National Institute of Standards and Technology

^{209}Po at NIST ; Now and Then

Radiation Physics Division Seminar -- 5 February 2014

R. Collé

Radioactivity Group

NIST Physics Measurement Lab

Thanks to:

Ryan Fitzgerald; Lizbeth Laureano-Perez; Iisa Outola;

Jerry LaRosa; Leticia Pibida; Lynn King

Polonium

42 known isotopes (A = 186 – 227)
maybe 45 more by nuclear theory

1898 – Marie & Pierre Curie -- ^{210}Po

1950s – ^{208}Po & ^{209}Po

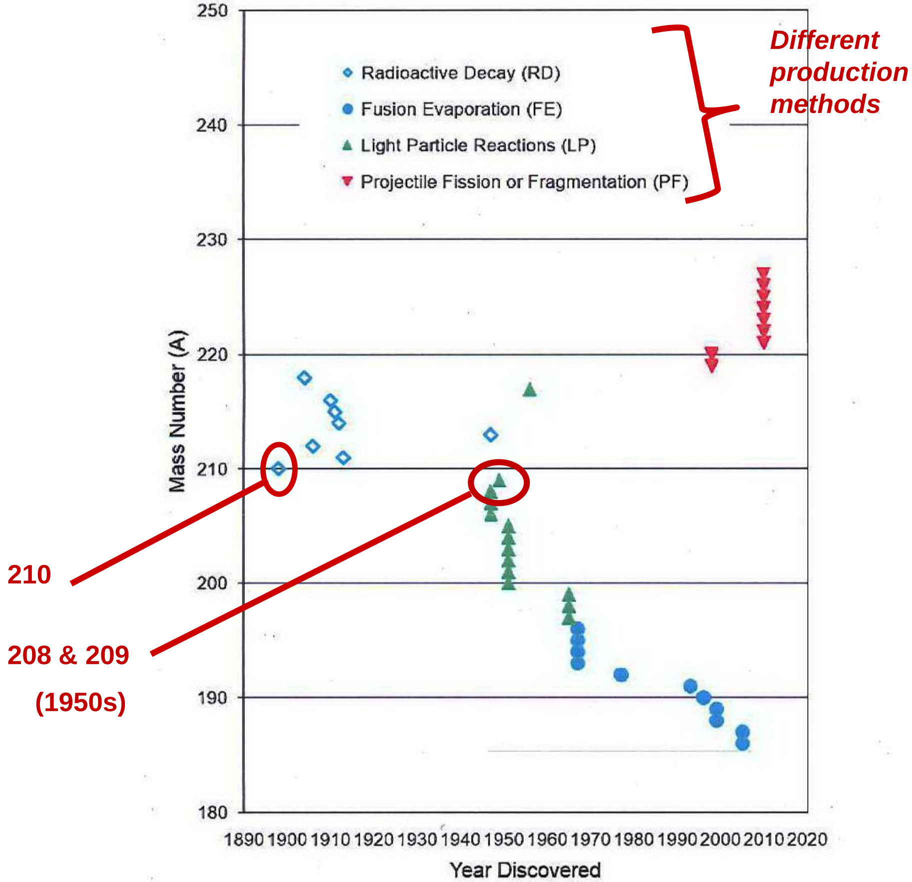
1955 – Andre, et al. ^{209}Po half-life

1975 – NBS / EPA ^{210}Po measurement comparison (NBS std)

1984 – ^{208}Po SRM 4327 (Noyce)

1990 – 2014 – ^{209}Po metrology (Collé era)

Polonium isotopes as a function of time when they were discovered



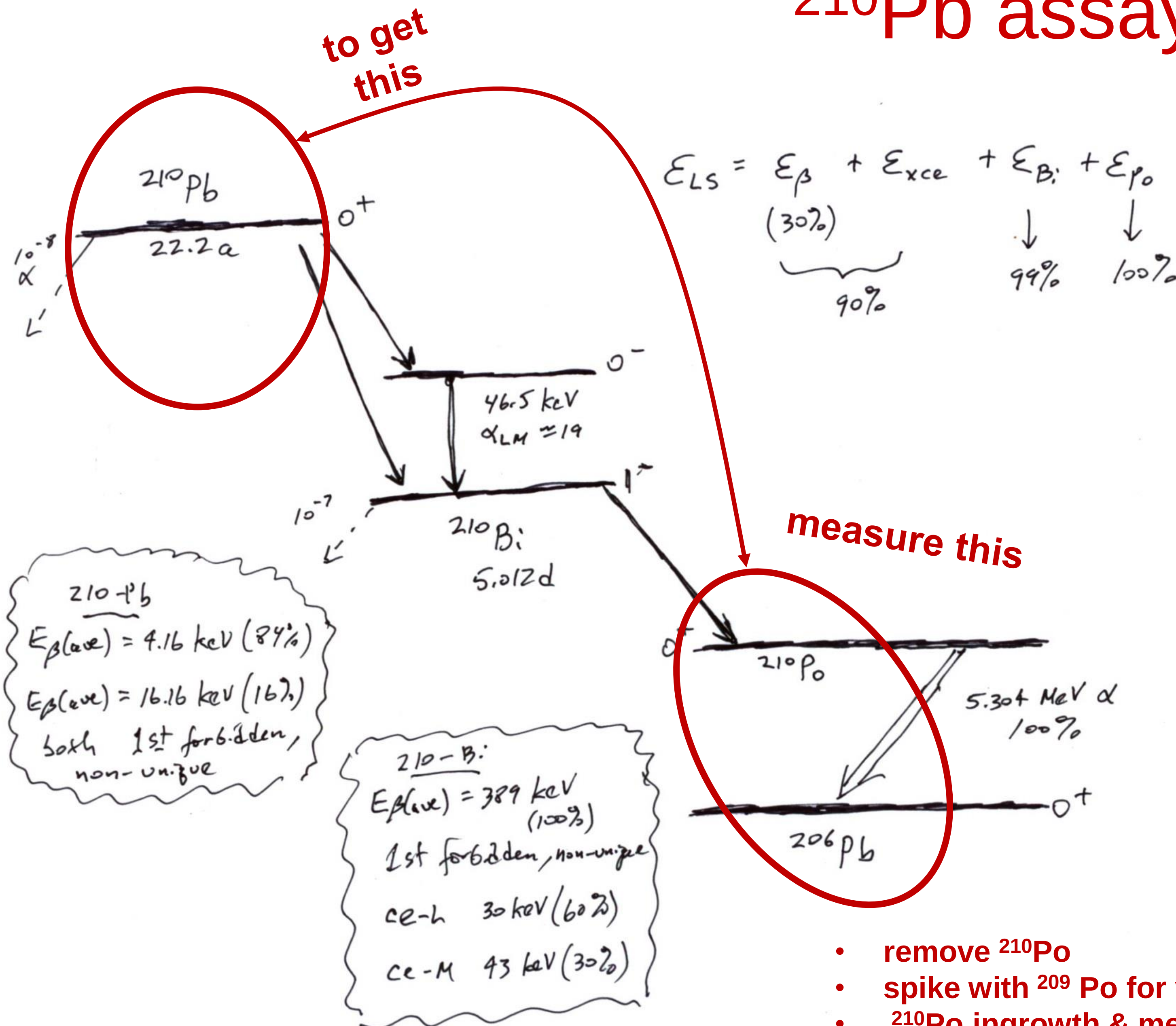
Fry & Thoennessen, *Atomic Data & Nucl. Data Tables* (preprint, 2001)

World needs a Po tracer standard !

One of most critical tracers needed by environmental, geochemical, bioassay, etc. measurement community

^{210}Po	0.4 a	5.3 MeV α
^{208}Po	2.9 a	5.1 MeV α
^{209}Po	~~102 a~~	4.9 MeV α + *junk*

^{210}Pb assay



- remove ^{210}Po
- spike with ^{209}Po for yield
- ^{210}Po ingrowth & measure both

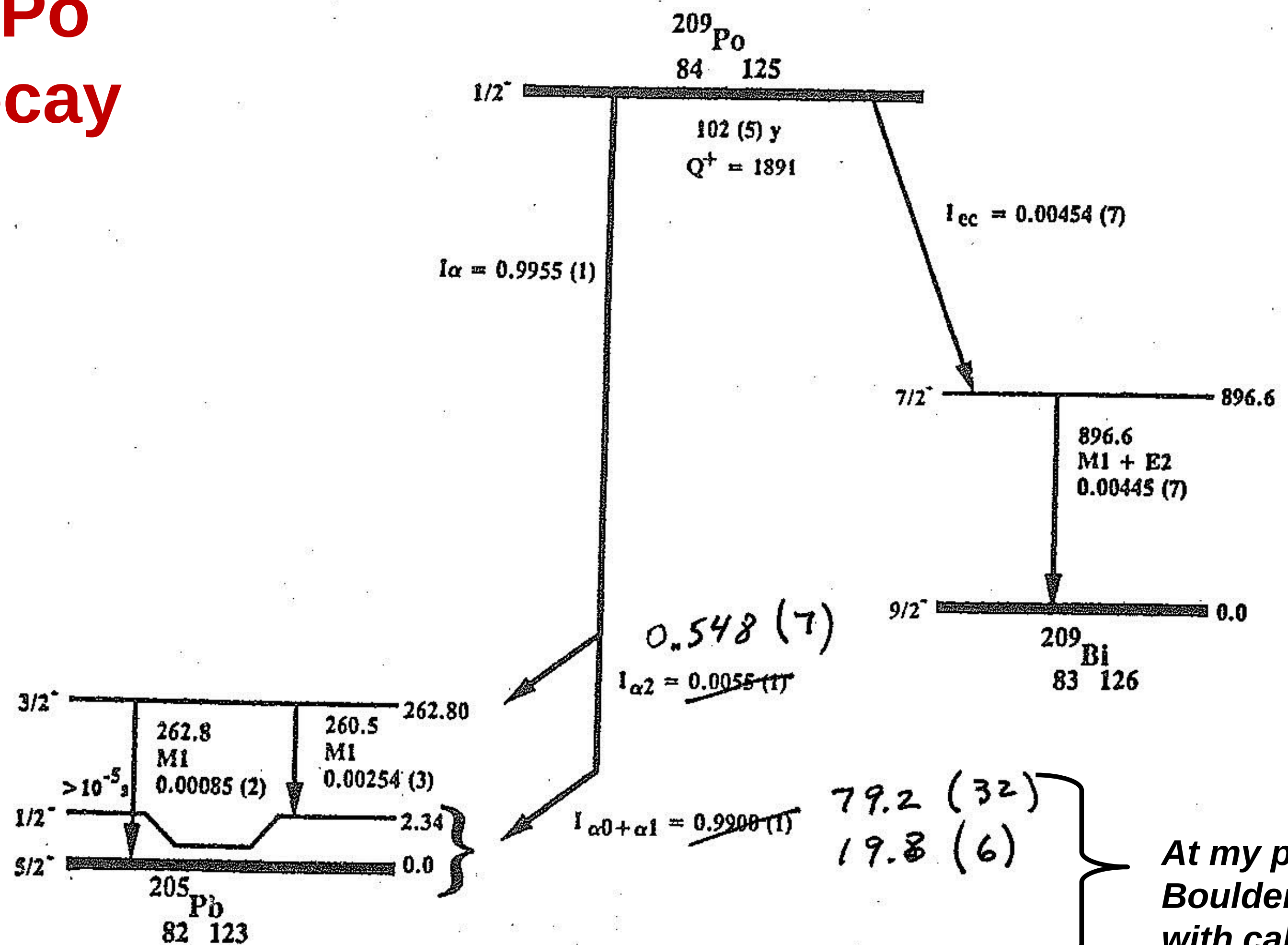
^{209}Po at NBS / NIST

1990 -- polonium solution stability	R. Collé, *Radioact. Radiochem*. **4**, no.2, 20-35 (1993).
1993 -- ^{209}Po SRM 4326	R. Collé, Z. Lin, et al., *J. Res. Natl. Inst. Std. Tech*. **100**, 1-36 (1995).
1994 -- ^{209}Po decay scheme	F.J Schima and R. Collé, *Nucl. Instrum. Meth, Phys. Res.* **A369**, 498-502 (1996).
1994 -- ^{205}Pb delayed isomeric state / LS implications	R. Collé, Z. Lin, et al., *Appl. Radiat. Isotop.* **45,** 1165-1175 (1994).
2005 -- ^{209}Po SRM 4326 recertification	R. Collé, L. Laureano-Perez , et al. [Certificate]
2005 -- ^{209}Po half-life discrepancy	R. Collé, L. Laureano-Perez, et al., *Appl. Radiat. Isotop*. **65,** 728-730 (2007).
2006 -- ^{210}Pb SRM 4337	L. Laureano-Perez, R. Collé, et al., *Appl. Radiat. Isotop*. **65,** 1368-1380 (2007).
2007 -- ^{209}Po & ^{210}Pb std methodology	R. Collé, L. Laureano-Perez, LSC 2008, Radiocarbon, 2009, pp.77-85
2008 -- ^{210}Pb intercomparison (NPL)	R. Collé, L. Laureano-Perez, LSC 2008, Radiocarbon, 2009, pp.77-85
2013 -- New 209Po standardization method	R. Collé, R. Fitzgerald, L. Laureano-Perez
2013 -- ^{209}Po SRM 4326a	R. Collé, L. Laureano-Perez,
2013 -- ^{209}Po definitive half-life	R. Collé, R. Fitzgerald, L. Laureano-Perez

[

^{209}Po decay

A369, 498 (1996)



At my prompt, NIST Boulder lab work with calorimeter (pre-Pu accident)

1

1990
stability

Polonium solution stability (c. 1990)

What known then !

"The Storage of Polonium Solutions", C. Rosenblum & E.W. Kaiser, *J. Phys. Chem.*, 1935, **39** (6), 797–802

exhibits colloidal behavior "under suitable acid conditions"
- dialyzability through membranes
- small diffusion velocity
- centerfugability
- Settling under gravity
- behavior in electric fields

solutions "show presence of large aggregations of radioactive atoms"
…."uncertain whether true colloids are involved", coined term "radiocolloid"
Wanted to find useful conditions to minimize colloid formation – none found to be total
"opinions as to cause of phenomena" (and existence) "are not in accord"

Radiochemistry of Polonium, P.E. Figgins, Radiochemistry of Elements Series, Natl. Acad Sciences, 1961

- no easy way to store long-term
- strange suggestions for storage – waxed glass

Lots of conflicting info on volatility, colloids & stability in acid

Polonium und Isotope, Gmelins Handbuch der Anorganisch Chemie, Verlag Chemie, 1941, 203 pages [NBS Library English Translation, 1945]

- questions volatility until very high temperatures
- chemistry in HCl and species in range 1 to 3 mol/L HCl range
- hydrolyzation issues in other solutions
- colloidal formation is rare or nonexistent in acidic solutions > 0.1 mol/L

Belief then

Po solutions at trace concentrations, under various alkaline, neutral,, or weakly acidic conditions are known to be unstable: being readily hydrolyzed, chemically deposited, or volatilized,; exhibiting “radiocolloidal” behavior; and undergoing “plate-out” or adsorption onto glass surfaces.

Stored Po solutions are generally considered by NIST to be stable in the acid range of 0.1 to 1.0 normality, but scant data exist on any possible long-term effects , particularly for very dilute, carrier free, aged solutions.

Looked like time to do some work …..

c. 1990 Collé study

(indebted to Jim Noyce for his meticulous records)

Carrier-free ^{208}Po and ^{210}Po solutions with "known" massic activity re-assayed after aged storage in flame-sealed glass ampoules storage

Number solutions studied	11
Range of solution ages	(1.2 to 8.8) years
Range of HCl normality	(0.09 to 2.0) mol/L
Range of massic activity	(< 0.1 to 3600) Bq/g
Range of recoveries found	(< 0.1 to 1.03) %
Typical recovery uncertainty (k = 1)	> 20 % at 0.1 Bq/g 1.2 % at 200 Bq/g

If 1 is Good;
2 is Better

Conclusions

< 0.3 mol/L	– "clearly unstable"
0.3 to 0.5 mol/L	– "somewhat equivocal"
$\geq$ 1 mol/L	– "appear to be stable over many years approaching a decade"

Therefore use 2 mol/L

R. Collé, *Radioact. Radiochem*. **4**, no.2, 20-35 (1993).

National Institute of Standards and Technology

2

1993 – 1994

SRM 4326

SRM 4326

Certified massic alpha emission rate
(NOT activity)

Measurements in periods

1993	March	4 days
1993	June	25 days
1994	March	15 days

Reference time -- 1200 EST 15 march 1994

(85.42 ± 0.29) $s^{-1} \cdot g^{-1}$ 1200 EST 15 march 1994

(5.1597 ± 0.0024) g solution in flame-sealed NIST ampoule

nominal 2 $mol \cdot L^{-1}$ HCl, carrier-free

(1.031 ± 0.004) $g^{-1} \cdot mL^{-1}$

^{208}Po impurity -- (0.106 ± 0.017) $s^{-1} \cdot g^{-1}$

R. Collé, Z. Lin, et al., *J. Res. Natl. Inst. Std. Tech*. **100**, 1-36 (1995).

methodology later – required 0.9988 correction for EC branch

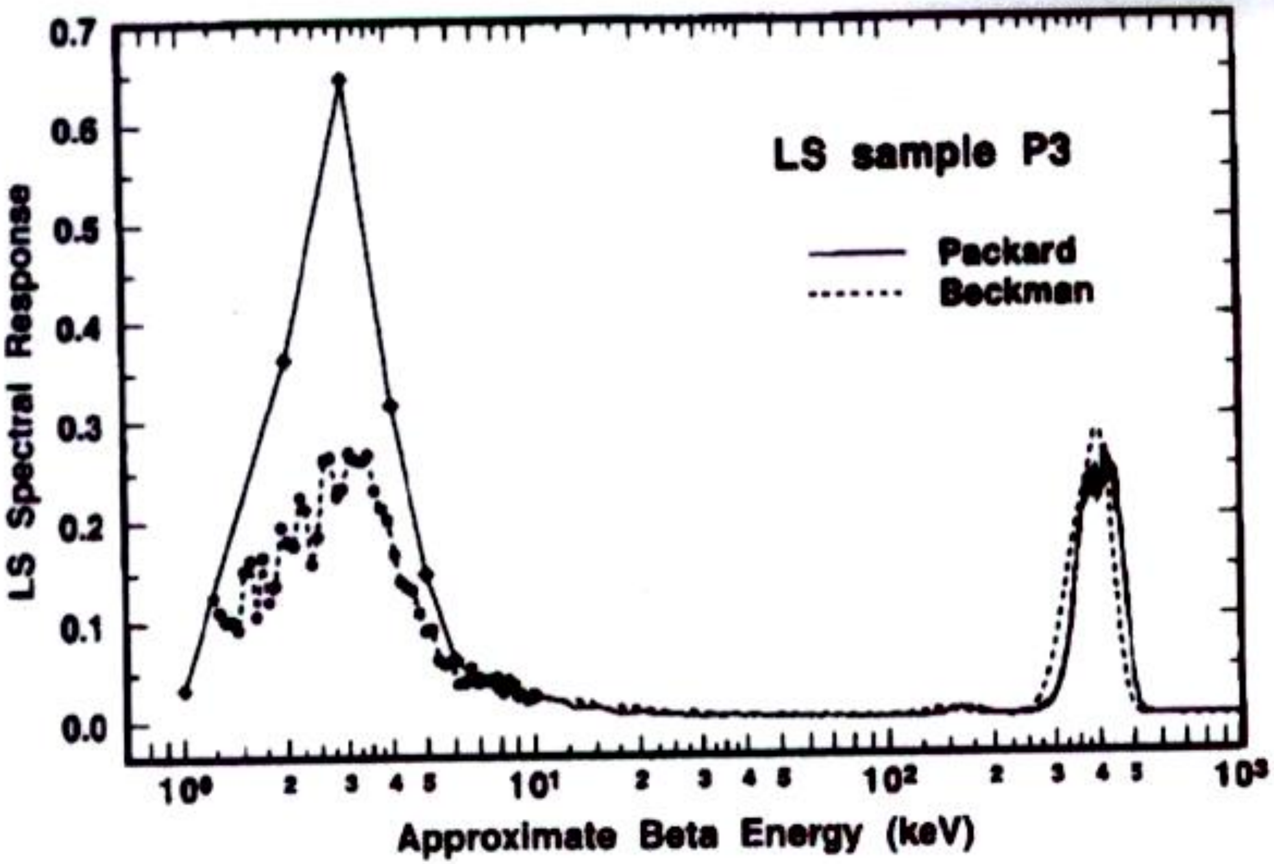


Fig. 6. Comparison of the ^{209}Po LS spectra obtained with the Beckman and Packard instruments.

