

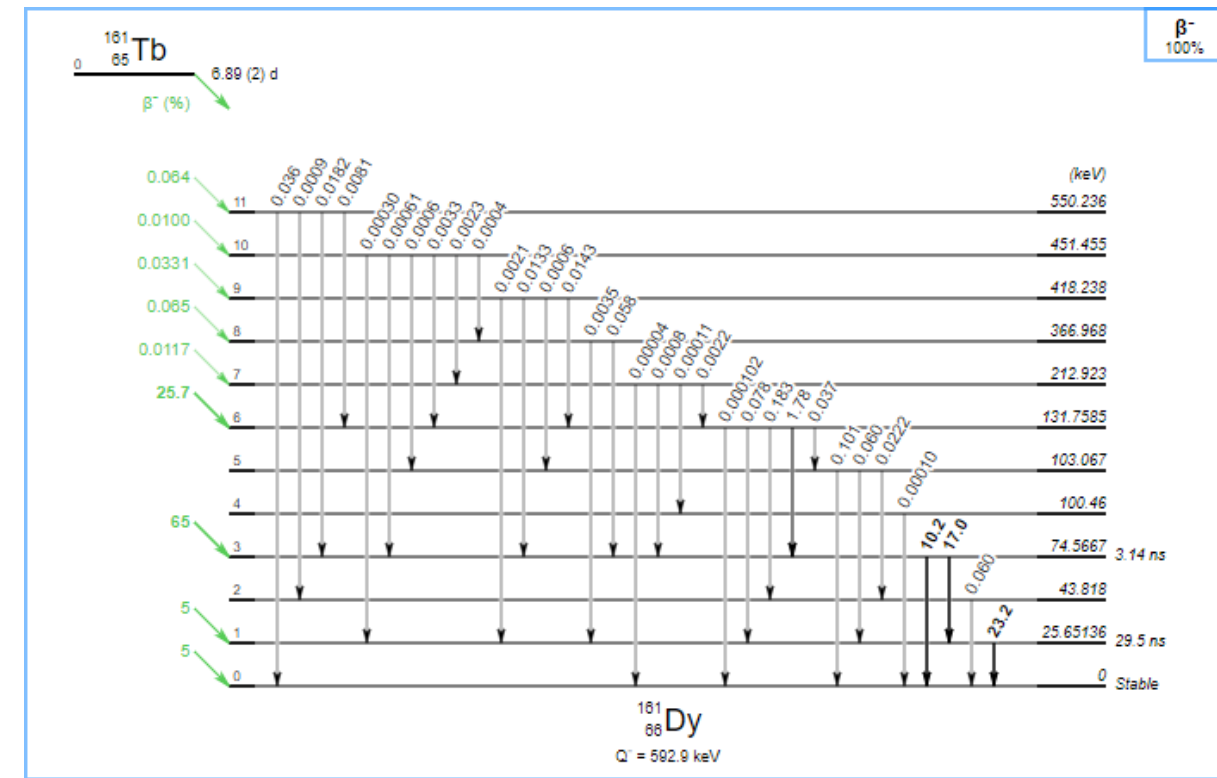
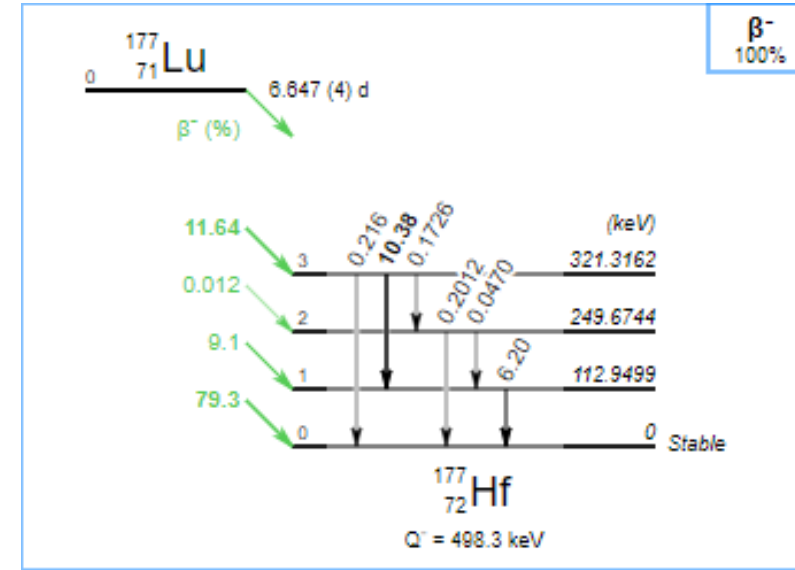
Decay Data: the importance and impact of metrology

Seán Collins

PRISMAP Radiolanthanides Workshop, 3 September 2024

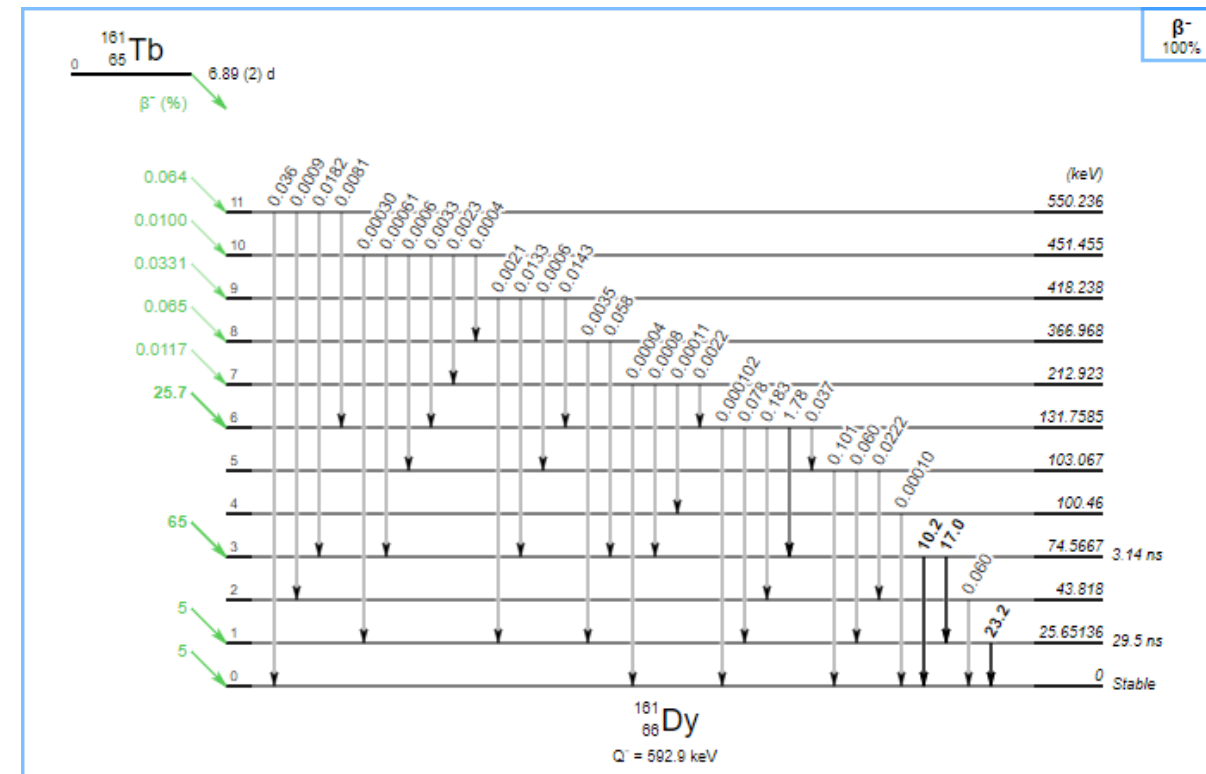
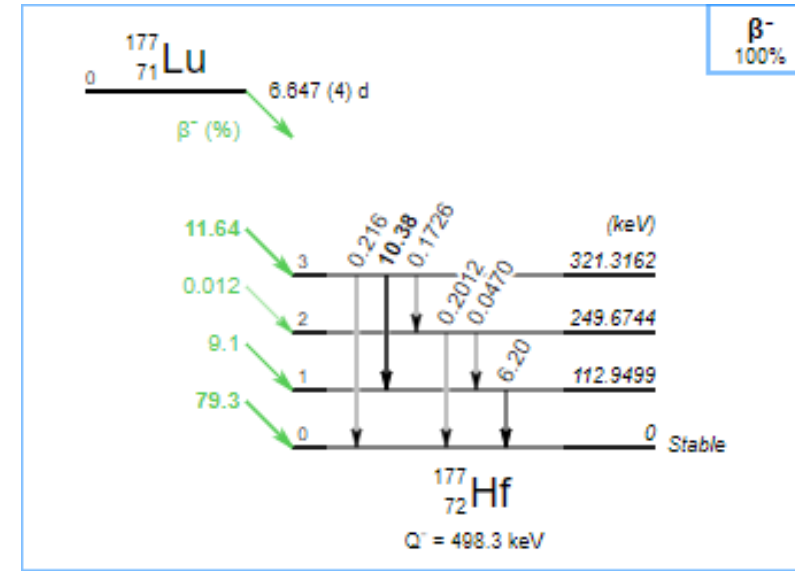
DECAY DATA

- A broad term that covers the unique values and arrangement associated with the decay process of a radionuclide
- Covers multiple parameters, such as:
 - Energy of excited states
 - Spin states (angular momentum)
 - Multipolarities
 - Emission probabilities
 - Half-lives



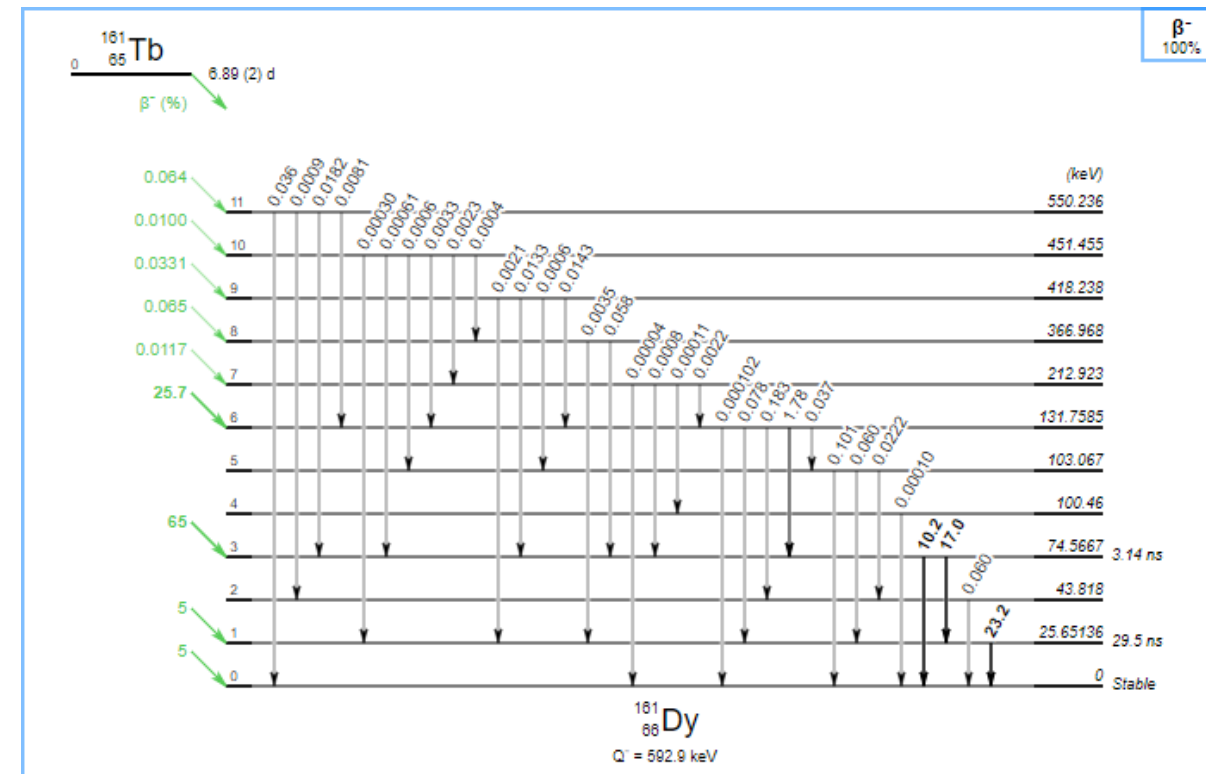
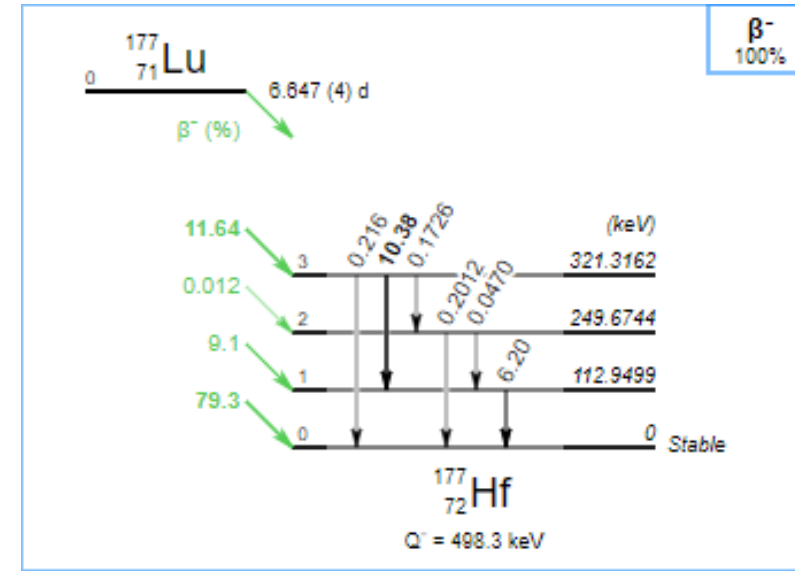
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- Atomic data – e.g., X-ray emission intensities, Auger-electron emission intensities



