^{209}Po and ^{210}Pb Radioactivity Standards:

A REVIEW

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Recent events may have led to increasing interest in Po & ^{210}Pb, but they have been of interest to our laboratory for many years ….

This is a review of about 20 years of work by me (and colleagues)

TOPICS

1992 * carrier free Po solution stability

1994 * ^{205}Pb (^{209}Po decay) confounding of LS measurements

1995 * ^{209}Po standard (SRM 4326)

1996 * ^{209}Po decay data -- photon emission probabilities (γ & K_α x)

2005 re-standardization of ^{209}Po

2007 * ^{210}Pb standard (SRM 4337)

2007 * ^{209}Po half-life error

2008 * ^{209}Po / ^{210}Pb links

2007 international comparison(s)

of UK (NPL) & USA (NIST) ^{210}Pb standards

2008-9 “

review not given in chronological order

* *publication dates*

NIST

Characteristic Elements of NIST Radioactivity Standardization Program *(R. Collé)*

Choice of nuclides / standards based on identified **<u>user needs</u>**

Standardization based on at least one **<u>primary method</u>**

Validity of primary method supported & confirmed by one or more **<u>independent confirmatory methods</u>**

Standardizations typically utilize **<u>many trials</u>**, with widely varying experimental conditions,

minimizing type-B uncertainty assessments

Any new standardization **<u>linked back</u>** to all previous ones (when possible)

through stored solutions or calibration factors for secondary instruments

<u>Disseminated standards</u> from primary methods used as SRM transfer standards

and/or employed as sources for quality assurance, & proficiency testing programs

Primary standardization **<u>uncertainties</u>** (k = 2) are typically < 1 % (few tenths at k = 1)

<u>Comparisons</u> with others metrology labs to demonstrate & ensure international consistency

World needs a Po tracer standard !

^{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*

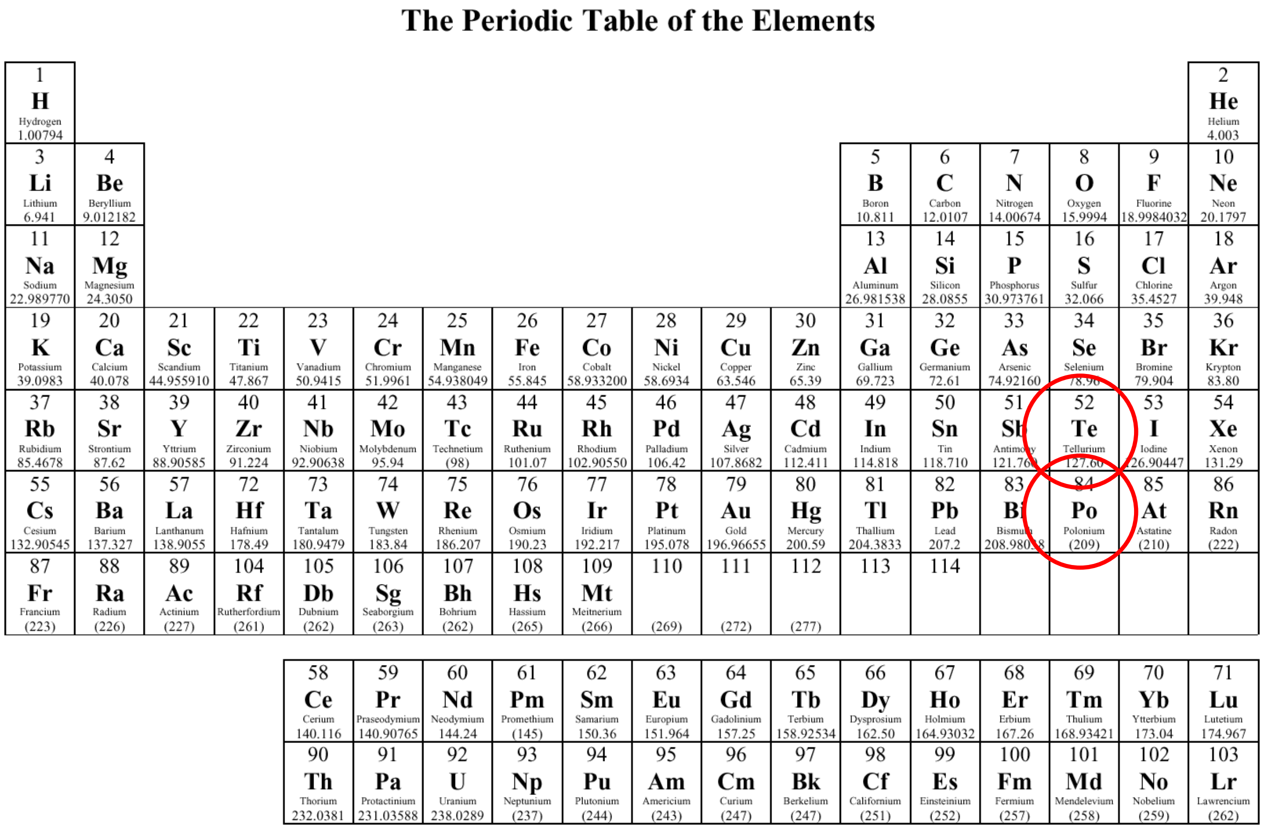
at NIST

< 1980	only ^{210}Po calibration sources
1984	^{208}Po standard (SRM 4327)
1994	^{209}Po standard (SRM 4326)
> 2007	no Po tracer standard available (now underway)

Carrier-free Po solutions & standards are stable

"Long-term stability of carrier-free polonium solution standards"

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



Carrier free --

How strong acid ?

In early 1990's not known (differing reports)

tests on ^{208}Po solutions over 1.2 to ≅ 9 years in range 0.1 to 1 mol L^{-1} HCl

< 0.3 N	unstable
0.3-0.5 N	equivocal
> 1 N	clearly stable

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

electronic homologues

Po [Xe] $4f^{14}$ $\underline{5d^{10}\ 6s^2\ 6p^4}$

Te [Kr] $\underline{4d^{10}\ 5s^2\ 5p^4}$

Solutions & standards – needs Te carrier or strong acid

Po at trace concentrations (under alkaline, neutral & weak acid conditions) known to be unstable: can be readily hydrolyzed, chemically deposited, volatilized; exhibit "radiocollodial behavior; "plate out & adsorption onto glass surfaces.