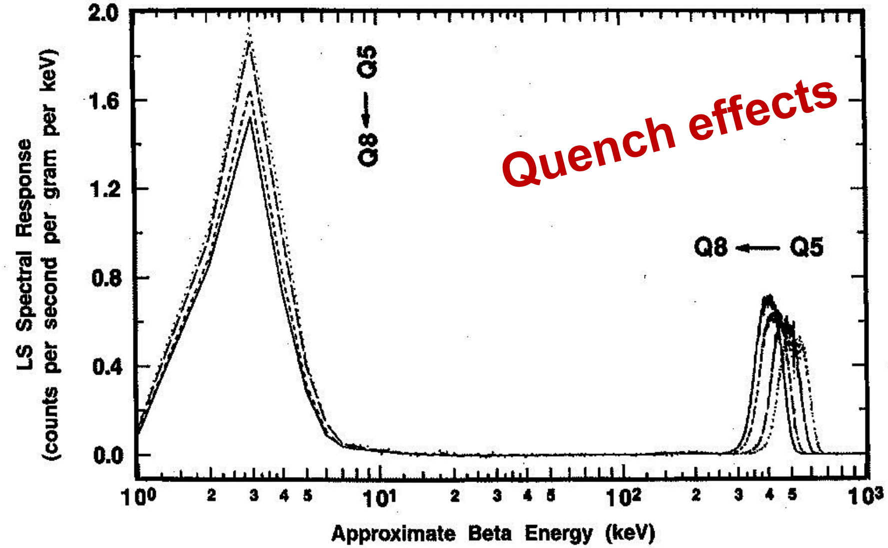


Fig. 14. LS spectra of increasingly quenched samples Q5 through Q8 obtained with the Packard counting system.

"anomalous bumps"

-- investigated as optical effects

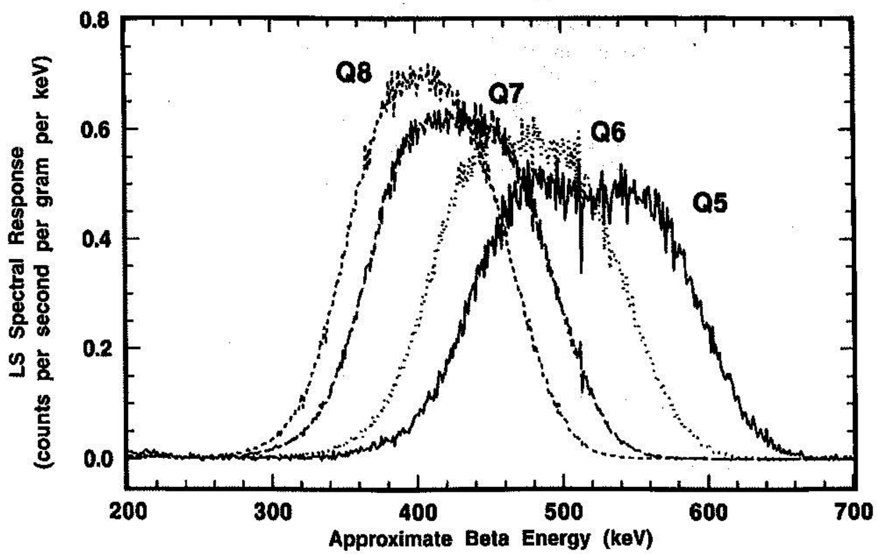


Fig. 15. Details of the broad alpha peaks shown in the full spectra of Fig. 14. The peak widths (FWHM) on a relative basis and peak areas are approximately equal in all four samples Q5 through Q8.

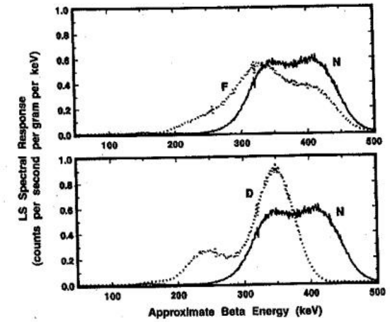


Fig. 21. Details of the broad alpha peaks for the three source configurations shown in the spectra of Fig. 20.

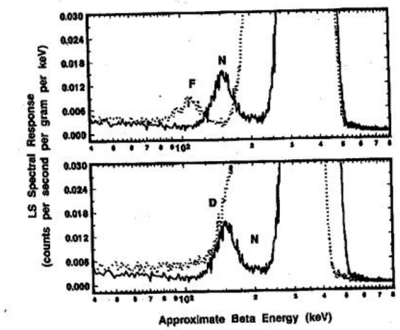


Fig. 22. Details of the "anomalous bumps" for the three source configurations shown in the spectra of Fig. 20.

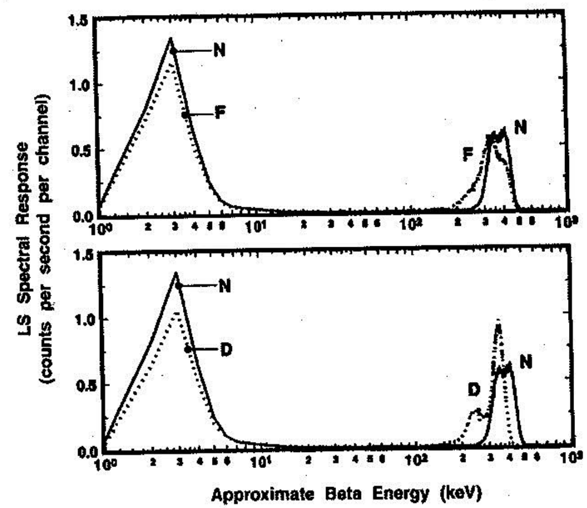


Fig. 20. Comparative ^{209}Po LS spectra for three very different source configurations N, F, and D. Refer to text for details. The upper trace compares the spectra for N and F; the lower compares N and D.

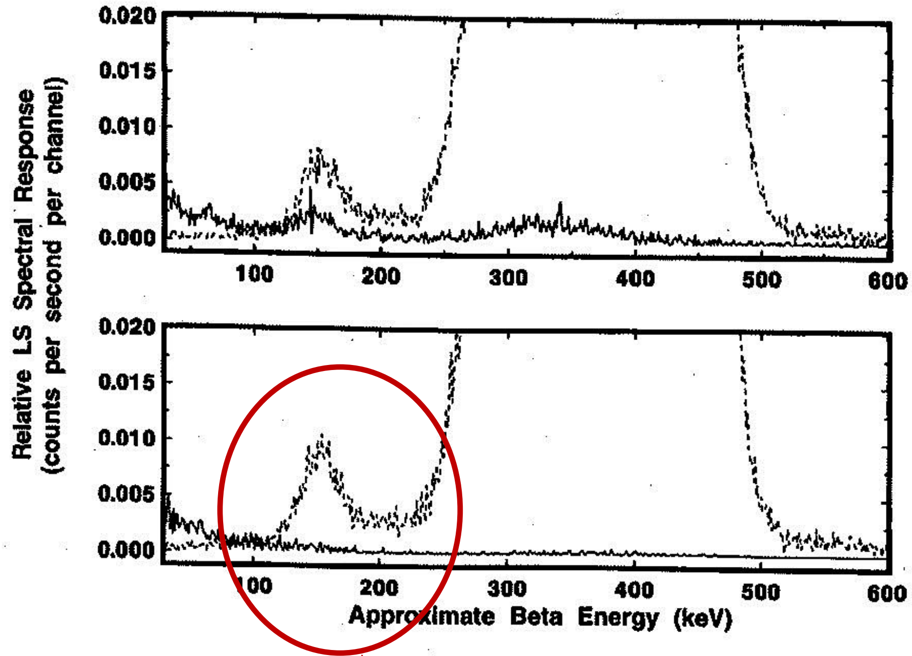


Fig. 19. Details of the LS spectrum of Fig. 18 showing the relative alpha and beta responses for the "anomalous bump" and α peak at two pulse discrimination settings.

must be alpha

from discrimination
&
from counting
consistency

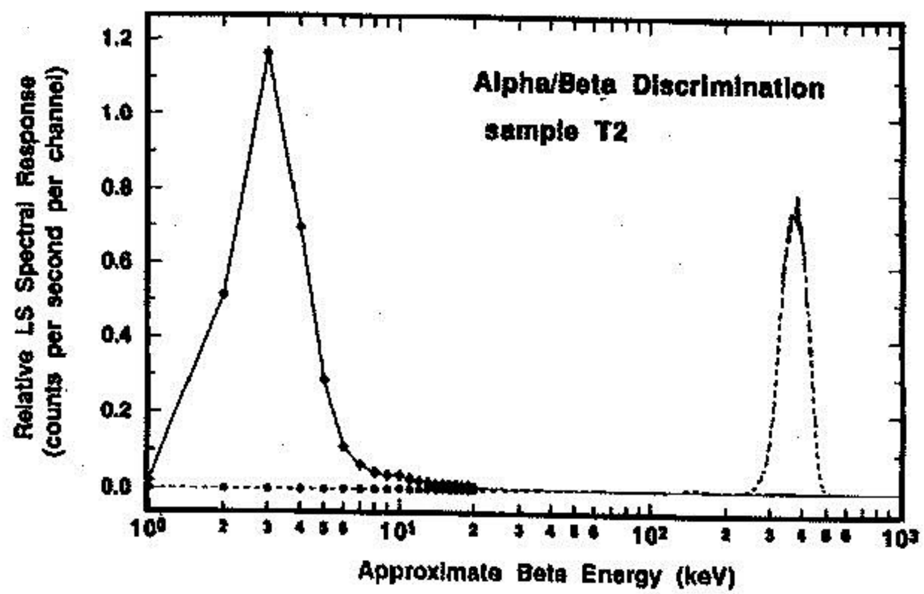


Fig. 18. LS spectrum of ^{209}Po obtained using alpha and beta pulse discrimination.

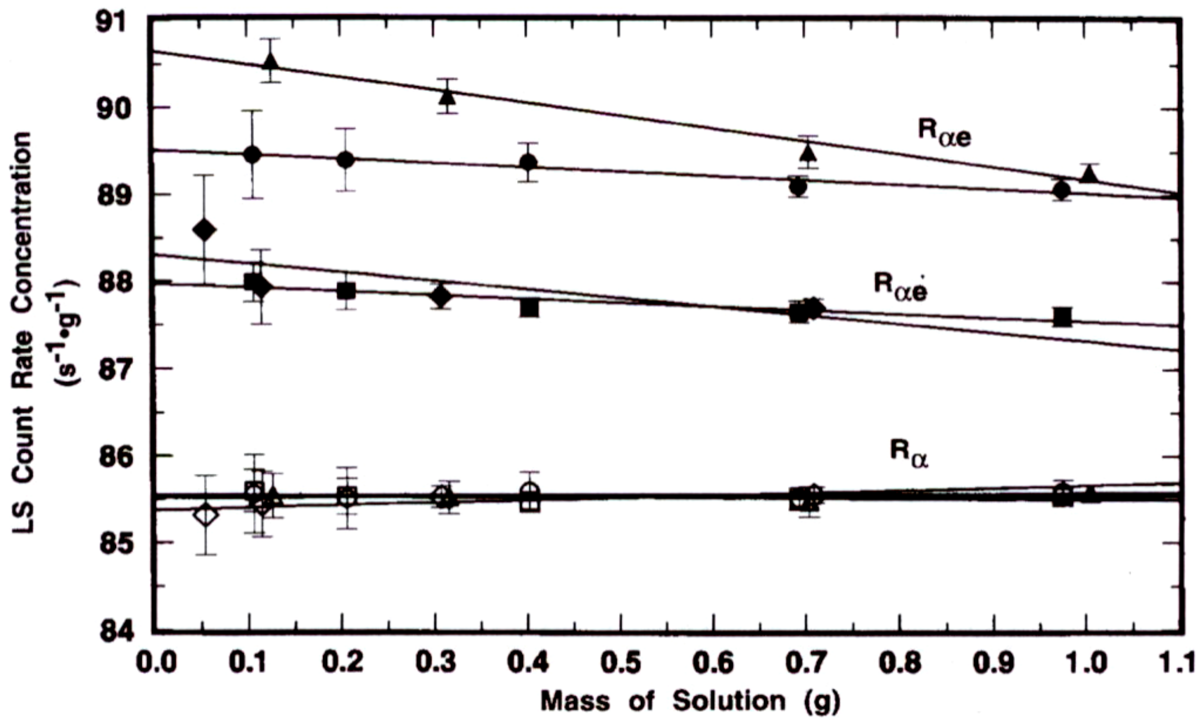


Fig. 13. LS counting rate concentrations $R_{\alpha e}$ and R_α obtained with the two LS systems for the P and Q series samples in 1994. Closed squares ($R_{\alpha e}$) and open squares (R_α) represent the mean values for samples Q5 through Q8 with the Packard; closed and open triangles represent $R_{\alpha e}$ and R_α, respectively, for samples P1 through P5 with the Packard; closed and open triangles ($R_{\alpha e}$ and R_α) are for samples Q1 through Q4 with the Beckman; and closed and open circles ($R_{\alpha e}$ and R_α) are for samples P1 through P5 with the Beckman. Each plotted value corresponds to the mean of 5 to 18 replicate measurements on each sample. The error bars represent standard deviation uncertainty intervals on the means. The solid lines are unweighted linear fits to the data. Although the $R_{\alpha e}$ values vary with the instrument used to perform the measurements (Packard or Beckman) and with sample compositions, all of the R_α values are statistically equivalent and invariant.

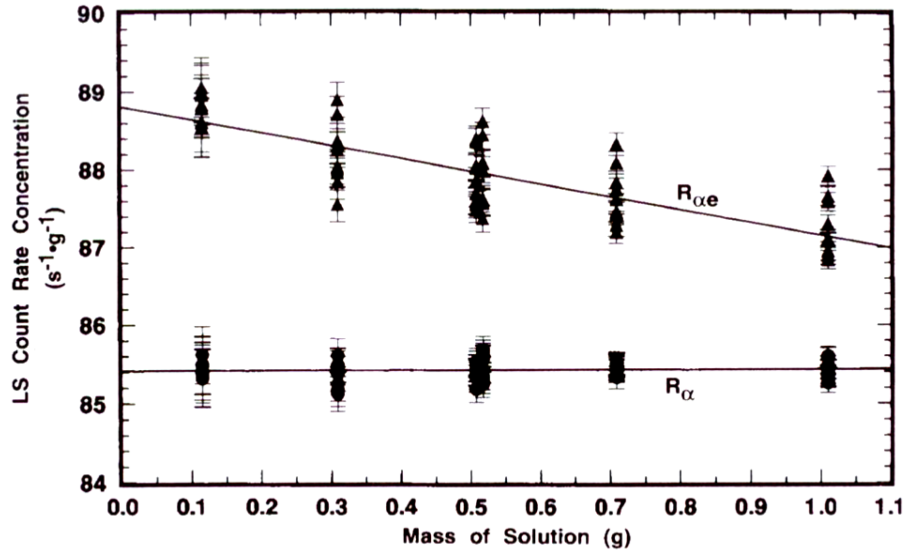


Fig. 12. LS counting rate concentrations $R_{\alpha e}$ and R_{α} as a function of m_s (analogous to that of Fig. 11) as obtained with the Packard instrument.

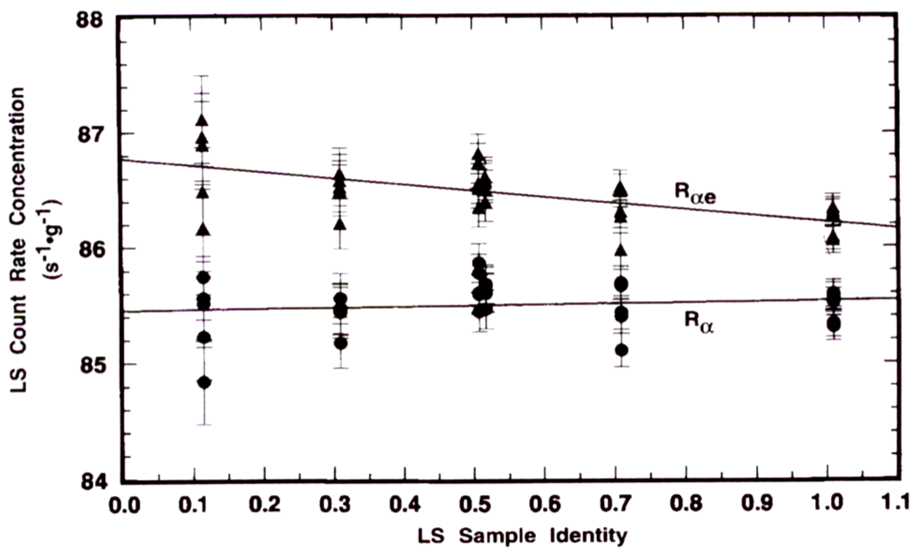


Fig. 11. LS counting rate concentrations $R_{\alpha e}$ (closed triangles) and R_{α} (closed circles) obtained with the Beckman instrument for the N series samples as a function of m_s (and sample quenching). The solid lines are linear regressions fitted to the data.