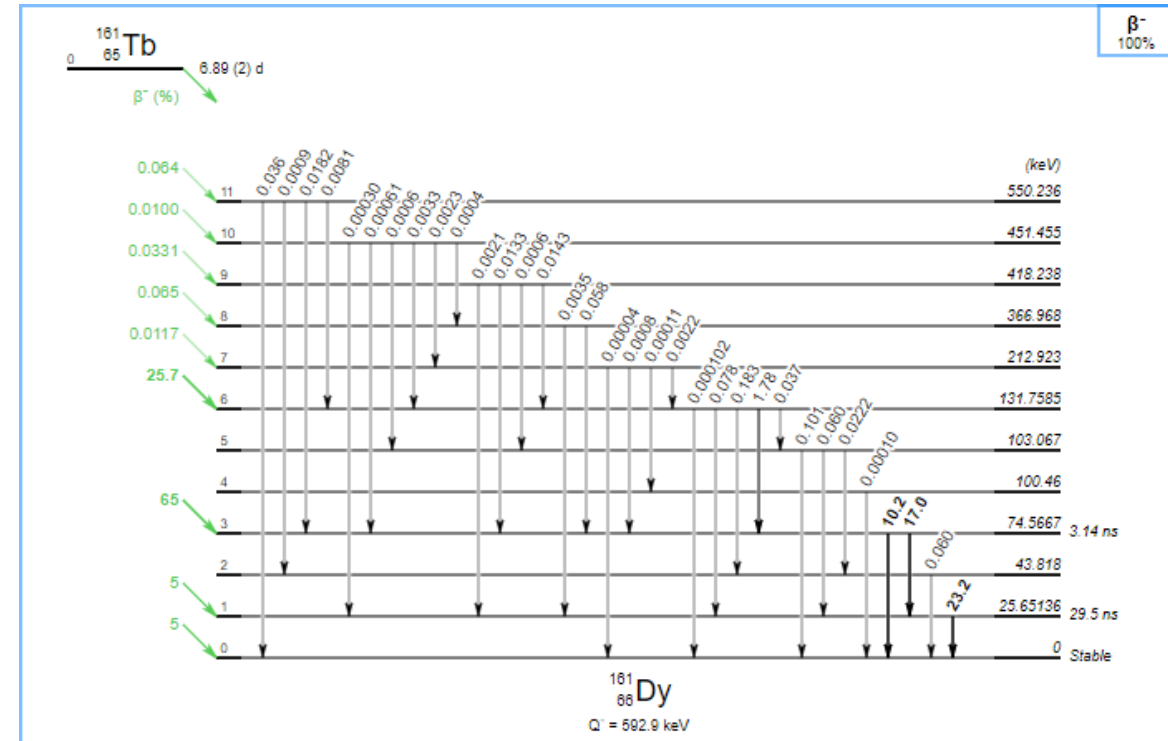
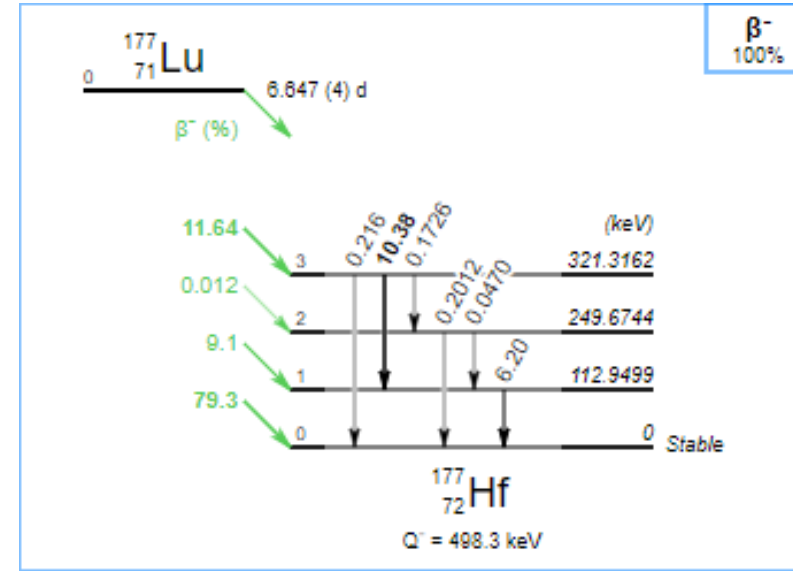
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- Cross-section data



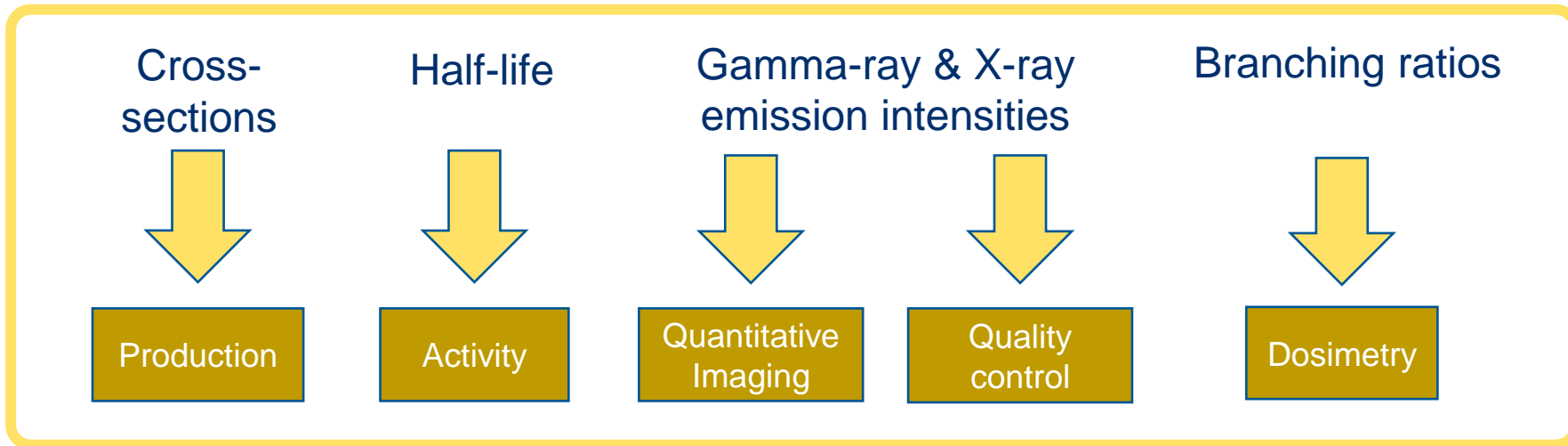
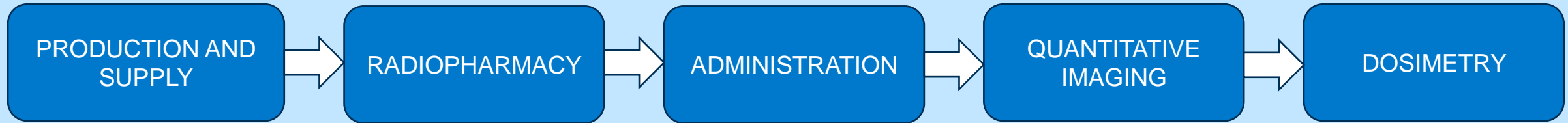
DECAY DATA

- Effectively the constants of radioactive decay of a radionuclide
- These are all measurable quantities!
- They have uncertainties!



IMPORTANCE IN NUCLEAR MEDICINE

CLINICAL RADIOPHARMACY PATHWAY



WHY CONFIDENCE IN DECAY DATA MATTERS



2010



Standardization of radium-223 by liquid scintillation counting

J.T. Cessna*, B.E. Zimmerman

Ionizing Radiation Division, Physics Laboratory, National Institute of Standards and Technology, 100 Bureau Drive MS 8462, Gaithersburg, MD 20899-8462, USA



2015



Standardisation of ^{223}Ra by liquid scintillation counting techniques and comparison with secondary measurements



John Keightley*, Andy Pearce, Andrew Fenwick, Sean Collins, Kelley Ferreira, Lena Johansson

National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, UK



2015

Volume 120 (2015) <http://dx.doi.org/10.6028/jres.120.004>
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10 % Revision of the NIST Standard for ^{223}Ra :
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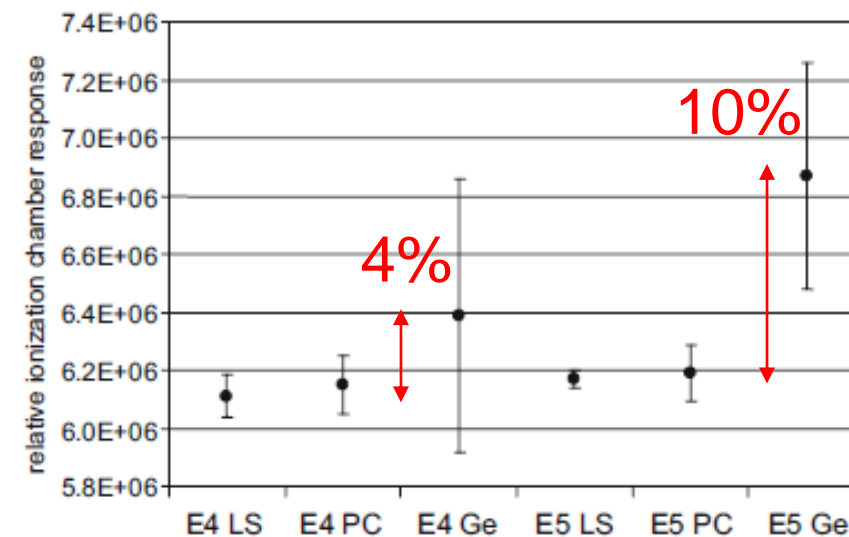


