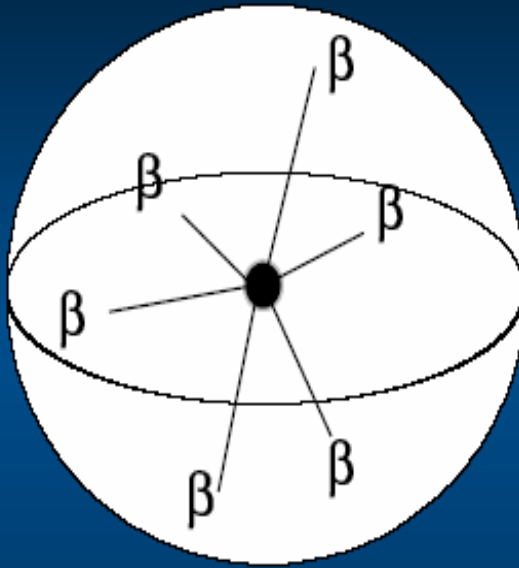


LSC SCHEMATIC



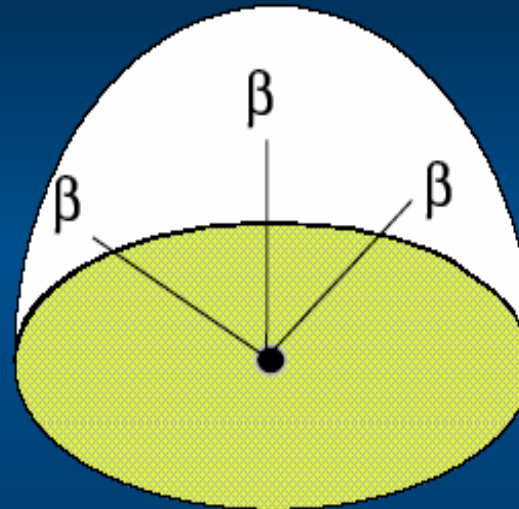
Advantage of 4π Geometry

**4π Geometry
(Homogeneous)**



Surface Area = $4\pi r^2$

**2π Geometry
(Heterogeneous)**

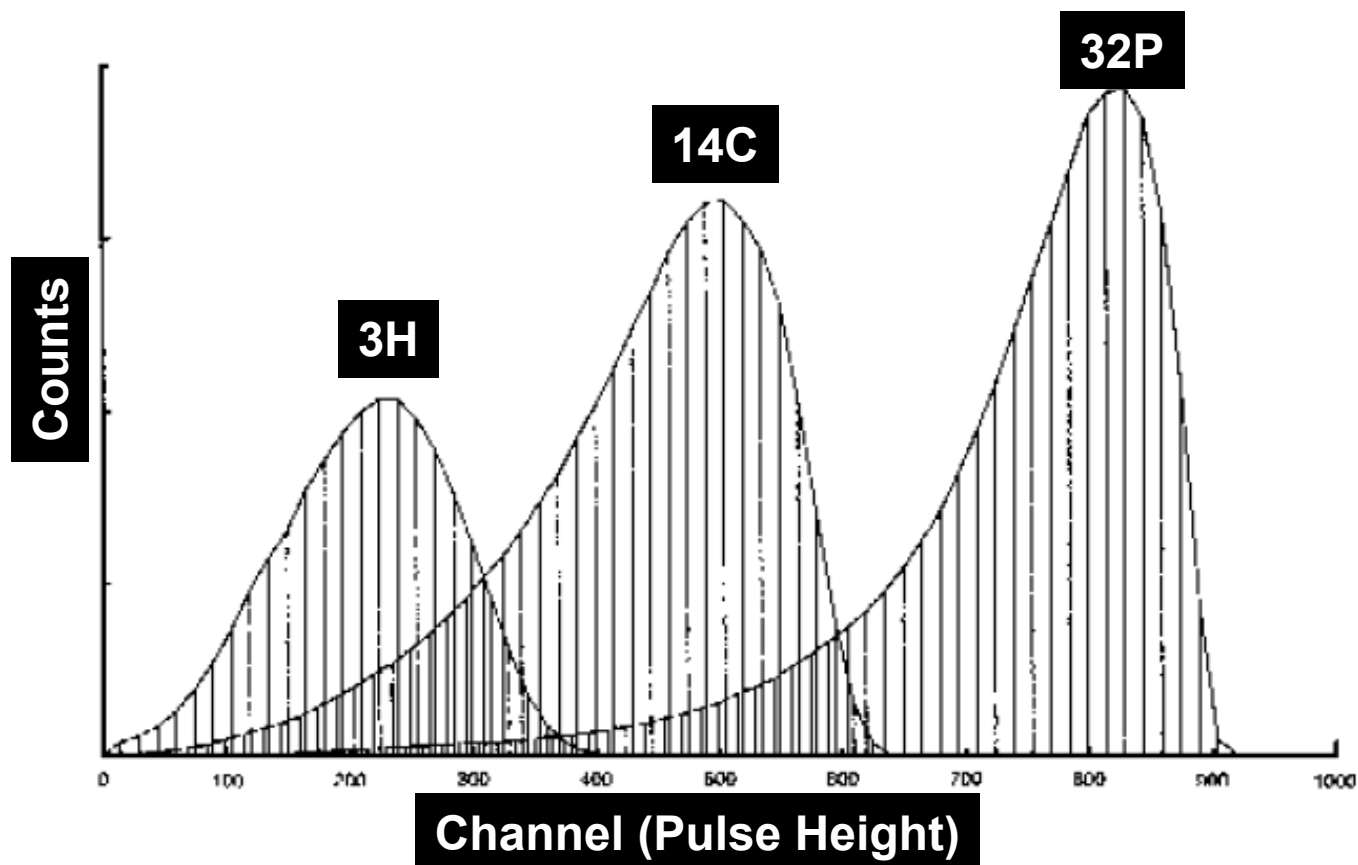


Surface Area = $2\pi r^2$

