Argonne NATIONAL LABORATORY

Member of the US Nuclear Data Program

Unexpected Uncertainty in the NIST $4\pi\beta-\gamma$ Ionization Chamber

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ICRM 2011

18th International Conference on Radionuclide Metrology and its Applications

September 19-23, 2011
Tsukuba, Japan

U.S. DEPARTMENT OF ENERGY

	Session: Nuclear Decay Data - I *Chairpersons: M.-M. Bé, Y. Hino*
13:50 – 14:10	**Gamma- and X-ray emission probabilities in decay of ^{177m}Lu** *F.G. Kondev, I. Ahmad and J.P. Greene on behalf of the ^{177m}Lu collaboration*
14:10 – 14:30	**Unexpected uncertainty for NIST 4πγ ionization chamber** *M.P. Unterweger, et.al.*
14:30 – 14:50	**Measurement of the ^{230}U half-life** *S. Pommé, T. Altzitzoglou, R. Van Ammel, G. Suliman, M. Marouli, V. Jobbagy, J. Paepen, H. Stroh, C. Apostolidis, K. Abbas, A. Morgenstern*
14:50 – 15:10	**Half life requirements for nuclear forensics measurements** *S.M. Jerome, K.G.W. Inn*
15:10 – 15:20	**Poster Introduction of ND (7 posters)** *Coordinating referee of ND: M.-M. Bé*
	Decay Data Evaluation Project (DDEP): Evaluation of the main ^{243}Cm and ^{245}Cm decay characteristics *V. Chechev*
	Measurements of ^{64}Cu and ^{68}Ga half-lives and gamma-rays emission intensities *A. Luca, M. Sahagia, A. Antohe*
	Photon emission probabilities of ^{176}Lu *O. Ott, K. Kossert, O. Sima*
	Standardization, decay data measurements and evaluation of ^{64}Cu *M.-M Bé, P. Cassette, M.-N. Amiot, M.C. Lépy, C. Bobin, K. Kossert, O.J. Nähle, [illegible] P. Dryak, G. Ratel, , M. Sahagia, A. Luca, A. Antohe, L. Johansson, J. Keightley, A. [illegible]earce*
	Measurements of relative photon emission intensities and nuclear decay data evaluation of ^{113}Sn *A. Luca and M.-C. Lépy*
	^{57}Co half-life determination *C. J. da Silva, A. Iwahara, R. S. Gomes*
	Half-life determination of ^{88}Kr and ^{138}Xe *Shi-lian Wang, Tao Bai, Qi Li, Zhan-ying Chen, [illegible] Xie, Yongfu Chang*

