NPL
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Decay Data for Alpha-emitters for medical applications

Seán Collins

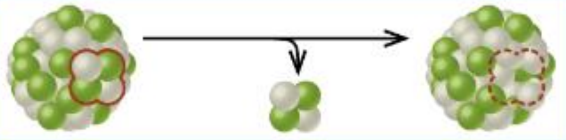
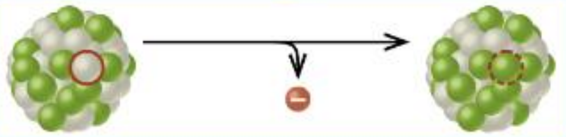
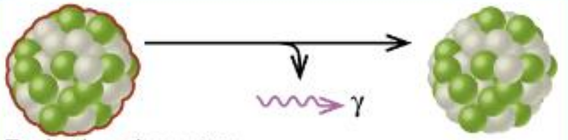
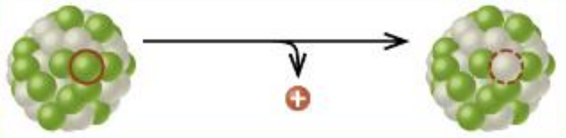
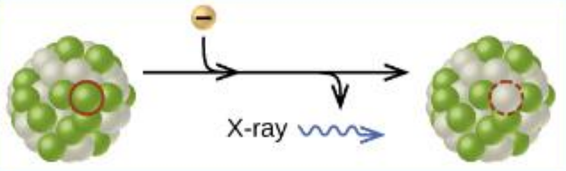
Workshop on Standards and Measurements for Alpha Emitting Nuclides in Therapeutic Nuclear Medicine,
22-23 February 2024

Radioactive decay

- Radioactive decay is the stochastic process by which an unstable atomic nucleus undergoes spontaneous transformation into one or more daughter nuclei

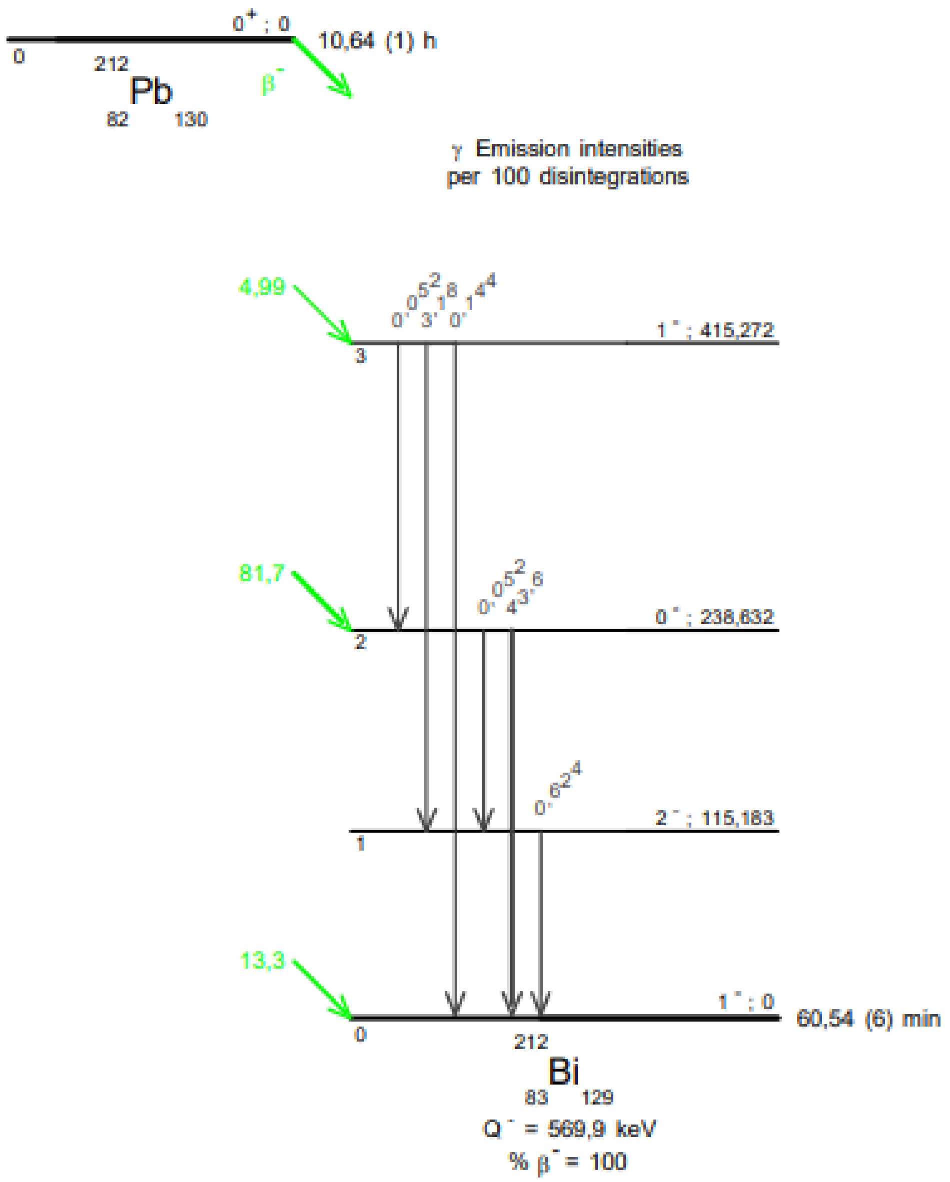
- Excess binding energy is removed via the emission of alpha- or beta-particles, and/or electromagnetic waves (gamma rays, X-rays) or by nuclear fission.