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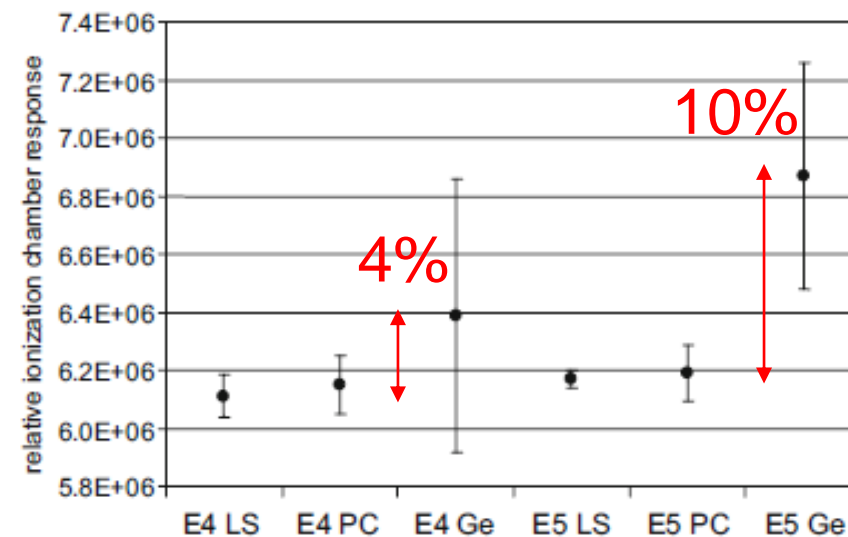


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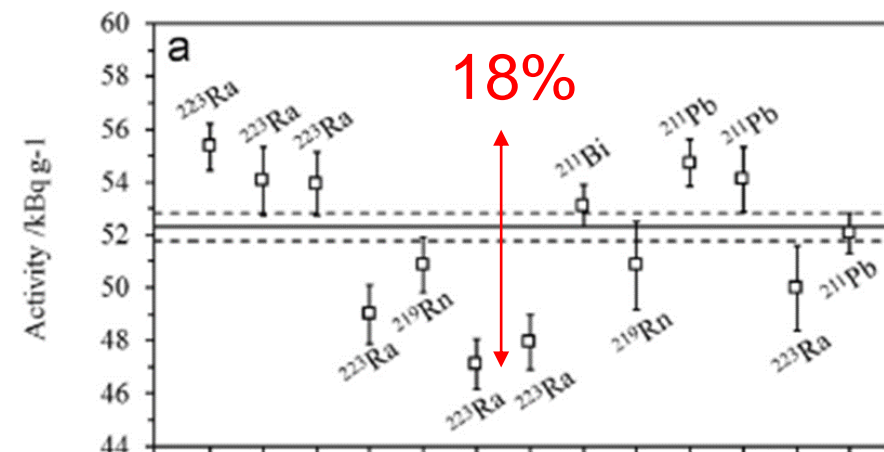
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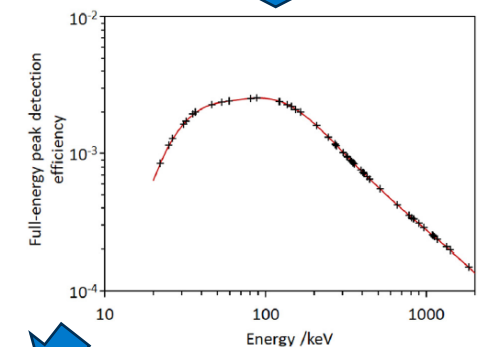
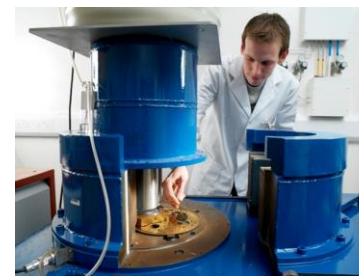
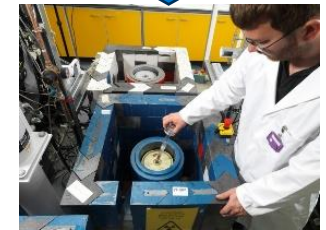
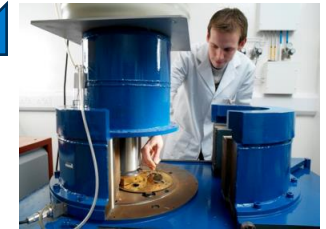
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Collins, S. M., et al. "Precise measurements of the absolute γ -ray emission probabilities of ^{223}Ra and decay progeny in equilibrium". *Applied Radiation and Isotopes* 102 (2015): 15-28.

METROLOGY UNDERPINNING DECAY DATA

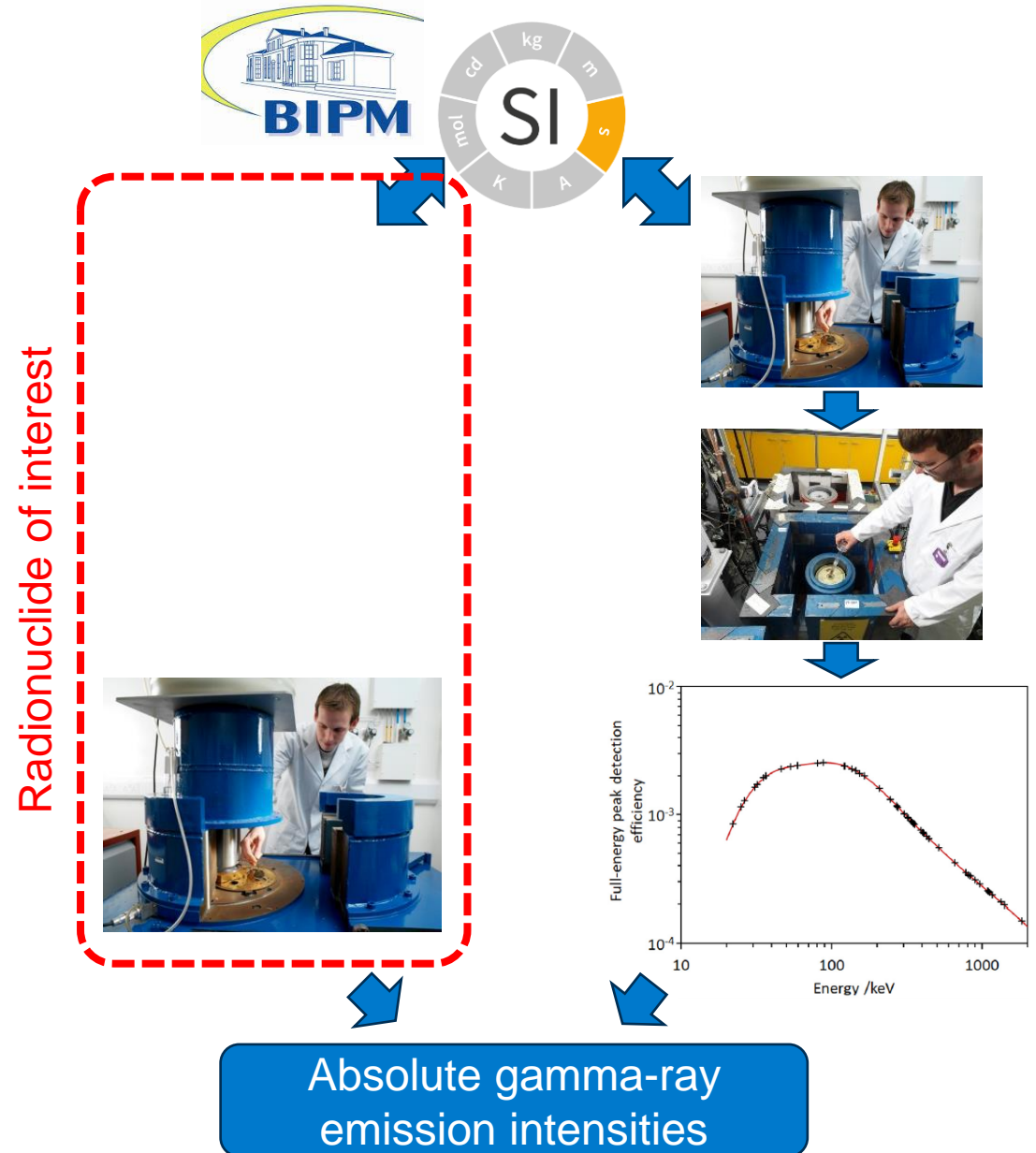
- Decay Data parameters are measurable quantities
- Traceability and standards can underpin these measurements