important consequences for decay data evaluation

improved half-life data for actinide nuclides

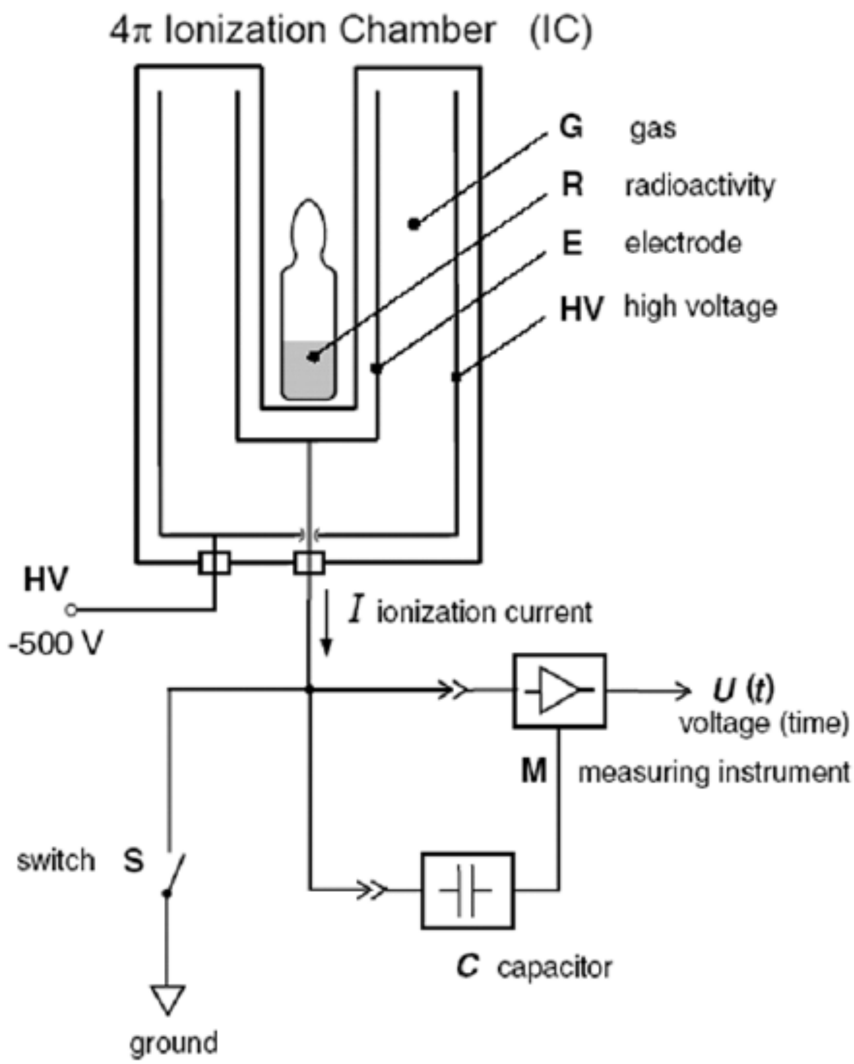
astrophysics

medical isotopes research

basic nuclear physics

	Session: Nuclear Decay Data -II *Chairpersons: F. Kondev, V. P. Chechev*
15:50 – 16:10	**Do radioactive half-lives vary with the earth-to-sun distance?** *J.C. Hardy, J.R. Goodwin, V.E. Iacob and V.V. Golovko*
16:10 – 16:30	**Assessment of actinide decay data: Findings of an IAEA coordinated research project** *M.A. Kellett et al.*
16:30 – 17:30	**Non-Neutron Nuclear Decay Data WG meeting**

Principles of operation



$I = \varepsilon_N A;\ A = A_0 e^{-\lambda \Delta t};\ \lambda = \ln 2/T_{1/2}$

ε_N – efficiency for a particular nuclide!

- ✓ activity determination
- ✓ lifetime determination

well-defined measurements conditions

- ✓ source geometry
- ✓ chemical composition and solution density
- ✓ corrections for various ampoule geometries and materials
- ✓ impurities
- ✓ **source holder parameters and source position in the chamber well**
- ✓ the shielding of the chamber - a 'stable' background

stability check (long-lived activities)

- ✓ ^{226}Ra ($T_{1/2}$=1600 a) or ^{166m}Ho ($T_{1/2}$=1200 a)