NIST

^{209}Po

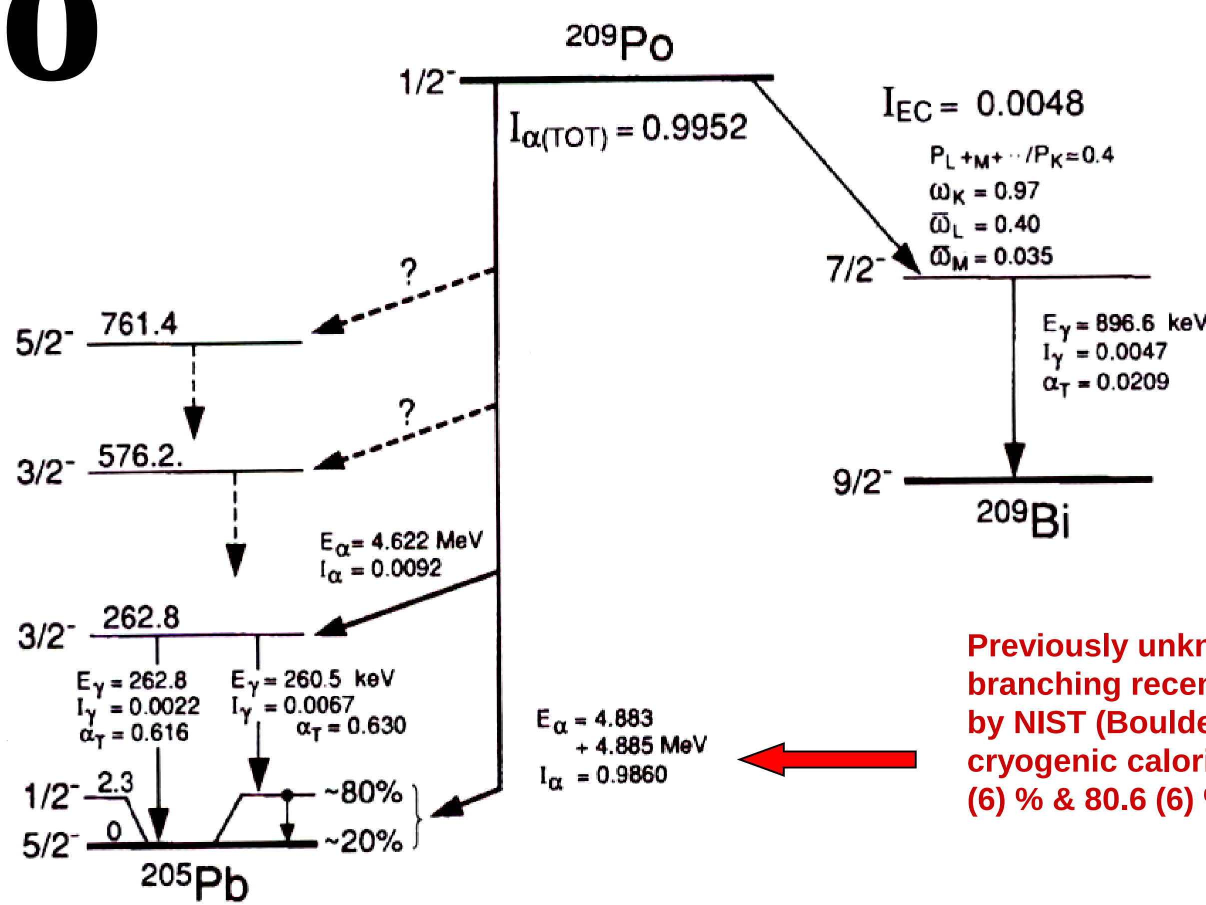


Fig. 2. Partial decay scheme for the ^{209}Po alpha and electron capture branch decays.

4

Previously unknown branching recently measured by NIST (Boulder Lab) by cryogenic calorimeter -- 19.4 (6) % & 80.6 (6) %

Collé, et al. *Appl. Radiat. Isot.* **45,** 1165 (1994);
Nucl. Instr. Meth. **A369,** 498 (1996).

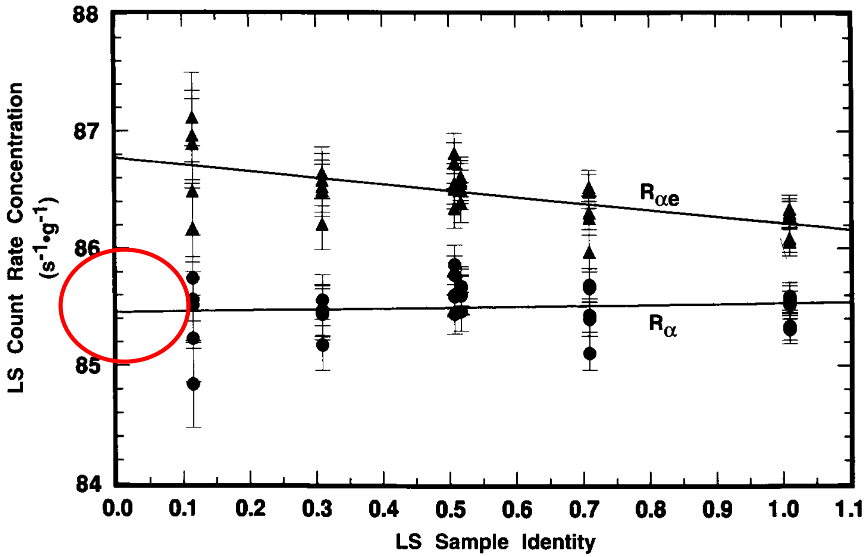


Fig. 11. LS counting rate concentrations $R_{\alpha e}$ (closed triangles) and R_α (closed circles) obtained with the Beckman instrument for the N series samples as a function of m_s (and sample quenching). The solid lines are linear regressions fitted to the data.

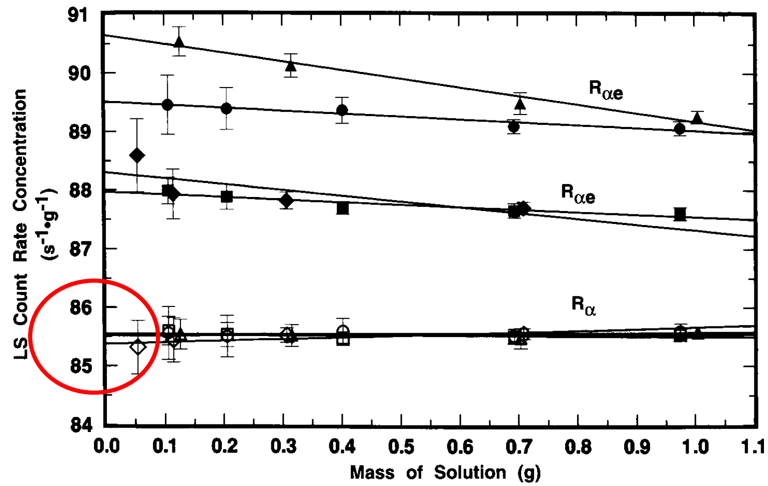


Fig. 13. LS counting rate concentrations $R_{\alpha e}$ and R_α obtained with the two LS systems for the P and Q series samples in 1994. Closed squares ($R_{\alpha e}$) and open squares (R_α) represent the mean values for samples Q5 through Q8 with the Packard; closed and open triangles represent $R_{\alpha e}$ 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.

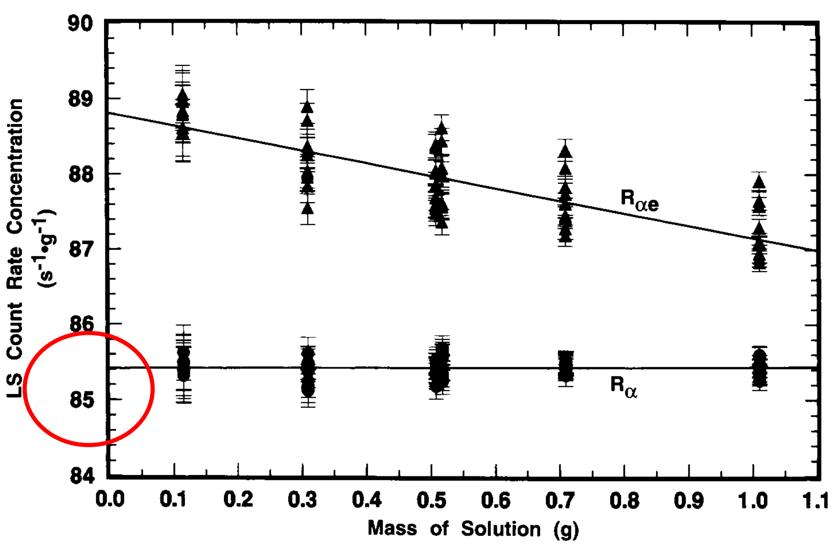


Fig. 12. LS counting rate concentrations $R_{\alpha e}$ and R_α as a function of m_s (analogous to that of Fig. 11) as obtained with the Packard instrument.

In 1994

LS result confirmed by $2\pi\alpha$ proportional counting

from

Collé, et al., *J. Res. Natl. Inst. Stds. Tech.* **100**, 1 (1995).

SAME IN 2005

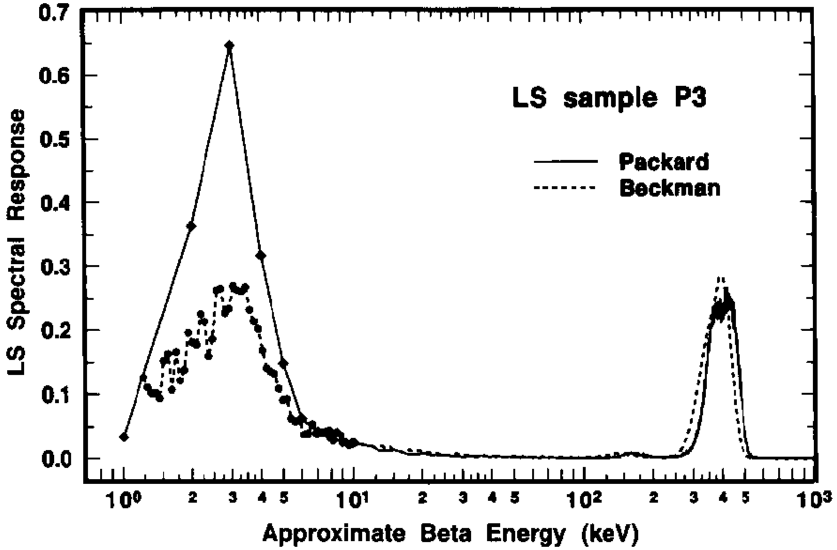


Fig. 6. Comparison of the ^{209}Po LS spectra obtained with the Beckman and Packard instruments.