Absolute gamma-ray
emission intensities

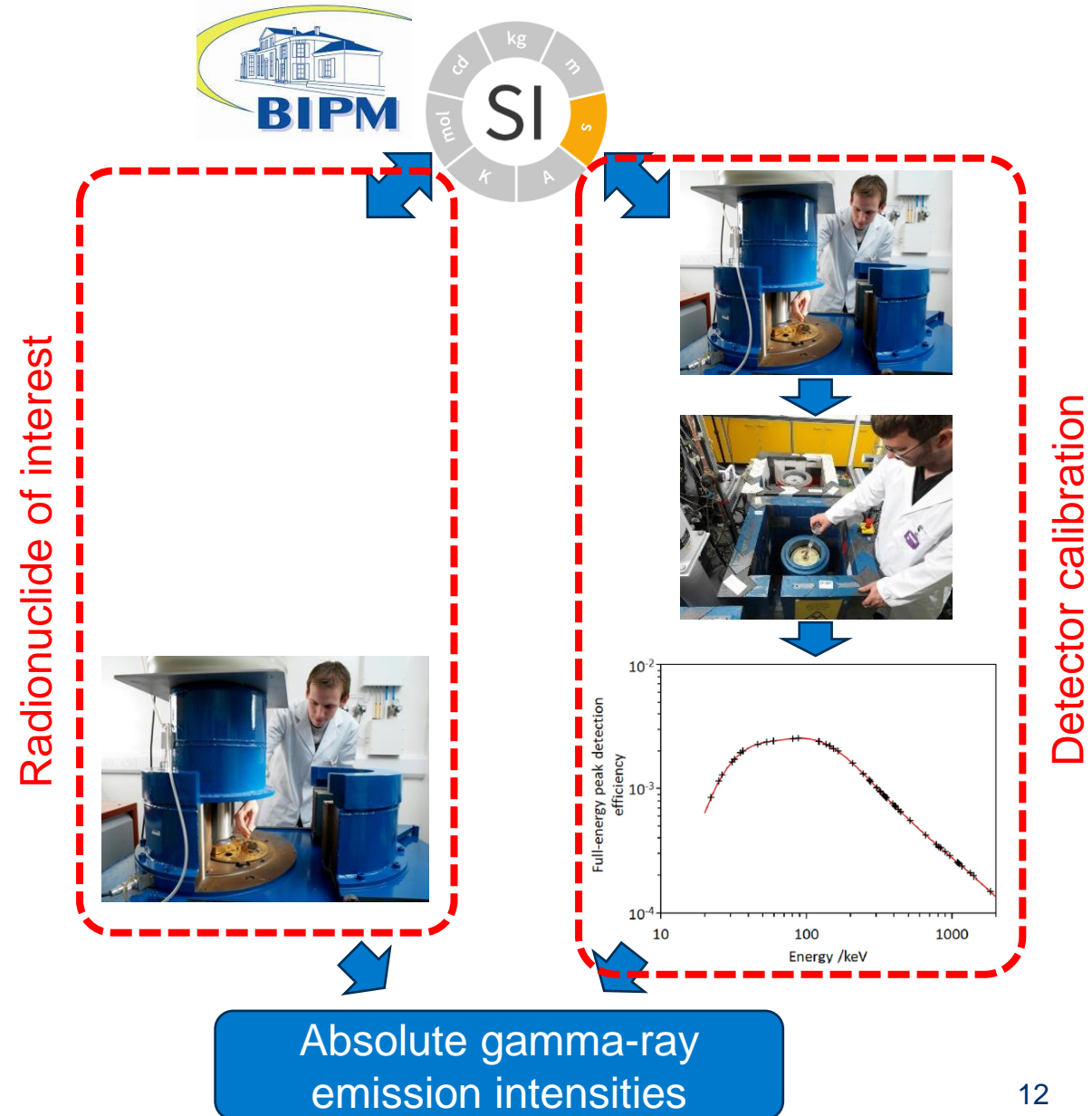
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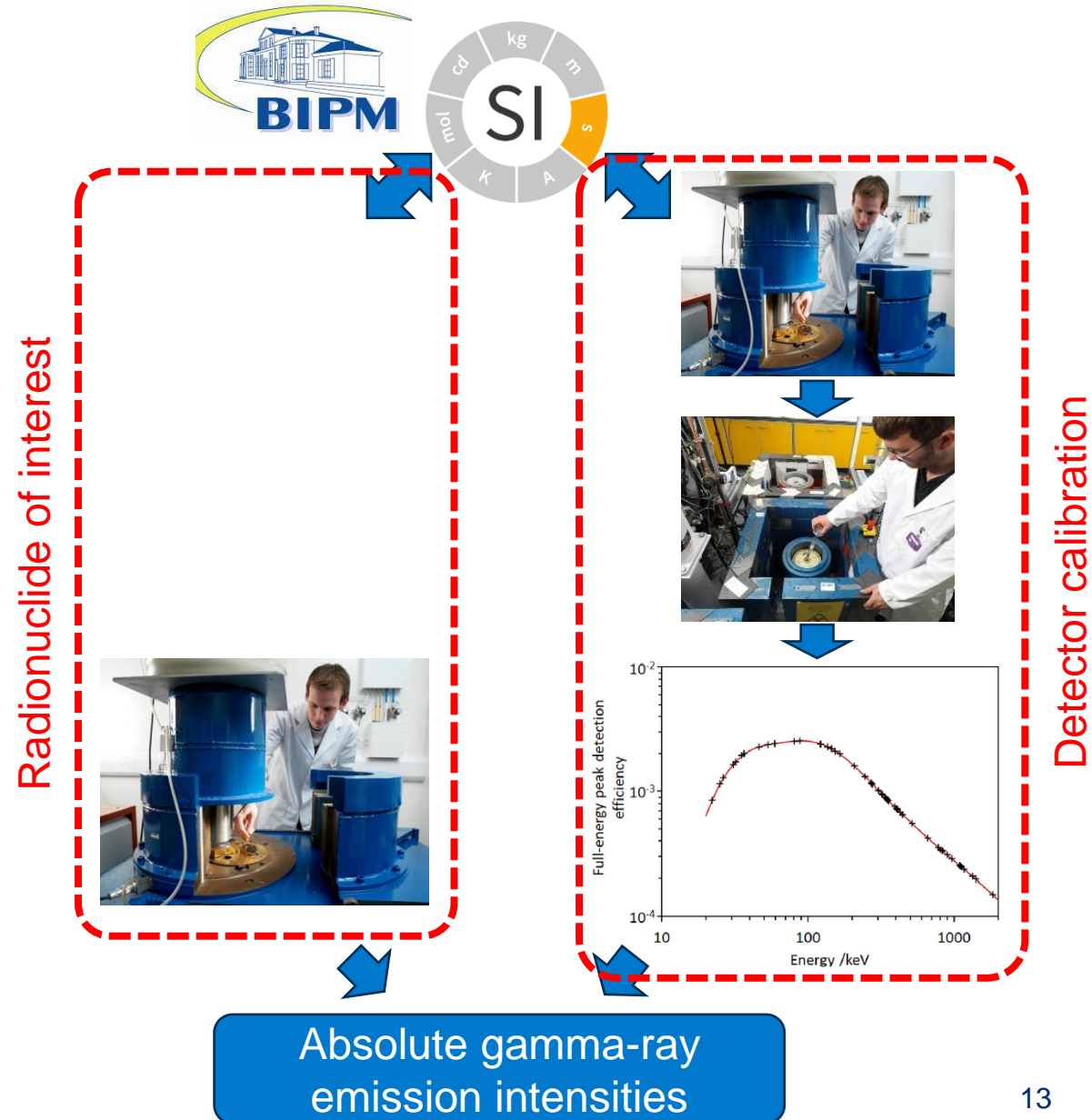
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METROLOGY UNDERPINNING DECAY DATA

- Decay Data parameters are measurable quantities
- Traceability and standards can underpin these measurements
- Good metrology requires more:
 - Quality control
 - Validation of methods
 - Comparison of techniques (or to other laboratories)
 - Robust uncertainty analysis



DECAY DATA EVALUATIONS

- Evaluated datasets provide the decay data that most use on a routine basis
- There are several databases of evaluated decay data, e.g.,
 - NNDC
 - Decay Data Evaluation Project (DDEP)
- Evaluators rely on the data being robust!



<https://www.nndc.bnl.gov/>



<http://www.lnhb.fr/home/nuclear-data/nuclear-data-table/>



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Half-life data—a critical review of TECDOC-619 update

M.J. Woods^{a,*}, S.M. Collins^b

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- 64 Radionuclides evaluated
- 48 recommendations for more measurements

Nuclide	Evaluated half-life (days)	Evaluated/required standard uncertainty	Observed/theoretical $\chi^2/(n-1)$	Number of data points		More measurements required?
				Used	Since 1968	
²² Na	950.57 (23)	0.84	2.15	3	3	Yes
²⁴ Na	0.62329 (9)	0.50	1.22	13	13	Yes
⁴⁰ K	4.563 (13) × 10 ¹¹	<0.01	1.13	3	2	Yes
⁴⁶ Sc	83.79 (4)	1.66	1.20	6	6	Yes
⁵¹ Cr	27.7009 (20)	0.25	0.62	6	6	No
⁵⁴ Mn	312.29 (26)	2.89	19.59	8	8	Yes
⁵⁵ Fe	1002.7 (23)	7.96	2.76	5	5	Yes
⁵⁶ Co	77.20 (8) d	3.60	0.90	5	5	Yes
⁵⁶ Mn	0.107449 (19)	0.61	5.29	6	6	Yes
⁵⁷ Co	271.80 (5)	0.64	1.24	6	6	Yes
⁵⁸ Co	70.86 (6)	2.94	1.19	6	6	Yes
⁵⁹ Fe	44.494 (13)	1.01	1.40	5	5	Yes
⁶⁰ Co	1925.23 (27)	0.49	0.09	5	5	No
⁶⁴ Cu	0.52929 (18)	1.18	0.96	9	9	Yes
⁶⁵ Zn	243.86 (20)	2.85	1.95	7	7	Yes
⁶⁶ Ga	0.3909 (25)	22.21	0.38	4	0	Yes
⁶⁷ Ga	3.2614 (4)	0.43	0.25	7	7	No
⁶⁸ Ga	0.04703 (7)	5.17	1.11	3	3	Yes
⁷⁵ Se	119.778 (29)	0.84	0.08	3	3	No



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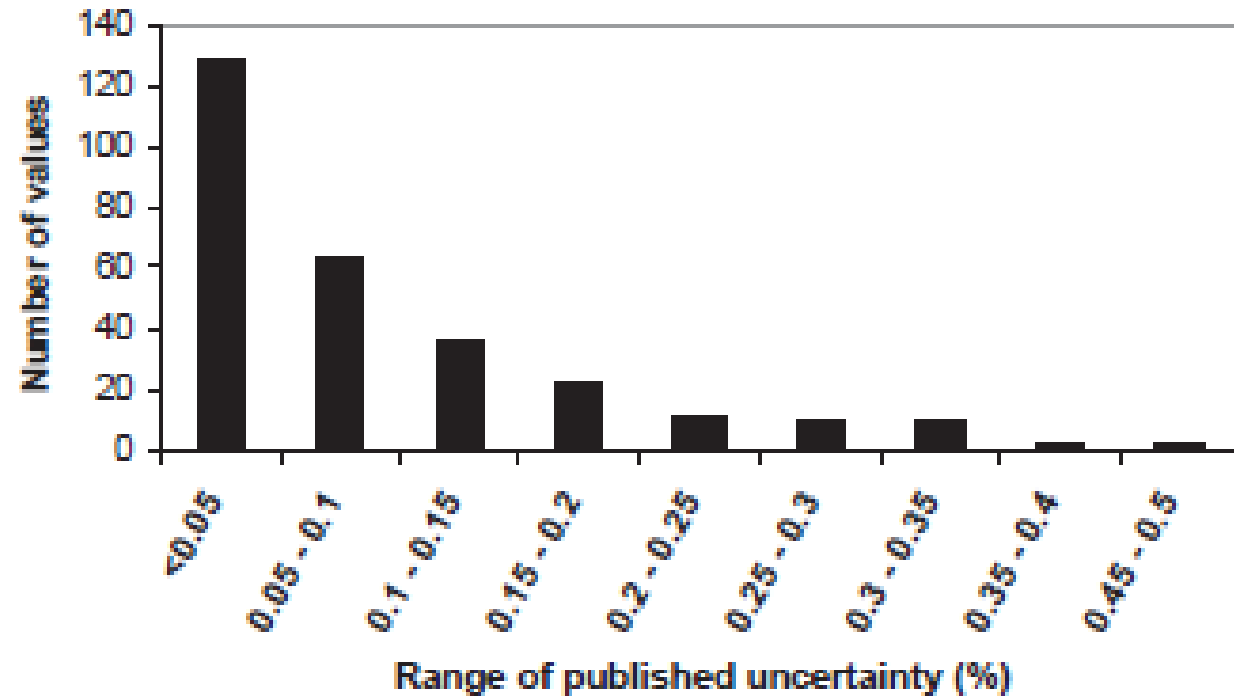


Fig. 1. Frequency of uncertainty range for published half-life values.

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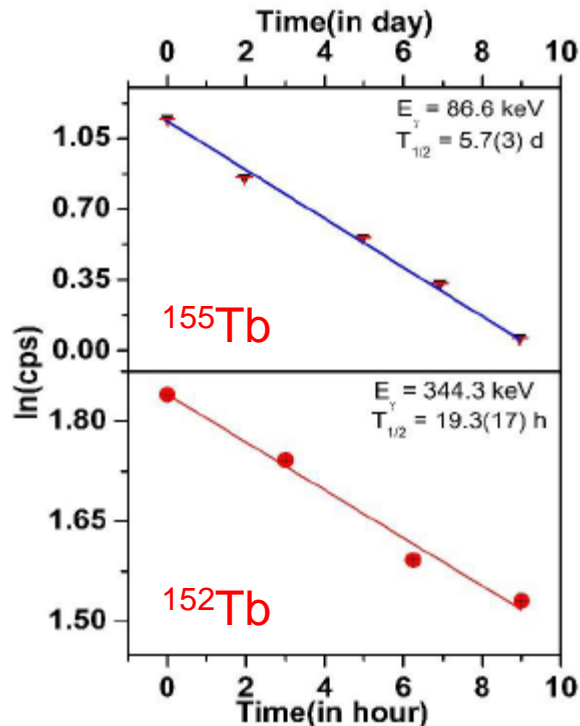


Fig. 2: Decay plots of ¹⁵²Tb and ¹⁵⁵Tb-isotopes.