IAEA - Austria

International Journal of Applied Radiation and Isotopes, Vol. 31, pp. 153 to 154

Half-lives of 35 Radionuclides

H. HOUTERMANS, O. MILOSEVIC and F. REICHEL
International Atomic Energy Agency, P.O. Box 100, A-1400 Vienna, Austria

35 radionuclides
from 1972 - 1980

- ✓ not many details provided
- ✓ in most cases followed several $T_{1/2}$

Radion	Measured points	Total decay time	Half-life	Uncertainty
^{22}Na	383	3108	950.34	0.13
^{24}Na	640	180	14.9590	0.0012
^{46}Sc	74	380	83.819	0.006
^{51}Cr	72	155	27.690	0.005
^{55}Fe	290	894	1000.4	1.3
^{59}Fe	82	300	44.496	0.007
^{57}Co	344	1331	271.77	0.05
^{58}Co	131	430	70.916	0.015
^{60}Co	940	1889	1925.2	0.4
^{67}Ga	204	3000	3.2607	0.0008
^{75}Se	386	1488	119.779	0.004
^{85}Sr	135	540	64.856	0.007
^{88}Y	360	850	106.612	0.014
^{95}Zr	187	321	64.030	0.006
^{95}Nb	104	141	34.979	0.009
^{99}Mo	24	405	65.945	0.003
^{99m}Tc	96	16	6.006	0.002
^{103}Ru	121	490	39.254	0.008
^{106}Ru	62	1740	373.59	0.15
^{110m}Ag	76	530	249.74	0.05
^{111}In	83	80	2.8071	0.0015
^{113}Sn	106	790	115.09	0.04
^{125}Sb	232	1698	1007.3	0.3
^{125}I	167	334	59.156	0.020
^{131}I	72	99	8.0213	0.0009
^{134}Cs	490	1943	754.50	0.07
^{137}Cs	1417	2765	11009	11
^{133}Ba	975	5063	3848.0	1.1
^{140}La	48	240	40.280	0.006
^{144}Ce	308	1052	285.8	0.1
^{169}Yb	160	359	32.022	0.008
^{180m}Hf	34	29	5.519	0.004
^{192}Ir	109	353	73.831	0.008
^{203}Hg	175	508	46.582	0.002
^{203}Pb	21	314	51.88	0.02

Chalk River - Canada

Nuclear Instruments and Methods in Physics Research A 390 (1997) 267–273

A measurement of the half-lives of ^{54}Mn, ^{57}Co, ^{59}Fe, ^{88}Y, ^{95}Nb, ^{109}Cd, ^{133}Ba, ^{134}Cs, ^{144}Ce, ^{152}Eu

Robert H. Martin[a,*], Kerry I.W. Burns[a], John G.V. Taylor[b]

10 radionuclides

from 1985 - 1996

Nuclide	Number of published values used	Unweighted average of published values	Weighted average of published values	Present work
^{54}Mn	4	312.14 ± 0.08	312.14 ± 0.04	312.11 ± 0.05
^{57}Co	6	271.7 ± 0.3	271.80 ± 0.04	271.68 ± 0.09
^{59}Fe	3	44.51 ± 0.02	44.503 ± 0.005	44.472 ± 0.008
^{88}Y	6	106.62 ± 0.06	106.61 ± 0.03	106.65 ± 0.13
^{95}Nb	4	35.02 ± 0.09	34.99 ± 0.02	34.997 ± 0.006
^{109}Cd	7	459 ± 5	462.5 ± 0.5	460.2 ± 0.2
^{133}Ba	6	3839 ± 33	3848 ± 3	3849.1 ± 0.6
^{134}Cs	5	753.4 ± 1.1	754.31 ± 0.14	754.52 ± 0.18
^{144}Ce	10	284.8 ± 0.6	284.90 ± 0.03	286.14 ± 0.09
^{152}Eu	7	4892 ± 74	4945 ± 3	4948 ± 7

PTB - Germany

Applied Radiation and Isotopes 60 (2004) 317–323

Half-life measurements with ionization chambers—systematic effects and results

H. Schrader*

Physikalisch-Technische Bundesanstalt, Bundesallee 100, Braunschweig 38116, Germany

Nuclide	n	$\Delta t/T_{1/2}$	$T_{1/2}$ (d)[a] Present work	$T_{1/2}$ (d)[a] Earlier work at PTB	$T_{1/2}$ (d)[a] ENSDF[1]	$T_{1/2}$ (d) TdeR[2]	$T_{1/2}$ (d) Unterweger[3]	$T_{1/2}$ (d) Others
^{65}Zn	2 × 30	4.7	243.66 (9)	243.9 (8)[4]	244.06 (10)	244.06 (10)	244.164 (99)	244.15 (10)[5] 243.8 (3)[6]
^{85}Kr	1531	2.6	3916.8 (25)	3915 (3)[7] 3909 (11)[4]	3934.4 (14)	3911.8 (73)	3935.7 (12)	—
^{90}Sr	1061	0.6	10557 (11)	10513 (14)[7]	10516 (22)	10516 (22)	—	10495 (4)[8] 10561(14)[9]
^{106}Ru	3 × 35	2.4	370.5 (6)	371.7 (27)[4]	373.59 (15)	372.6 (10)	—	—
^{108}Ag^{m}	1071	0.05	159900 (3200)	153000 (5500)[7]				
			437.7 (88) a	418 (15) a[7]	418 (21) a	—	—	—
^{109}Cd	⩽249	2.25	459.6 (17)	—	461.4 (12)	461.4 (12)	463.26 (63)	460.2 (2)[10]
^{133}Ba	1613	2.7	3840.5 (65)	3841 (5)[7] 3842 (18)[4]	3848.9 (7)	3849.9 (25)	3854.7 (28)	3849.1 (6)[10]
^{137}Cs	420	0.2	10970 (20)	10921 (17)[4]	10983 (11)	10964 (9)	11018.3 (5)	10967.8 (45)[11] 10940.8 (69)[12]
^{152}Eu	1636	2.1	4934.1 (23)	4936 (2)[7] 4936.6 (20)[13]	4936.7 (22)	4940 (5)	4947.2 (11)	4943 (4)[14] 4948 (7)[10]
^{154}Eu	1534	3.0	3138.1 (11)	3139 (2)[7] 3138.1 (16)[13]	3138.6 (15)	3136.8 (29)	3145.2 (11)	3138 (2)[14]