- This excess energy depends on the difference in mass of the parent atom and the daughter atom plus the particle emission (Q-value).

Type	Nuclear equation	Representation	Change in mass/atomic numbers
Alpha decay	${}^{A}_{Z}X \quad {}^{4}_{2}He + {}^{A-4}_{Z-2}Y$		A: decrease by 4 Z: decrease by 2
Beta decay	${}^{A}_{Z}X \quad {}^{0}_{-1}e + {}_{Z+1}^{A}Y$		A: unchanged Z: increase by 1
Gamma decay	${}^{A}_{Z}X \quad {}^{0}_{0}\gamma + {}^{A}_{Z}Y$	 Excited nuclear state	A: unchanged Z: unchanged
Positron emission	${}^{A}_{Z}X \quad {}^{0}_{+1}e + {}_{Y-1}^{A}Y$		A: unchanged Z: decrease by 1
Electron capture	${}^{A}_{Z}X \quad {}^{0}_{-1}e + {}_{Y-1}^{A}Y$		A: unchanged Z: decrease by 1

Radioactive Decay

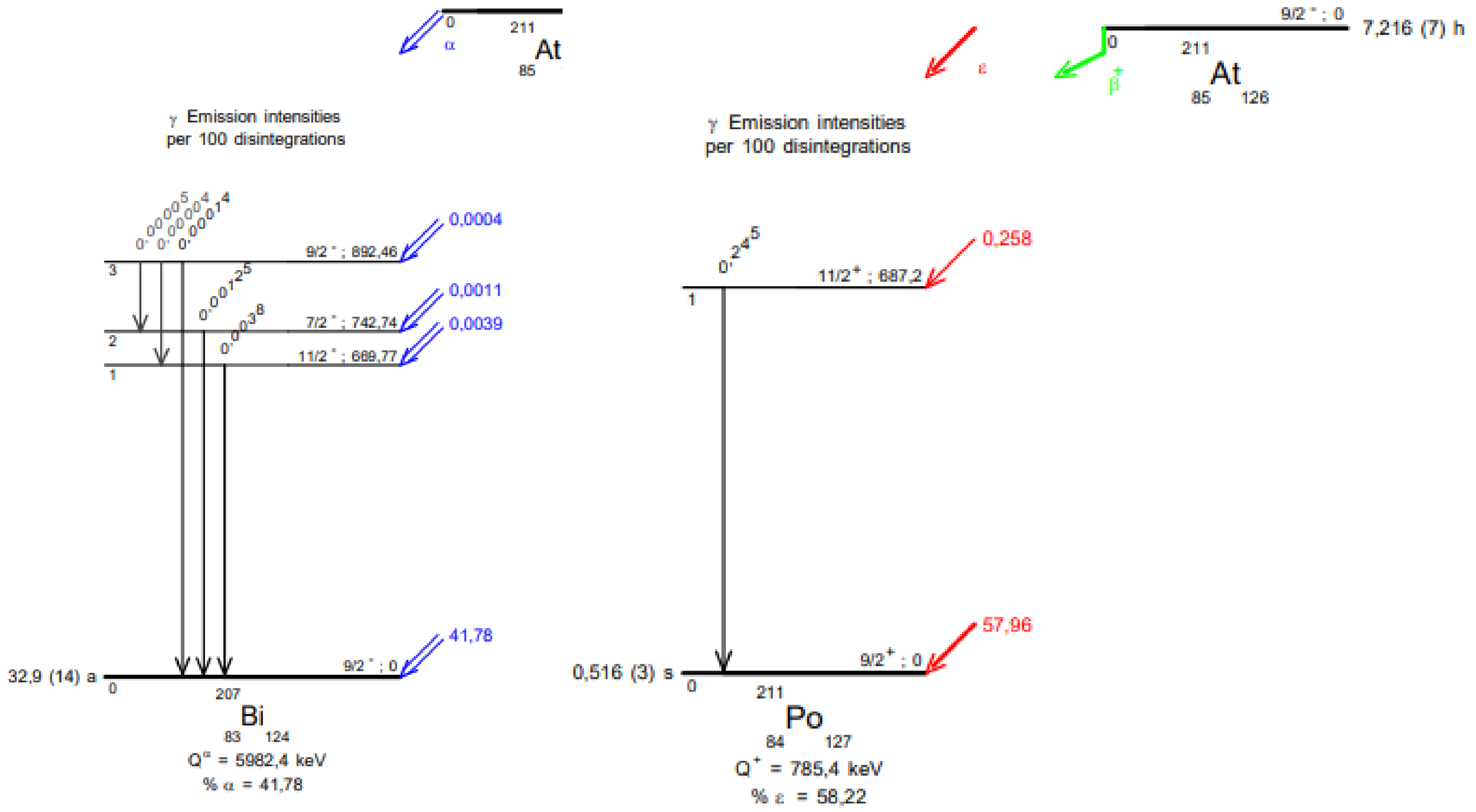
- Each radionuclide undergoes its own unique process to lose this excess binding energy
- This process could be achieved through multiple pathways
- Though the decay of a single atom will only go through one
- This process is illustrated by its “Decay scheme”