Proceedings of the DAE Symp. on Nucl. Phys. 66 (2022) 184

Measurement of Decay Half Lives in Tb-Isotopes

A. Pal^{1,3*}, D. Banerjee², S. Basak^{1,3}, D. Kumar^{1,3} and T. Bhattacharjee^{1,3}

¹Variable Energy Cyclotron Centre, 1/AF Bidhan Nagar, Kolkata – 700 064, India
²Radiochemistry Division (BARC), VECC, 1/AF, Bidhan Nagar, Kolkata-700064, India
³Homi Bhabha National Institute, Mumbai, India, PIN – 400094

[*ag.pal@vecc.gov.in](mailto:ag.pal@vecc.gov.in)

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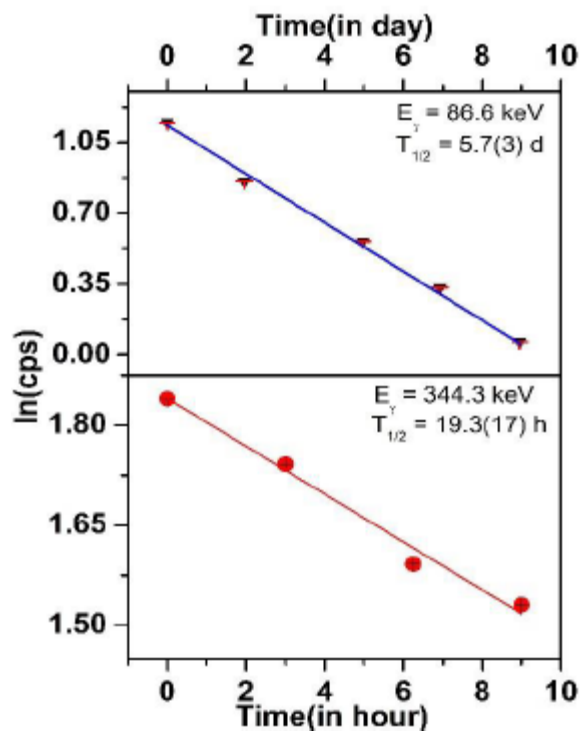
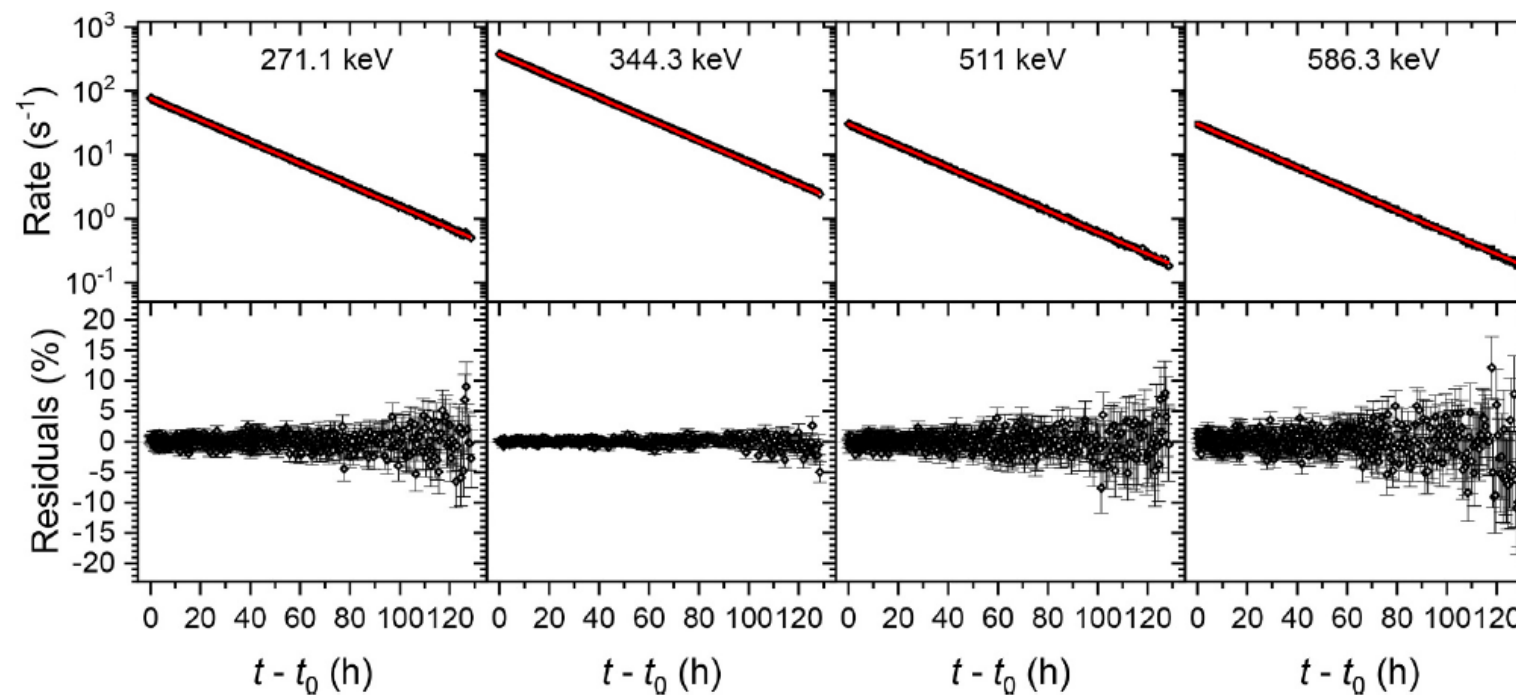


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The uncertainty of the half-life

S Pommé

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Abstract

Half-life measurements of radionuclides are undeservedly perceived as ‘easy’ and the experimental uncertainties are commonly underestimated. Data evaluators, scanning the literature, are faced with bad documentation, lack of traceability, incomplete uncertainty budgets and discrepant results. Poor control of uncertainties has its implications for the end-user community, varying from limitations to the accuracy and reliability of nuclear-based analytical techniques to the fundamental question whether half-lives are invariable or not. This paper addresses some issues from the viewpoints of the user community and of the decay data provider. It addresses the propagation of the uncertainty of the half-life in activity measurements and discusses different types of half-life measurements, typical parameters influencing their uncertainty, a tool to propagate the uncertainties and suggestions for a more complete reporting style. Problems and solutions are illustrated with striking examples from literature.

$$\frac{\sigma_{T_{1/2}}}{T_{1/2}} \approx \frac{2}{\lambda T} \sqrt{\frac{3(n-1)}{n(n+1)}} \frac{\sigma_A}{A}$$

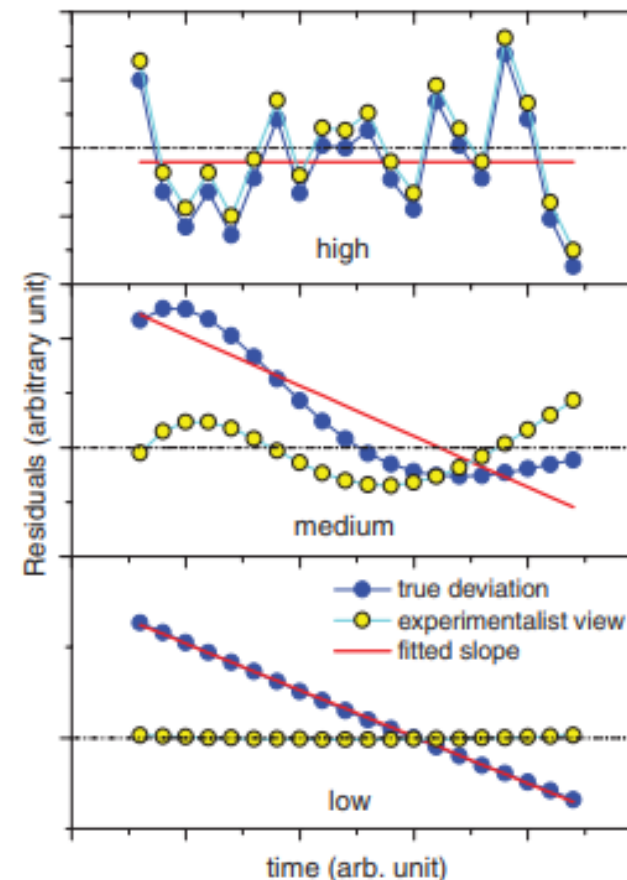
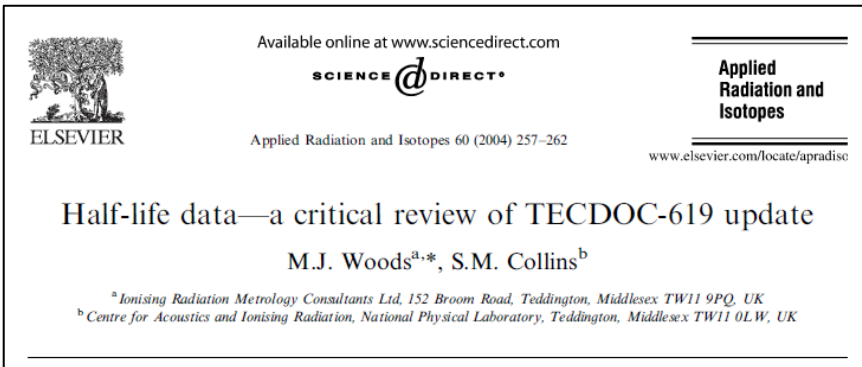


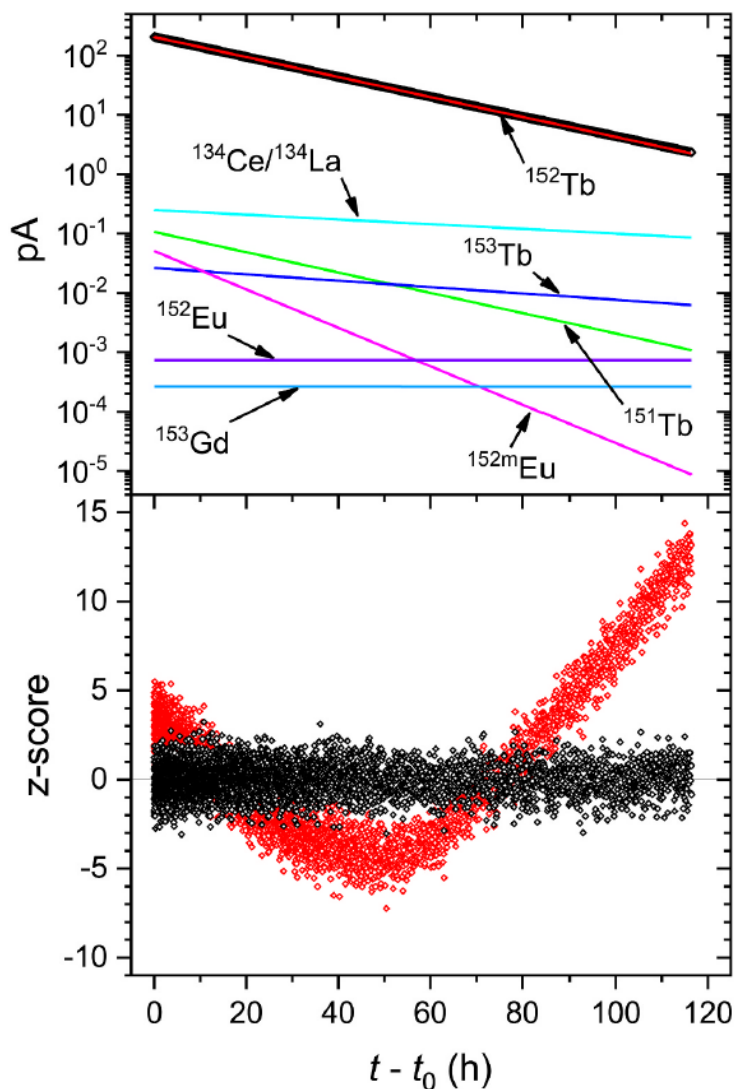
Figure 1. True (dark dots) and perceived (light dots) residuals from a fit of a decay curve through hypothetical data affected by high (top), medium (middle) and low (bottom) frequency instabilities. Systematic deviations are not fully observed by the experimentalist, as the fit tends to minimise them.



- 64 Radionuclides evaluated
- 48 recommendations for more measurements
- Many issues arising due to “precise” measurements disagreeing

Again, a closer examination of these new data is interesting. For many of the replacement data, the uncertainty quoted is larger than that previously published. This no doubt indicates that the relevant uncertainty budgets have been considered in more depth and more robustly than before. Some of these new data have uncertainties that are significantly larger than the rest of the data in the evaluation set for the radionuclide of concern. When the evaluation is conducted, the resulting weighting factor for these data mean that they have little or no significant impact on the final result. In essence, these values add little or nothing to the improvement in evaluated half-lives.