Nuclide	n	$\Delta t/T_{1/2}$	$T_{1/2}$ (d)[a] Present work	$T_{1/2}$ (d)[a] ENSDF[1]	$T_{1/2}$ (d)[a] TdeR[2]	$T_{1/2}$ (d)[a] Unterweger[3]	$T_{1/2}$ (d)[a] Others
^{18}F	123 and 55	⩽9.2	1.82914 (35) h	1.8295 (8) h	1.82857 (30) h	1.82951 (34) h	1.8283 (19) h[4]
^{67}Ga	161 and 44	⩽2.0	3.2623 (15)	3.2612 (6)	3.2612 (5)	3.26154 (54)	—
^{81}Rb	⩽344	≈15	4.570 (4) h	4.576 (5) h	—	—	—
^{82}Rb^{m}	⩽344	≈15	6.472 (5) h	6.472 (6) h	—	—	—
^{99}Mo	732	7.9	2.7489 (6)	2.7475 (5)	2.7478 (4)	2.74683 (24)	—
^{99}Tc^{m}	2 × 120	⩽11.2	0.25024 (5)	0.25030 (4)	0.25033 (9)	0.250299 (36)	0.25026 (8)[5]
^{111}In	⩽159	⩽2.3	2.8063 (7)	2.8047 (5)	2.8047 (4)	2.80477 (53)	—
^{123}I	2 × 140	⩽7.4	0.55135 (27)	0.5529 (33)	0.5504 (13)	0.550979 (79)	—
^{131}I	1039	5.3	8.0252 (6)	8.02070 (11)	8.0210 (5)	8.0197 (22)	8.0207 (9)[5]
^{153}Sm	783	11.4	1.9284 (29)	1.92850 (17)	1.92849 (11)	1.928554 (58)	1.92808 (24)[6]
^{169}Er	454	⩽2.7	9.36 (4)	9.40 (2)	9.40 (2)	—	—
^{177}Lu	1074	⩽5.2	6.6475 (20)	6.734 (12)	6.647 (40)	6.64 (1)	6.646 (5)[7]
^{186}Re	⩽656	⩽10.7	3.7186 (5)	3.7183 (11)	3.775 (7)	3.7187 (29)	3.7183 (11)[8]
^{188}Re	⩽331	⩽9.9	0.70846 (14)	0.70850 (10)	0.70854 (17)	0.7084 (10)	0.70848 (9)[7]
^{201}Tl	⩽666	⩽6.8	3.0486 (30)	3.0380 (7)	3.0409 (29)	3.0456 (15)	3.043 (3)[4]
^{222}Rn	1530	⩽5.2	3.8195 (30)	3.8235 (3)	3.8235 (4)	—	3.8224 (18)[9]
^{224}Ra	⩽184	4.0	3.6319 (23)	3.66 (4)	3.66 (4)	—	—