Oh yes,

for a **good read**

see the **34 page** paper in *NIST J. Res*.

for “anomalous bumps” – still unexplained optical effects ?

and a just wonderful & creative appendix on the EC correction

Photonic emission work
with Frank Schima (1994)

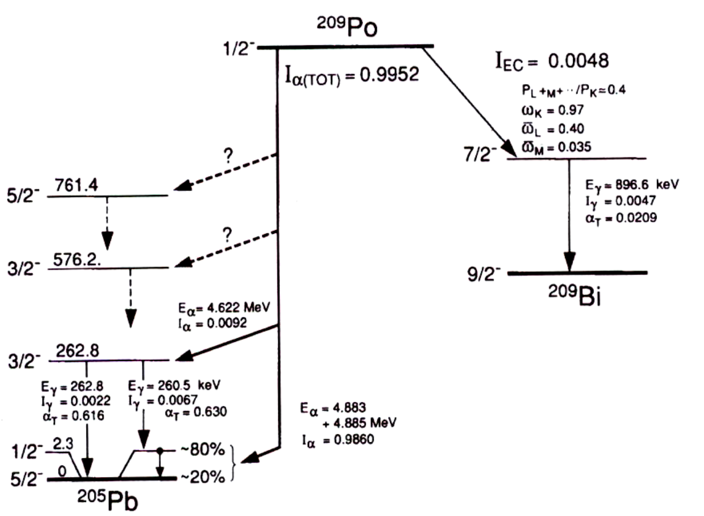
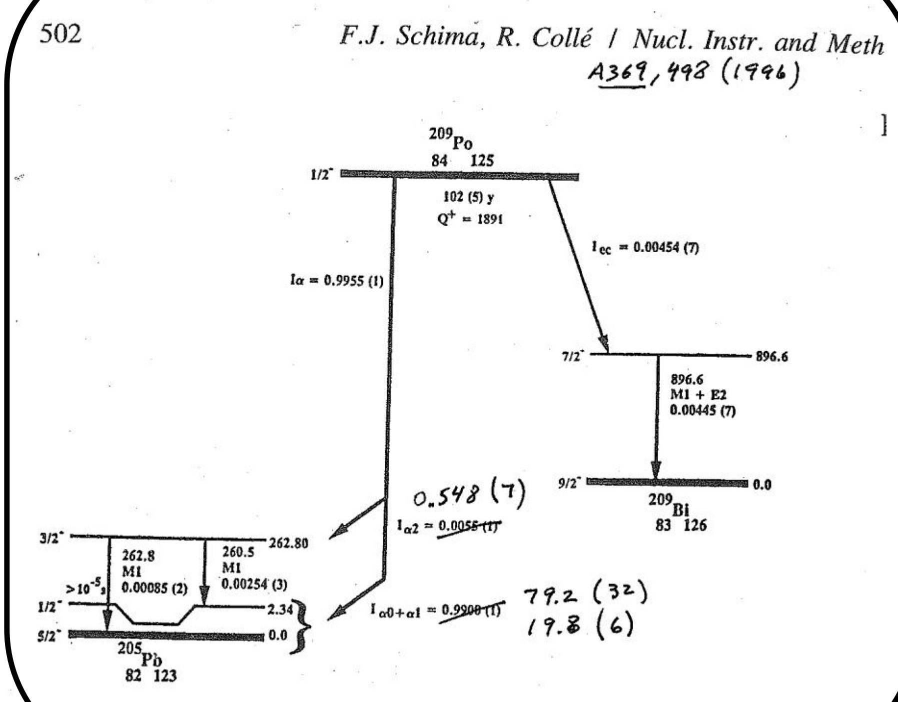


Fig. 2. Partial decay scheme for the ^{209}Po alpha and electron capture branch decays.

4

BEFORE



AFTER

3

2005
re-certify ! ! !

November 2005

Re-certify SRM 4326

Identical Methodology – same kinds of results

found

1.6 % discrepancy

with decay-corrected 1994 value using extant (102 $\pm$ 5) half-life

from two measurements each with uncertainties of about 0.2 % (k = 1) made approximately 12 years apart

! ! ! !

didn't initially believe

15 march 1994

R_α = (85.42 ± 0.18) $s^{-1}g^{-1}$

15 November 2005

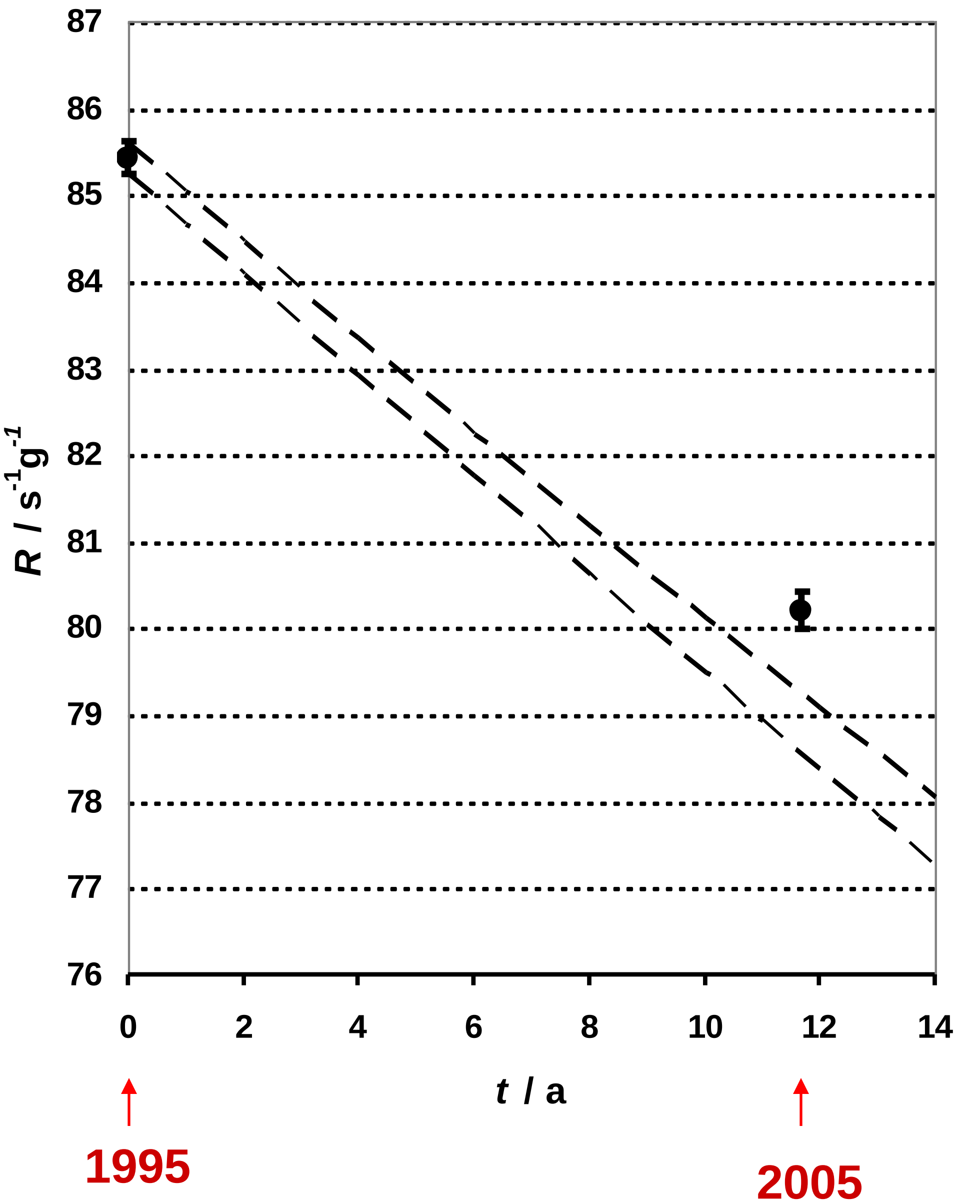
R_α = (80.20 ± 0.22) s-1g-1

2 point fit gives

$T_{1/2}$ = 128 a

U = 5.5 % (7 a)

<u>Not</u> considered a new determination



Only ^{209}Po Half-life Determination

Andre, Huizenga, et al. **1956** *Phys Rev.* 101, 645-651

^{208}Po/^{209}Po mass ratio 1.14 %

^{208}Po/^{209}Po activity ratios 5 %

"private communication"

with $T_{1/2}(^{208}\mathrm{Po}) = (2.93 \pm 0.03)$ a,

got $T_{1/2}(^{208}\mathrm{Po}) = 103$ a

Compiler M. Martin,1991

with $T_{1/2}(^{208}\mathrm{Po}) = 2.898 \pm 0.002$ a,

got $T_{1/2}(^{208}\mathrm{Po}) = (102 \pm 5)$ a 4.9 %

Based on 2005 finding must be wrong

R. Collé, L. Laureano-Perez, et al., *Appl. Radiat. Isotop*. **65,** 728-730 (2007).

^{209}Po half-life in error by 25 % !!

Result supported by work on ^{210}Pb – another long story

$$A / N = \lambda$$

New determination was urgently needed

Collaboration with Polish Academy of Sciences labs

Institute of Nuclear Physics (Krakow)

Institute of Geological Sciences (Warsaw)

failed

OUTCOME

from Data Evaluation by V. Christie & M.M. Bé (Oct.2009)

Experimental ^{209}Po half-life values (in years) are given in Table 1.

Table 1: Experimental values of ^{209}Po half-life.

Reference	Experimental value (a)	Comments
C. G. Andre (1956An05)	102 (5)	From $^{209}Po/^{208}Po$ mass and activity ratios and $T_{1/2}(^{208}Po)$ = 2.898 (2) a (see 1991Ma16).
R. Collé (2007Co07)	128 (7)	Decay data from two separate primary standardizations of a ^{209}Po solution standard, carried out ~ 12 years apart.
Recommended value	115 (13)	$\chi^2 = 6.9$

The value from 2007Co07 is not a direct measurement of the ^{209}Po half-life. R. Collé said in a private communication: "My paper which stated the value **128 a** was not a new determination… The whole point was to show that there was evidence to suggest and support that the extant **102 a** value is very wrong, perhaps by 25 %".
However, to take into account all scarce information available, the evaluators have chosen to adopt the simple mean of the two existing values (1956An05 and 2007Co07) with an uncertainty covering them. Then, the recommended value is 115 (13) a.

LSC meeting in Davos (2008) – word spread quickly through environmental community – great concern & shock – totally uncertain half-life & no available standard

4

2013

SRM 4326a

Next best thing to one's success

Is someone else's failure !

NPL experience –

started preparing ^{209}Po standards and standardization in 2008 .... about the time we got our new stock material

their results made us nervous – they had ^{209}Po from same Oak Ridge batch --we began to re-think the chemistry

they still have problems as of December 2013

2013

- For half-life data on OLD SRM
  used identical 1993 – 94 methodology again

- For standardization of NEW SRM
  used a new & improved method

- No observable or "statistically significant" difference
  between methods

- New method is just more defensible in withstanding
  critical scrutiny in terms of spectral analyses
  and magnitude of EC correction

DIFFERENCE IN

1994 & 2013

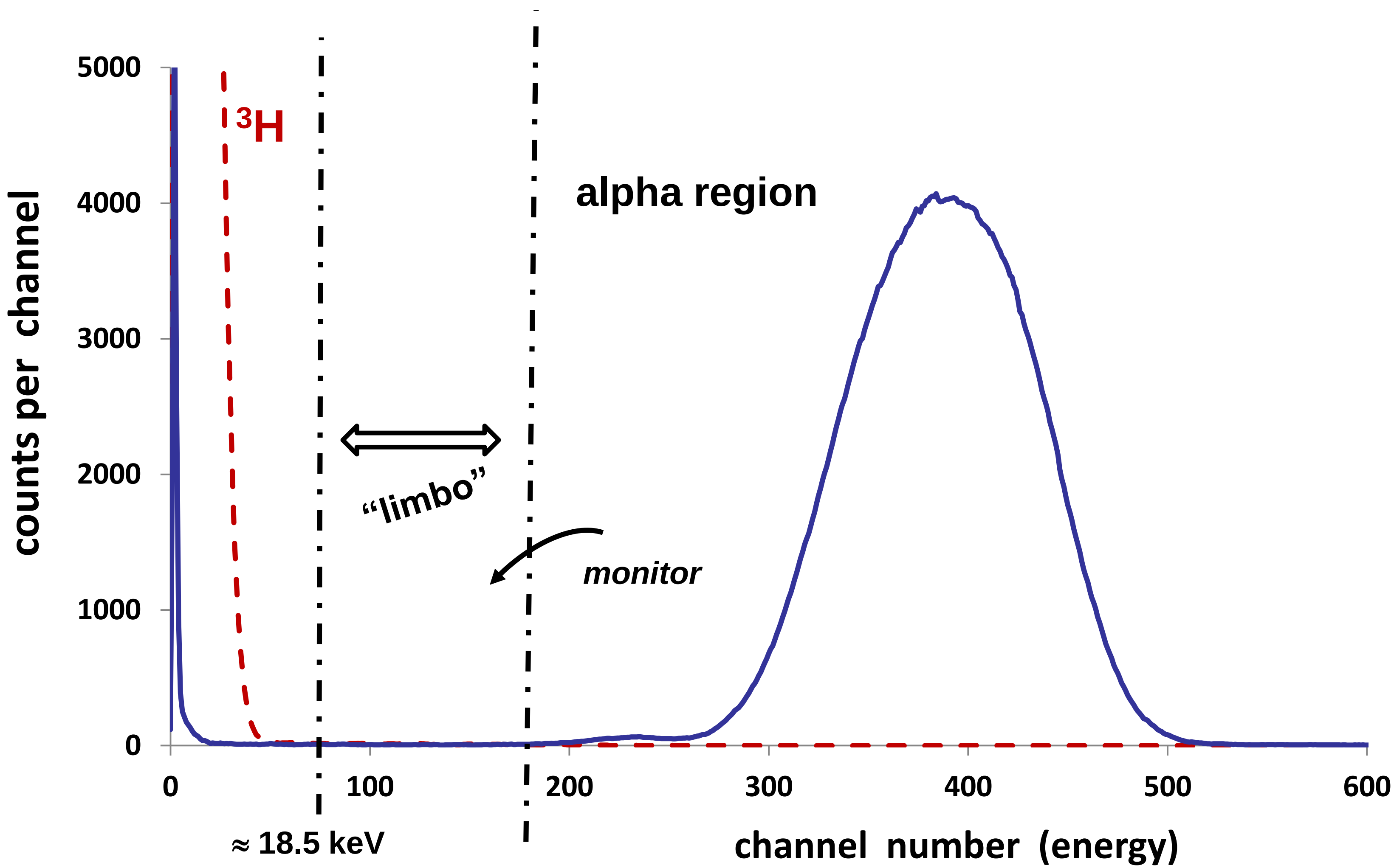
STANDARDIZATION

METHODS

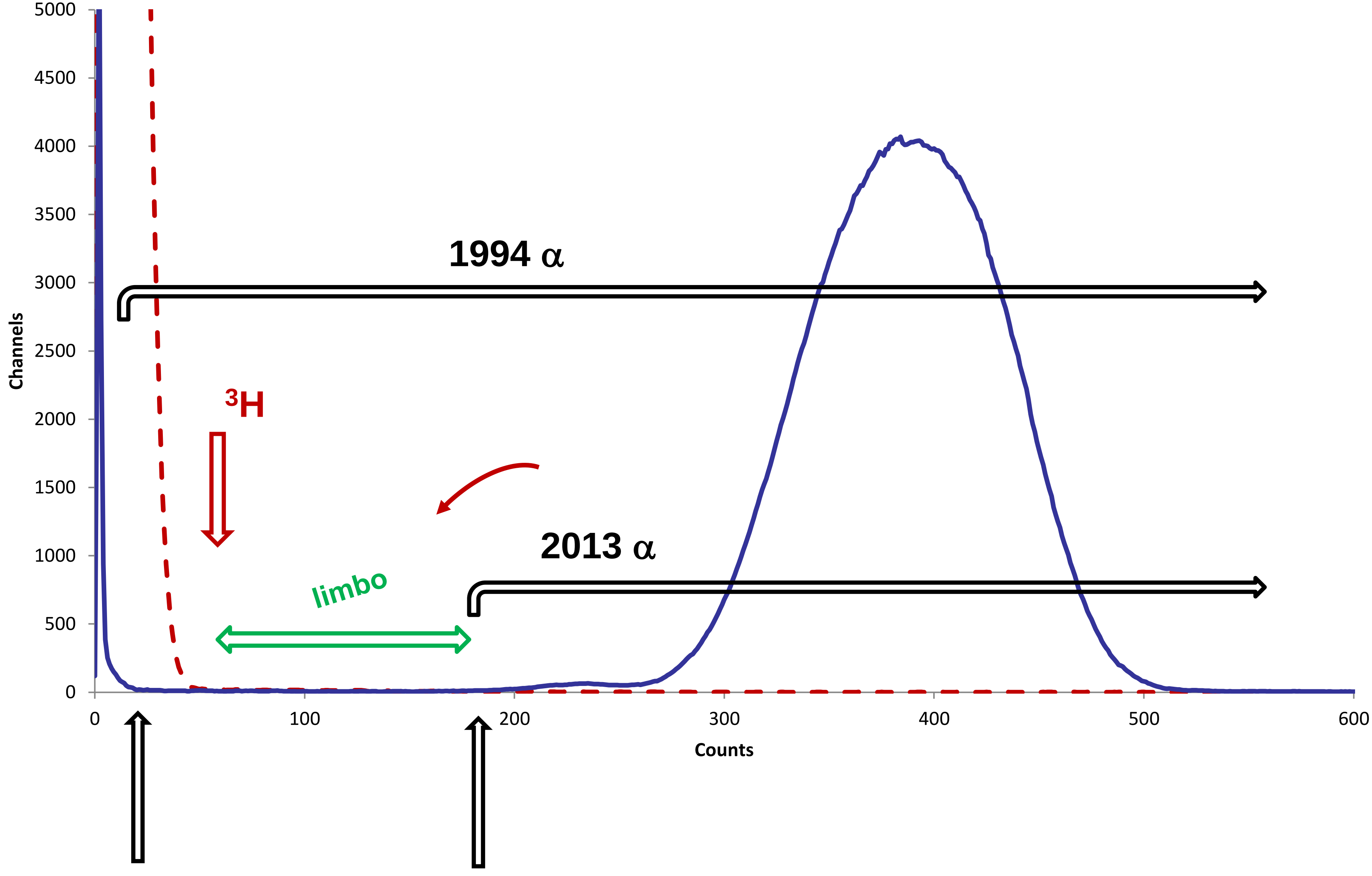
main important difference :

1994 = No analytical help

2013 = Fitzgerald helped

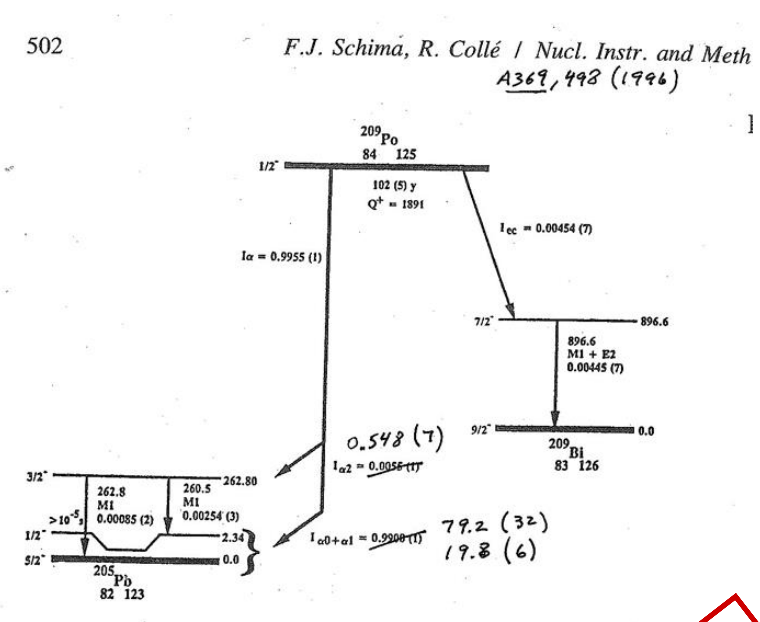


counts per channel
5000
4000
3000
2000
1000
0
³H
alpha region
"limbo"
monitor
0
100
200
300
400
500
600
≈ 18.5 keV
channel number (energy)



5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
Channels
0
100
200
300
400
500
600
Counts
1994 α
2013 α
^{3}H
limbo

all known transitions in ^{209}Po decay



α	4622	0.548	
	4883	79.2	
	4885	19.8	
eA	5.4 - 16.3	0.224	L (Bi)
	57.5 - 63.4		**KLL**
	70.0 - 77.1	**0.012**	**KLX**
	82.5 - 90.5		**KXY**
	5.3 - 15.8	0.104	L (Pb)
	56.0 - 61.7		KLL
	68.2 - 75.0	0.006	KLX
	80.3 - 88.0		KXY
ec	172.5	0.128	γ 2-1 K (Pb)
	174.8	0.043	γ 2-0 K (Pb)
X	9.4 - 15.7	0.141	L (Bi)
	74.8 - 90.4	**0.317**	**K (Bi)**
	9.2 - 15.2	0.063	L (Pb)
	72.8 - 87.9	0.164	K (Pb)
γ	260.5	0.254	γ 2-1 (Pb)
	262.8	0.085	γ 2-0 (Pb)
	896.28	**0.445**	**γ 1-0 (Bi)**

most excluded in LS

because of

coincident transitions

or use of ^{3}H cut-off

Need to consider corrections for these in bold

	f			
Collé (1994)	0.9988	(note1)		
Collé (2013)	0.9995	(note2)	"lucky"	"same"
Fitzgerald (2013)	0.99934	(note3)	★	

Minor Differences Part 2

Note1:

*based on analysis in Appendix of Collé, et al., J.Res. NIST **100,** 1 (1995).*

This assumed L-capture detection with $\varepsilon_L = 0.5$, whereas Fitzgerald (2013) calculation assumed $\varepsilon_L = 0$ since used ^3H spectral cut-off & $E_L < 18$ keV

Note2:

This result was characterized as being "lucky" and the "same" as Fitzgerald (2013).