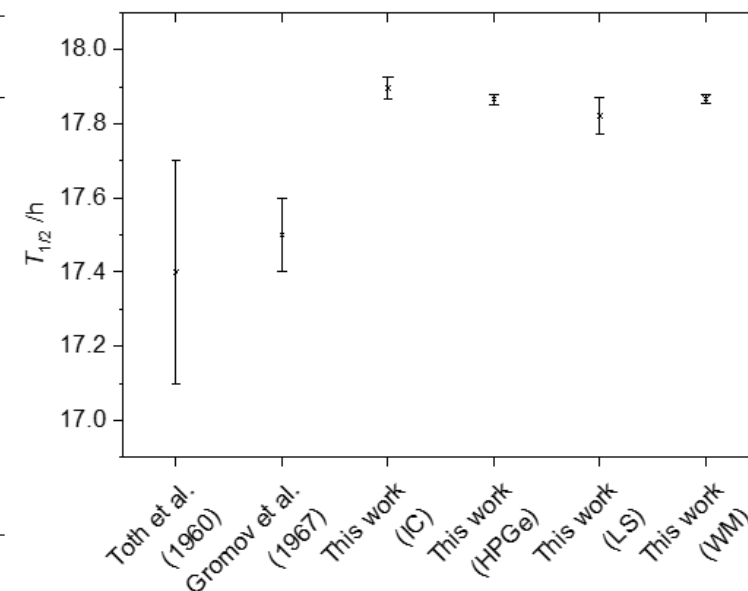
HALF-LIFE UNCERTAINTY MODEL – TB-152



$$\frac{\sigma_{T_{1/2}}}{T_{1/2}} \approx \frac{2}{\lambda T} \sqrt{\frac{3(n-1)}{n(n+1)}} \frac{\sigma_A}{A}$$

Uncertainty budget for the half-life of ^{152}Tb determined by the VINTEN 671 ionisation chamber.

Component	$u(A)/A$ (%)	n	Propagation Factor	$u(T_{1/2})/T_{1/2}$ (%)
Statistical (C_1)	–	–	–	0.0030
Medium-frequency (C_2)	–	–	–	0.0017
Linearity	0.10	1	0.8639	0.086
Stability	0.020	1	0.8639	0.017
Background	0.0052	1	0.8639	0.0045
Contaminant (^{151}Tb)	0.0078	1	0.8639	0.0067
Contaminant (^{153}Tb)	0.0045	1	0.8639	0.0039
Contaminant (^{134}Ce)	0.15	1	0.8639	0.13
Contaminant (^{152m}Eu)	0.0035	1	0.8639	0.0030
Contaminant (^{152}Eu)	0.00012	1	0.8639	0.00011
Contaminant (^{153}Gd)	0.00016	1	0.8639	0.00014
Combined standard uncertainty				0.16



Collins, S. M., et al. "Determination of the Terbium-152 half-life from mass-separated samples from CERN-ISOLDE and assessment of the radionuclide purity". *Applied Radiation and Isotopes* 202 (2023): 111044.

RA-223 AND DECAY PROGENY GAMMA-RAY EMISSION INTENSITIES

- Total of 148 gamma-rays from the decay series
- No previous absolute gamma-ray emission measurements just normalised values available
- Absolute emission intensities calculated from decay scheme
- Gamma-ray spectrometry showed agreement with the primary techniques.....so no problem?



Standardisation of ^{223}Ra by liquid scintillation counting techniques and comparison with secondary measurements

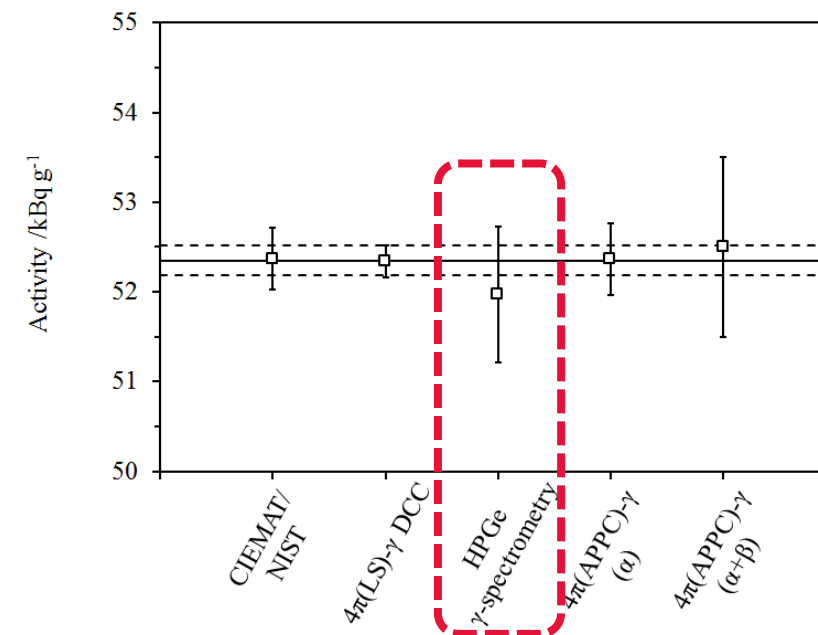


John Keightley*, Andy Pearce, Andrew Fenwick, Sean Collins, Kelley Ferreira, Lena Johansson

National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0BW, UK

HIGHLIGHTS

- An aqueous solution of ^{223}Ra chloride was standardised by liquid scintillation counting.
- CIEMAT/NIST efficiency tracing and Digital Coincidence Counting were utilised.
- Calibration factors for a variety of radionuclide calibrators were calculated.
- A discrepancy of around 9% was identified utilising existing published calibration factors.
- γ -spectrometry measurements exhibited a large spread (18.3%) in the individual activity estimations using published γ -emissions.



RA-223 AND DECAY PROGENY GAMMA-RAY EMISSION INTENSITIES

- 18% range in the activities determined from the main gamma-ray emissions
- No confidence in this data

Applied Radiation and Isotopes 102 (2015) 15–28



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Precise measurements of the absolute γ -ray emission probabilities of ^{223}Ra and decay progeny in equilibrium



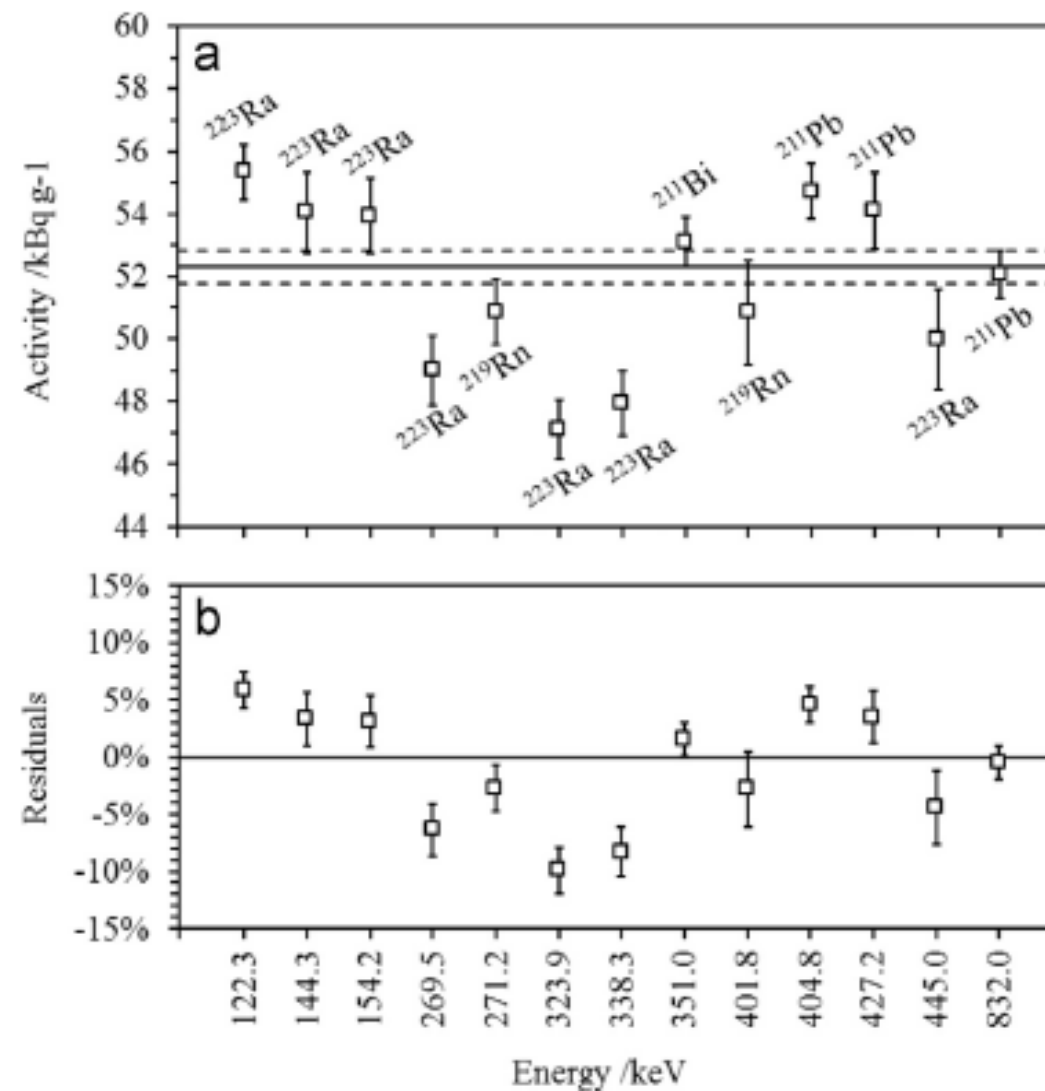
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HIGHLIGHTS


- Discrepancies found within currently published γ -ray emission probabilities.
- Absolute γ -ray emission probabilities of decay series in equilibrium determined.
- Significant improvement in precision of measured values.
- Closer agreement between deduced and experimental α transition probabilities.
- Correlation coefficients presented for γ -emissions of ^{223}Ra , ^{219}Rn and ^{211}Pb .