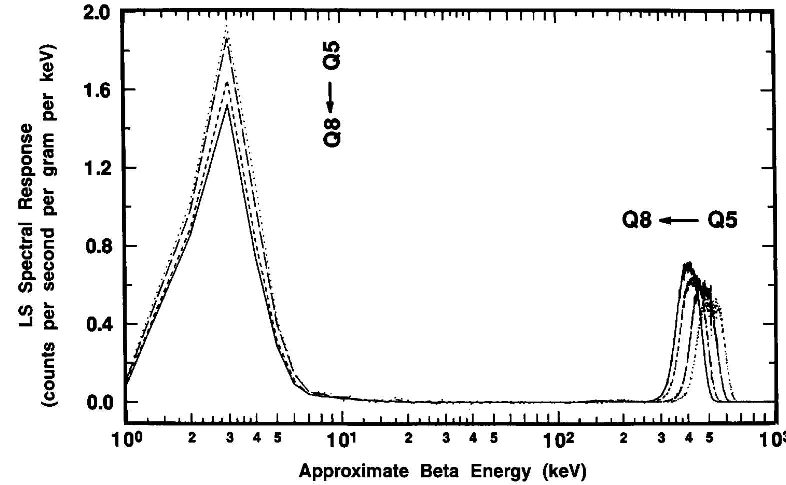


Fig. 14. LS spectra of increasingly quenched samples Q5 through Q8 obtained with the Packard counting system.

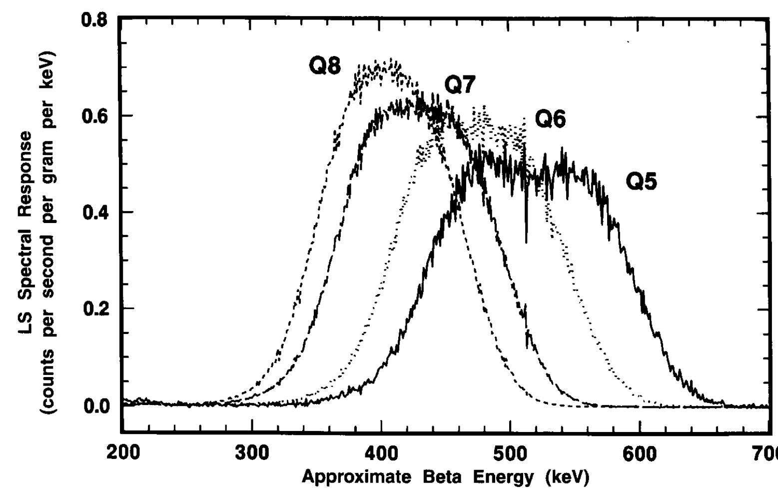


Fig. 15. Details of the broad alpha peaks shown in the full spectra of Fig. 14. The peak widths (FWHM) on a relative basis and peak areas are approximately equal in all four samples Q5 through Q8.

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

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

15 march 1994

R_α = (85.42 ± 0.18) $s^{-1}g^{-1}$

15 November 2005

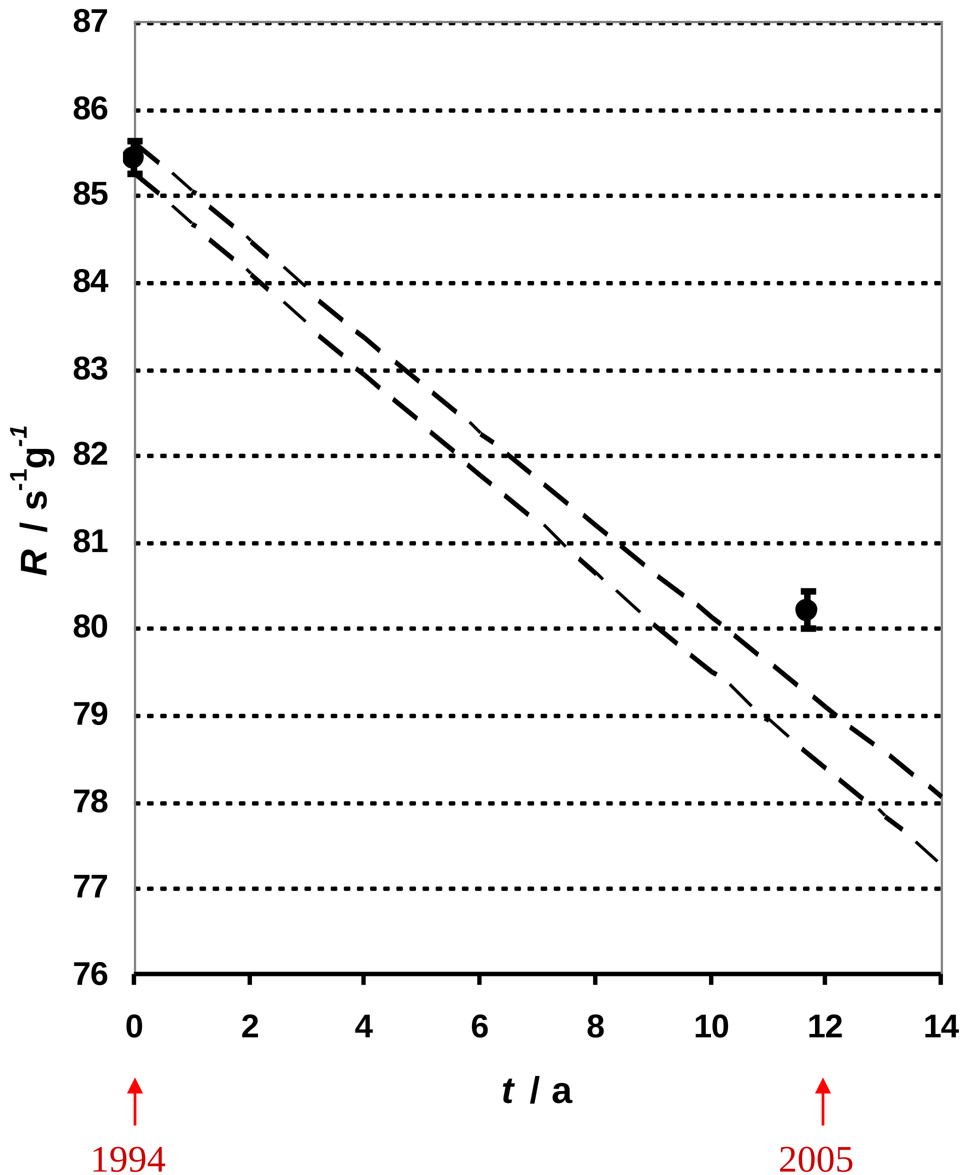
R_α = (80.20 ± 0.22) s^{-1} g^{-1}

2 point fit gives

$T_{1/2}$ = 128 a

U = 5.5 % (7 a)