It was based on calculating the probability of photon detection and that their location in the LS spectrum was proportioned by the size of windows.

Note3:

If Fitzgerald calculation assumes $\varepsilon_L = 0.5$ as in 1994 assumption, then 1994 f value is recovered.

12 DEC '13 RPF

The definative ^{209}Po EC branch correction

item	GEANT efficiency ε ($E_{LS} > 18$ keV)
X_K	3%
X_{K+L}	11%
A_K	100%
γ_{896}	7%
$C.\varepsilon._{896}$	100%.
else	ϕ

DDEP

$$\begin{bmatrix} B_{EC} = 0.00454 \\ P_K = 0.70796 \\ \omega_K = 0.964 \\ n_{KL} = 0.809 \\ \alpha_{K\,896} = 0.017 \end{bmatrix}$$

- effective **K efficiency**, including X-rays & Auger

$$\varepsilon_K = \omega_K n_{KL} \overset{(11\%)}{\varepsilon_{X_{K+L}}} + \omega_K(1-n_{KL}) \overset{(3\%)}{\varepsilon_{X_K}} + (1-\omega_K) \overset{(100\%)}{\varepsilon_{A_K}}$$

$$\varepsilon_K = 0.0858 + 0.0055 + 0.0370$$

$$\varepsilon_K = 0.1284$$

- effective **γ-transition efficiency**, including conversion

$$\varepsilon_\gamma' = \frac{1}{1+\alpha}\varepsilon_\gamma + \frac{\alpha}{1+\alpha}\varepsilon_{CE}$$

$$\varepsilon_\gamma' = 0.98(0.07) + 0.02(1)$$

$$\varepsilon_\gamma' = 0.089$$

∴ **efficiency per K capture**, including γ

$$\varepsilon_K' = \varepsilon_K + (1-\varepsilon_K)\varepsilon_\gamma'$$

$$\varepsilon_K' = 0.1284 + (1-0.1284)\,0.089$$

$$\varepsilon_K' = 0.1284 + 0.0776$$

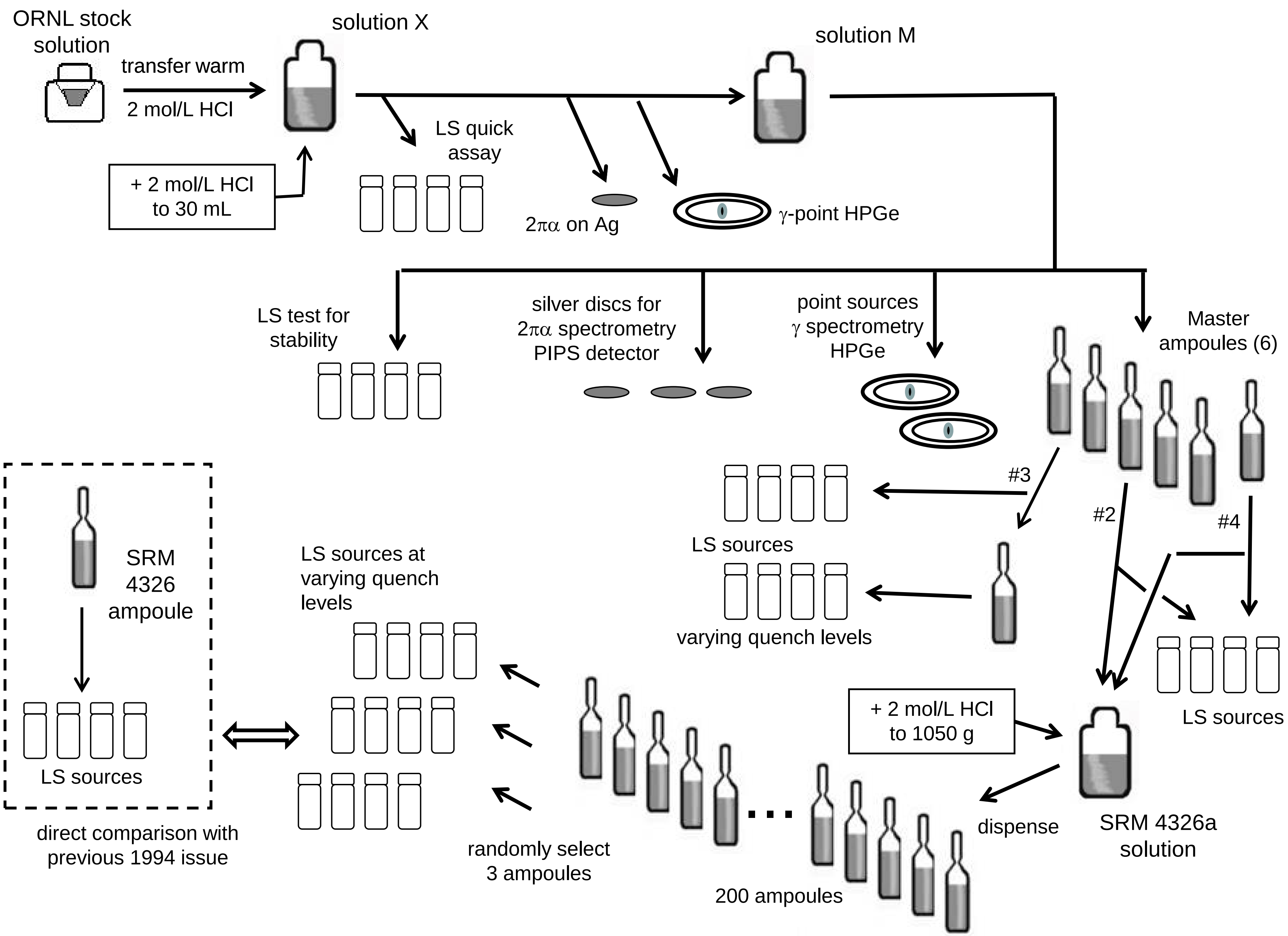
$$\varepsilon_K' = 0.2060 \longleftarrow \text{efficiency per K capture}$$

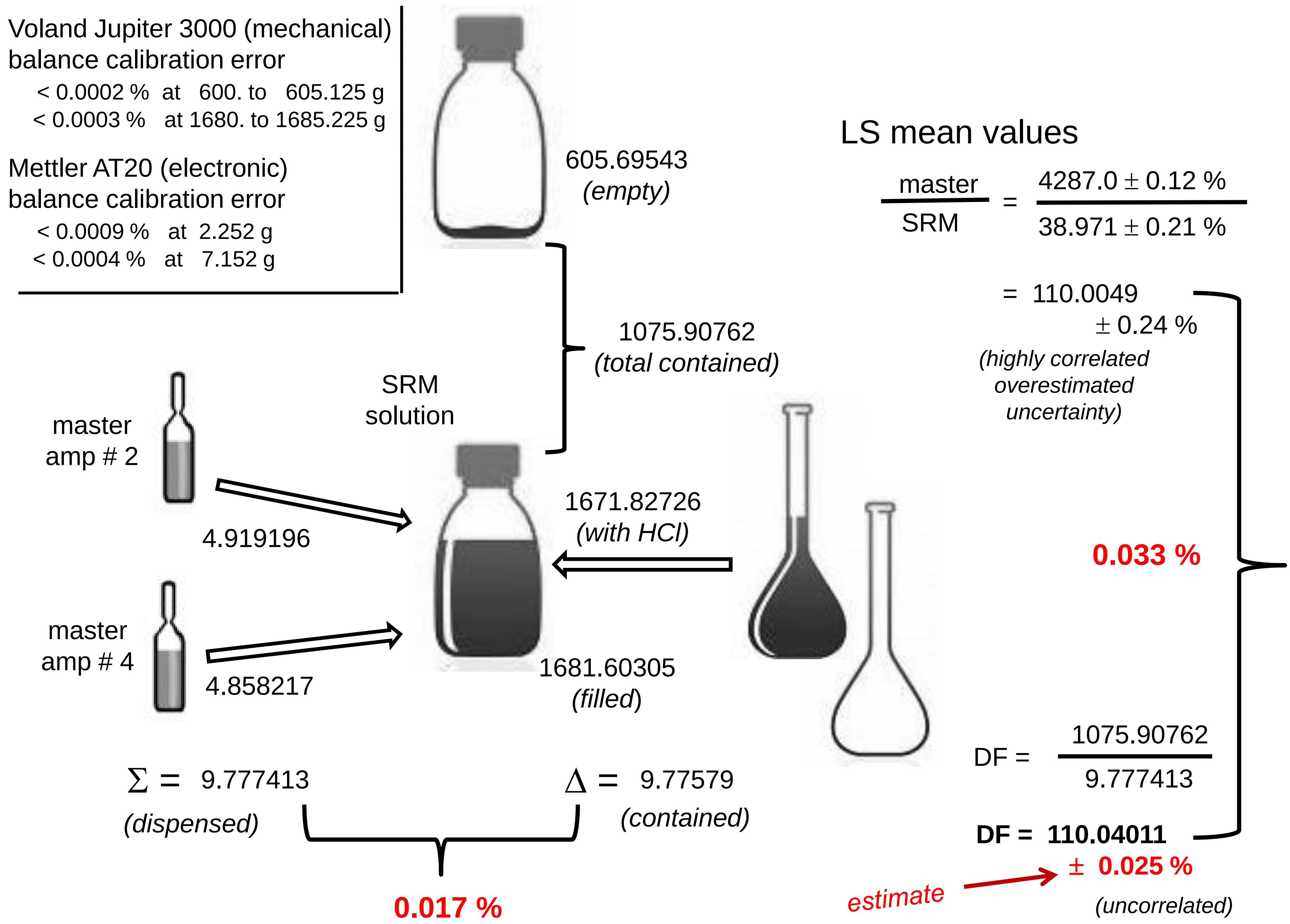
- **Absolute efficiency per ^{209}Po decay**

$$\varepsilon = B_{EC} \cdot P_K \cdot \varepsilon_K' = 0.00454(0.70796)(0.2060), \quad \varepsilon = 0.00066$$

$$\boxed{F = 0.99934}$$

$F = \frac{1}{1+\varepsilon}$


ORNL stock solution
transfer warm
2 mol/L HCl
solution X
+ 2 mol/L HCl to 30 mL
LS quick assay
2πα on Ag
γ-point HPGe
solution M
LS test for stability
silver discs for 2πα spectrometry PIPS detector
point sources γ spectrometry HPGe
Master ampoules (6)
#3
#2
#4
LS sources
varying quench levels
LS sources
SRM 4326 ampoule
LS sources
direct comparison with previous 1994 issue
LS sources at varying quench levels
randomly select 3 ampoules
200 ampoules
+ 2 mol/L HCl to 1050 g
dispense
SRM 4326a solution



Voland Jupiter 3000 (mechanical)
balance calibration error
< 0.0002 % at 600. to 605.125 g
< 0.0003 % at 1680. to 1685.225 g
Mettler AT20 (electronic)
balance calibration error
< 0.0009 % at 2.252 g
< 0.0004 % at 7.152 g
605.69543
(empty)
1075.90762
(total contained)
SRM
solution
master
amp # 2
4.919196
master
amp # 4
4.858217
1671.82726
(with HCl)
1681.60305
(filled)
Σ = 9.777413
(dispensed)
Δ = 9.77579
(contained)
0.017 %
LS mean values
master / SRM = 4287.0 ± 0.12 % / 38.971 ± 0.21 %
= 110.0049
± 0.24 %
(highly correlated
overestimated
uncertainty)
0.033 %
DF = 1075.90762 / 9.777413
DF = 110.04011
± 0.025 %
estimate
(uncorrelated)

2013 ^{209}Po Measurements for New SRM

- aliquots from **13** different solutions
  - **3** LS sources from each solution
  - (excepting **6** sources from one solution for quench test)

- **42** LS sources in all

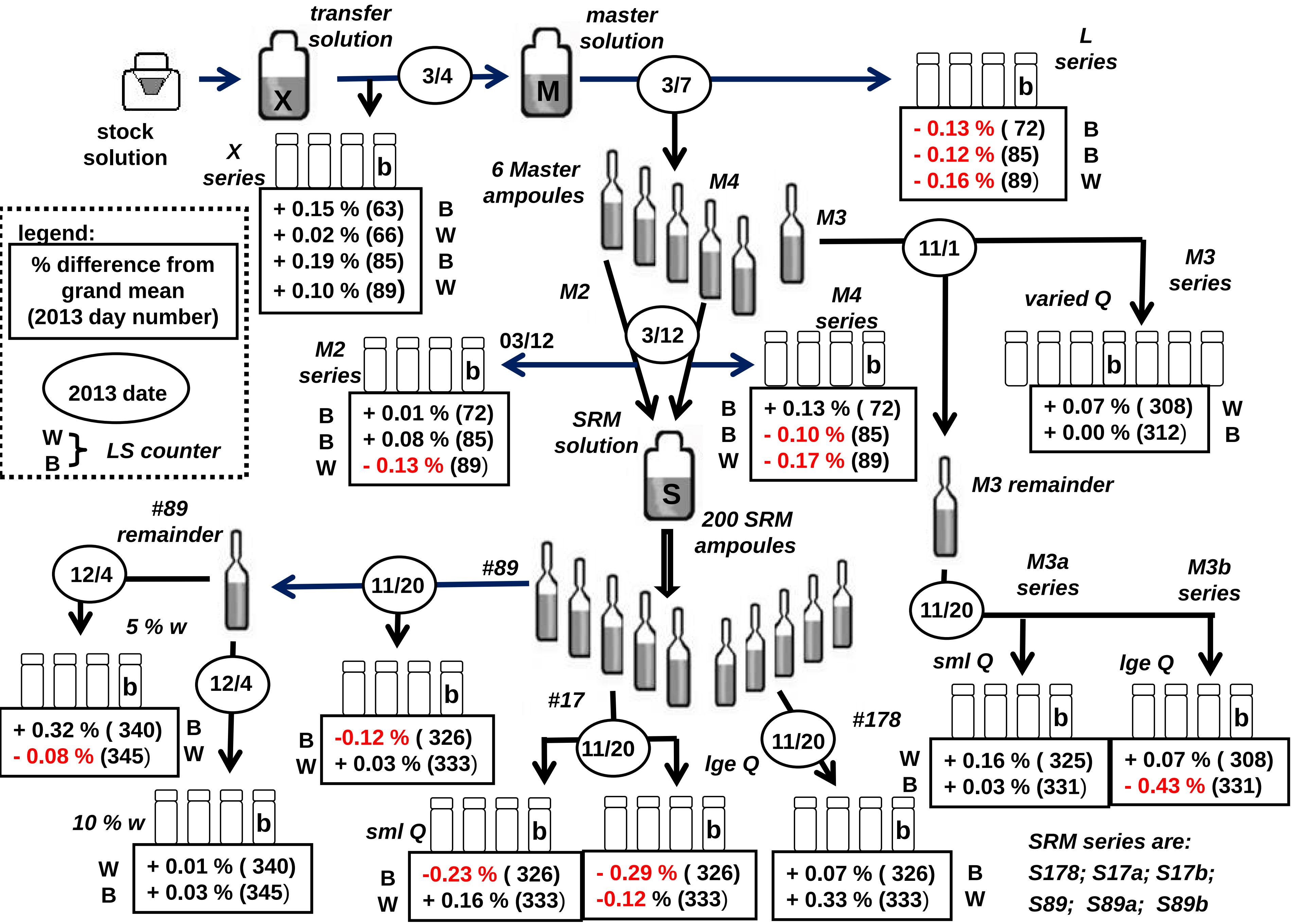
- each source measured n = **3** cycles in **2** counters
  on either **1** or **2** measurement occasions

- **297** measurements in all

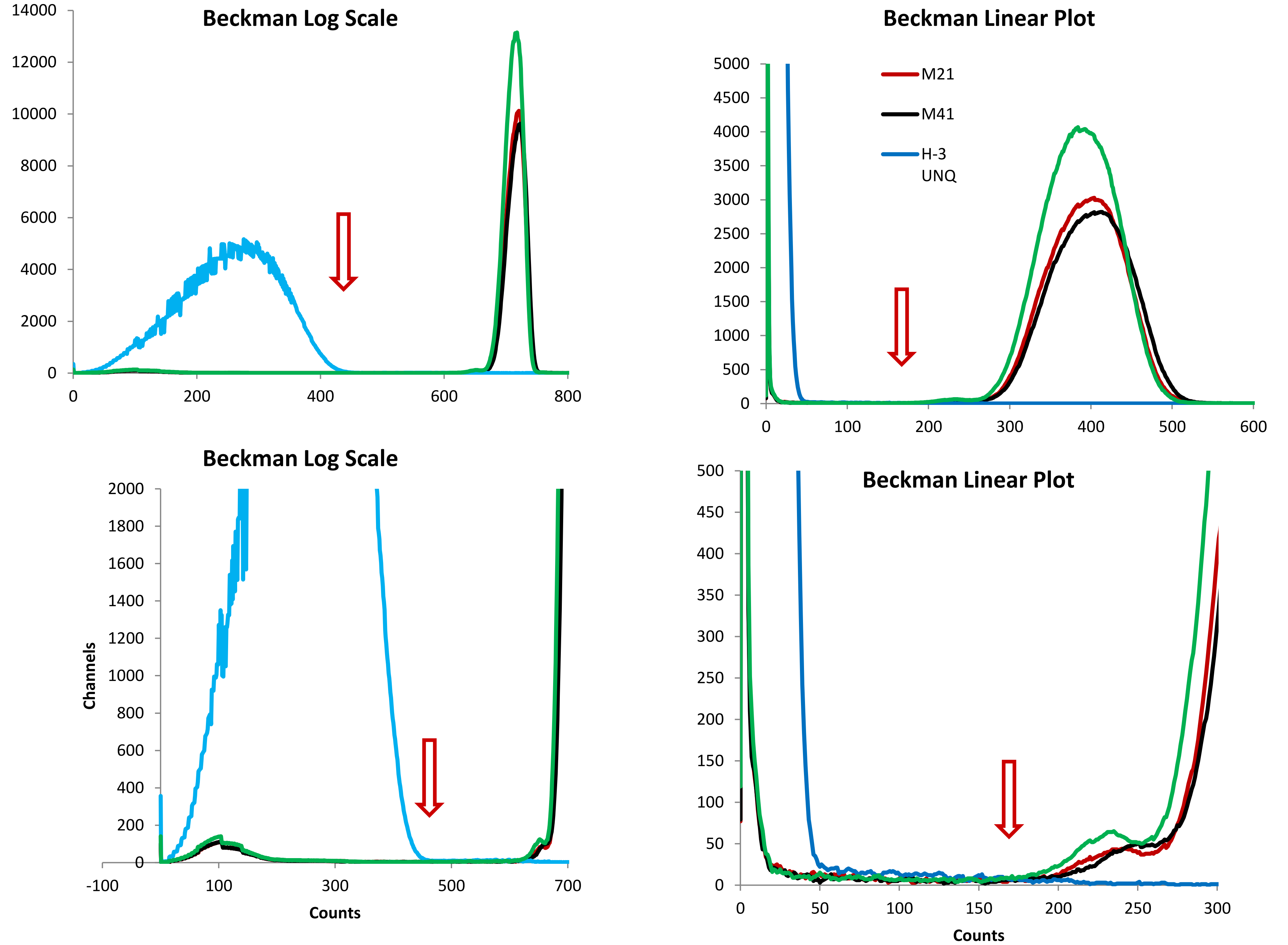
- measurements performed over period of **276** days
  - **25** days in March 2013
  - **31** days in 31 days in November – December 2013