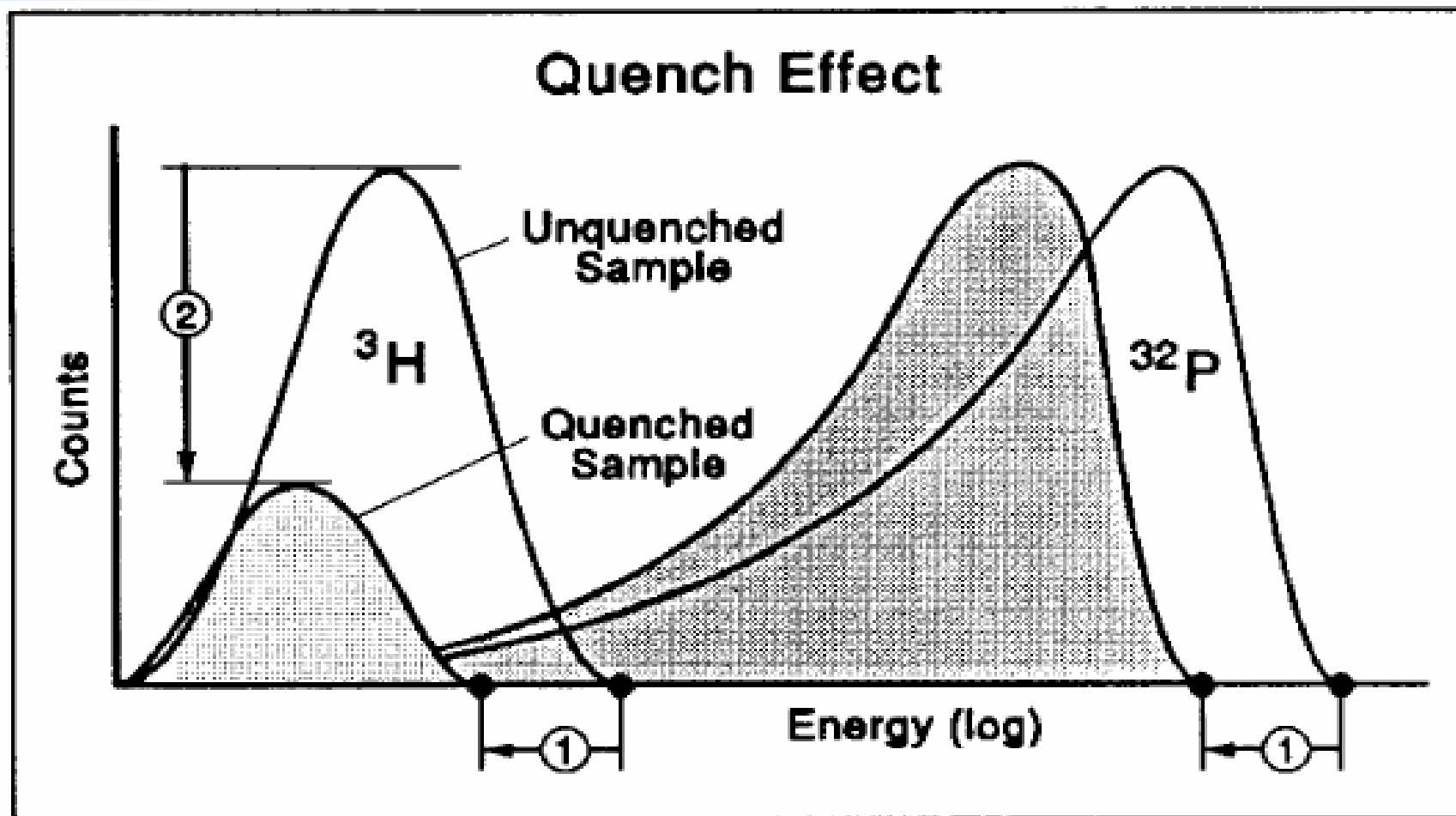


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Typical Beta Spectrum



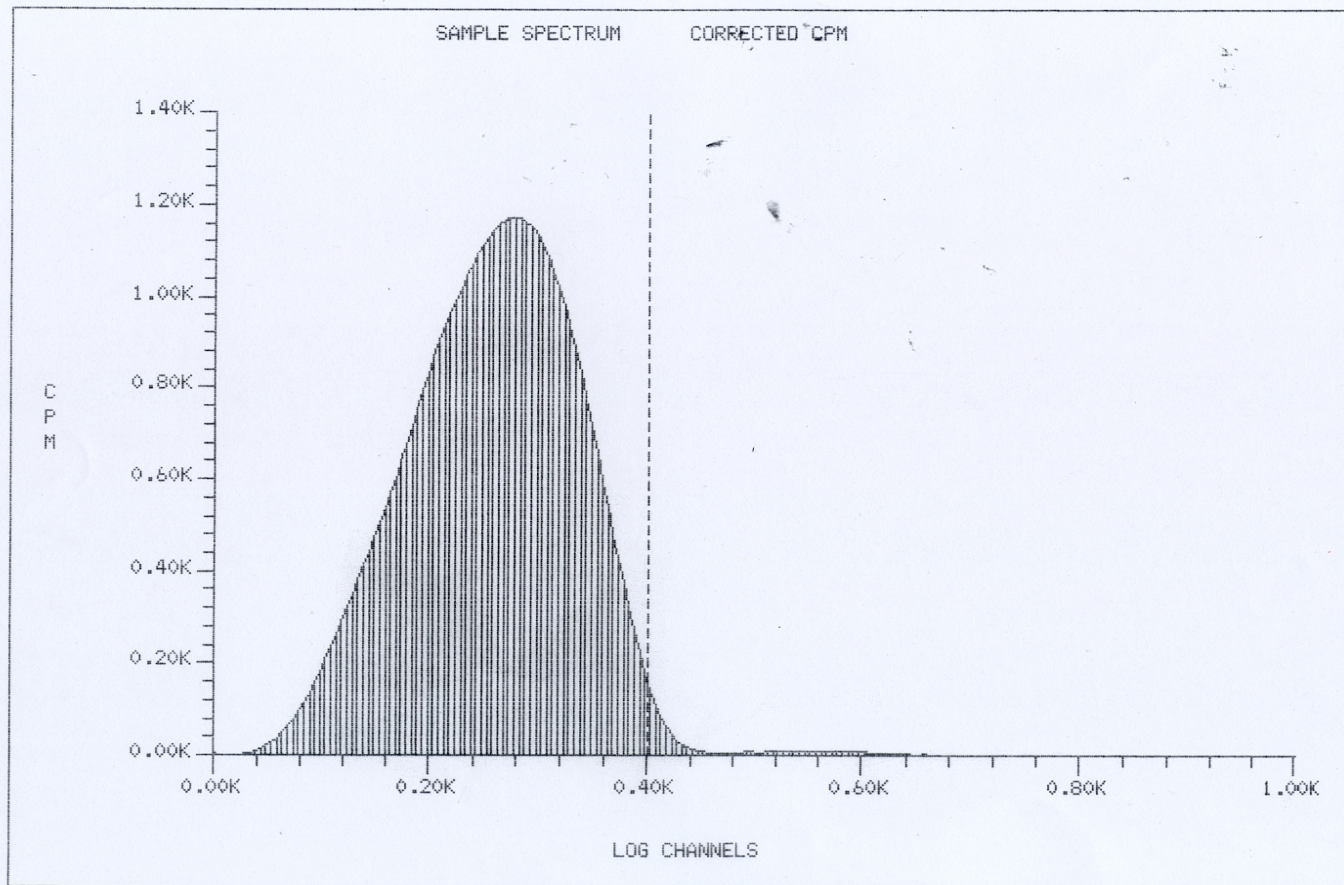
Quenching Effect



Unquenched 3H

User: 11 Sample: 1 Position: **1

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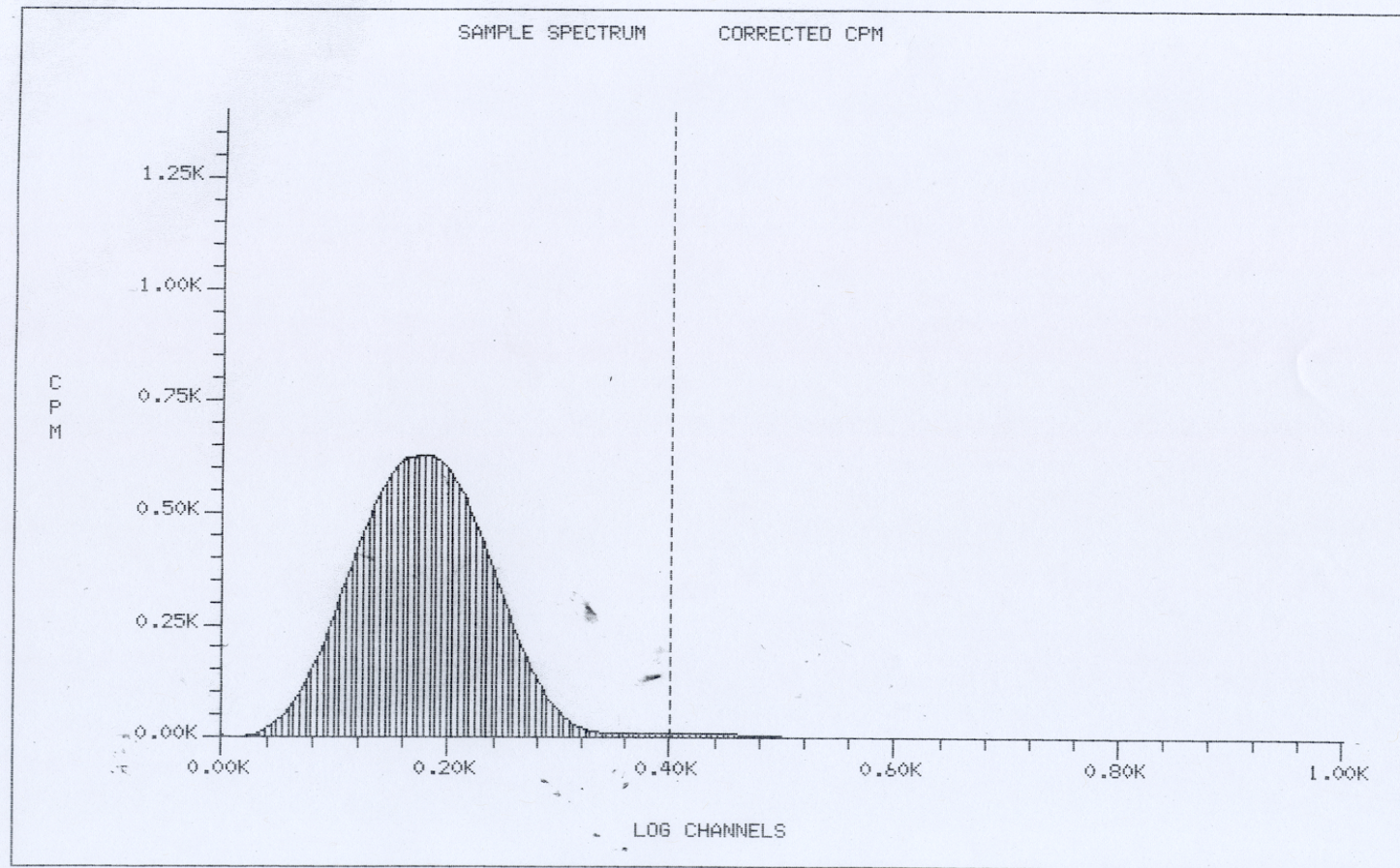