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



R. Collé, et al., *Appl. Radiat. Isot.* 65, 728 (2007)

^{210}Pb

Difficult case

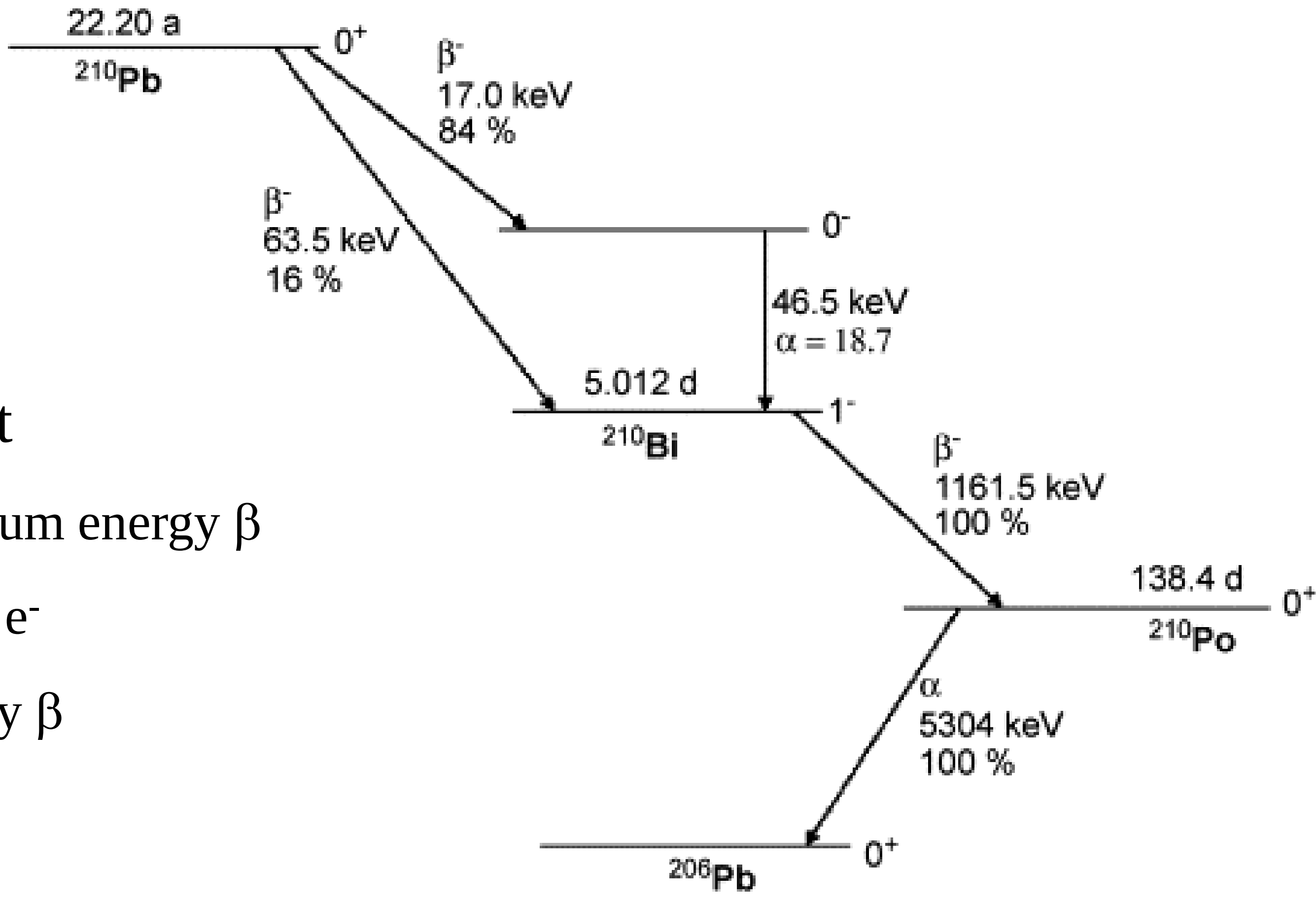
rarely done by metrology labs

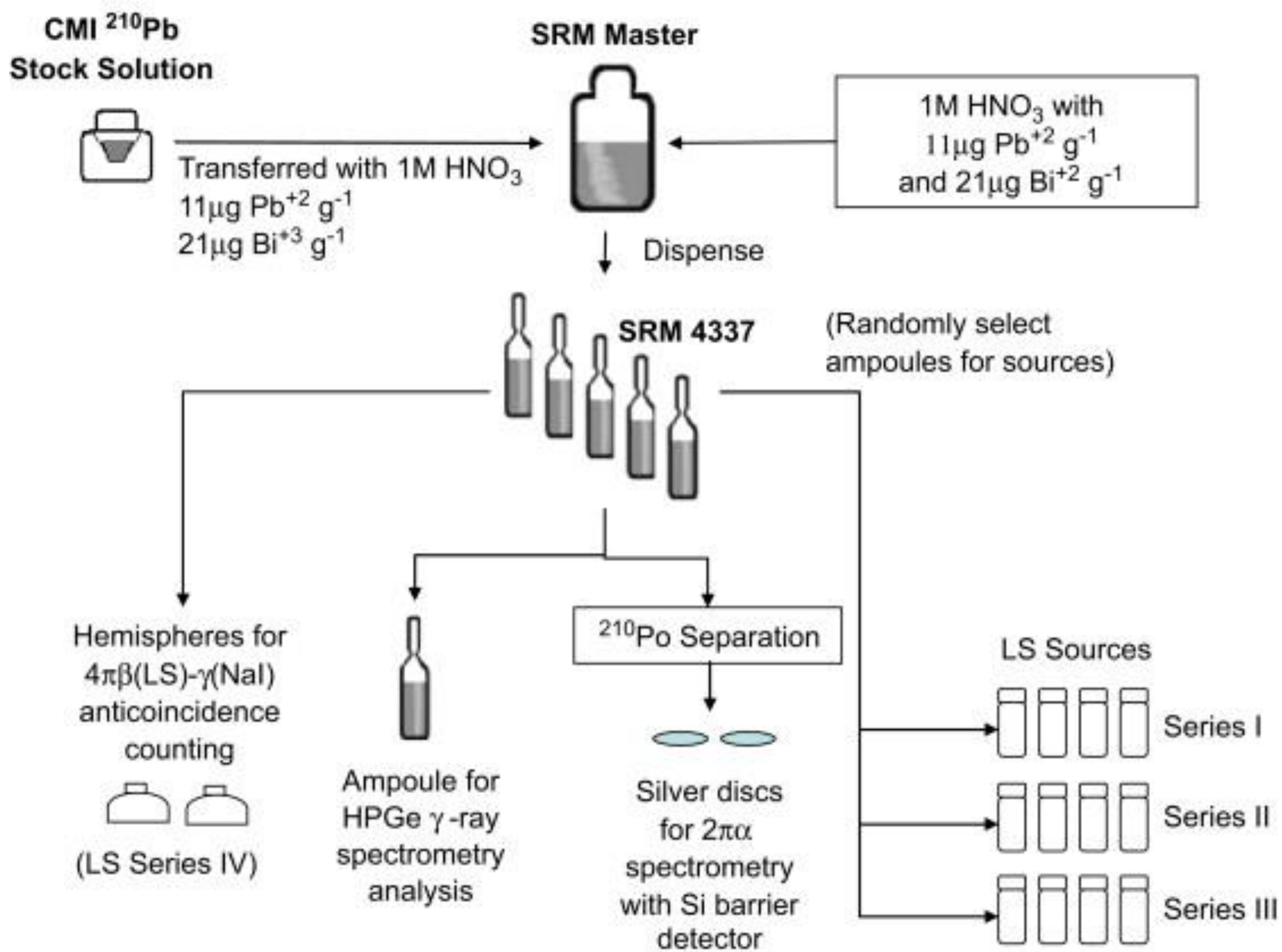
different methods used

Comparisons with NPL standard (via PTB too)

Need to detect

- low – medium energy β
- conversion e^-
- High energy β
- α





LTAC

γ spect

α spect

CNET

INADEQUATE TIME TO COVER

^{210}Pb standardization methods & data are published

L. Laureano-Perez, R. Collé, et al., *Appl. Radiat. Isot.* 65, 1368 (2007).

Ask or see me if interested -- I have 6 back-up slides of information !