27 nuclides

17 short-lived

10 long-lived

NIST - USA

Applied Radiation and Isotopes 56 (2002) 125–130

Half-life measurements at the National Institute of Standards and Technology

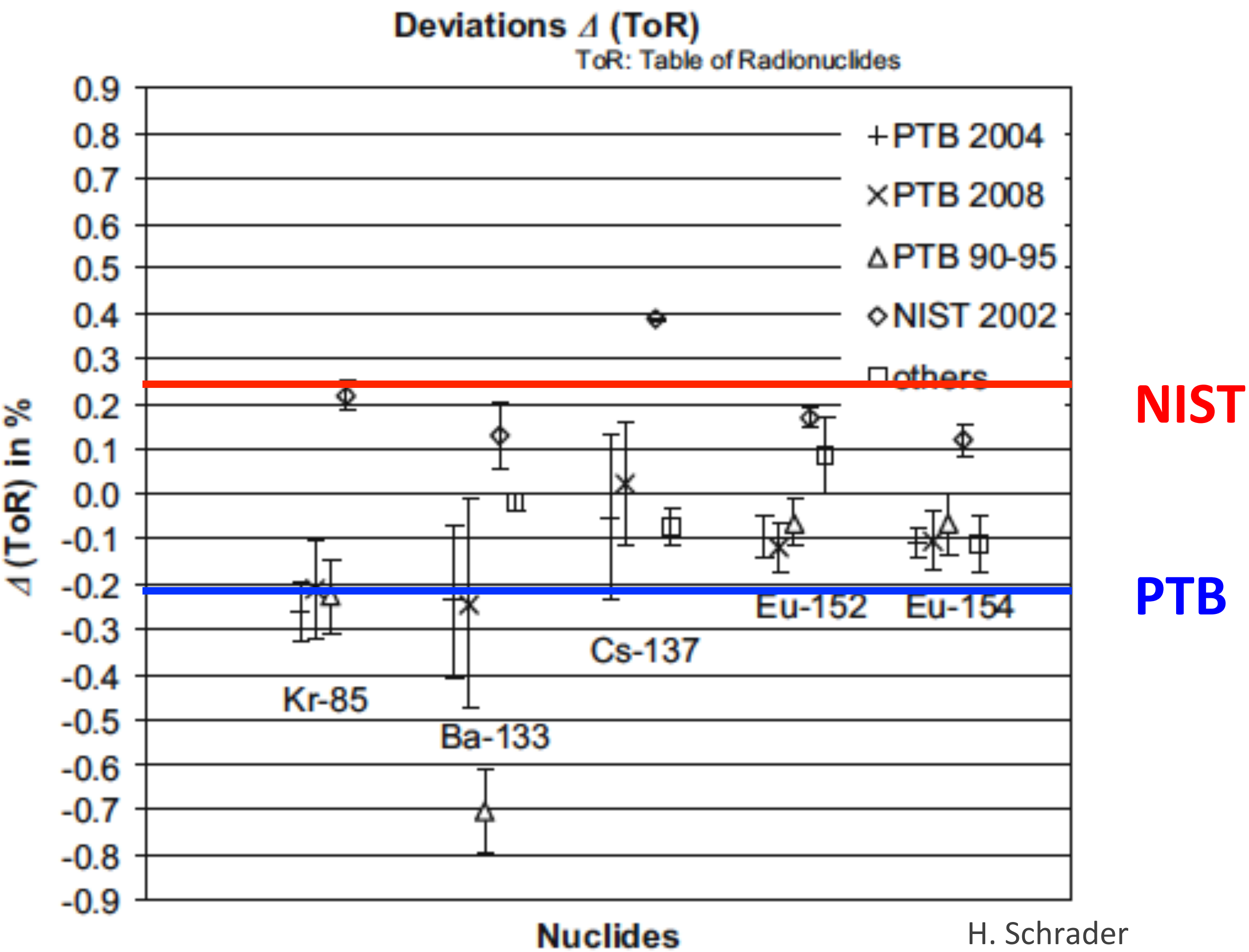
M.P. Unterweger*

Radio-nuclide	Number of sources	Number of half-lives followed	Half-life
^{188}W[b]	1	4.0	(69.783 ± 0.048) d
^{192}Ir	1	2.4	(73.810 ± 0.019) d
^{195}Au	5	0.6 — 6.0	(186.098 ± 0.047) d
^{198}Au	4	4.5 — 7.4	(2.69517 ± 0.00021) d
^{201}Tl	12	2.6 — 11.5	(3.0456 ± 0.0015) d
^{202}Tl	1	1.4	(12.466 ± 0.081) d
^{203}Hg	14	1.7 — 6.6	(46.619 ± 0.027) d
^{203}Pb	7	1.8 — 2.8	(51.923 ± 0.037) h
^{207}Bi[e]	2	0.9	$(11523 \pm 15.)$ d
^{228}Th	6	1.7 — 8.3	(698.60 ± 0.36) d