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Moderately Quenched 3H

User: 11 Sample: 5 Position: **5

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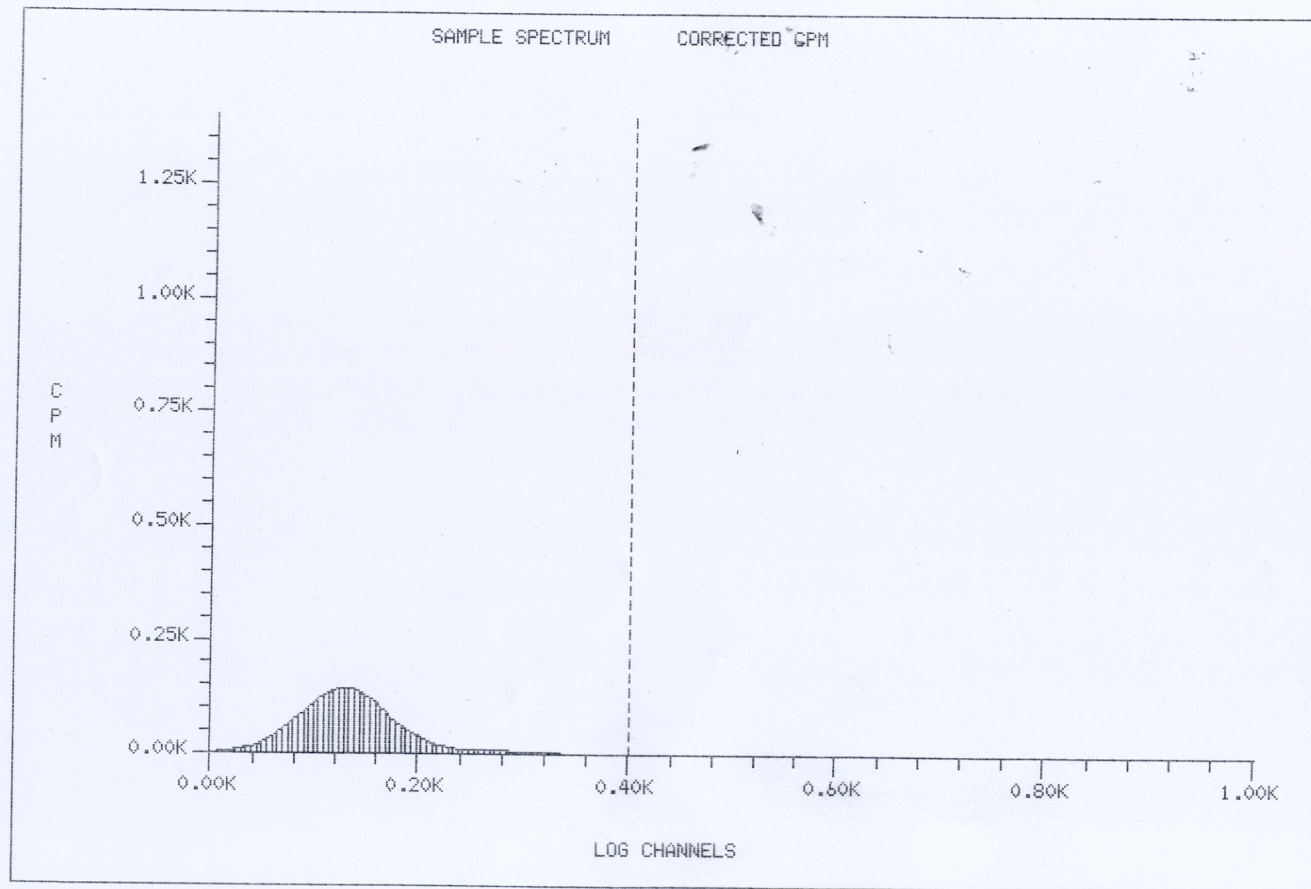


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Highly Quenched 3H

User: 11 Sample: 1 Position: **-1

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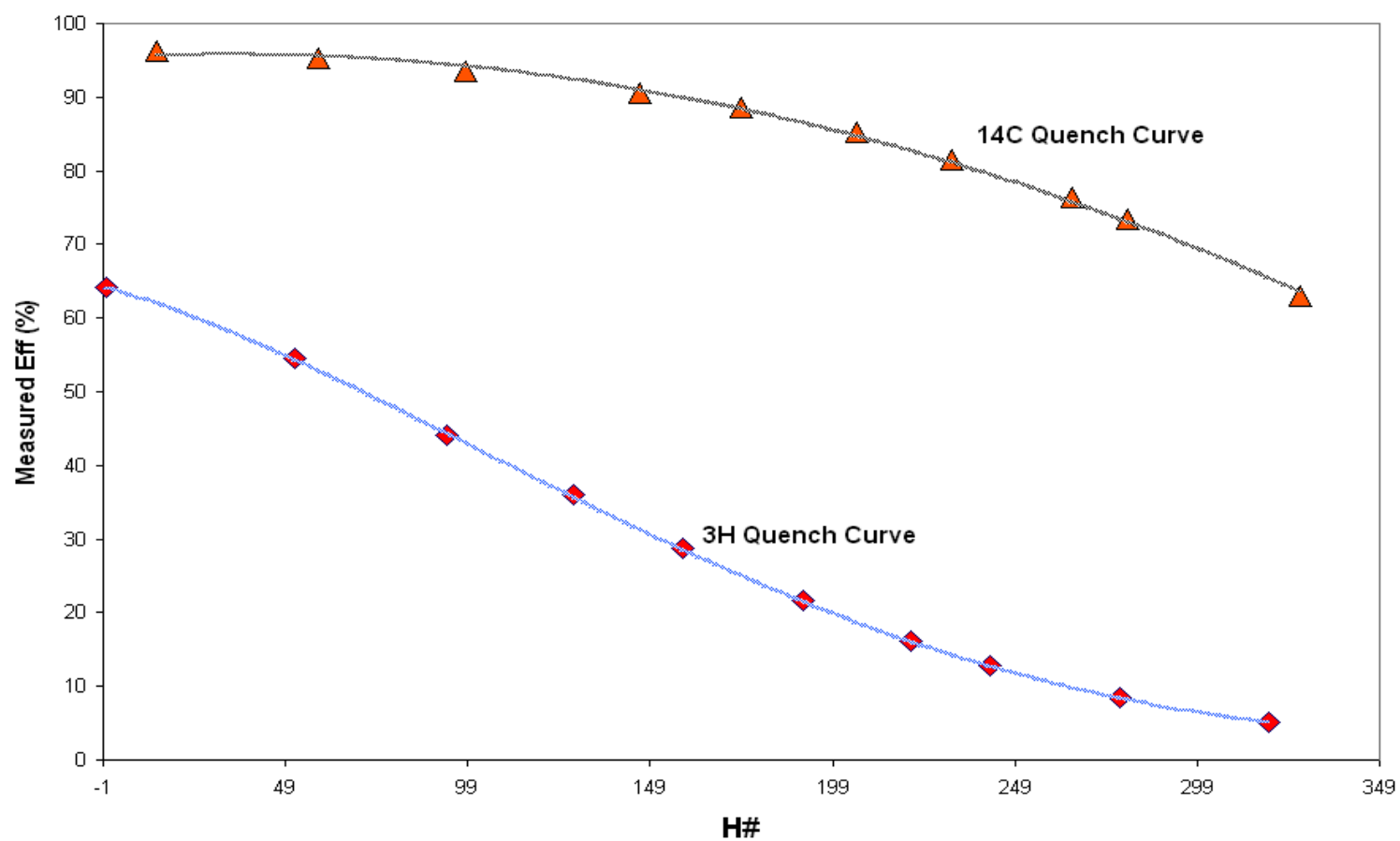
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3H Quench Curve