^{210}Pb standardization results (SRM 4337)

	primary method	relative standard uncertainty	confirmatory measurements	percent differences
^{210}Pb	4παβ LS CNET	1.2 %	4παβ(LS)-γ(NaI) anticoin. Counting (± 1.7 %)	+ 0.7
			^{210}Po α spect. (102 a ^{209}Po tracer) (± 1 %)	- 3.0
			^{210}Po α spect. (128 a ^{209}Po tracer) (± 1 %)	- 1.3
			HPGe photon spect. (± 4.2 %)	+ 4.7

many assumptions, big extrapolation

^{209}Po half-life link

* big unc. if don't use some other ^{210}Pb std

uncertainty atypically large compared to most NIST radioactivity standardizations

* *probable error due to ^{210}Bi bremsstrahlung*

Relatively large 2.4 % uncertainty (k = 2) because of

(1) LS cocktail composition effects

(2) tracing code differences & assumptions,

(3) lack of good confirmatory measurements,

L. Laureano-Perez, R. Collé, et al., *Appl. Radiat. Isot.* **65**, 1368 (2007).

NIST

^{209}Po

^{209}Po

^{210}Pb

LINKED

In applications

– use of ^{209}Po tracer for ^{210}Po (^{210}Pb assays)

In our standardization measurements

^{210}Pb standardization results (SRM 4337)

	primary method	relative standard uncertainty	confirmatory measurements	Percent differences
^{210}Pb	$4\pi\alpha\beta$ LS CNET	1.2 %	$4\pi\alpha\beta$(LS)-γ(NaI) anticoin. Counting	+ 0.7
			^{210}Po α spect. (102 a ^{209}Po tracer)	- 3.0
			^{210}Po α spect. (128 a ^{209}Po tracer)	- 1.3
			HPGe photon spect.	+ 4.7 *

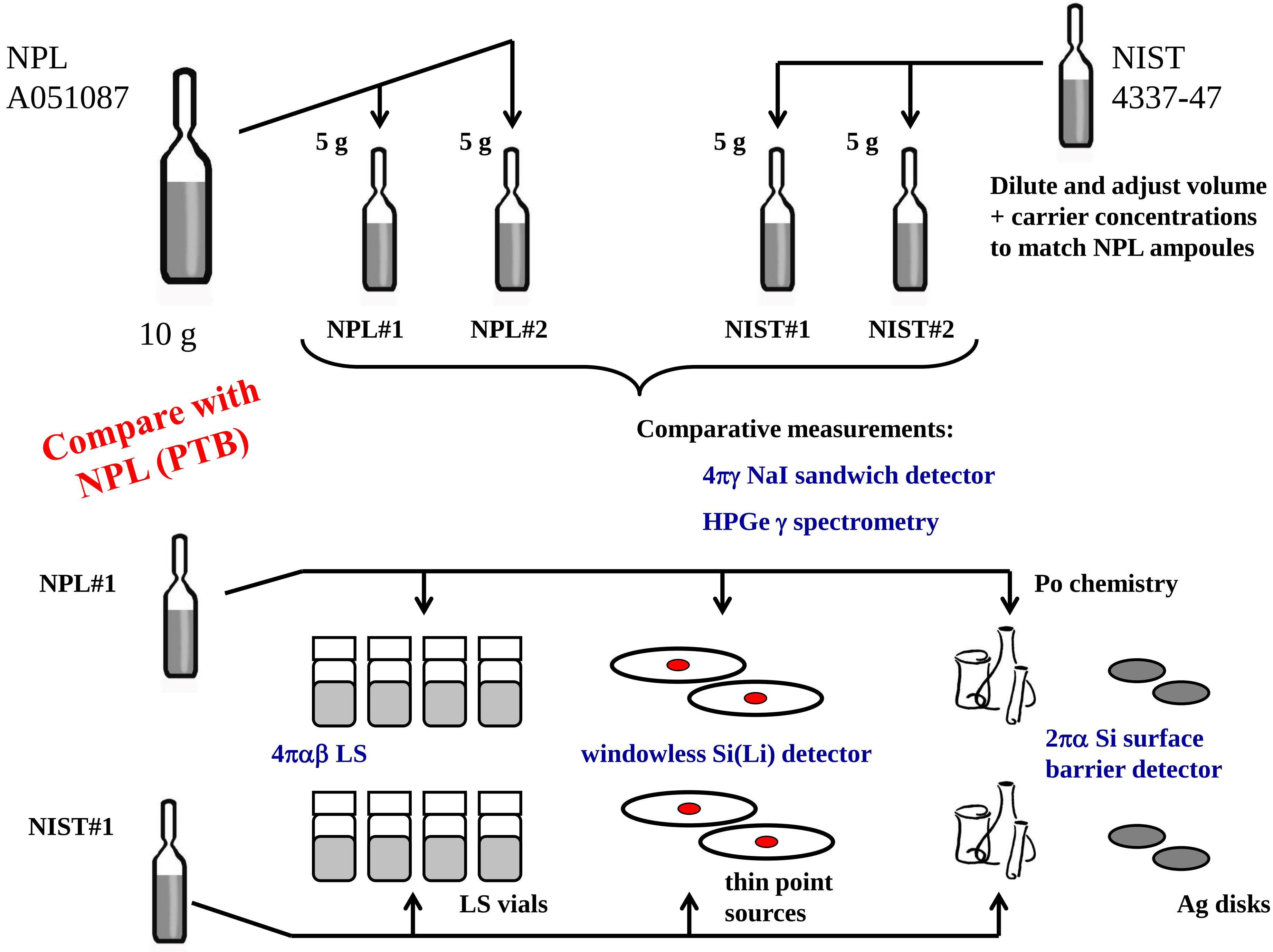
Needs longer half-life for consistency

* *probable error due to ^{210}Bi bremsstrahlung*

uncertainty atypically large compared to most NIST radioactivity standardizations

L. Laureano-Perez, R. Collé, et al., *Appl. Radiat. Isot.* **65**, 1368 (2007).

NIST

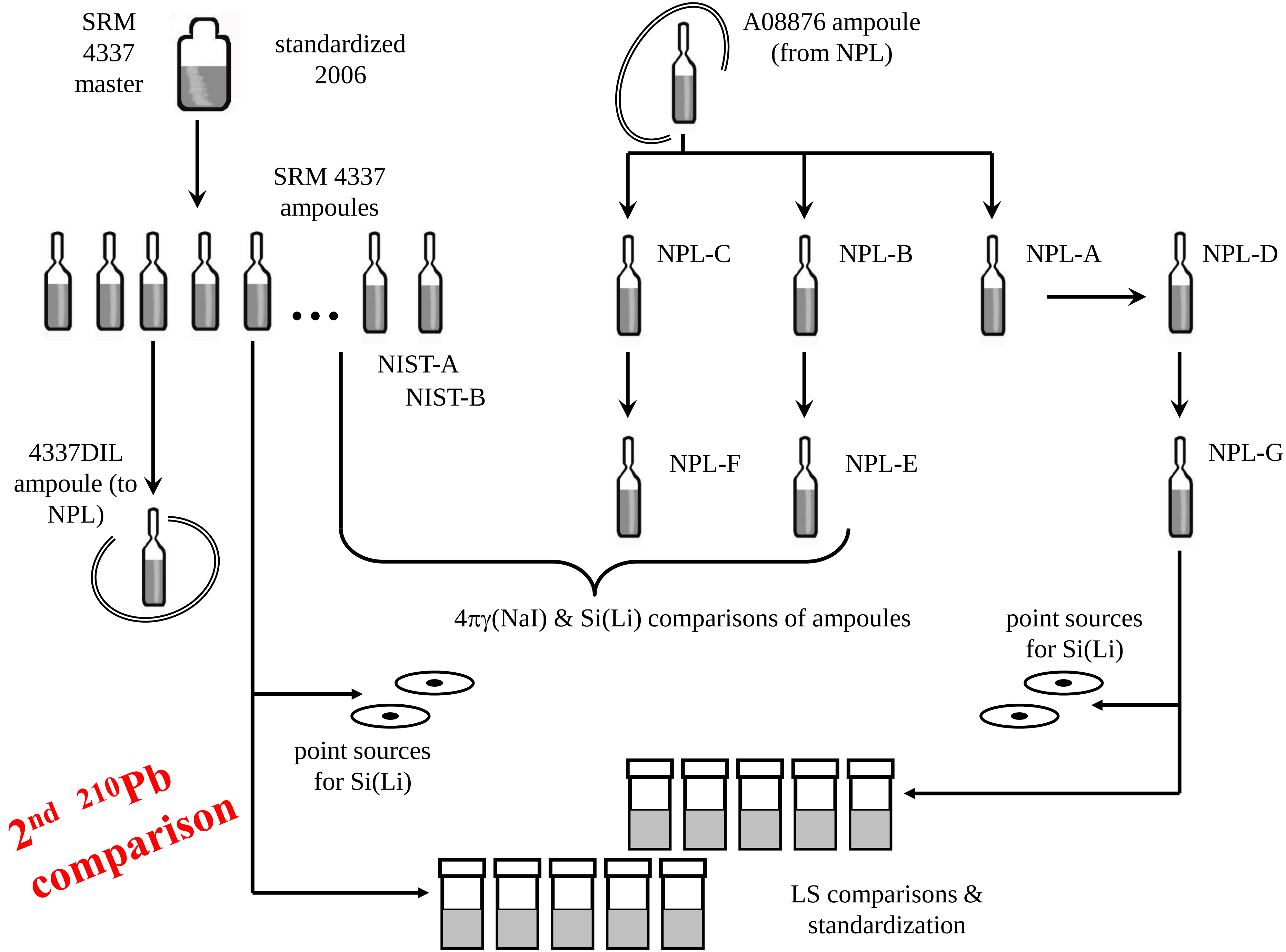

NPL
A051087
10 g
5 g
5 g
NPL#1
NPL#2
5 g
5 g
NIST#1
NIST#2
NIST
4337-47
Dilute and adjust volume
+ carrier concentrations
to match NPL ampoules
Comparative measurements:
4πγ NaI sandwich detector
HPGe γ spectrometry
Compare with
NPL (PTB)
NPL#1
Po chemistry
4παβ LS
windowless Si(Li) detector
2πα Si surface
barrier detector
NIST#1
LS vials
thin point
sources
Ag disks