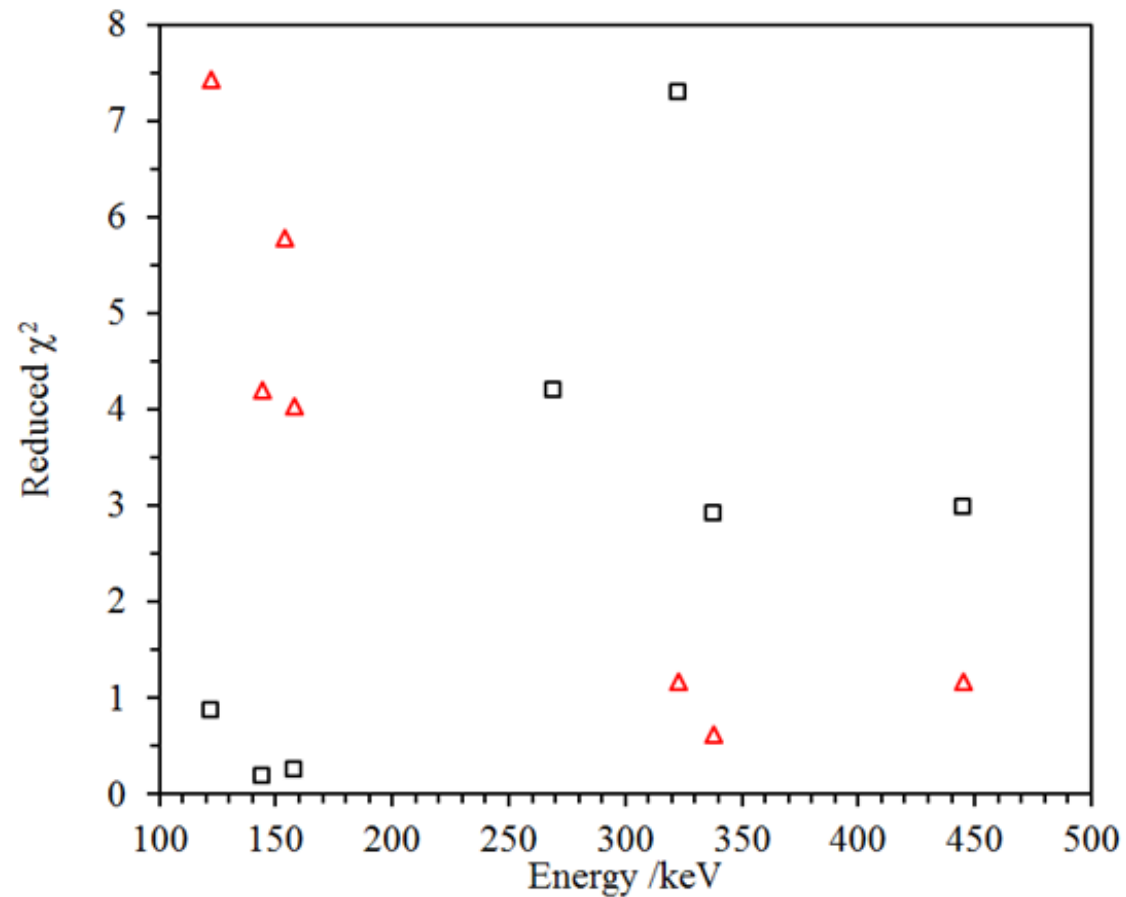
COMPARISON TO DDEP RECOMMENDED VALUES.

Radionuclide	Energy /keV	I_γ (This work) %	I_γ (DDEP) %	z-score	Difference %
²²³ Ra	269.5	13.37 (7)	14.23 (32)	-2.6	-6.0
²¹⁹ Rn	271.2	10.75 (6)	11.07 (22)	-1.4	-2.9
²¹⁵ Po	438.8	0.0533 (7)	0.058 (19)	-0.3	-8.1
²¹¹ Pb	404.8	4.011 (9)	3.83 (6)	3.0	4.7
²¹¹ Bi	351.0	13.17 (7)	13.00 (19)	0.8	1.3
²⁰⁷ Tl	897.7	0.2725 (15)	0.263 (9)	1.0	3.6

COMPARISON OF RA-223 RESULTS


 normalised to the 154.2 keV γ -ray emission

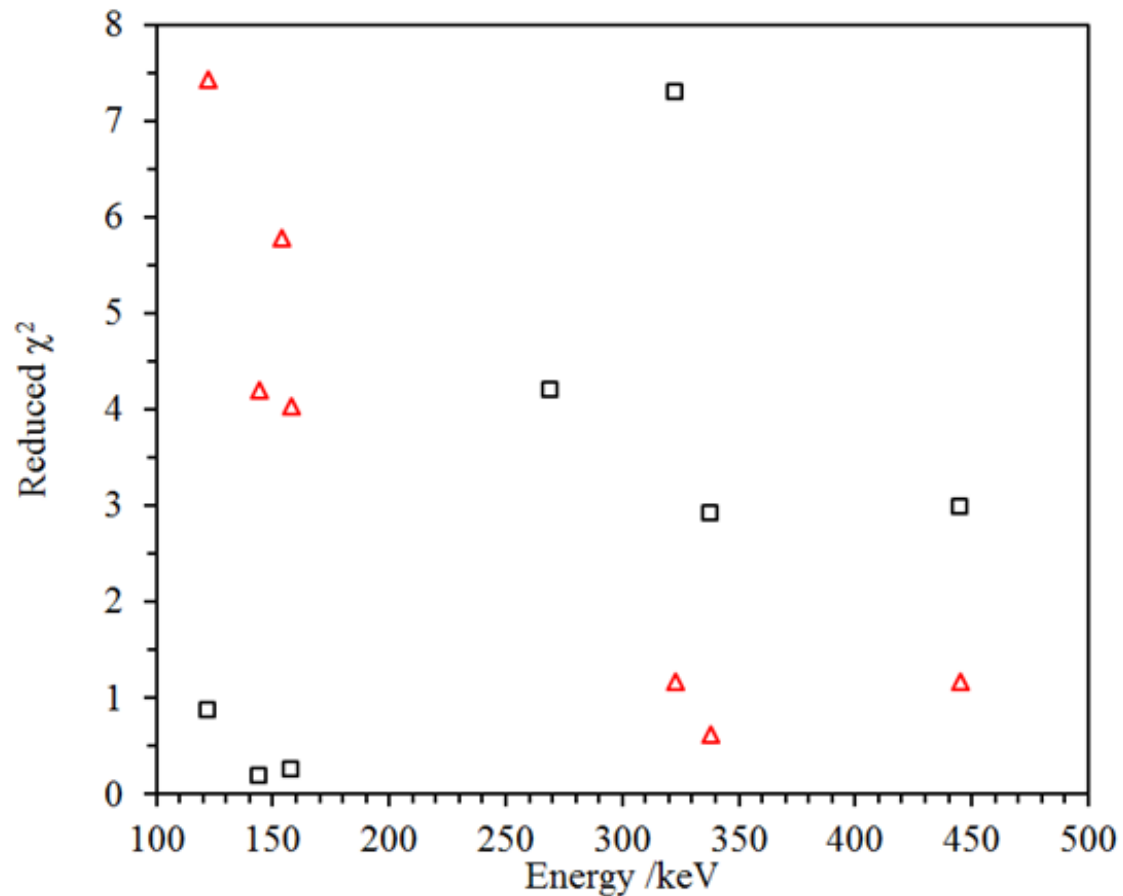
 normalised to the 269.5 keV γ -ray emission



COMPARISON OF RA-223 RESULTS

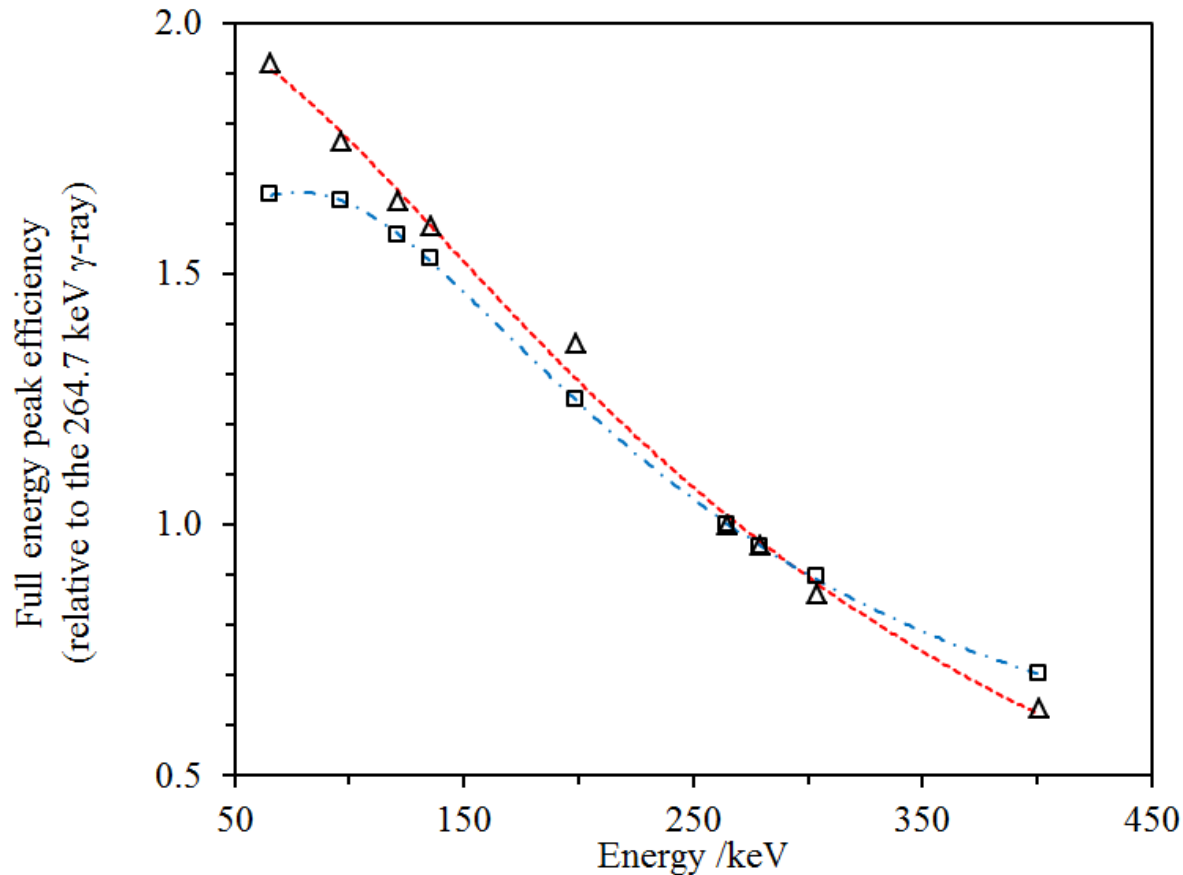
 normalised to the 154.2 keV γ -ray emission



 normalised to the 269.5 keV γ -ray emission



- There appears to be a ‘switching’ point in the discrepancies.
- What could cause this?
- Errors in efficiency calibrations?