Radioactive Decay

- Some radionuclides can decay by more than one type of decay process

- Which can have different decay branch probabilities

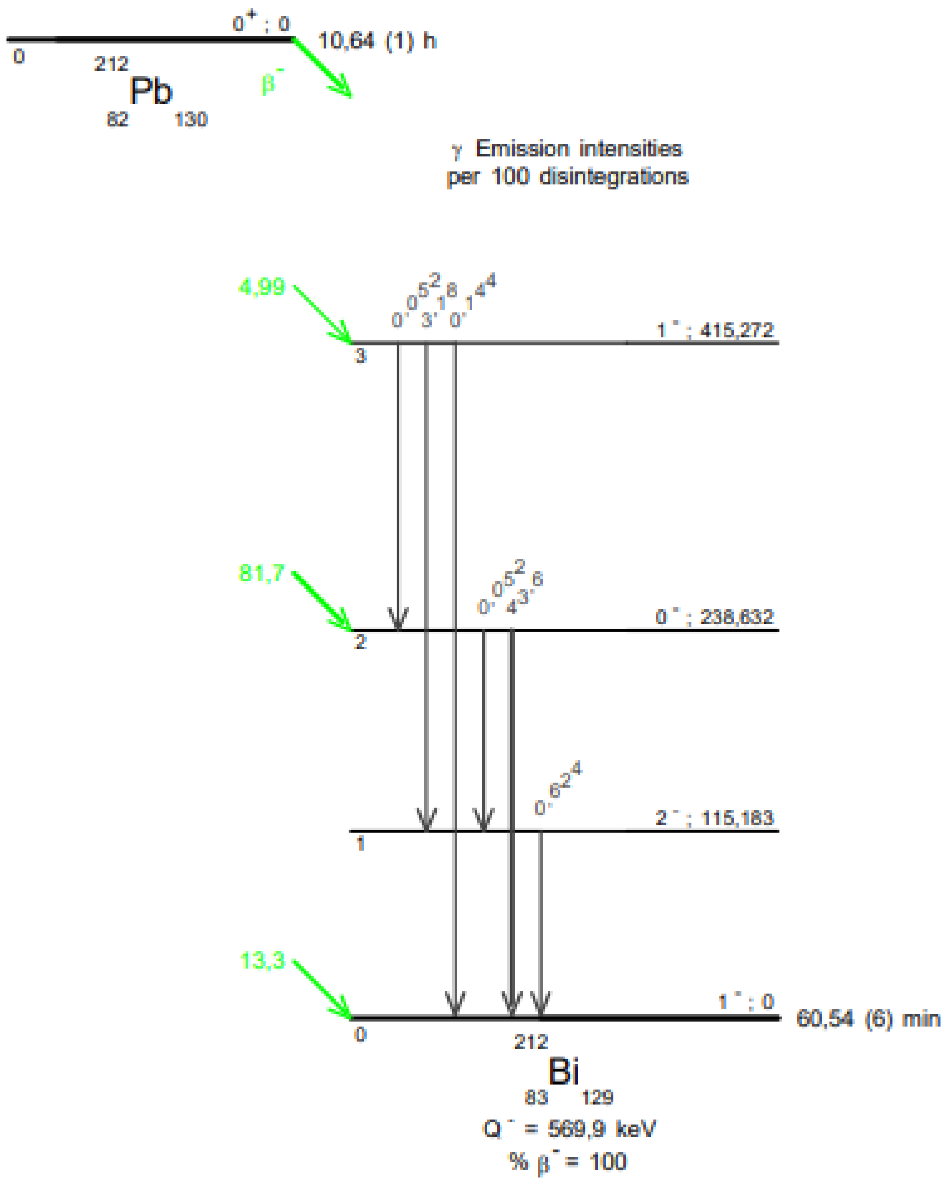


Nuclear Decay Data

- Decay Data is a broad term that covers the values and arrangement associated with the decay processes of a radionuclide

- It covers multiple parameters, such as:
    - Decay scheme
    - Energy of excited states