2007 Comparison of the NIST and NPL ^{210}Pb standards by five measurement methods.

method	NPL / NIST ratio	relative standard uncertainty	difference
NPL and NIST certified values from primary standardizations	**0.037484**	1.5 %	--
$4\pi\gamma$(NaI)	0.037373	0.56 %	- 0.30 %
HPGe spectrometry.	0.036542	0.71 %	- 2.6 % ⇦ *
$4\pi\alpha\beta$(LS)	0.037249	0.17 %	- 0.63 %
^{210}Po assay ($2\pi\alpha$ spect.)	0.03736	0.75 %	- 0.33 %
Si(Li) low-energy spectrometry	0.0381	1.9 %	+ 1.6 %

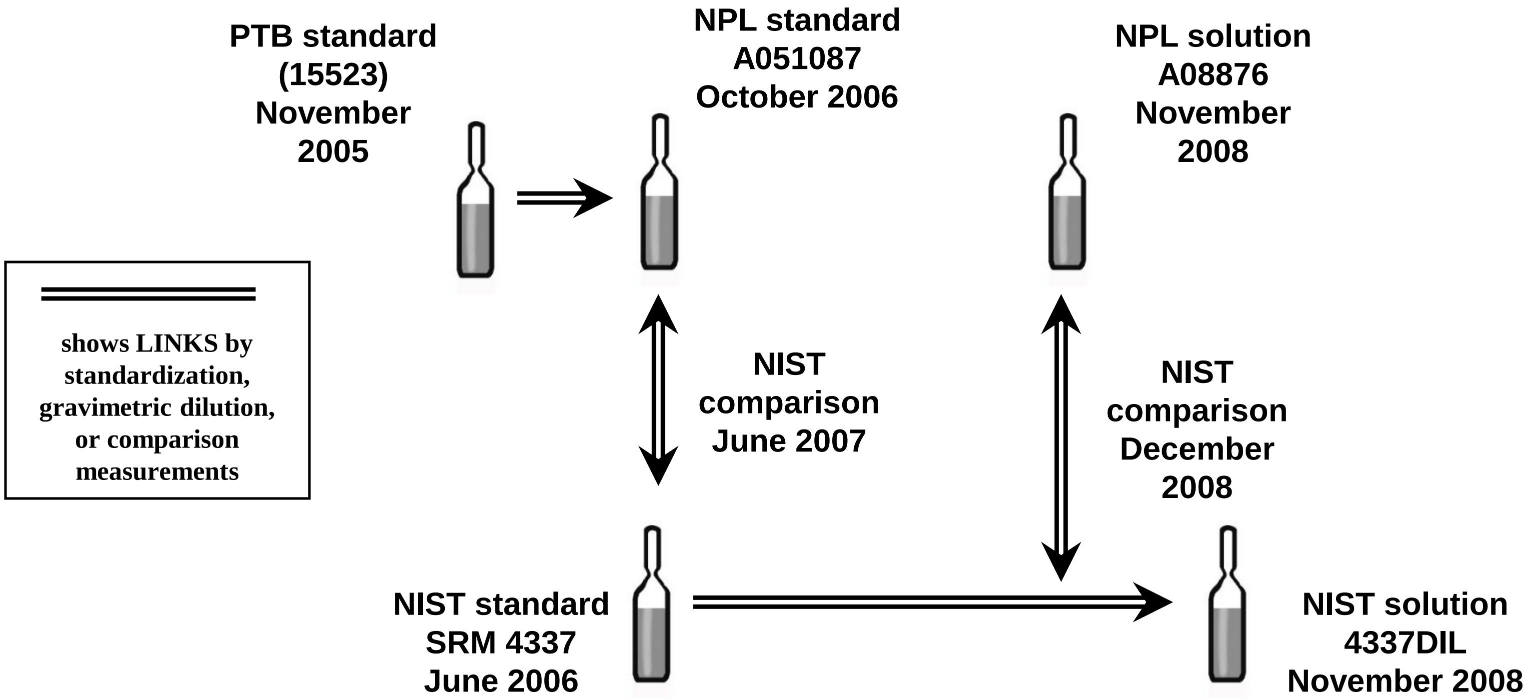
***** *probable error due to ^{210}Bi bremsstrahlung*

NIST



SRM 4337 master
standardized 2006
A08876 ampoule (from NPL)
SRM 4337 ampoules
NPL-C
NPL-B
NPL-A
NPL-D
NIST-A
NIST-B
NPL-F
NPL-E
NPL-G
4337DIL ampoule (to NPL)
4πγ(NaI) & Si(Li) comparisons of ampoules
point sources for Si(Li)
point sources for Si(Li)
2nd 210Pb comparison
LS comparisons & standardization

^{210}Pb NPL / NIST comparisons



similar 0.6 % NPL / NIST agreement – consistent with previous comparison

(for ≅ 1 % combined standard uncertainty in two standards)

For data, see **Lena Johansson** poster – ***this conference***

NIST

Summary important points:

- carrier-free Po solutions, ≥ 1 mol L^{-1} HCl
- ^{205}Pb isomeric state may confound ^{209}Po standardization / measurements
- ^{209}Po standard will be limited by half-life uncertainty
- ^{209}Po & ^{210}Pb linked through standards
- ^{210}Pb standard uncertainty may be over-estimated
- reasonable international agreement on ^{210}Pb standards ^{210}Pb

Thank you.

Acknowledgements & Special Thanks to

Lizbeth Laureano-Perez
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Leticia Pibida
Arzu Arinc (NPL)
Chris Gilligan (NPL)
Philippe Cassette (LNHB)
Frank Schima
Zhichao Lin
Pamela Hodge
J.M.R. (Robin) Hutchinson
Joy Thomas
Bert Coursey
and probably others I inadvertently forgot

for their contributions over the years

FIN

Backup slides only follow

^{210}Pb standardization data

^{210}Pb LS results (CN2003 code)