<u>Std #</u>	<u>H#</u>	<u>Measured Eff (%)</u>
1	0.3	64.27
2	51.7	54.43
3	93.3	44.19
4	128.1	35.93
5	157.8	28.68
6	190.8	21.57
7	220.4	16.09
8	242.3	12.74
9	277.8	8.56
10	318.5	5.13



Quench Curves



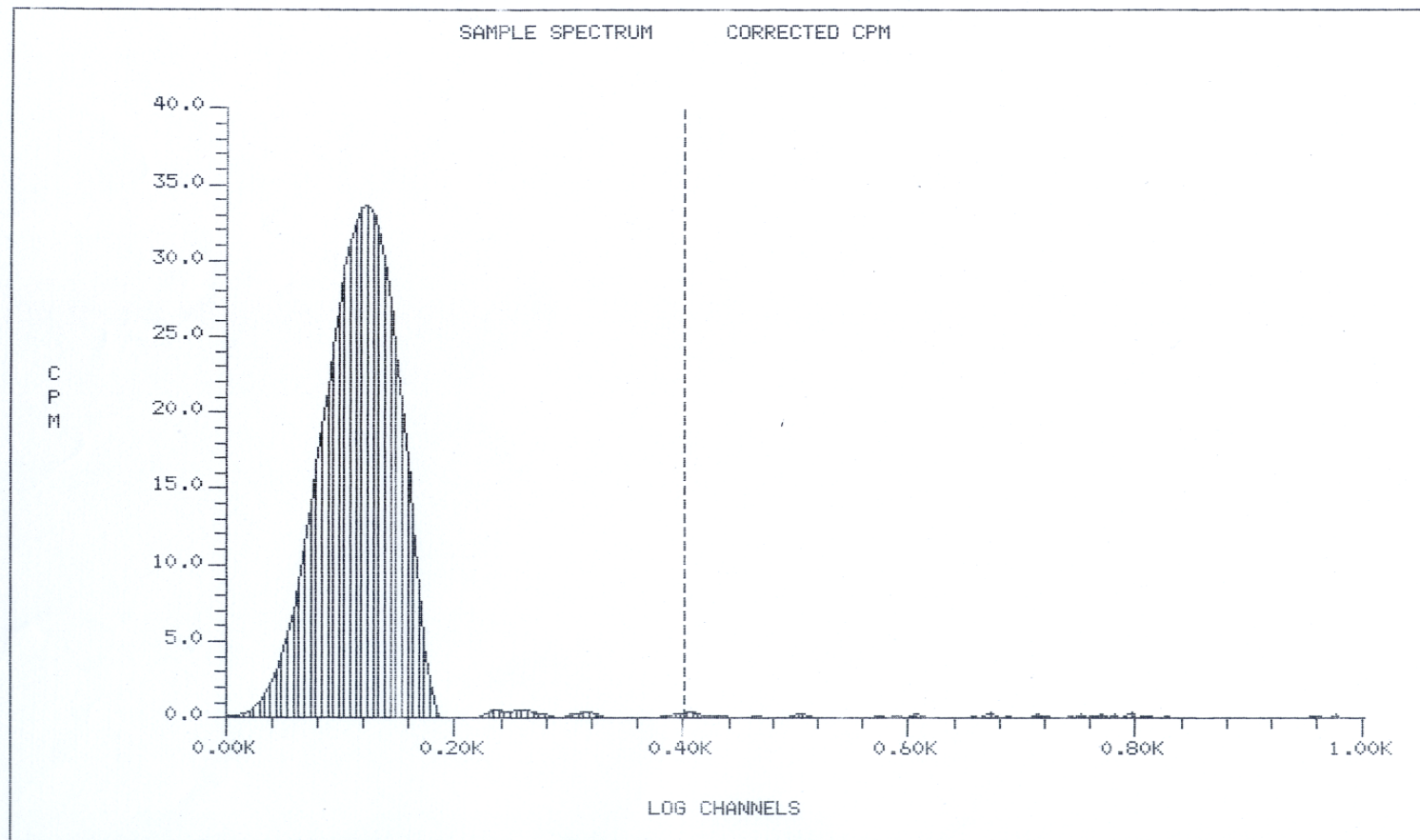
Chemiluminescence

- **Sample may contain substances which can be excited by absorption of ambient light or other interactions.**
- **Lifetimes in the seconds to 10's of minutes, relax by emitting light.**
- **Can translate into false high activity.**
- **Show up at very low energy.**
- **Recounting, dark adapting, or changing counting parameters is sometimes necessary to differentiate between true radioactivity and chemiluminescence.**

Chemiluminescence

User: 2 Sample: 2 Position: **2

5 APR 2007 11:16



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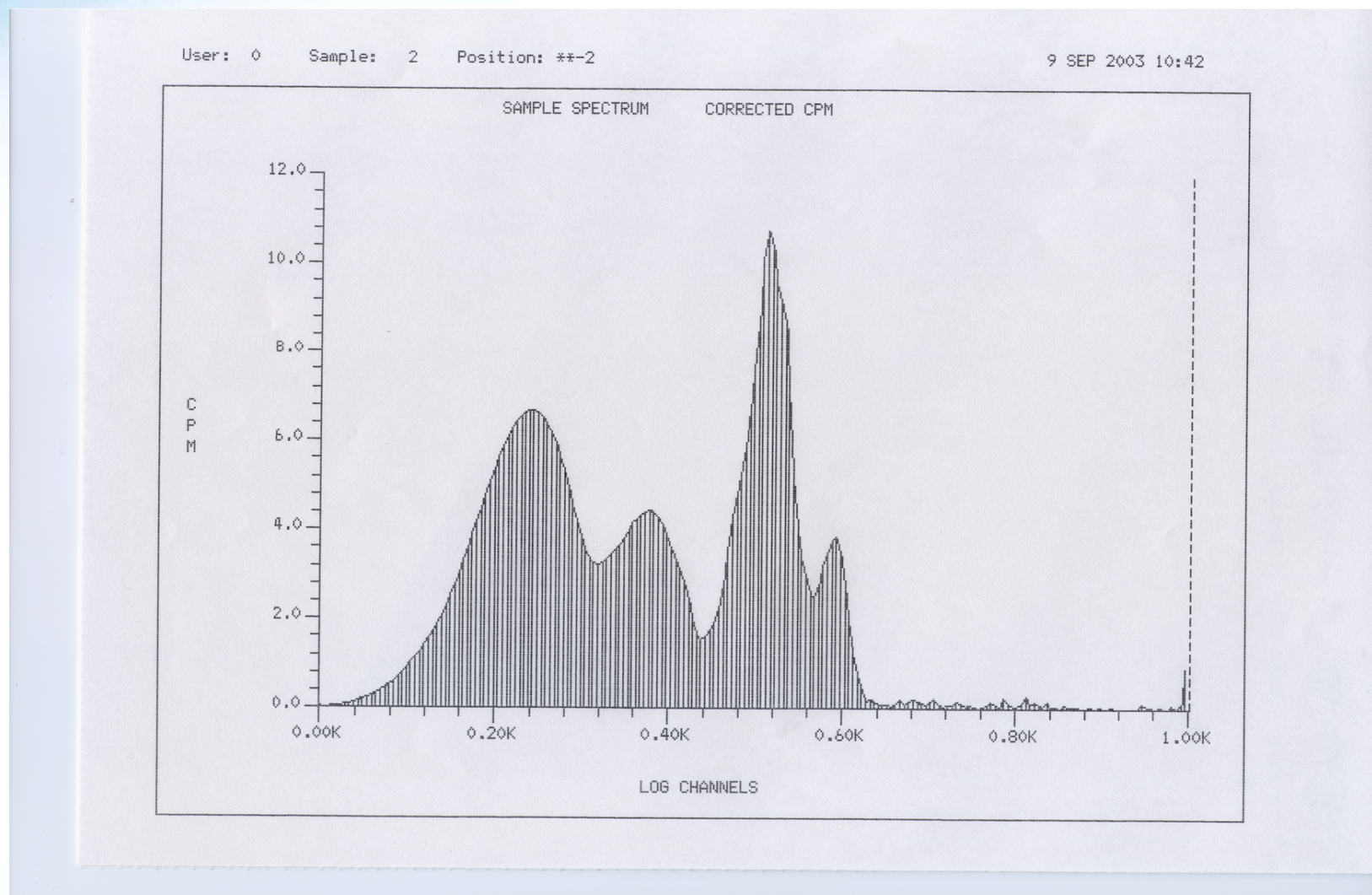
Beckman's LUMEX® Correction