- ❑ TPA Mk II IC, filled with 20 atm of argon
- ✓ in use since 1967
- ✓ data used for $T_{1/2}$ after 1973 (earlier values rejected due to IC instability)
- ❑ the most comprehensive series of measurements – never doubted them!

Radio-nuclide	Number of sources	Number of half-lives followed	Half-life	Statistical uncertainty[a]	Other uncertainty[a]
^{3}H[b,c,d]	18	3	(4500 ± 8) d	8	0
^{18}F	3	13.1	(1.82951 ± 0.00034) h	0.00024	0.00024
^{22}Na	5	1.9 — 4.7	(950.97 ± 0.15) d	0.09	0.12
^{24}Na	14	1.1 — 7.6	(14.9512 ± 0.0032) h	0.0009	0.0031
^{32}P[b]	2	2.8	(14.263 ± 0.003) d	0.003	0.0
^{46}Sc	4	3.6 — 10.3	(83.831 ± 0.066) d	0.030	0.059
^{51}Cr	11	2.3 — 8.9	(27.7010 ± 0.0012) d	0.0007	0.0009
^{54}Mn	2	3.3 — 7.4	(312.028 ± 0.034) d	0.034	0.0
^{57}Co	7	4.7 — 10.4	(272.11 ± 0.26) d	0.09	0.25
^{58}Co	1	9.1	(70.77 ± 0.11) d	0.11	0.0
^{59}Fe	6	4.0 — 9.3	(44.5074 ± 0.0072) d	0.0048	0.0053
^{60}Co[e]	8	3.7 — 5.3	(1925.20 ± 0.25) d	0.10	0.23
^{62}Cu[b,f]	3	2.7 — 3.9	(9.6725 ± 0.0080) m	0.0080	0.0
^{65}Zn	1	3.2	(244.164 ± 0.099) d	0.099	0.0
^{67}Ga	13	1.8 — 8.3	(3.26154 ± 0.00054) d	0.00015	0.00052
^{75}Se	19	2.4 — 8.7	(119.809 ± 0.066) d	0.014	0.065
^{85}Kr[e]	1	1.9	(3935.7 ± 1.2) d	1.2	0.0
^{85}Sr	8	1.1 — 4.8	(64.8530 ± 0.0081) d	0.0039	0.0071
^{88}Y	8	1.3 — 8.1	(106.626 ± 0.044) d	0.017	0.041
^{99}Mo	14	3.6 — 9.5	(65.9239 ± 0.0058) h	0.0031	0.0049
^{99m}Tc[g]	33	2.1 — 12.0	(6.00718 ± 0.00087) h	0.00015	0.00086
^{99m}Tc (acid)	17	1.9 — 8.0	(6.0123 ± 0.0032) h	0.0007	0.0031
^{103}Ru[b]	7	0.4	(39.310 ± 0.044) d	0.044	0.0
^{109}Cd	2	3.4 — 5.3	(463.26 ± 0.63) d	0.36	0.51
^{110m}Ag	1	9.3	(249.950 ± 0.024) d	0.024	0.0
^{111}In	11	1.4 — 9.3	(2.80477 ± 0.00053) d	0.00017	0.00051
^{113}Sn	11	2.3 — 11.0	(115.079 ± 0.080) d	0.025	0.076
^{117m}Sn[b,h]	10	1.5 — 4.0	(14.00 ± 0.05) d	0.05	0.0
^{123}I	3	5.4 — 12.7	(13.2235 ± 0.0019) h	0.0019	0.0
^{125}I	18	1.4 — 6.2	(59.49 ± 0.13) d	0.03	0.12
^{125}Sb[e]	1	5.4	(1007.56 ± 0.10) d	0.10	0.0
^{127}Xe	5	1.1 — 11.5	(36.3446 ± 0.0028) d	0.0028	0.0
^{131}I	21	1.0 — 10.9	(8.0197 ± 0.0022) d	0.0005	0.0021
^{131m}Xe	2	1.8	(11.934 ± 0.021) d	0.014	0.016
^{133}Ba[e]	4	2.0	(3854.7 ± 2.8) d	1.3	2.5
^{133}Xe	3	4.8 — 11.2	(5.24747 ± 0.00045) d	0.00045	0.0
^{134}Cs	5	1.7 — 3.0	(753.88 ± 0.15) d	0.11	0.11
^{137}Cs[e]	6	0.7 — 0.9	(11018.3 ± 9.5) d	3.5	8.8
^{139}Ce	9	1.5 — 6.4	(137.734 ± 0.091) d	0.029	0.086
^{140}Ba	10	1.8 — 4.4	(12.7527 ± 0.0023) d	0.0009	0.0022
^{140}La	2	4.2	(40.293 ± 0.012) h	0.008	0.009
^{141}Ce	1	6.1	(32.510 ± 0.024) d	0.024	0.0
^{144}Ce[e]	1	3.9	(284.534 ± 0.032) d	0.032	0.0
^{153}Sm	1	7.3	(46.2853 ± 0.0014) h	0.0014	0.0
^{154}Eu[e]	4	2.4	(3145.2 ± 1.1) d	1.1	0.0
^{155}Eu[e]	2	3.1 — 4.3	(1739.06 ± 0.45) d	0.45	0.0
^{166}Ho[e]	2	5.4 — 7.2	(26.794 ± 0.023) h	0.013	0.019
^{169}Yb	14	3.4 — 9.5	(32.0147 ± 0.0093) d	0.0026	0.0089
^{177}Lu[b,i]	4	2 — 5	(6.64 ± 0.01) d	0.01	0.0
^{181}W	3	5.9 — 6.6	(121.095 ± 0.064) d	0.042	0.048
^{186}Re	2	5.7 — 6.5	(89.248 ± 0.069) h	0.018	0.067
^{188}Re[e]	3	4.2 — 7.1	(17.001 ± 0.022) h	0.09	0.021