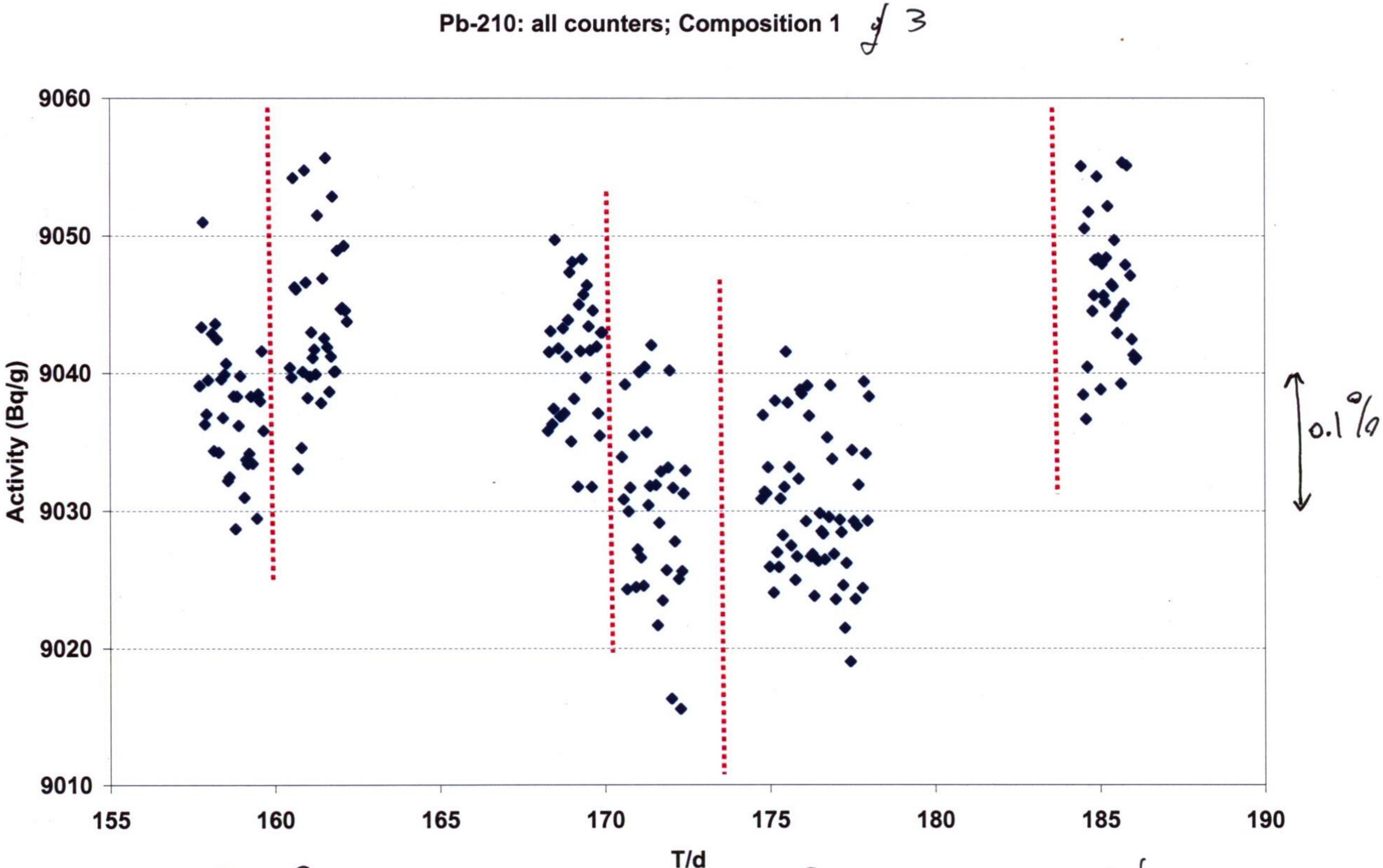
Pb-210

Series	Average	sd (%)	Normal	nc	ns	Counter	Scint	Age start	Age end	f_{H2O}	ε_{H-3}
1	9037.397	0.029	Y	3	11	Packard	HS	0.21	2.12	0.1	0.36-0.30
	9043.779	0.008	Y	3	11	Beckman	HS	2.95	4.65		
	9041.030	0.014	Y	3	11	Wallac	HS	10.76	12.4		
	9030.169	0.021	Y*	3	11	Packard	HS	13	14.9		
	9030.377	0.017	Y*	5	11	Packard	HS	17.22	20.46		
	9046.129	0.007	Y*	3	11	Wallac	HS	26.93	28.57		
2	9034.269	0.031	N	5	7	Packard	PCS	0.11	4.06	0.01	0.40-0.22
	9035.597	0.035	Y	5	7	Packard	PCS	0.11	4.06	0.04	
	9039.466	0.027	N	3	7	Wallac	PCS	4.78	6.91	0.01	
	9044.048	0.014	Y	3	7	Wallac	PCS	4.78	6.91	0.04	
	9040.539	0.026	no	3	7	Beckman	PCS	10.74	12.83	0.01	
	9041.935	0.026	yes	3	7	Beckman	PCS	10.74	12.83	0.04	
	9032.072	0.056	no	5	7	Packard	PCS	14.17	18.6	0.01	
	9026.263	0.034	yes	5	7	Packard	PCS	14.17	18.6	0.04	

* Data normal after removing sample with unstable cocktail

436 determinations

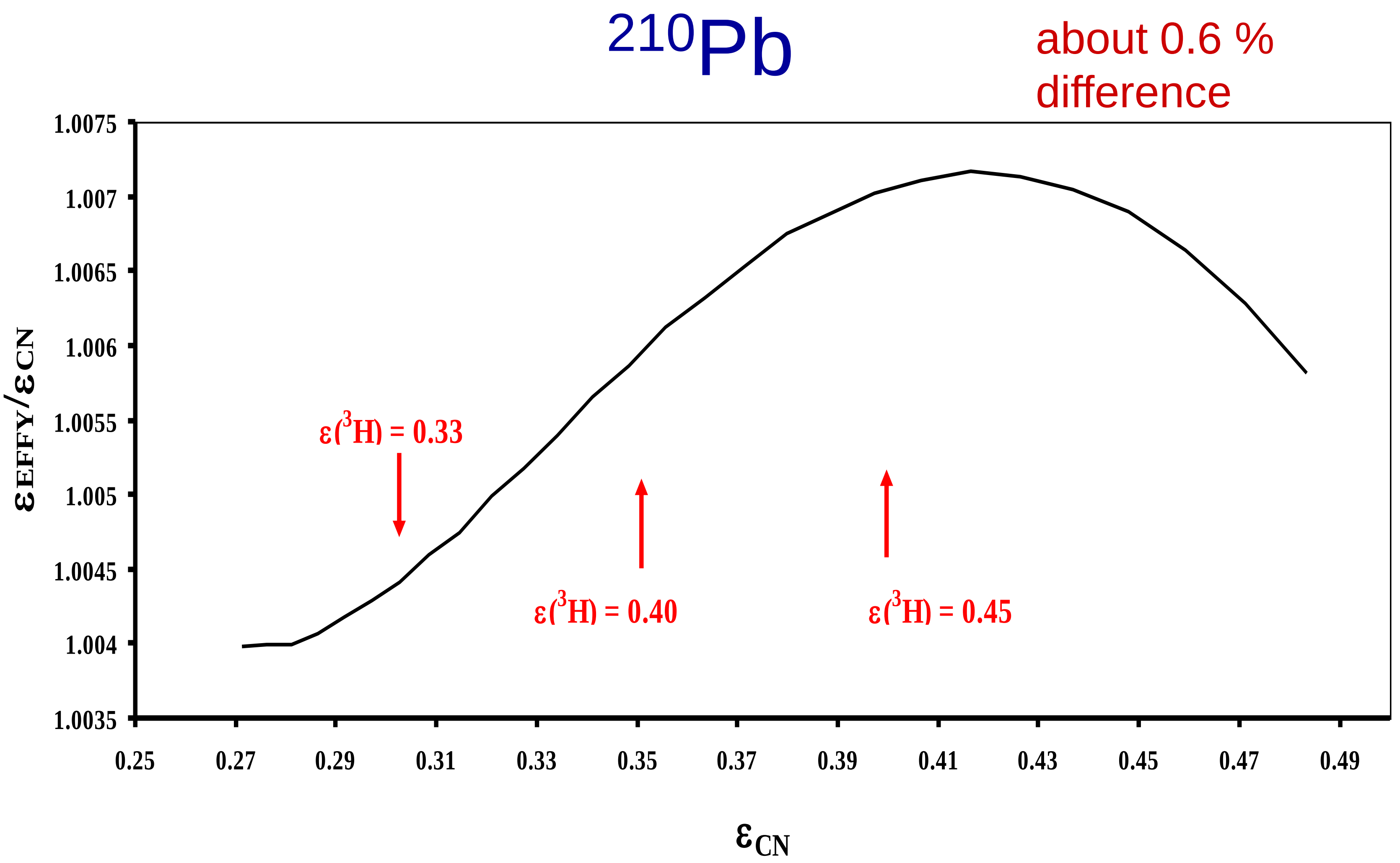
Series	Average	SD	SD (%)	Normal
1	9038.147	6.7577	0.07477	Yes
2	9036.774	5.8702	0.06496	Yes
Total	9037.362	6.0511	0.06696	Yes