- **Random single photon events which are generated as a result of the chemical interaction of the sample components.**
- **Chemiluminescence events are excluded by the coincidence circuit (LUMEX®).**

Medical Isotope

- **Very small quantity of radioactive substance used in safe, cost-effective imaging and treatment of disease.**
- **Usually weak gamma emitters**
- **Short Half Life**
- **Examples: I-131, Tc-99m, Tl-201, F-18**
- **Photon Emission Tomography (PET scanner) or Gamma Camera Imaging**

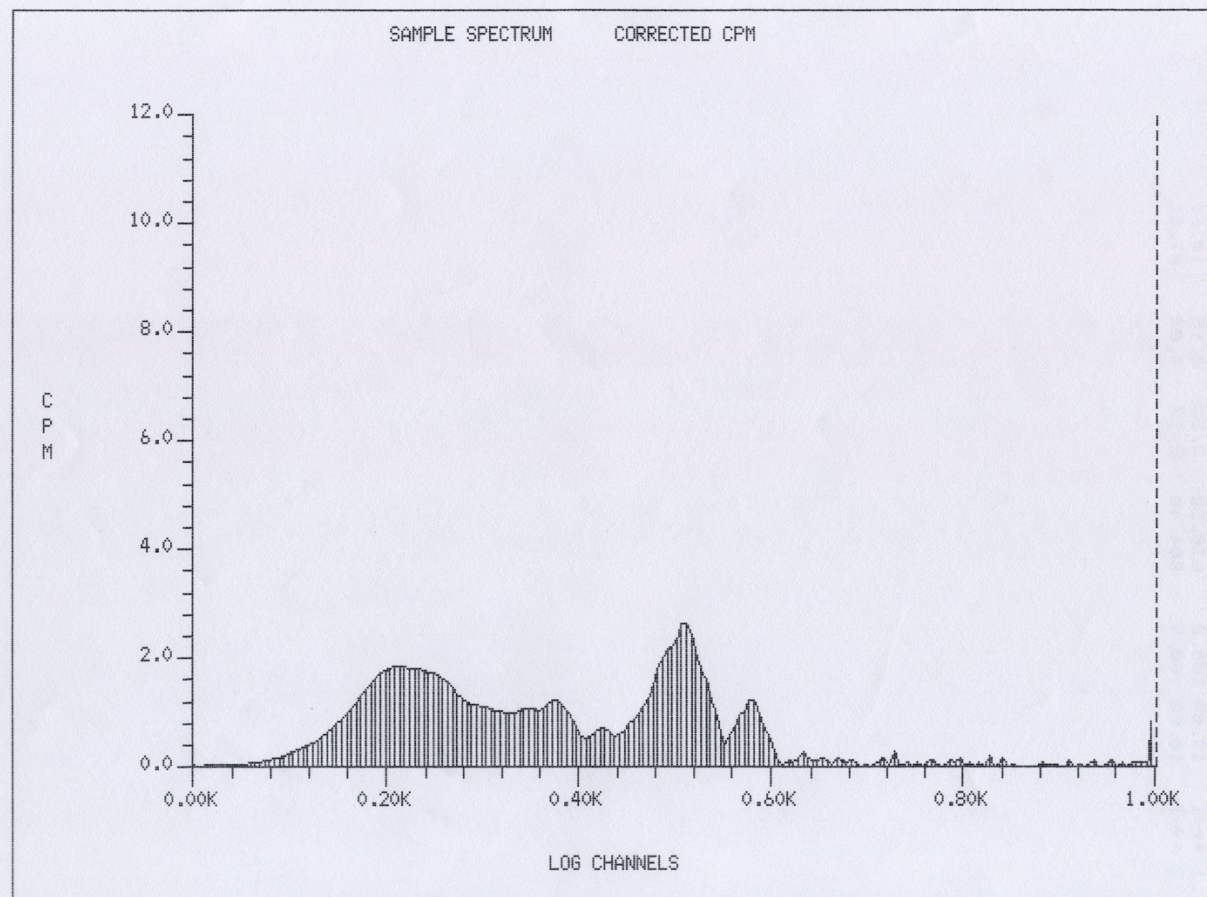
Medical Isotope



Decayed Medical Isotope Spectrum

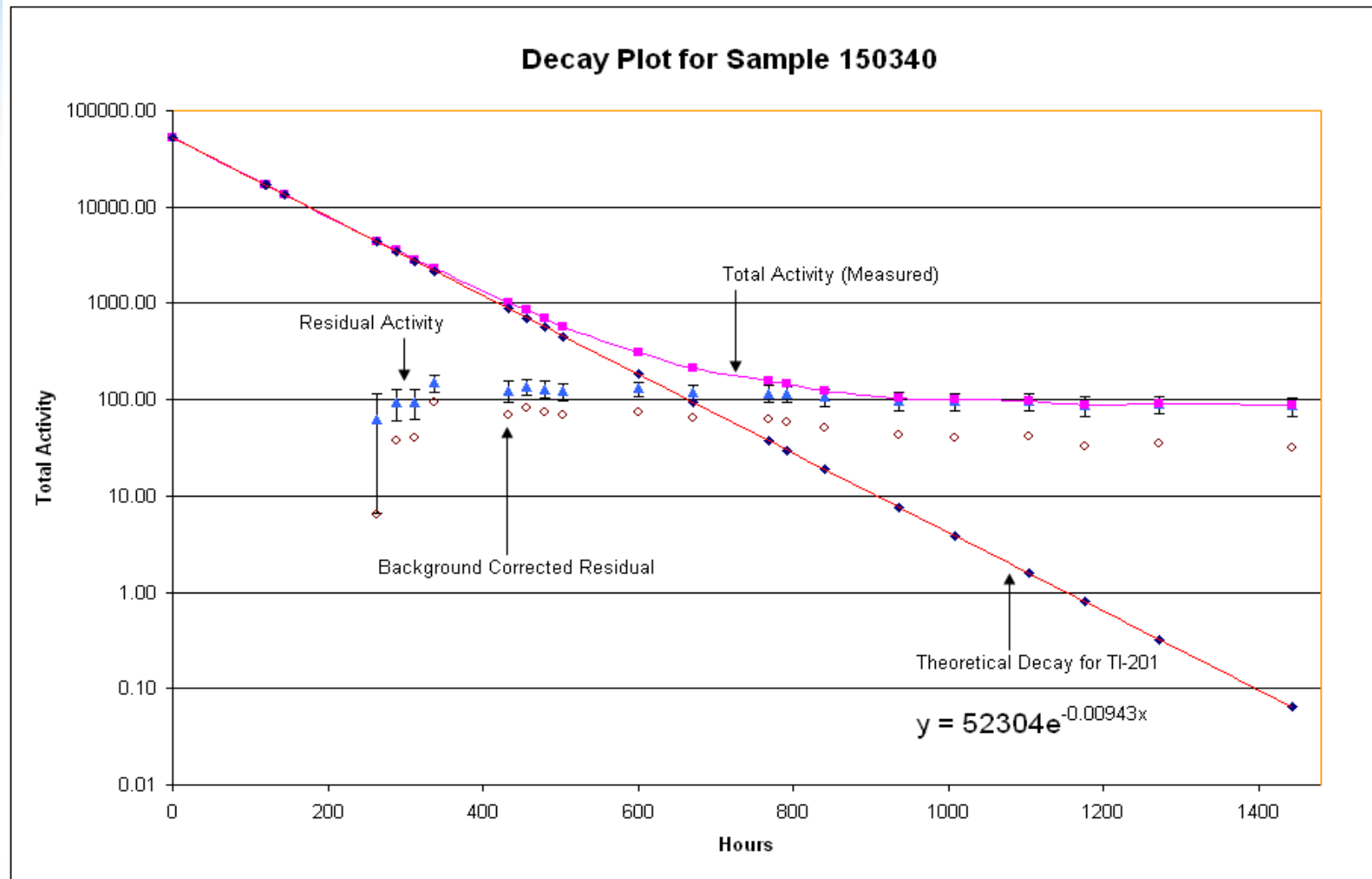
User: 0 Sample: 1 Position: #-1

15 SEP 2003 10:07



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Medical Isotope



Medical Isotope

- $N = N_0 e^{-\lambda t}$