- **31** mean values

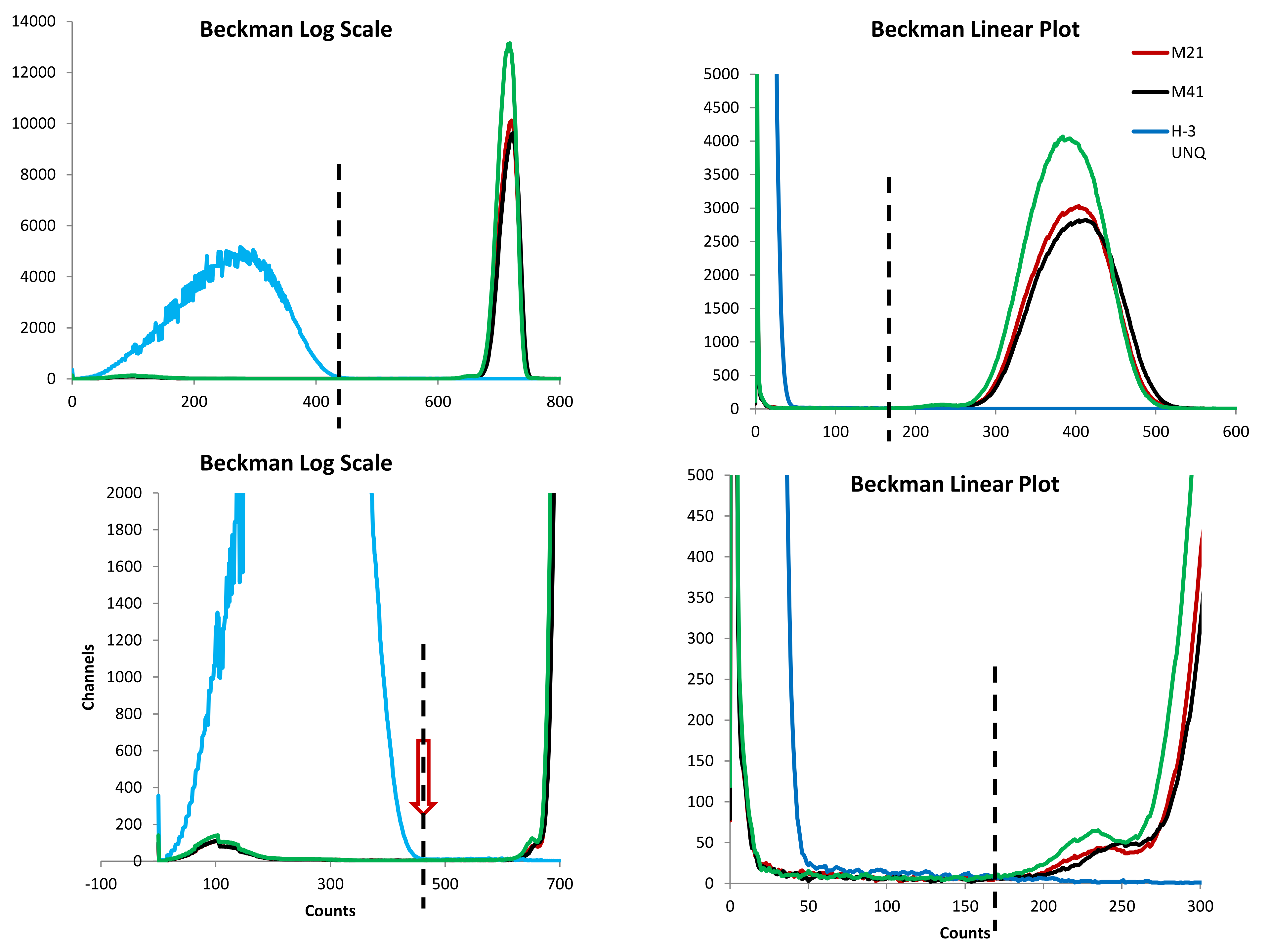
- Typical precision on means = **0.24 %**
  - combined standard deviation (within & between)
    for n = **3** cycles with n = **3** sources)



stock solution
transfer solution
X
3/4
master solution
M
3/7
L series
- 0.13 % (72) B
- 0.12 % (85) B
- 0.16 % (89) W
X series
+ 0.15 % (63) B
+ 0.02 % (66) W
+ 0.19 % (85) B
+ 0.10 % (89) W
6 Master ampoules
M4
M3
11/1
M3 series
varied Q
+ 0.07 % (308) W
+ 0.00 % (312) B
legend:
% difference from grand mean (2013 day number)
2013 date
W
B
LS counter
M2
3/12
M4 series
B + 0.13 % (72)
B - 0.10 % (85)
W - 0.17 % (89)
M2 series
03/12
B + 0.01 % (72)
B + 0.08 % (85)
W - 0.13 % (89)
SRM solution
S
M3 remainder
200 SRM ampoules
#89 remainder
12/4
11/20
#89
5 % w
M3a series
M3b series
11/20
sml Q
lge Q
+ 0.32 % (340) B
- 0.08 % (345) W
12/4
B -0.12 % (326)
W + 0.03 % (333)
#17
11/20
11/20
#178
lge Q
W + 0.16 % (325)
B + 0.03 % (331)
+ 0.07 % (308)
- 0.43 % (331)
10 % w
W + 0.01 % (340)
B + 0.03 % (345)
sml Q
B -0.23 % (326)
W + 0.16 % (333)
- 0.29 % (326)
-0.12 % (333)
+ 0.07 % (326) B
+ 0.33 % (333) W
SRM series are:
S178; S17a; S17b;
S89; S89a; S89b
b

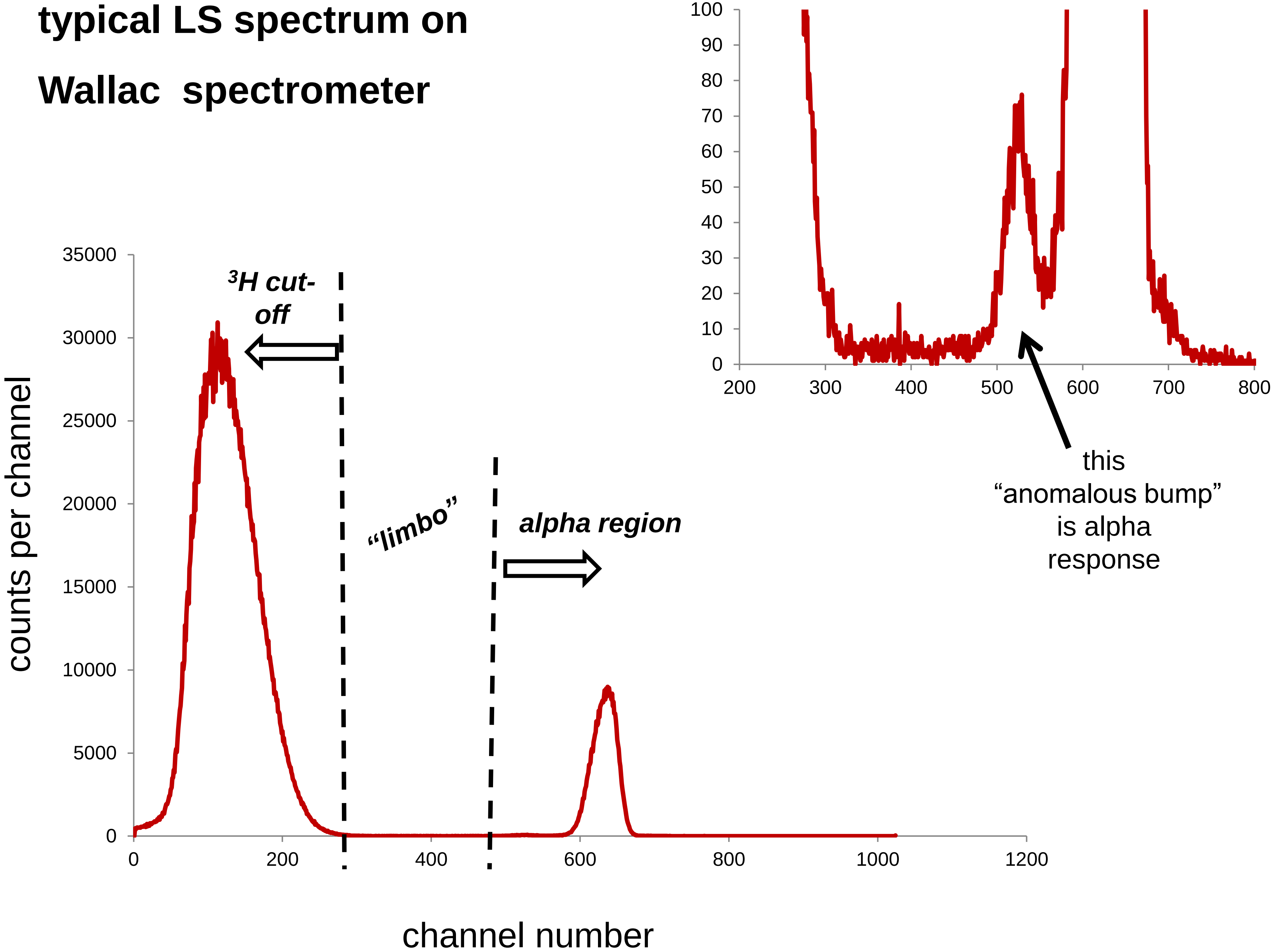


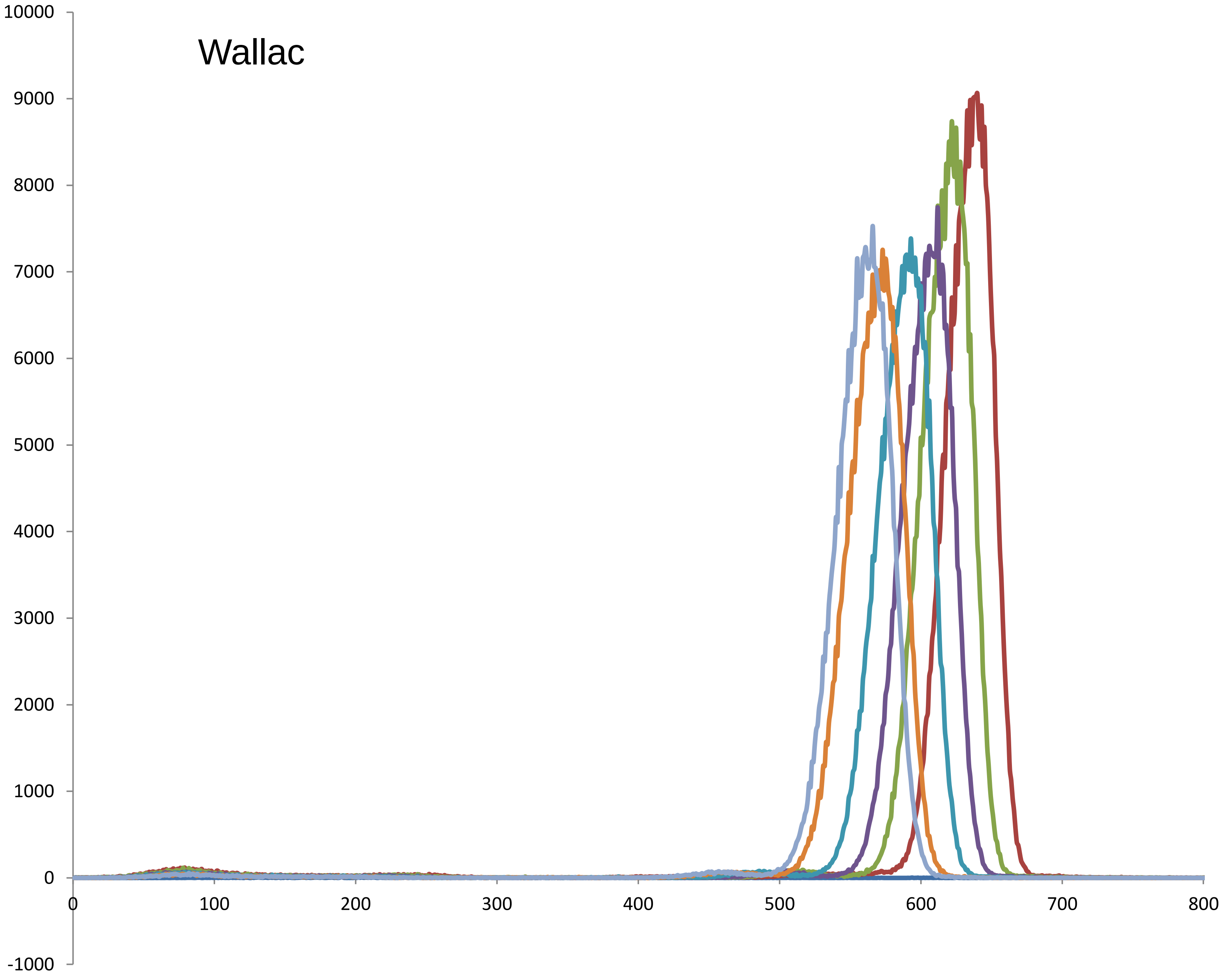
Beckman Log Scale
14000
12000
10000
8000
6000
4000
2000
0
0
200
400
600
800
Beckman Linear Plot
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
0
100
200
300
400
500
600
M21
M41
H-3 UNQ
Beckman Log Scale
2000
1800
1600
1400
1200
1000
800
600
400
200
0
-100
100
300
500
700
Channels
Counts
Beckman Linear Plot
500
450
400
350
300
250
200
150
100
50
0
0
50
100
150
200
250
300
Counts

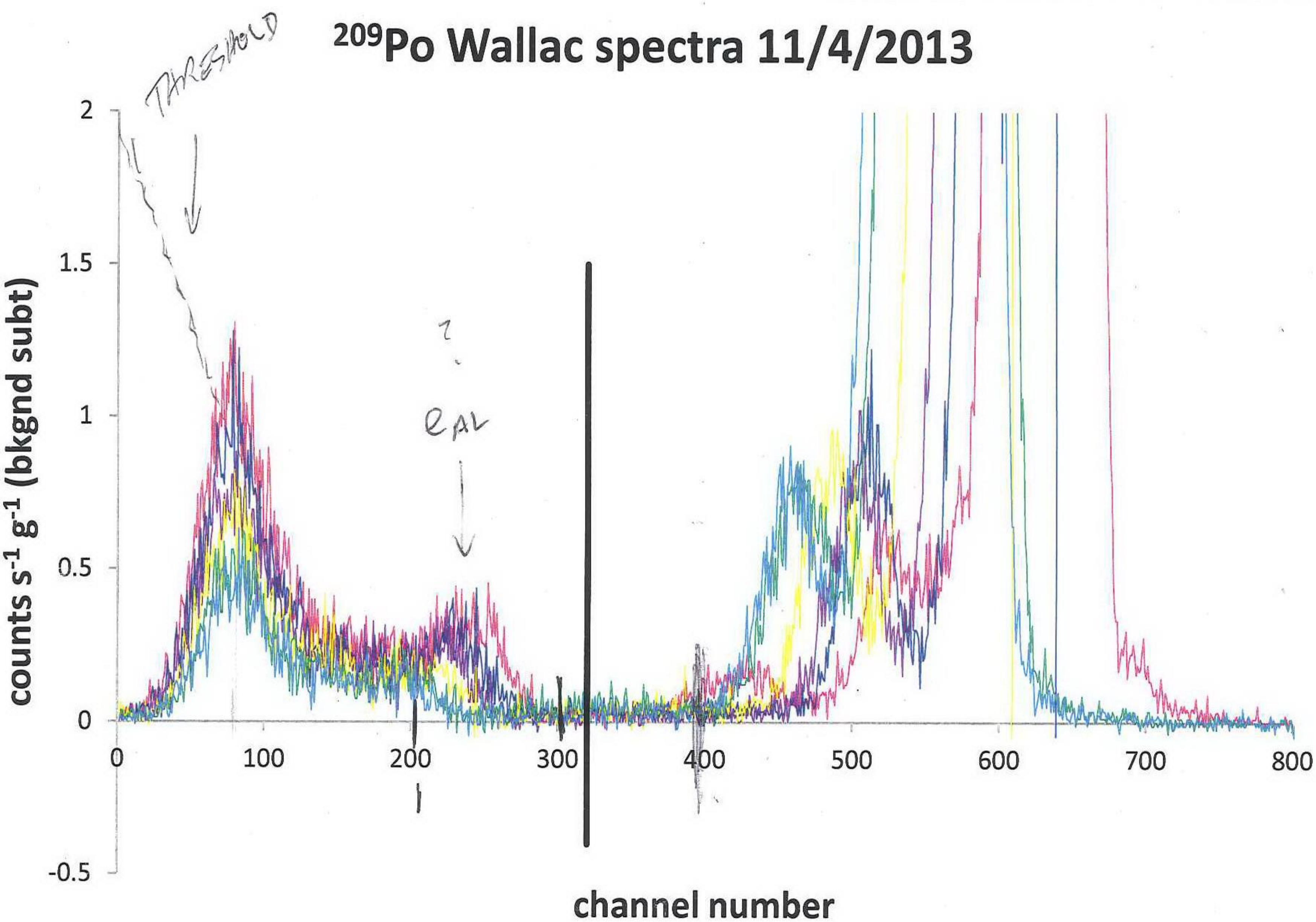


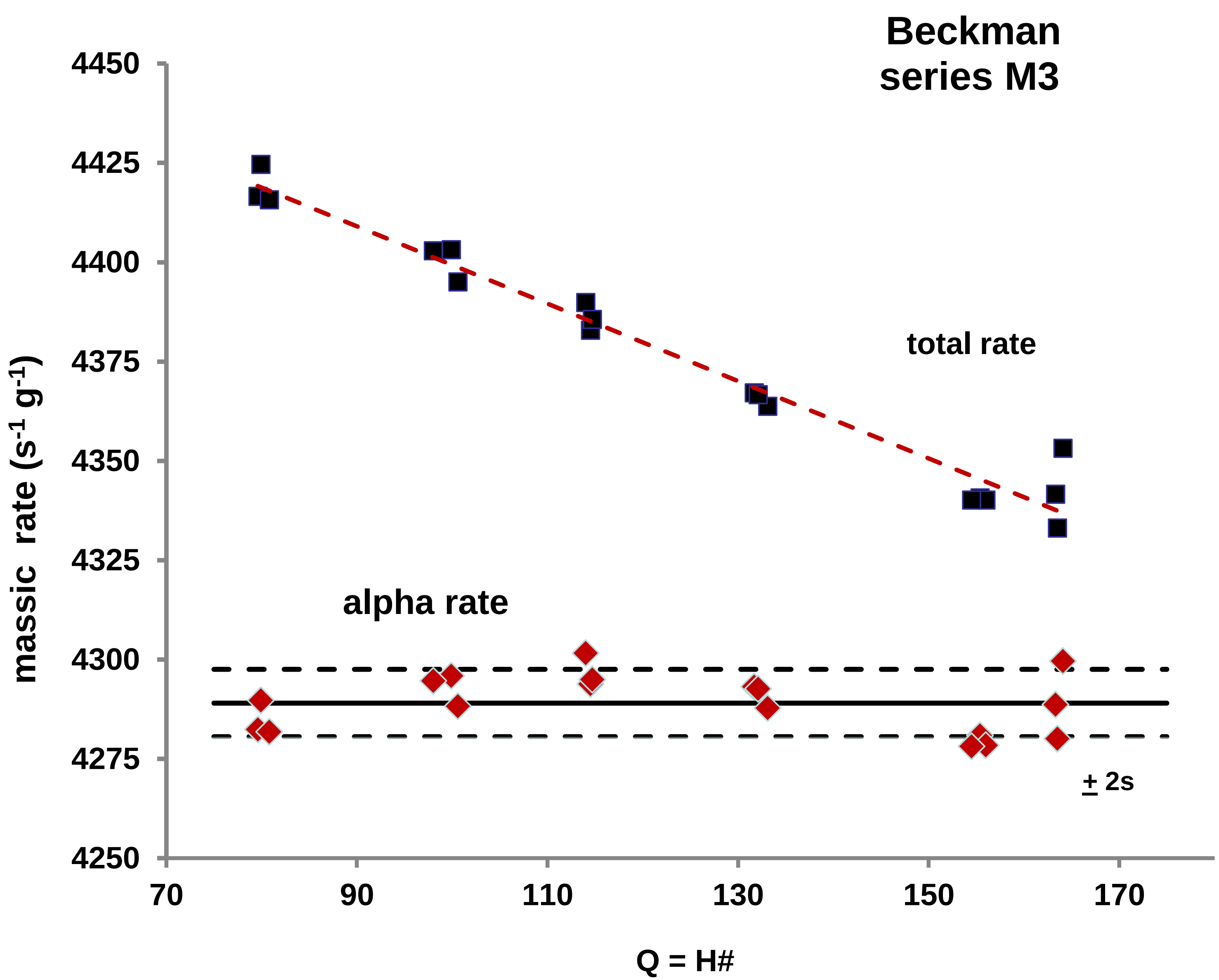
Beckman Log Scale
Beckman Linear Plot
M21
M41
H-3 UNQ
Beckman Log Scale
Channels
Counts
Beckman Linear Plot
Counts