NIST vs. PTB



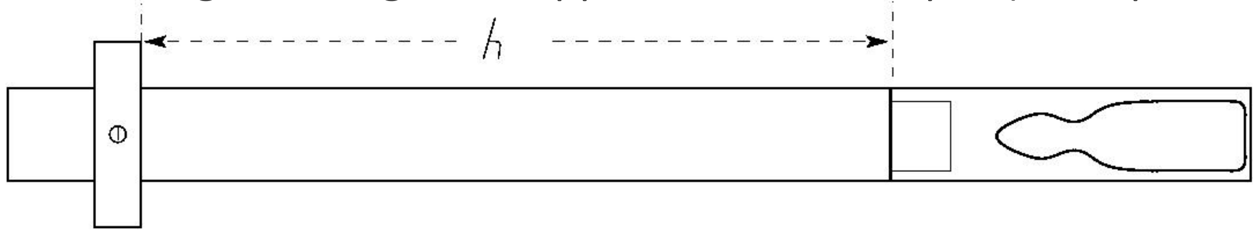
~0.4% difference

H. Schrader

Applied Radiation and Isotopes 68 (2010) 1583–1590

What went wrong?

M. P. Unterweger & Fitzgerald, Appl. Radiat. Isotopes (to be published)



- in January 2010 it was found that the source holder used for calibrations at NIST was not stable
- led to a slow change in calibration factors for various nuclides
  - ✓ dependent on the gamma-ray energy and the time IC was calibrated
  - ✓ changes most significant for long-lived gamma emitters: ^{60}Co, ^{85}Kr, ^{125}Sb, ^{133}Ba, ^{137}Cs, ^{152,154,155}Eu, ^{207}Bi

- best possible recalibration for the longer-lived radionuclides
- publication expected within ~6 months