- $\lambda = 0.00943 = \ln(2)/t_{1/2}$
- Unk $t_{1/2} = 73.62$ hours
- $\text{TI-201} = 73.5$ hours

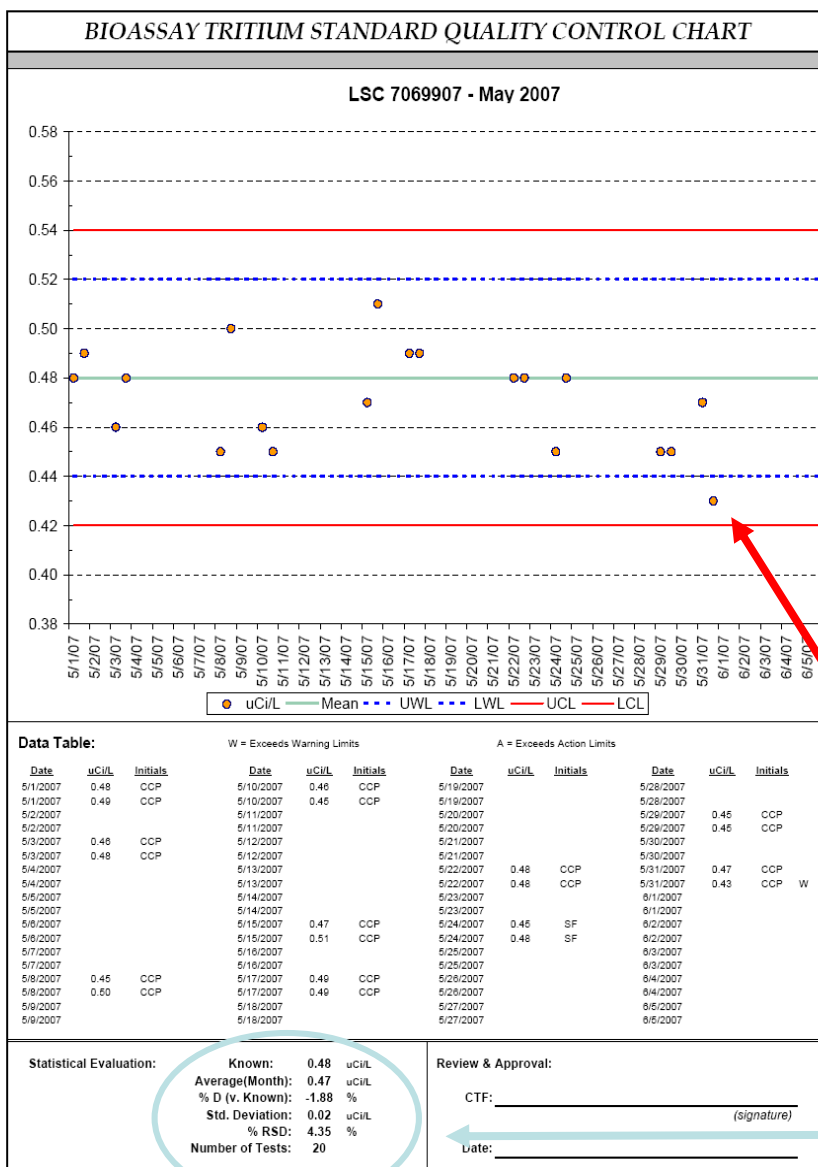
Error Analysis

- **Independent Random Errors (1σ):**
 - NIST: $\pm 1.3\%$
 - Half-Life: $\pm 0.5\%$
 - Counting: $\pm 4.2\%$ (0.25 $\mu\text{Ci/L}$ standard)
 - Pipette (2): $\pm 0.3\%$
- **Total RMS Error: $\pm 4.45\%$**
- **Observed Performance: $\pm 4.51\%$**

Quality Control

- **Daily Background Check**
- **Daily QC Check (~ 0.25 $\mu\text{Ci/L}$, in duplicate)**
 - Decay Corrected to reference date, Control Charted
- **MDL Based on Daily Background**
 - Approx. 0.02 $\mu\text{Ci/L}$
- **Bracketed Batches (QC duplicates)**
- **Every 10th Sample Duplicated**
- **Delta $\leq$ Calculated Allowable Variance**
- **Concern Level of 0.5 $\mu\text{Ci/L}$**
- **Action Level of 5.0 $\mu\text{Ci/L}$**