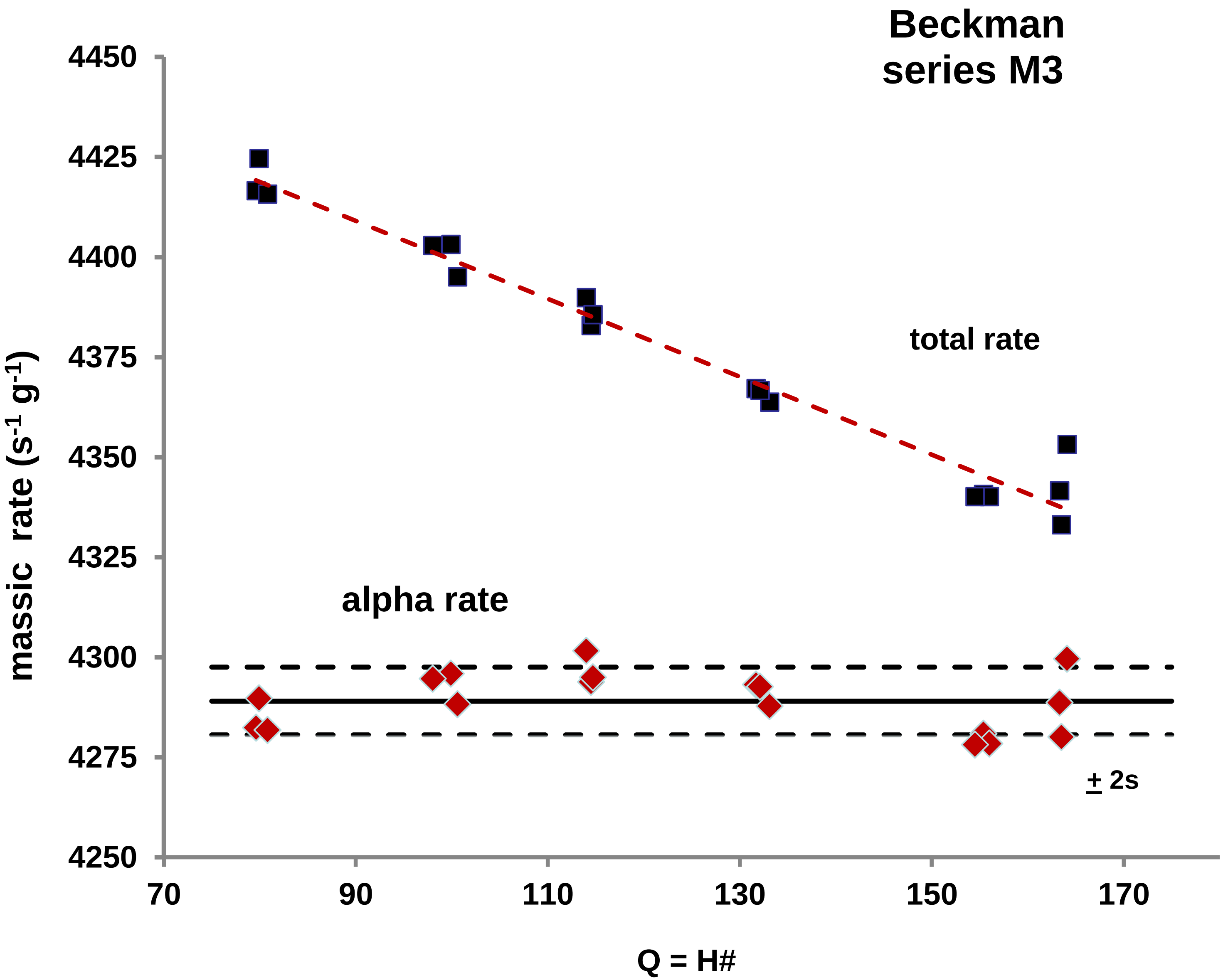
typical LS spectrum on Wallac spectrometer

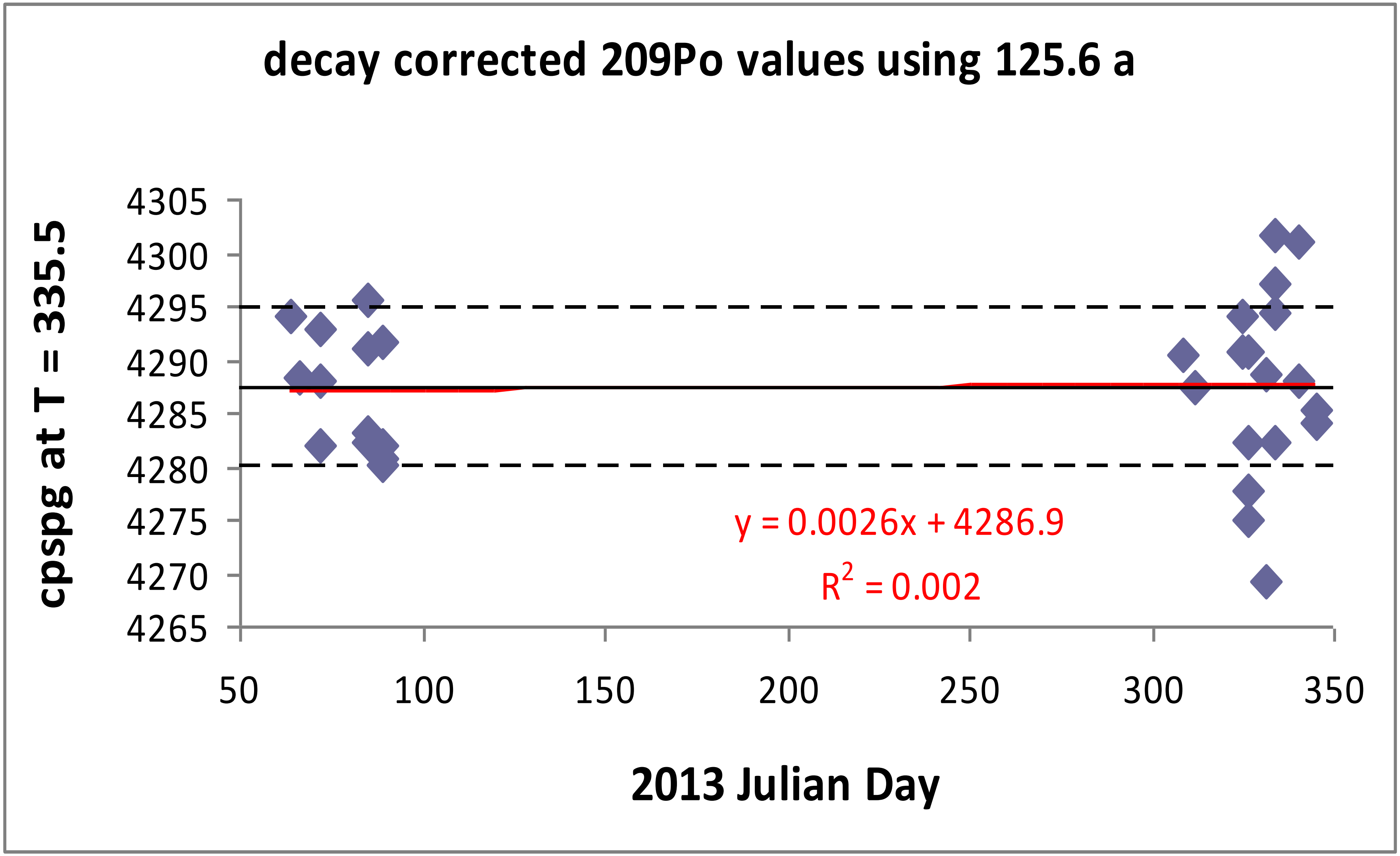



Wallac
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
-1000
0
100
200
300
400
500
600
700
800

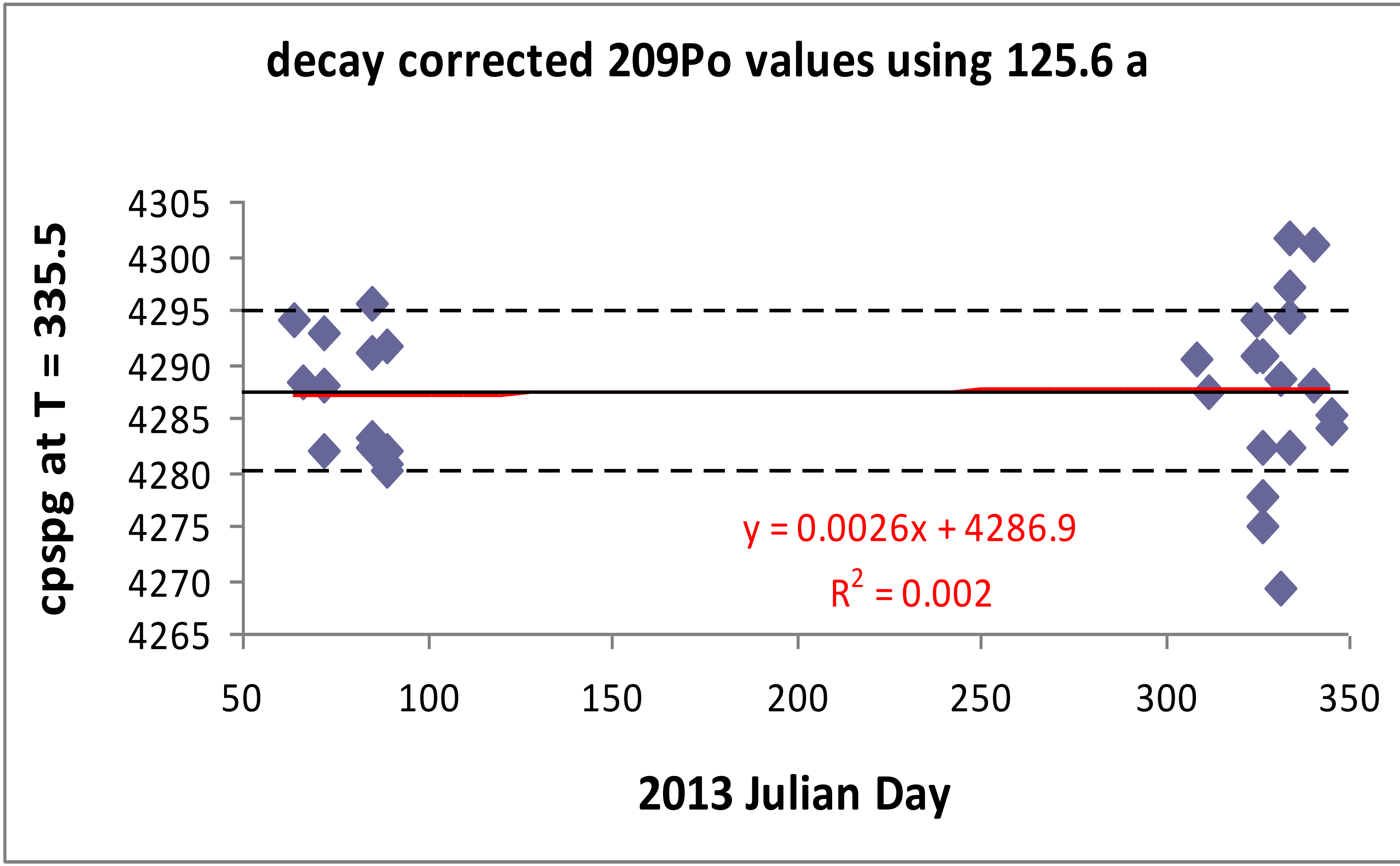

^{209}Po Wallac spectra 11/4/2013
THRESHOLD
? eAL
counts s^{-1} g^{-1} (bkgnd subt)
channel number
2
1.5
1
0.5
0
-0.5
0
100
200
300
400
500
600
700
800


Beckman
series M3
total rate
alpha rate
± 2s
massic rate (s-1 g-1)
Q = H#
4450
4425
4400
4375
4350
4325
4300
4275
4250
70
90
110
130
150
170


Beckman
series M3
total rate
alpha rate
± 2s
massic rate (s-1 g-1)
4450
4425
4400
4375
4350
4325
4300
4275
4250
70
90
110
130
150
170
Q = H#