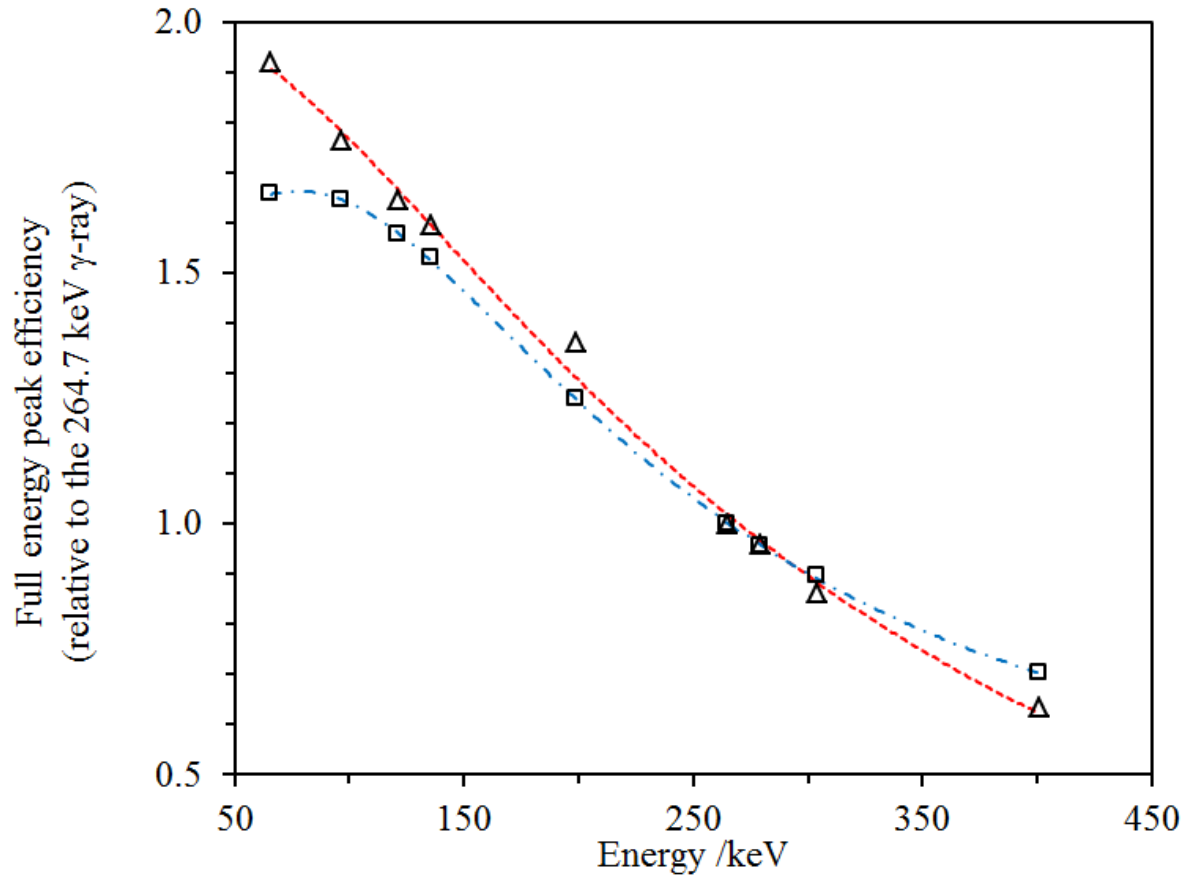
COMPARISON OF ^{75}Se DECAY DATA



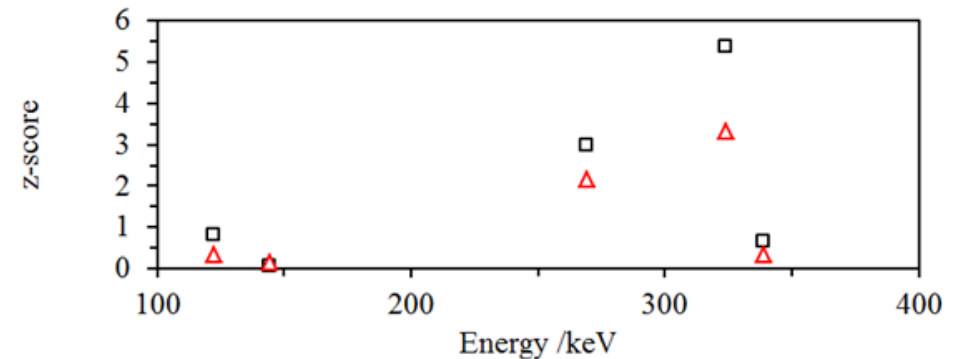
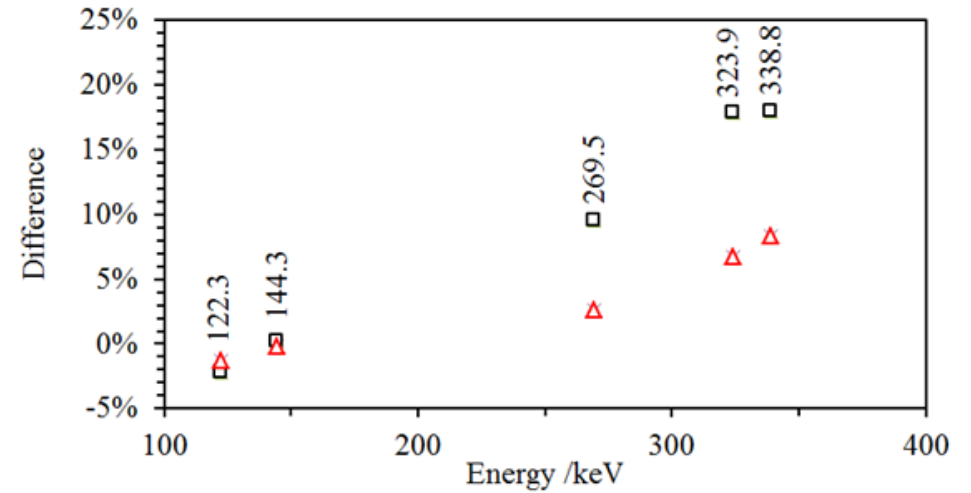
 Krien *et al.* (1970).  ENSDF (2013)

- Only one publication (before 2015) provides any detector calibration details
- This also happens to be the one that quoted the best uncertainties
- Krien et al. (1970) used ^{75}Se to determine the full-energy peak efficiency calibration.
 - The γ -ray emission probabilities used at the time are provided.
 - These are significantly different from the recommended values now.
- This allowed us to do some investigation work....

COMPARISON OF ^{75}Se DECAY DATA



△ Krien *et al.* (1970).
 □ ENSDF (2013)



□ Before
 △ After

RA-223 AND DECAY PROGENY GAMMA-RAY EMISSION INTENSITIES



Absolute emission intensities

Review
Realization and dissemination of activity standards for medically important alpha-emitting radionuclides
Denis E. Bergeron^{a,*}, Karsten Kossert^b, Sean M. Collins^{c,d}, Andrew J. Fenwick^e

Energy /keV	Radionuclide	DDEP (Bé et al., 2011) /I _γ	Pibida et al. (2015) (I _{ref} /I _{DDEP})/%	Collins et al. (2015a)	Kossert et al. (2015b)	Marouli et al. (2019)	Simões et al. (2021)	χ ² / (ν-1)
122.3	²²³ Ra	1.238(19)	5.0	6.0	5.3	12.3	-1.0	2.1
144.3	²²³ Ra	3.36(8)	4.5	3.6	3.2	10.4	-1.2	2.7
154.2	²²³ Ra	5.84(13)	4.1	3.1	3.3	10.1	-0.2	2.5
269.5	²²³ Ra	14.23(32)	-7.0	-6.0	-7.5	-0.9	-1.5	5.9
271.2	²¹⁹ Rn	11.07(22)	-3.4	-2.9	-1.8	2.1	-4.0	1.1
323.9	²²³ Ra	4.06(8)	-10.6	-10.0	-9.8	-5.2	-6.2	2.4
338.3	²²³ Ra	2.85(6)	-9.1	-8.6	-8.3	-3.2	0.0	6.6
351.0	²¹¹ Bi	13.00(19)	0.9	1.3	1.9	6.9	3.62	1.2
401.8	²¹⁹ Rn	6.75(22)	-2.8	-2.7	-1.9	3.4	-2.1	0.9
404.8	²¹¹ Pb	3.83(6)	4.7	4.7	5.7	9.7	10.2	0.9
427.2	²¹¹ Pb	1.81(4)	4.4	4.4	5.6	8.3	0.0	1.8
445.0	²²³ Ra	1.28(4)	-4.9	-4.8	-4.5	0.0	0.8	0.6
832.0	²¹¹ Pb	3.5(5)	-0.6	-1.5	-2.0	6.3	0.6	1.8

Pibida, L., Zimmerman, B., Fitzgerald, R., King, L., Cessna, J.T., Bergeron, D.E., 2015. Determination of photon emission probabilities for the main gamma-rays of ²²³Ra in equilibrium with its progeny. Appl. Radiat. Isot. 101, 15–19.

Collins, S.M., Pearce, A.K., Regan, P.H., Keightley, J.D., 2015. Precise measurements of the absolute γ-ray emission probabilities of ²²³Ra and decay progeny in equilibrium. Appl. Radiat. Isot. 102, 15–18.

Kossert, K., Bokeloh, K., Dersch, R., N'ahle, O.J., 2015. Activity determination of ²²⁷Ac and ²²³Ra by means of liquid scintillation counting and determination of nuclear decay data. Appl. Radiat. Isot. 95, 143–152.

Marouli, M., Lutter, G., Pommé, S., Van Ammel, R., Hult, M., Pierre, S., Dry'ak, P., Carconi, P., Fazio, A., Bruchertseifer, F., Morgenstern, A., 2019. Measurement of absolute γ-ray emission probabilities in the decay of ²²⁷Ac in equilibrium with its progeny. Appl. Radiat. Isot. 144, 34–46.

Simoes, R.F.P., da Silva, C.J., da Silva, R.L., de S'á, L.V., Poledna, R., de Oliveira, A.E., Iwahara, A., da Cruz, P.A.L., Delgado, J.U., 2021. Standardization of ²²³Ra by live-time anticoincidence counting and gamma-ray emission determination. Appl. Radiat. Isot. 170, 109559.

RA-223 AND DECAY PROGENY GAMMA-RAY EMISSION INTENSITIES



Review

Realization and dissemination of activity standards for medically important alpha-emitting radionuclides

Denis E. Bergeron ^{a,*}, Karsten Kossert ^b, Sean M. Collins ^{c,d}, Andrew J. Fenwick ^e

Relative emission intensities

Energy /keV	Radionuclide	Pibida et al. (2015) $(I_{\gamma}/I_{\gamma,154 \text{ keV}})/100$ decays	Collins et al. (2015)	Kossert et al. (2015)	Marouli et al. (2019)	Simões et al. (2021)	$\chi^2/(\nu-1)$
122.3	²²³ Ra	21.38(27)	21.79(15)	21.63(27)	21.6(12)	21.03(68)	0.7
144.3	²²³ Ra	57.73(75)	57.82(39)	57.53(58)	57.7(31)	56.95(13)	0.1
269.5	²²³ Ra	217.8(29)	222.1(16)	218.2(31)	219(11)	240.5(42)	5.8
271.2	²¹⁹ Rn	175.8(24)	178.6(13)	180.3(25)	175.7(88)	182.3(32)	0.8
323.9	²²³ Ra	59.70(67)	60.71(43)	60.71(61)	59.9(34)	65.4(14)	3.5
338.3	²²³ Ra	42.60(53)	43.27(31)	43.35(42)	42.9(22)	48.9(11)	7.0
351.0	²¹¹ Bi	215.6(26)	218.8(16)	219.6(21)	216(11)	231.1(51)	1.9
401.8	²¹⁹ Rn	107.9(13)	109.14(74)	109.8(11)	108.6(56)	113.4(29)	0.9
404.8	²¹¹ Pb	65.95(82)	66.63(46)	67.2(10)	65.3(33)	72.4(29)	1.3
427.2	²¹¹ Pb	31.09(35)	31.40(22)	31.71(31)	30.5(15)	31.05(96)	0.6
445.0	²²³ Ra	20.02(24)	20.23(14)	20.27(20)	19.9(11)	22.1(14)	0.7
832.0	²¹¹ Pb	57.24(75)	57.28(39)	56.88(55)	57.9(30)	60.4(15)	1.3

Pibida, L., Zimmerman, B., Fitzgerald, R., King, L., Cessna, J.T., Bergeron, D.E., 2015. Determination of photon emission probabilities for the main gamma-rays of ²²³Ra in equilibrium with its progeny. *Appl. Radiat. Isot.* 101, 15–19.

Collins, S.M., Pearce, A.K., Regan, P.H., Keightley, J.D., 2015. Precise measurements of the absolute γ -ray emission probabilities of ²²³Ra and decay progeny in equilibrium. *Appl. Radiat. Isot.* 102, 15–18.

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RA-223 AND DECAY PROGENY GAMMA-RAY EMISSION INTENSITIES



Review
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 Denis E. Bergeron ^{a,*}, Karsten Kossert ^b, Sean M. Collins ^{c,d}, Andrew J. Fenwick ^e

Energy	DDEP (Bé et al., 2011)	ENSDF (Singh et al., 2021)	I_{DDEP}/I_{ENSDF}	Precision improvement factor ($u(I_{DDEP})/u(I_{ENSDF})$)
/keV	I_{γ} /per 100 decay	I_{γ} /per 100 decay	/%	
122.3	1.238(19)	1.3045(93)	5.4	2.1
144.3	3.36(8)	3.474(25)	3.4	3.2
154.2	5.84(13)	6.020(57)	3.1	2.3
269.5	14.23(32)	13.304(94)	-6.5	3.4
323.9	4.06(8)	3.642(26)	-10.3	3.1
338.3	2.85(6)	2.601(18)	-8.7	3.3
445.0	1.28(4)	1.2184(86)	-4.8	4.7

CONCLUSION

- Decay data has an important role in nuclear medicine
- ‘Good’ decay data relies on ‘good’ metrology practice
- Robust uncertainty analysis is key
- Historical data should be treated with caution
- Poor data in = Poor data out



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