BIOASSAY TRITIUM STANDARD QUALITY CONTROL CHART



Typical Monthly Control Chart

2 σ

Known value

3 σ

Exceeds Warning Limit

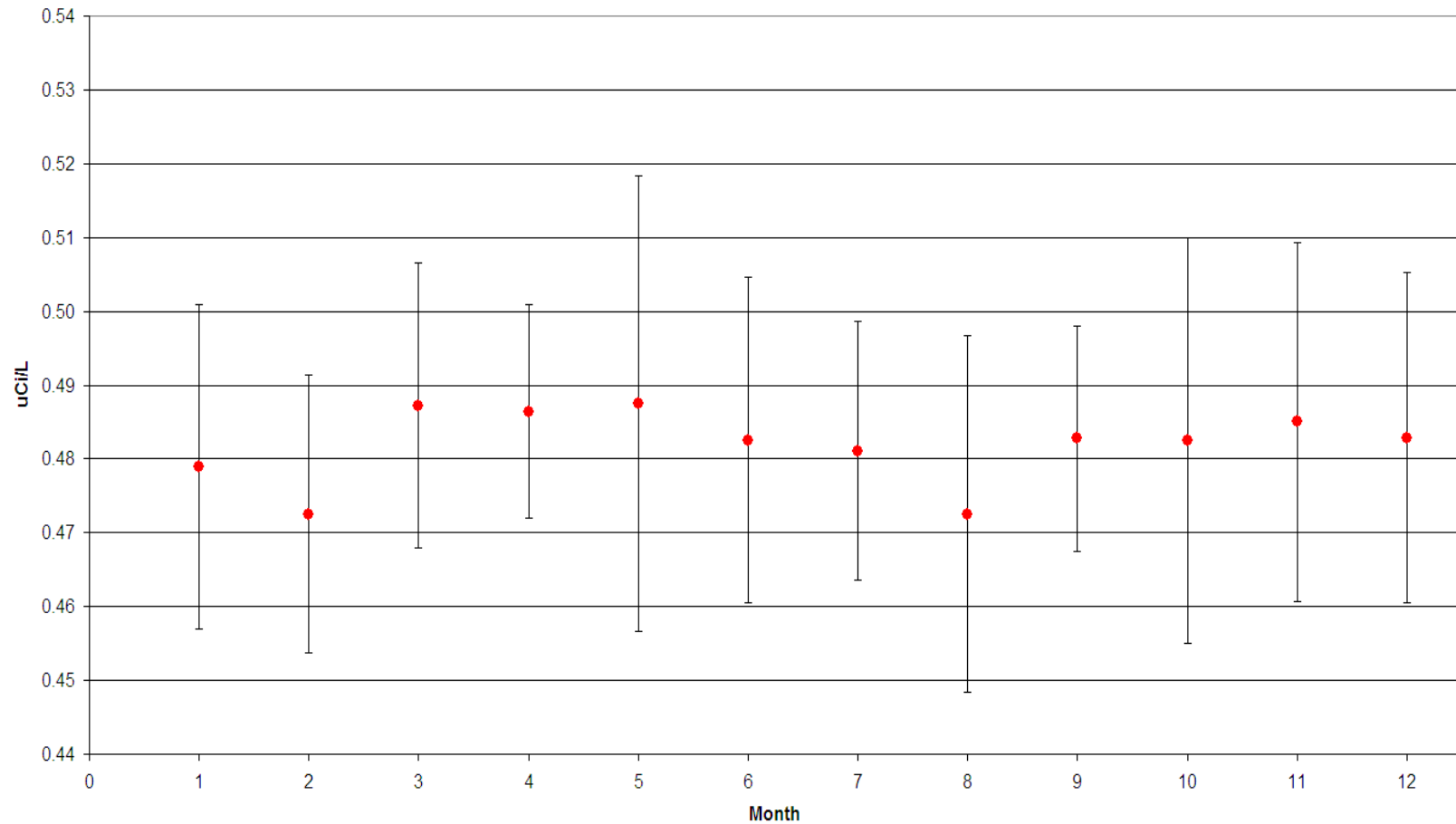
Statistical Evaluation



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CY2008 PERFORMANCE BY MONTH

LSC-07 STANDARD PERFORMANCE CY-2008



ORNL Intercomparison

- **Quarterly Samples.**
- **Run using approved method.**
- **Historical Performance:**

Average % Bias 2005 – 2008 (n = 58): -3.1%

ORNL Tritium Results 2005

<u>MM/YY</u>	<u>K</u>	<u>2sigma</u>	<u>M</u>	<u>2 sigma</u>	<u>%B</u>
January-05	249	12	243	34	-2.36
	249	12	220	31	-11.7
March-05	245	12	231	34	-5.65
	245	12	229	33	-6.53
April-05	252	13	254	34	0.78
	252	13	242	35	-4.13
June-05	251	13	241	36	-4.03
	251	13	255	33	1.67
July-05	269	13	251	34	-6.85
	269	13	261	33	-2.86
September-05	253	13	260	33	2.76
October-05	261	13	282	37	7.95
	261	13	290	35	11.01
December-05	267	13	276	37	3.39
	267	13	267	35	0.13



ORNL Tritium Results 2006

<u>MM/YY</u>	<u>K</u>	<u>2sigma</u>	<u>M</u>	<u>2 sigma</u>	<u>%B</u>
January-06	273	14	264	36	-3.05
	273	14	265	35	-2.99
March-06	250	12	243	37	-2.77
	250	12	224	33	-10.5
April-06	259	13	269	37	3.7
	259	13	265	35	2.16
June-06	260	13	240	36	-7.89
	260	13	256	33	-1.58
July-06	256	13	281	37	9.75
	256	13	250	33	-2.56
September-06	266	13	271	37	1.71
	266	13	261	34	-1.99
October-06	259	13	251	35	-3
	259	13	244	33	-5.49
December-06	248	12	249	36	0.65
	248	12	243	33	-1.76



ORNL Tritium Results 2007

<u>MM/YY</u>	<u>K</u>	<u>2sigma</u>	<u>M</u>	<u>2 sigma</u>	<u>%B</u>
January-07	268	13	268	42	-0.03
	268	13	238	37	-10.93
March-07	248	12	224	34	-9.72
	248	12	210	33	-15.22
April-07	249	12	235	35	-5.91
	249	12	231	33	-7.23
June-07	268	13	266	37	-0.75
	238	13	262	36	-2.38
July-07	250	12	211	35	-15.59
	250	12	233	35	-6.68
September-07	264	13	254	38	-3.51
	264	13	227	30	-13.97
October-07	253	13	247	37	-2.49
	253	13	240	34	-5.14
December-07	251	13	226	41	-10.25

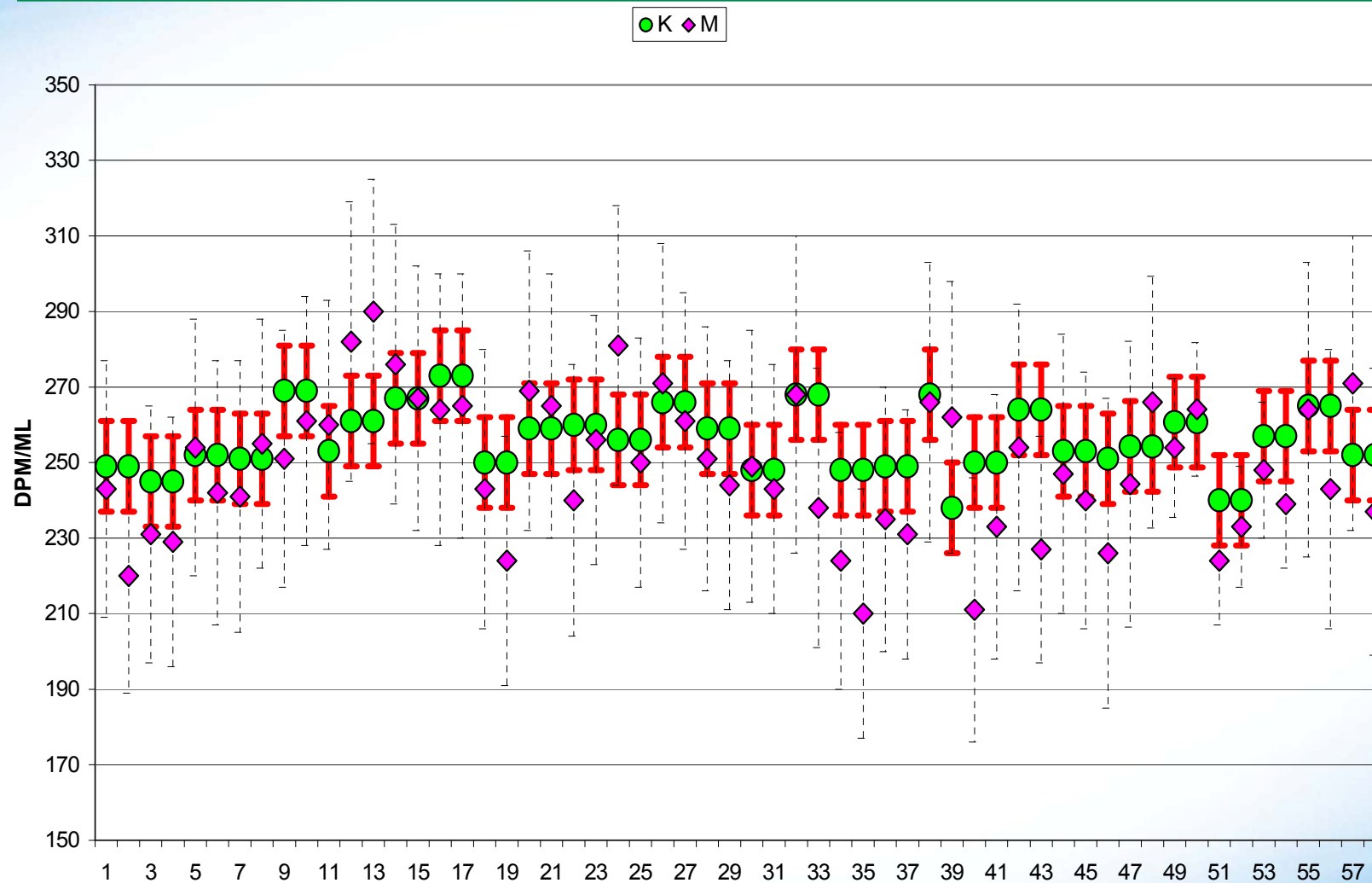


ORNL Tritium Results 2008

<u>MM/YY</u>	<u>K</u>	<u>2sigma</u>	<u>M</u>	<u>2 sigma</u>	<u>%B</u>
January-08	254.3	12.7	244.29	37.82	-3.9
	254.3	12.7	266.01	33.34	4.6
March-08	260.7	13	253.91	18.37	-2.6
	260.7	13	264.1	17.65	1.3
April-09	240	12	224	17	-6.5
	240	12	233	16	-3.03
June-08	257	13	248	18	-3.55
	257	13	239	17	-7.11
July-08	265	13	264	39	-0.09
	265	13	243	37	-8.29
September-08	252	13	271	39	7.41
	252	13	237	38	-5.93