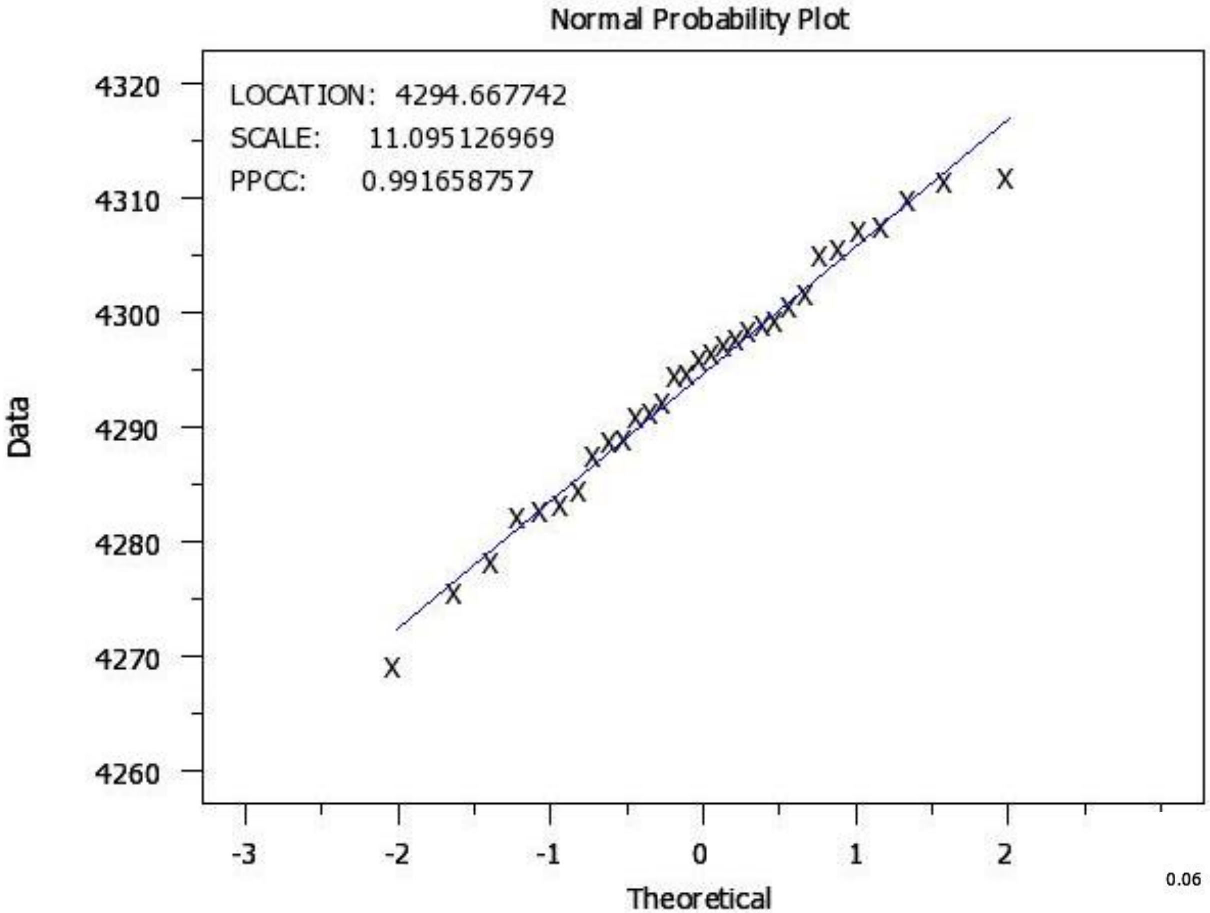


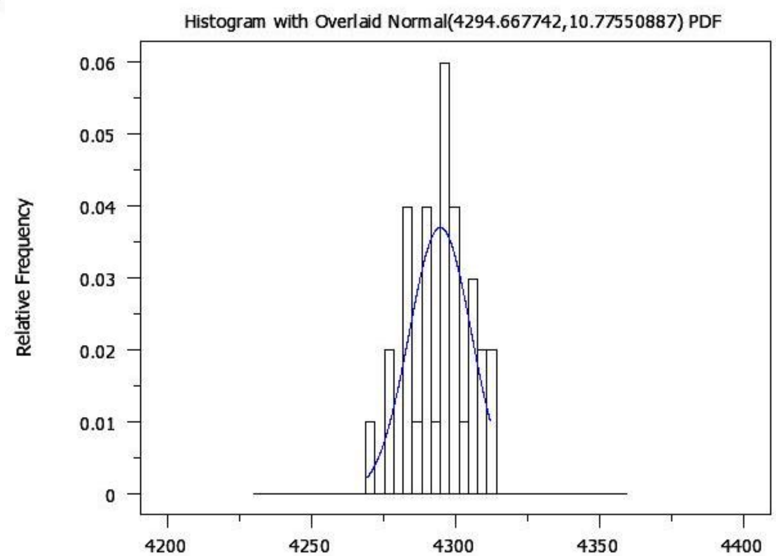
31 mean values

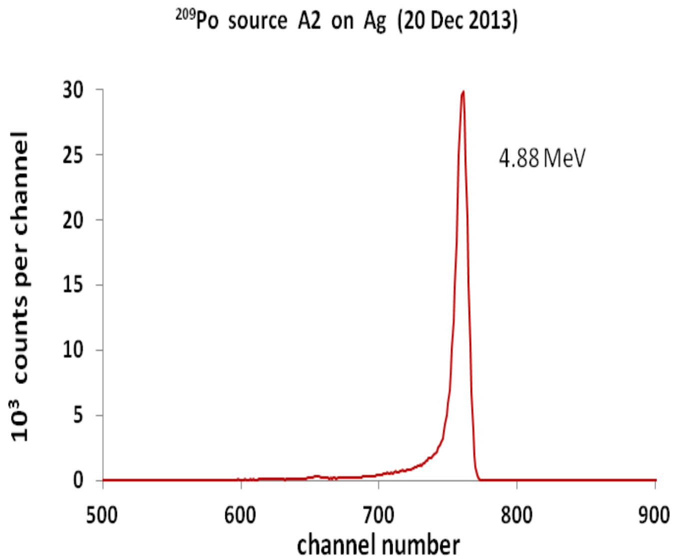


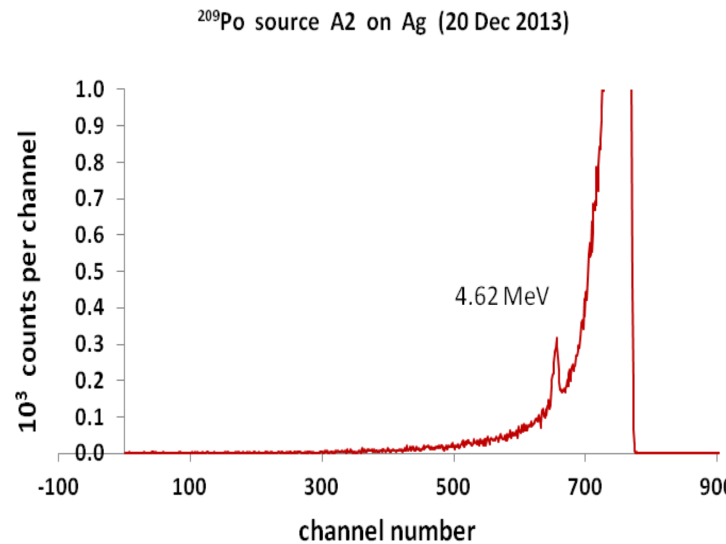
decay corrected 209Po values using 125.6 a
cpspg at T = 335.5
4305
4300
4295
4290
4285
4280
4275
4270
4265
50
100
150
200
250
300
350
2013 Julian Day
y = 0.0026x + 4286.9
R2 = 0.002

n = 31 means


Normal Probability Plot
LOCATION: 4294.667742
SCALE: 11.095126969
PPCC: 0.991658757
Data
Theoretical
4320
4310
4300
4290
4280
4270
4260
-3
-2
-1
0
1
2


Histogram with Overlaid Normal(4294.667742,10.77550887) PDF
Relative Frequency
0.06
0.05
0.04
0.03
0.02
0.01
0
4200
4250
4300
4350
4400





NO alpha impurities

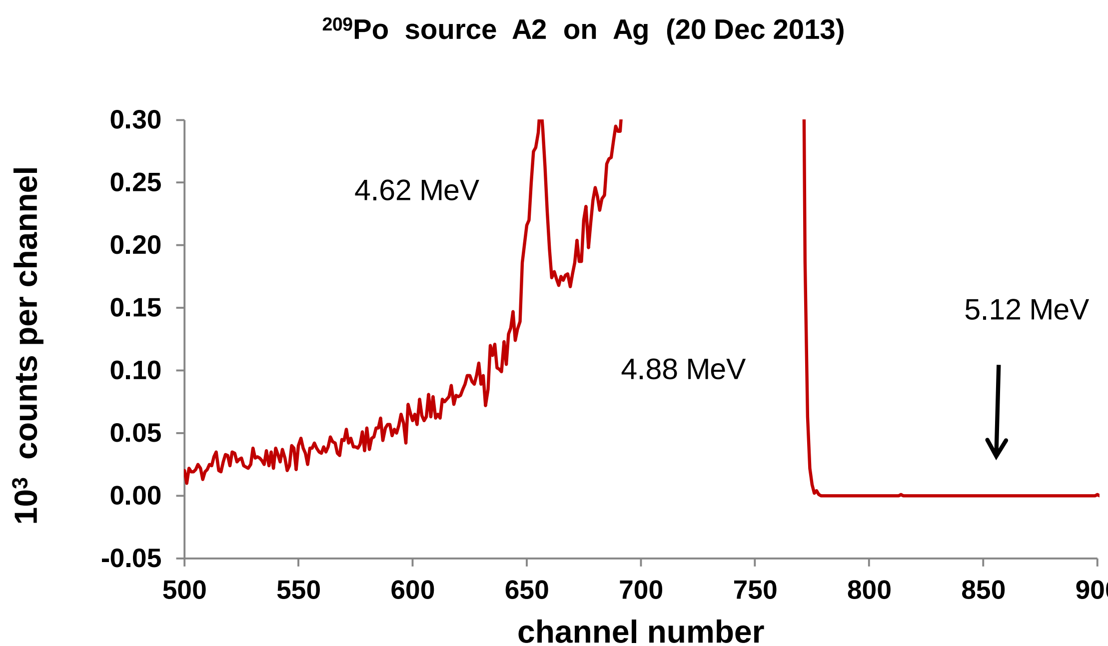
activity ratio

^{208}Po/^{209}Po $< 10^{-7}$

limit / ^{209}Po

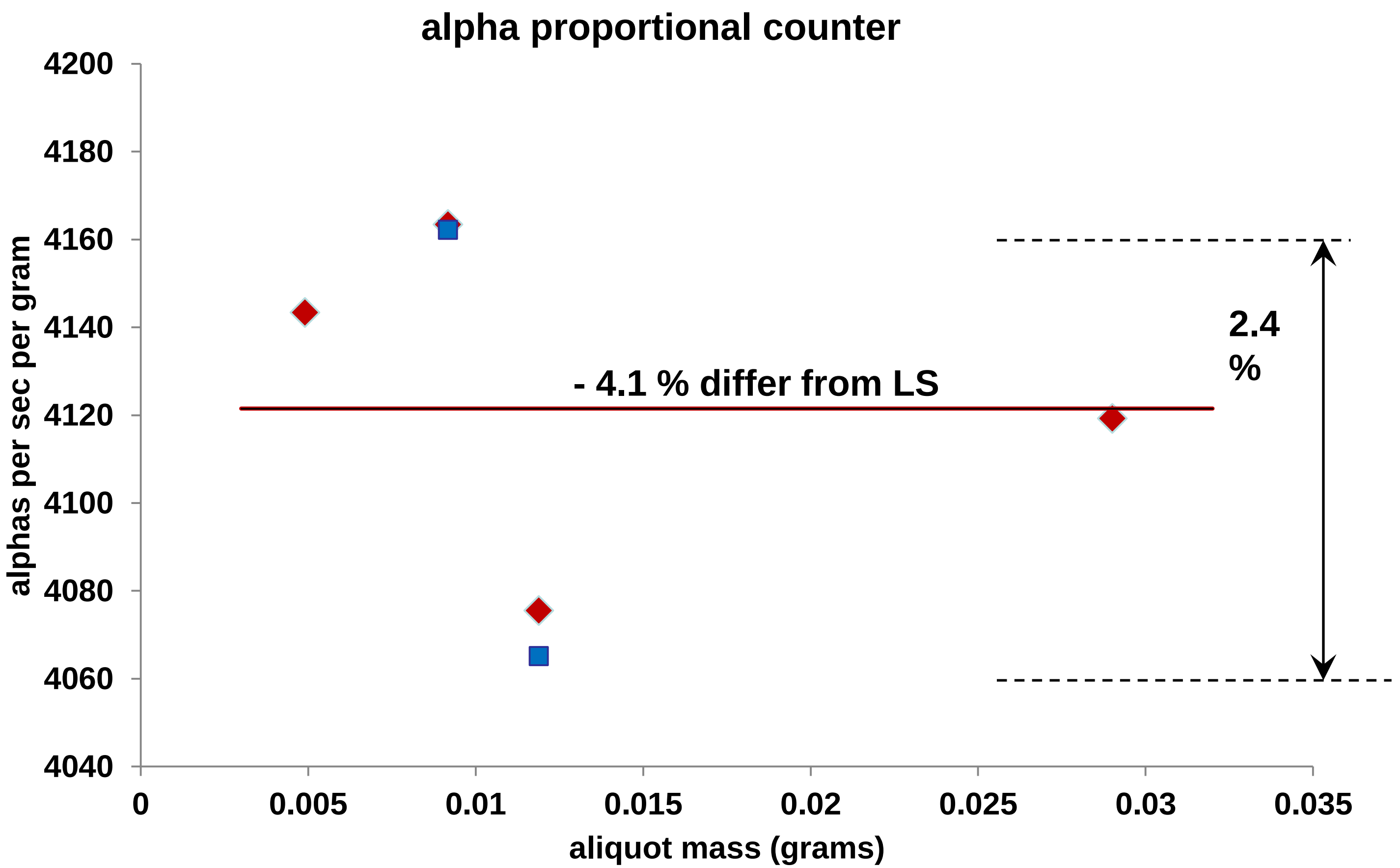
$< 10^{-3}$ for $E_\alpha \leq 4.5$ MeV

$< 2(10^{-5})$ for $E_\alpha \geq 5.0$ MeV

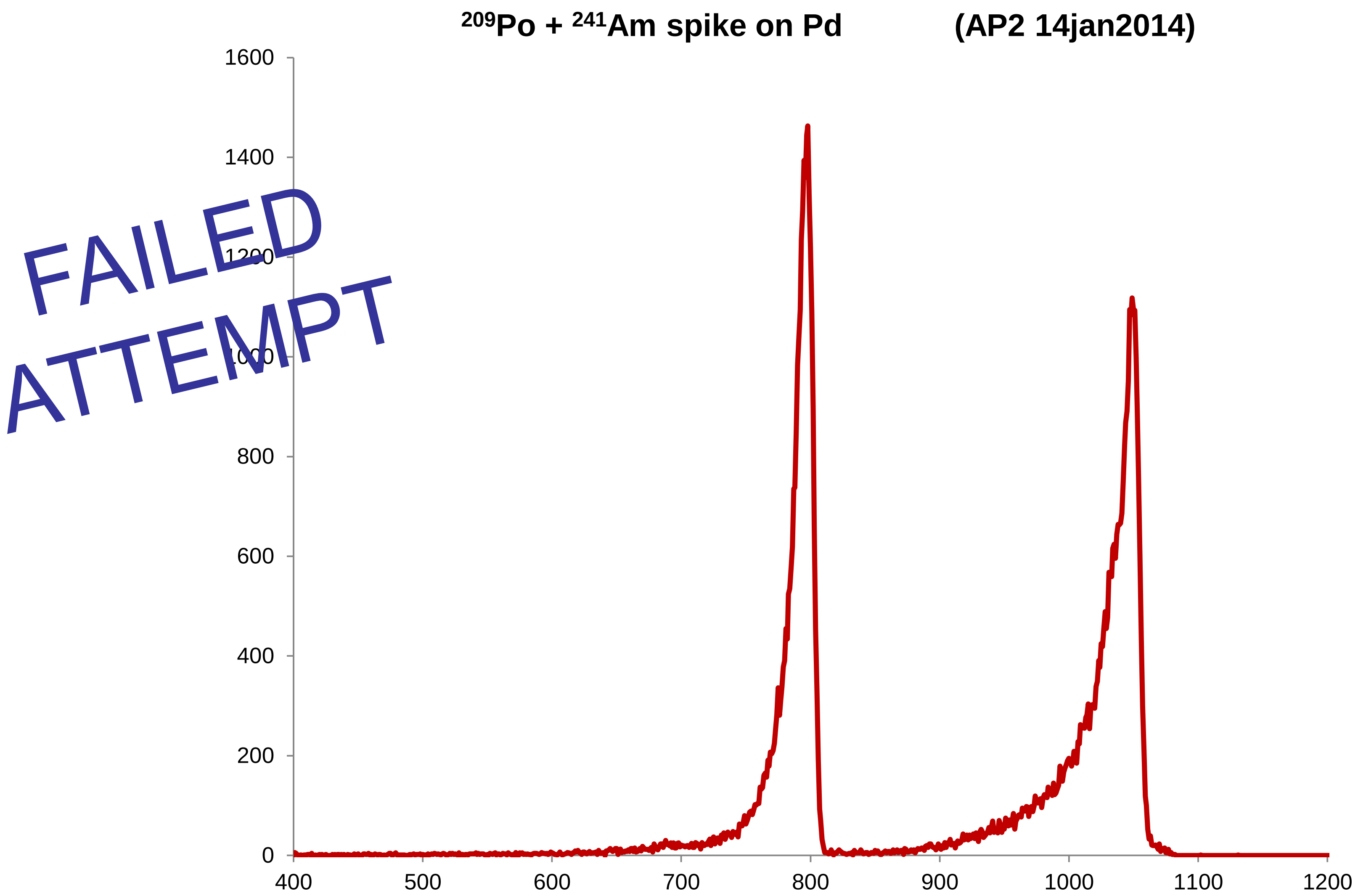


Discrepant result
(Po loss probable)

^{209}Po on Ag sources



results
L. King,
April-May 2013



^{209}Po + ^{241}Am spike on Pd (AP2 14jan2014)
FAILED ATTEMPT
1600
1400
1200
1000
800
600
400
200
0
400
500
600
700
800
900
1000
1100
1200

Photonic emission impurities

L. Pibida, 14 Jan 2014

photons from ^{209}Po decay

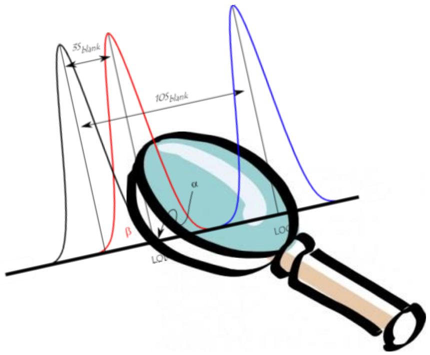
Pb & Bi x-rays	< 88 KeV
260.50 keV (Pb)	– 0.254 %
262.80 keV (Pb)	– 0.085 %
896.28 keV (Bi)	– 0.445 %

No other photons observed

Detection limits in the energy region
30 keV ≤ E ≤ 2000 keV
ranged from 0.001 to 0.002 s^{-1}▪g^{-1}
(excepting in the region 880 keV ≤ E ≤ 910 keV
where limit was 0.003 s^{-1}▪g^{-1})

Therefore,

ratio of detection limit to 209Po activity
is **0.003 to 0.008 %**
in all energy regions 30 keV ≤ E ≤ 2000 keV



SRM 4326a

“*certified*” massic alpha emission rate

$$(38.94 \pm 0.18)\ \mathrm{s^{-1} \cdot g^{-1}}$$

1 December 2013

SRM uncertainty assessment

	component	method	percent relative uncertainty
1	LS measurement precision; standard deviation for *n* = 31 mean determinations, includes both within- and between-variability for (i) *n* = 3 measurements on each LS counting source, (ii) use of *n* = 2 different LS counters on either one or two separate measurement occasions; (iii) *n* = 3 to 6 sources for each solution / LS cocktail composition; (iv) *n* = 11 separate sets of sources / solutions of varying compositions	A	0.17
2	LS spectral analysis method	B *	0.15
3	LS alpha detection efficiency, includes wall effect	B *	0.01
4	LS response correction for electron capture branch decay	B	0.015
5	gravimetric dilution factor	B	0.025
6	counting source aliquot mass determinations, includes mass measurement precision	B *	0.02
7	^{209}Po decay corrections for half-life uncertainty of 2.6 %	B	0.011
8	potential alpha or photon emitting impurities	B	0.005
	combined uncertainty		**0.23**
	***k* = 2 expanded uncertainty**		**0.46**

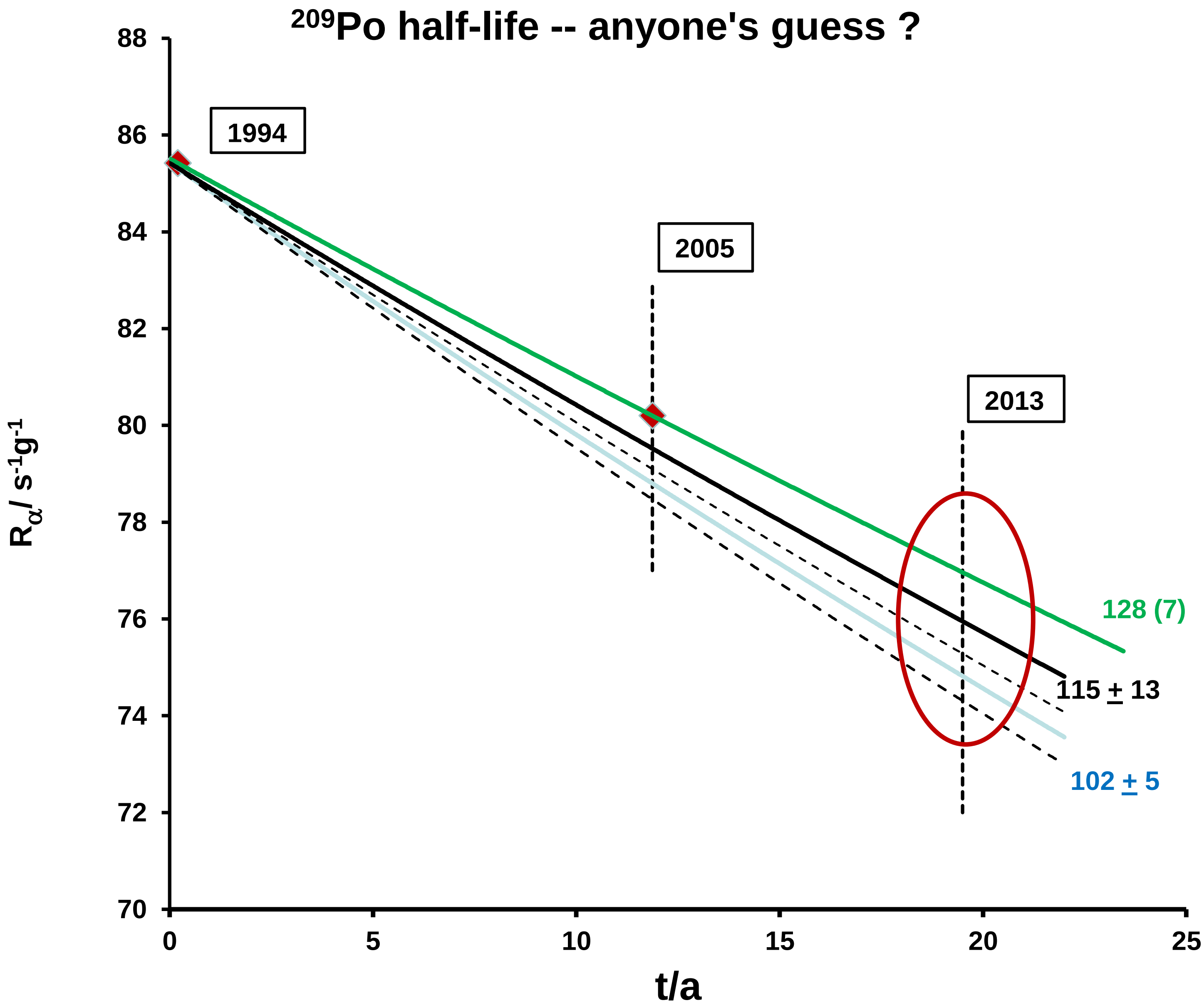
* uncertainty is partially embodied within component 1