	P	B	W	P	P	W
cycles	3	3	3	3	5	3
×12 samples.	36	36	36	36	60	36

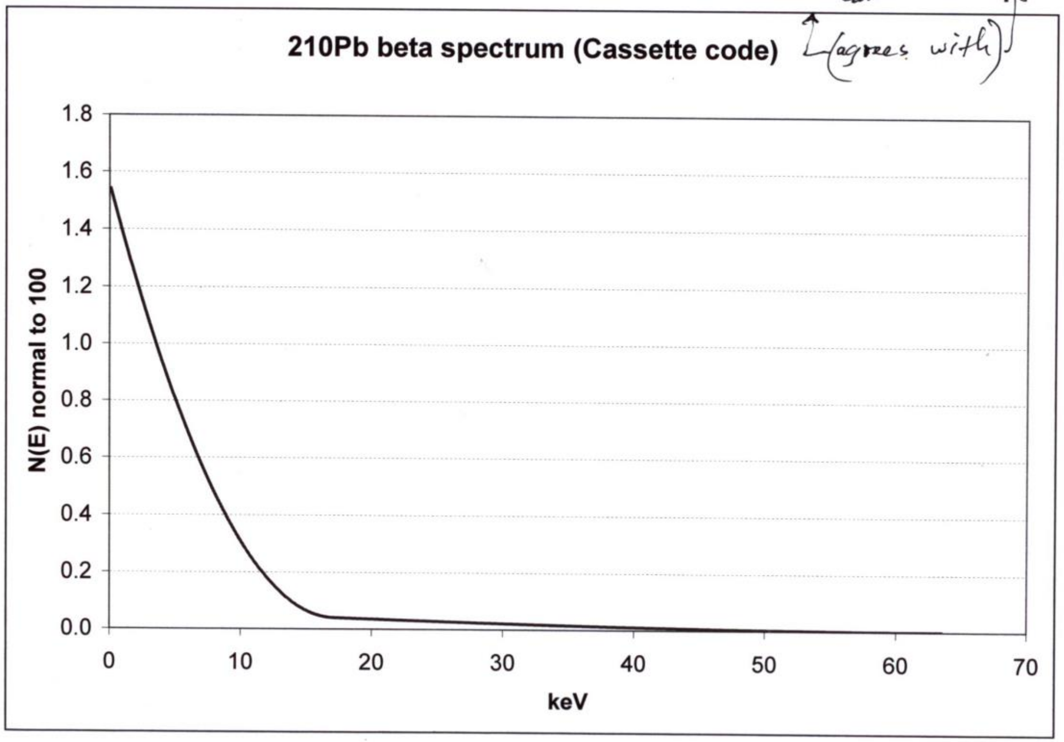
CN2003 vs EFFY4 code differences

(just Beta efficiency part)

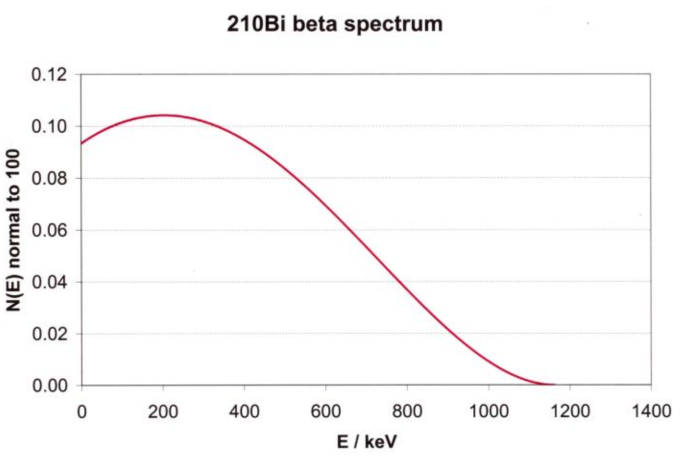


EFFY 4
CN2003 ← low E cutoff
Carine

(agrees with)



differences not due to spectra

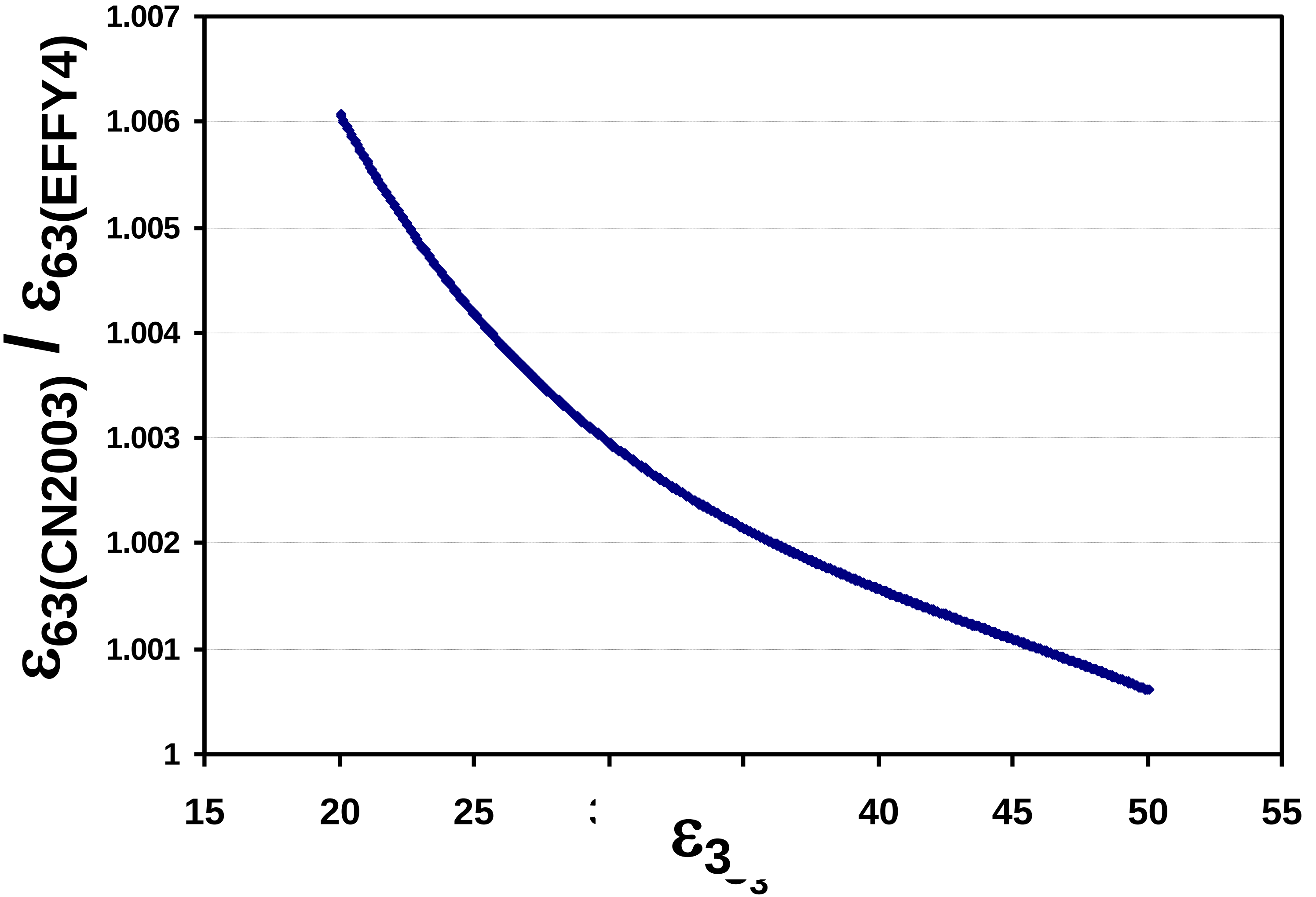


CN2003 vs EFFY4 code differences – due to assumed Quench function

EFFY4 – for toluene cocktails
CN2003 – for DIN (like HS3 used)
but also used xylene based (PCS)

Effect seen for other nuclides, like

^{63}Ni -- 17 keV $E_{\beta(ave)}$ allowed



^{210}Pb massic activity results

	kBq/g (k = 2)	diff from LS	
LS (CN2003)	9.037 ± 2.4 %	----	
γ-spect (HPGe)	9.46 ± 8.3 %	+ 4.7 %	big unc. if don't use ^{210}Pb γ std
$4\pi\beta$(LS)-γ(NaI) anticoincidence *(attempt)*	9.10 ± 3.3 %	+ 0.7 %	lots of assumptions, big extrapolation
α-spect (Po tracer)	8.77 ± 1 % 8.92	- 3.0 % - 1.3 %	$T_{1/2}$ = 102 a $T_{1/2}$ = 128 a

Relatively large 2.4 % uncertainty because of

(1) LS cocktail composition effects

(2) tracing code differences & assumptions,

(3) lack of good confirmatory measurements,