ORNL BLINDS



DOELAP Accreditation

- Audited every 3 years
- Performance test by Blind Analysis
- Historical Performance Excellent
- **Session 2005:**
 - Known 7.79 +/- 0.19 E4 pCi/L
 - Measured 7.8 +/- 0.7 E4 pCi/L
- **Session 2008:**
 - Known 5.50 +/- 0.11 E4 pCi/L
 - Measured 6.1 +/- 0.3 E4 pCi/L

DEPARTMENT OF ENERGY LABORATORY

ACCREDITATION PROGRAM

INDIVIDUAL NUCLIDE REPORT

Reference Date: 3/1/2005

Radionuclide: H-3

RESL Log # DL34

Laboratory: LAB 9

Matrix: SU

Session: 0205

Sample #	Known Values and 1s Uncertainty	Experimental Results and 1s Uncertainty	Bias
DL4SU0205LB-1	7.79 +/- 0.19 E4 pCi/L	6.7 +/- 0.5 E4 pCi/L	-0.140
DL4SU0205LB-2	7.79 +/- 0.19 E4 pCi/L	8.0 +/- 0.6 E4 pCi/L	0.027
DL4SU0205LB-3	7.79 +/- 0.19 E4 pCi/L	8.6 +/- 0.6 E4 pCi/L	0.104
DL4SU0205LB-4	7.79 +/- 0.19 E4 pCi/L	7.4 +/- 0.5 E4 pCi/L	-0.050
DL4SU0205LB-5	7.79 +/- 0.19 E4 pCi/L	7.5 +/- 0.5 E4 pCi/L	-0.037
DL4SU0205LB-6	7.79 +/- 0.19 E4 pCi/L	8.6 +/- 0.6 E4 pCi/L	0.104

Mean (Br) = 0.001

St. Dev (Sb) = 0.096

Acceptance Criteria: -0.25 =< Br <= 0.5

Sb =< 0.4



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DEPARTMENT OF ENERGY LABORATORY ACCREDITATION PROGRAM INDIVIDUAL NUCLIDE REPORT

Reference Date: 2/15/2008

Radionuclide: H-3

RESL Log # DL267

Laboratory: LAB 4

Matrix: SU

Session: 0108

Sample #	Known Values and 1s Uncertainty	Experimental Results and 1s Uncertainty	Bias
DL5SU0108LB-1	5.50 +/- 0.11 E4 pCi/L	6.4 +/- 0.3 E4 pCi/L	0.164
DL5SU0108LB-2	5.50 +/- 0.11 E4 pCi/L	6.4 +/- 0.3 E4 pCi/L	0.164
DL5SU0108LB-3	5.50 +/- 0.11 E4 pCi/L	5.8 +/- 0.3 E4 pCi/L	0.055
DL5SU0108LB-4	5.50 +/- 0.11 E4 pCi/L	6.0 +/- 0.3 E4 pCi/L	0.091
DL5SU0108LB-5	5.50 +/- 0.11 E4 pCi/L	6.0 +/- 0.3 E4 pCi/L	0.091
DL5SU0108LB-6	5.50 +/- 0.11 E4 pCi/L	6.1 +/- 0.3 E4 pCi/L	0.109

Mean (Br) = 0.112

St. Dev (Sb) = 0.044

Acceptance Criteria: -0.25 <= Br <= 0.50

Sb <= 0.4



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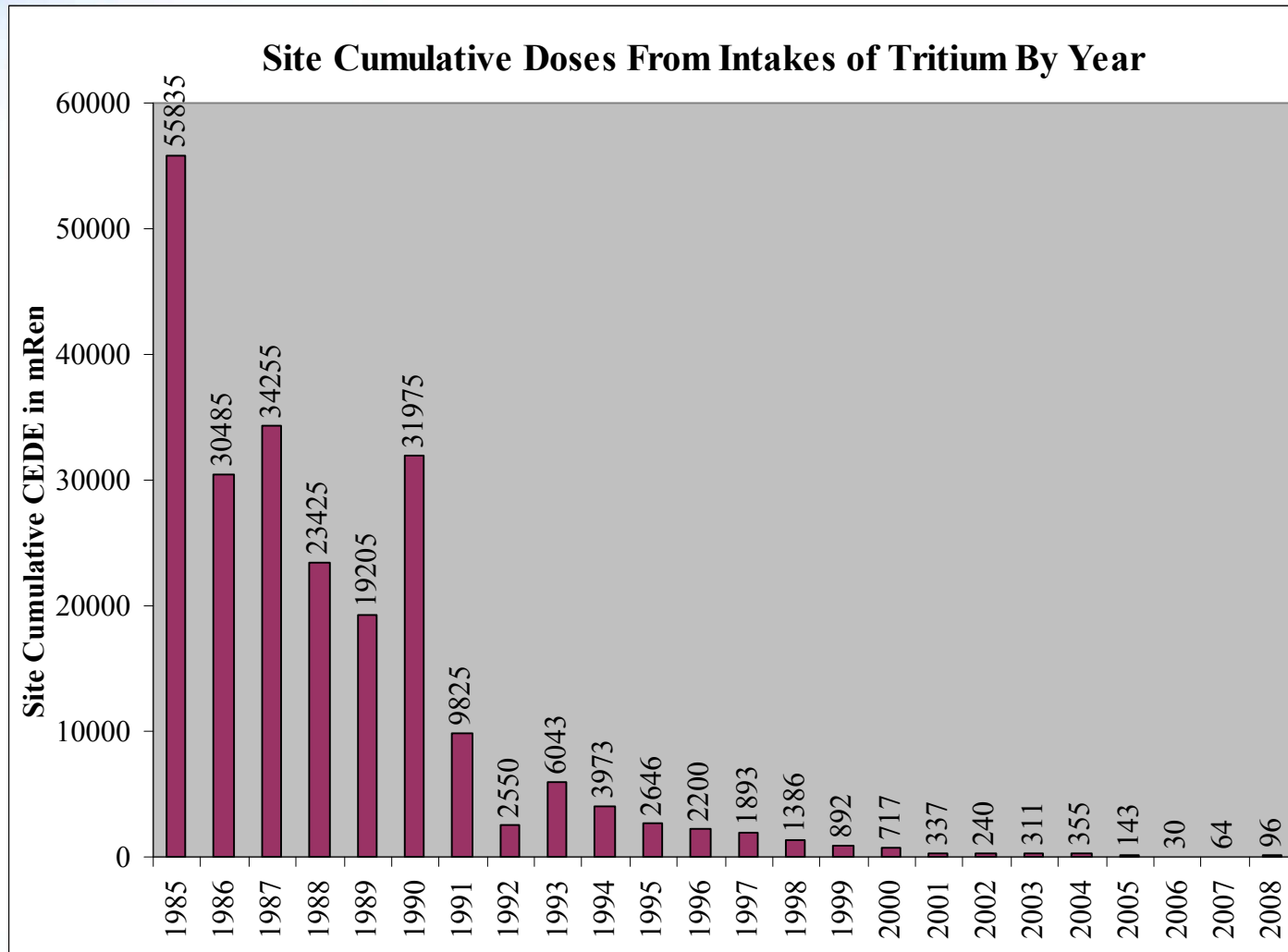
SRS Tritium Exposure History

(Courtesy of Mitch Findley – HPS)



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SRS Tritium Exposure History



SRS Tritium Exposure History

- **End of Cold War ended workers exposure to tritiated moderator at the reactor facilities.**
- **Analyzing about 110,000 tritium urine samples annually (1990).**
- **~ 5,000 samples this year.**
- **Since 2001 doses have been about the same.**
- **PRORAD cut the number of tritium samples submitted in half.**
- **1990 was the last year we had an individual worker exposed to more than 100 mRem of tritium in a calendar year.**

QUESTIONS?



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