5

2013

half-life

as of early 2013



^{209}Po measurement conditions all years

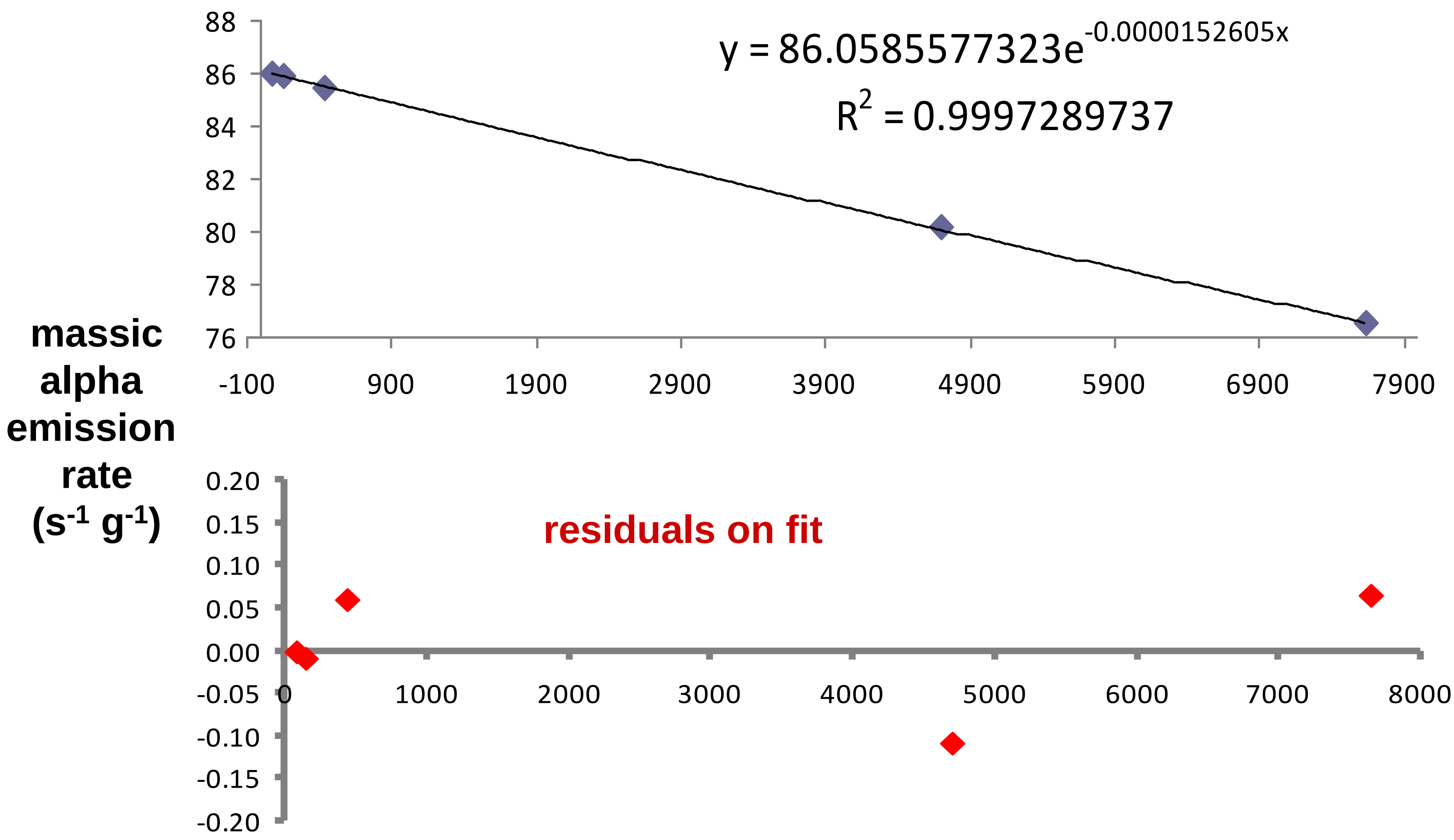
	1993 (a)	1993 (b)	1994	2005	2013
Number of different cocktail compositions used	4 quench varied	1 quench varied	2 quench varied	2 quench varied	2 2 quench conditions for each
Number sources per composition	3	6	4	5	3
Scintillator(s) used (1)	RS	RS	RS, UGAB	RS	UGAB
LS counters used (2)	B & P	B & P	B & P	P & B2	B2 & W
Number measurements of each source in each counter	10	5 in B 10 in P	5 to 18	5	3
Duration of measurements (days)	2.98	25.37	25.66	5.3	21.86
Total number of measurements	240	90	206	100	72
Number of determinations (data sets)	8	4	4	4	8

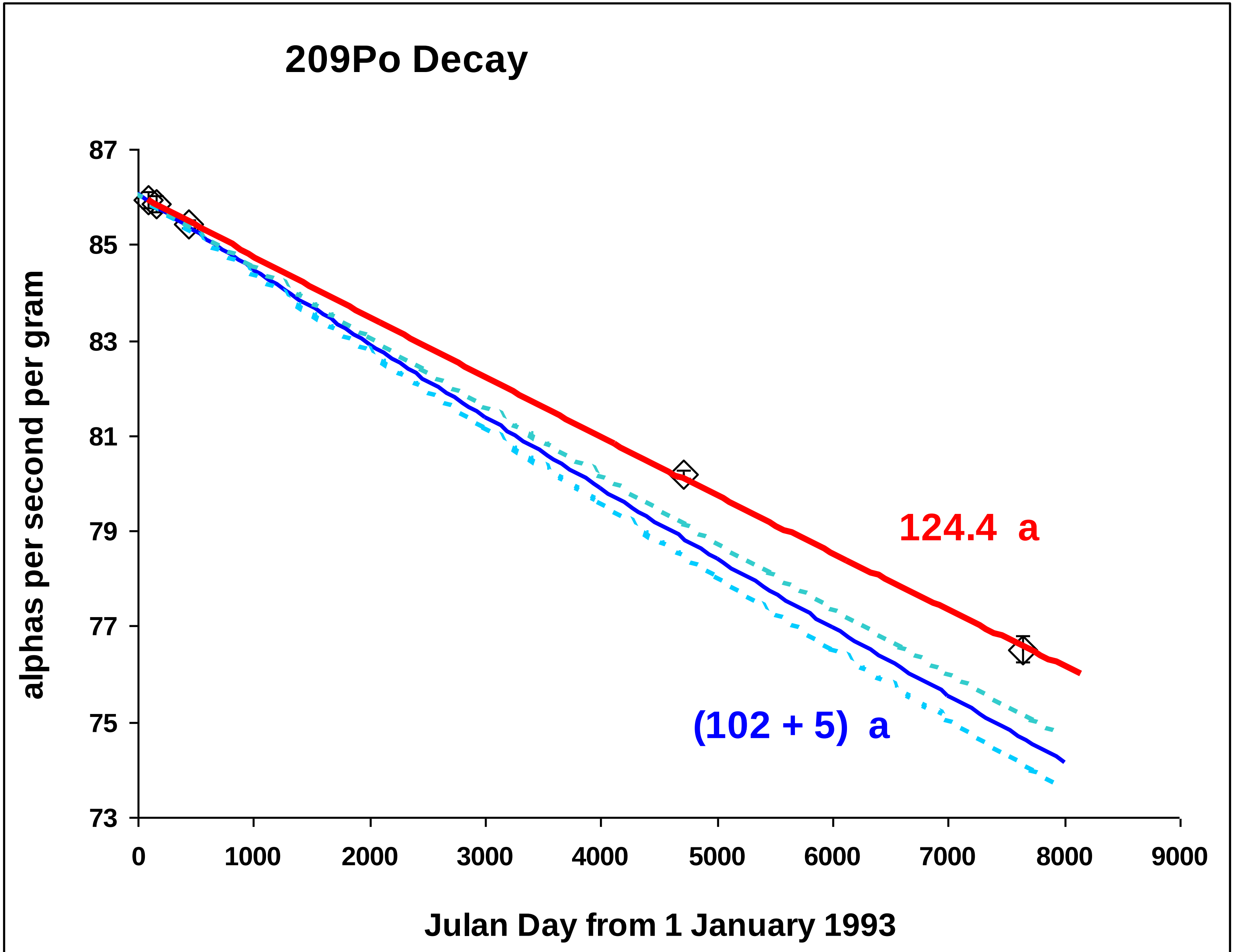
^{209}Po Decay Data

YEAR	T for YEAR	delta T	alphas	%s	n
1993	78.5123	1.4890	85.9571	0.192	8
1993	151.1836	12.6875	85.8692	0.196	4 (2)
1994	67.3984	7.5212	85.4343	0.120	4
2005	315.8840	3.1715	80.2100	0.104	4
2013	336.2601	9.3156	76.5263	0.342	8

half-life from all 5 values **(over values 1993-94 data)**

$T_{1/2} = 124.4$ a





209Po Decay
alphas per second per gram
87
85
83
81
79
77
75
73
0
1000
2000
3000
4000
5000
6000
7000
8000
9000
Julan Day from 1 January 1993
124.4 a
(102 + 5) a

Fitzgerald massaging

(*private communication, 16 Dec 2013*)

RESULTS FROM LEAST-SQUARES FITS USING DATAPLOT.

THE 68 % CONFIDENCE INTERVAL IS LISTED AS CL68.

***FOR CASES D AND F, I GUESSED AT S_I, BASED ON THE S_I'S THAT WERE BEING AVERAGED.**

prefer

Case	*n*	u_i	$T_{1/2}$/a	$u_{T1/2}$ /a	CL68/a (ν)	χ^2/ν
A	5	ERU (0.1 %)	124.4	1.2	1.4 (3)	0.97
B	5	s_i (Collé)	126.5	1.3	1.6 (3)	0.55
C	4	ERU	124.4	1.6	2.1 (2)	1.20
D	4	s_i (Collé*)	126.7	1.7	2.2 (2)	0.66
E	3	ERU	124.0	2.3	4.3 (1)	1.5
F	3	s_i (Collé*)	126.2	2.0	3.6 (1)	0.80

125.5

125.6

± 0.3 a

overestimated variances

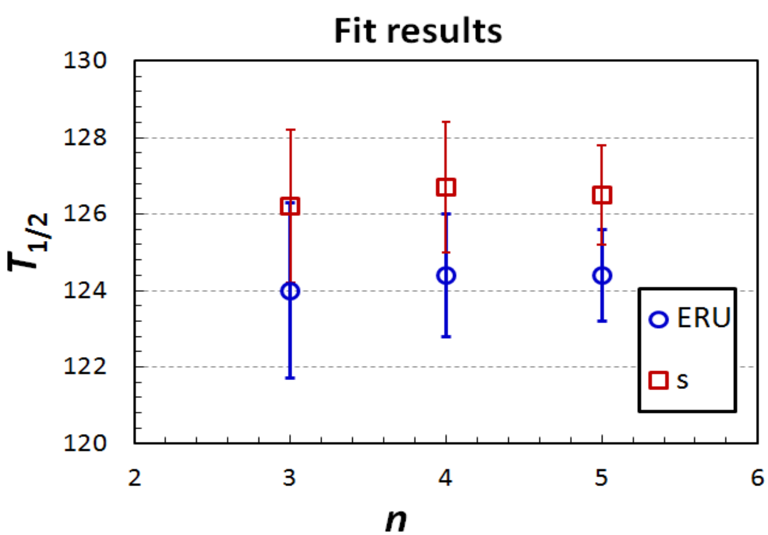


TABLE 2. RESULTS FROM LEAST-SQUARES FITS USING DATAPLOT. SAME AS TABLE 1, EXCEPT THE PRIMED CASES HAVE BEEN ADDED.

Case	*n*	u_i	$T_{1/2}$/a	$u_{T1/2}$ /a	CL68/a (ν)	χ^2/ν
A	5	ERU	124.4	1.2	1.4 (3)	0.97
B	5	s_i (Collé)	126.5	1.3	1.6 (3)	0.55
B'	5	$s_i/\sqrt{n}$	125.9	1.4	1.7 (3)	**1.3**
C	4	ERU	124.4	1.6	2.1 (2)	1.20
D	4	s_i (Collé)	126.7	1.7	2.2 (2)	0.66
D'	4	$s_i/\sqrt{n}$	125.3	1.6	1.9 (2)	**1.6**

Half-life uncertainty assessment

125.6 ± 3.2 a

component		***u* (a)**	**%**
Spectral analysis method	B	0.2	0.2
Least-squares fit	A	2.2	1.8
Weighting model choice	B	0.3	0.2
Long-term effects	B	2.2	1.8
combined		**3.2**	**2.6**

Fitzgerald support from nuclear theory

(private communication, 31 Dec 2013)

Most-complete account of closed-shell (Z=84) effect for α-decay half-life calculations, as of Dec 2013 – RGF

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Nuclear Physics A 852 (2011) 82–91

NUCLEAR PHYSICS A

www.elsevier.com/locate/nuclphysa

Systematic calculations on exotic α-decay half-lives of nuclei with $N = 125, 126, 127$

Yibin Qian [a,*], Zhongzhou Ren [a,b,c]

[a] Department of Physics, Nanjing University, Nanjing 210093, China
[b] Center of Theoretical Nuclear Physics, National Laboratory of Heavy-Ion Accelerator, Lanzhou 730000, China
[c] Kavli Institute for Theoretical Physics China, Beijing 100190, China

Received 9 December 2010; received in revised form 4 January 2011; accepted 7 January 2011

agrees with 126 a NOT 102 a

Half-life from Theoretical Partial Half-lives

Γ = total width (s^{-1})
$\Gamma_{0,1}$ = partial width of g.s., excited state
Γ_x = width to α_3 & electron capture
ρ_x = prob to α_3 & EC (1%)

$$T = 1/\Gamma$$

$$\Gamma = \Gamma_0 + \Gamma_1 + \Gamma_x$$

$$\Gamma = \Gamma_0 + \Gamma_1 + \rho_x \Gamma$$

$$\Gamma = \frac{\Gamma_0 + \Gamma_1}{1-\rho_x}$$

$$T = \frac{1-\rho_x}{T_0^{-1} + T_1^{-1}}$$

$$T = 4.4588 \cdot 10^9\ s$$

$$\boxed{T = 141.\ y}$$

INPUT DATA	Qian & Ren (2011)
$T_0 = 2.04 \cdot 10^{10}\ s$	theory
$T_1 = 5.78 \cdot 10^9\ s$	theory
$1-\rho_x = 0.98998 \pm 0.00014$ (NDS)	

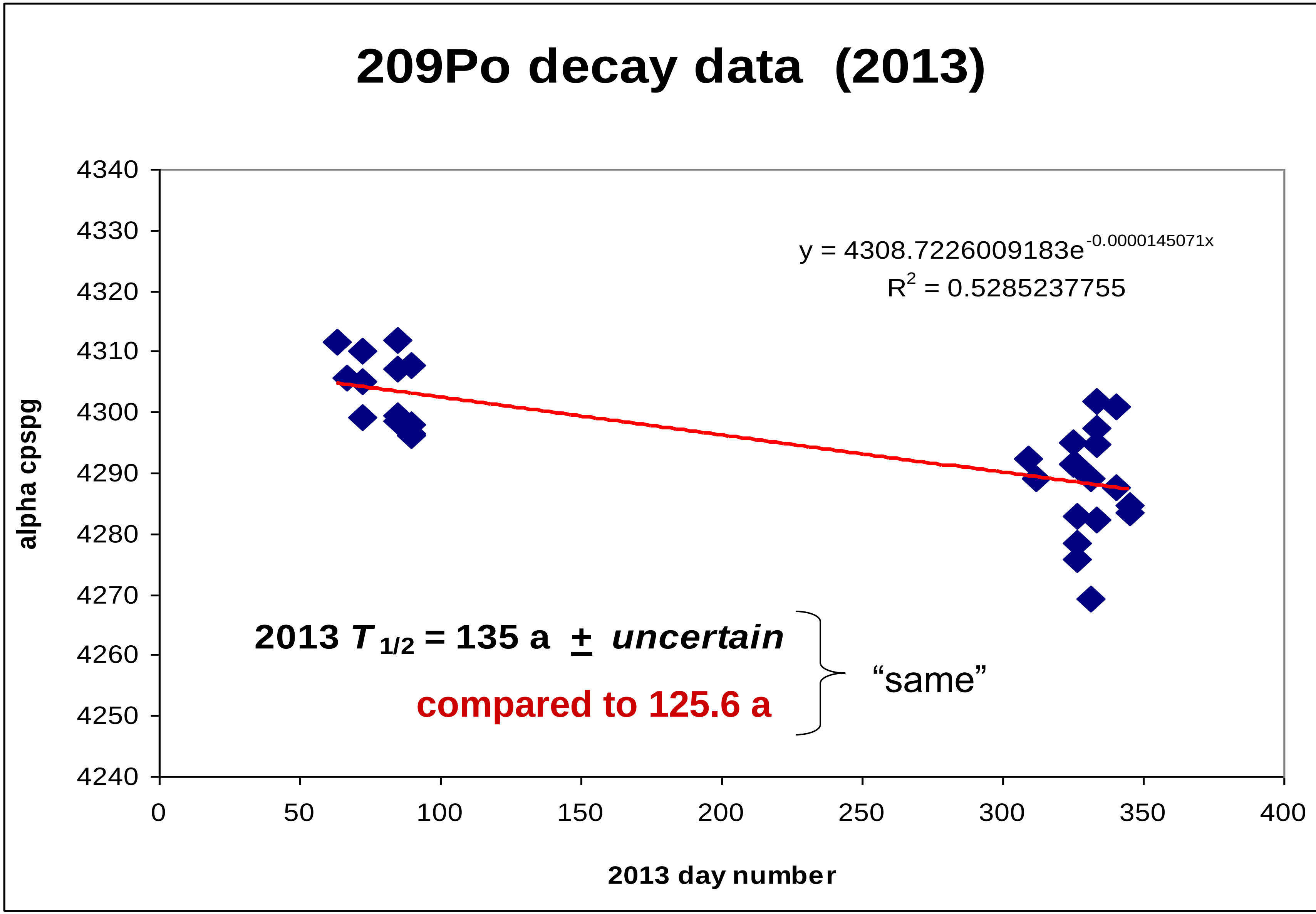
As uncertainty estimate, look at RMS residuals for similar nuclides.

R	N	Data Set
41%	42	all data in paper
12%	11	N=125,126; odd A
12%	6	N=125; odd A
9%	3	N=125; odd A; ground state trans.

call this "representative"

$$\boxed{T \approx (140 \pm 20)\ y}$$

RGF 12/31/13



31 mean values from New SRM

CREDITS

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Jerry LaRosa

Leticia Pibida

Lynn King

“Having goals and seeking specific results are desirable, and establishing rules & procedures may seem necessary, if you don't let them deprive you of interesting detours.”

R. Collé

FIN

with expectation of huge round of applause