    - Spin states (angular momentum)
    - Emission probabilities
    - Half-lives

- These have to be determined experimentally



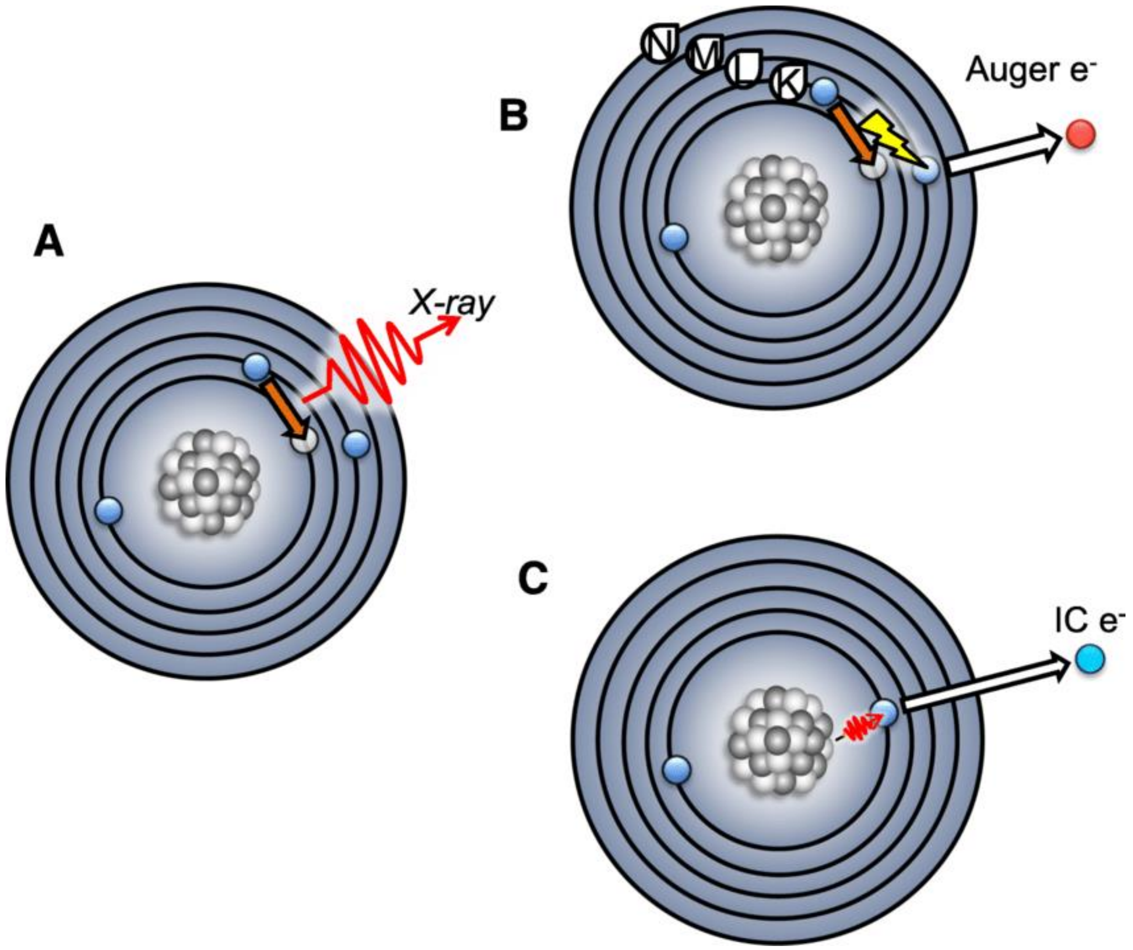
Atomic Decay Data

- The term "Decay Data" can also get used to cover atomic data

- This atomic data is associated from radiations occurring from rearrangements of the atomic electrons

- Such as X-ray emissions, Auger electron emissions, internal conversion electrons

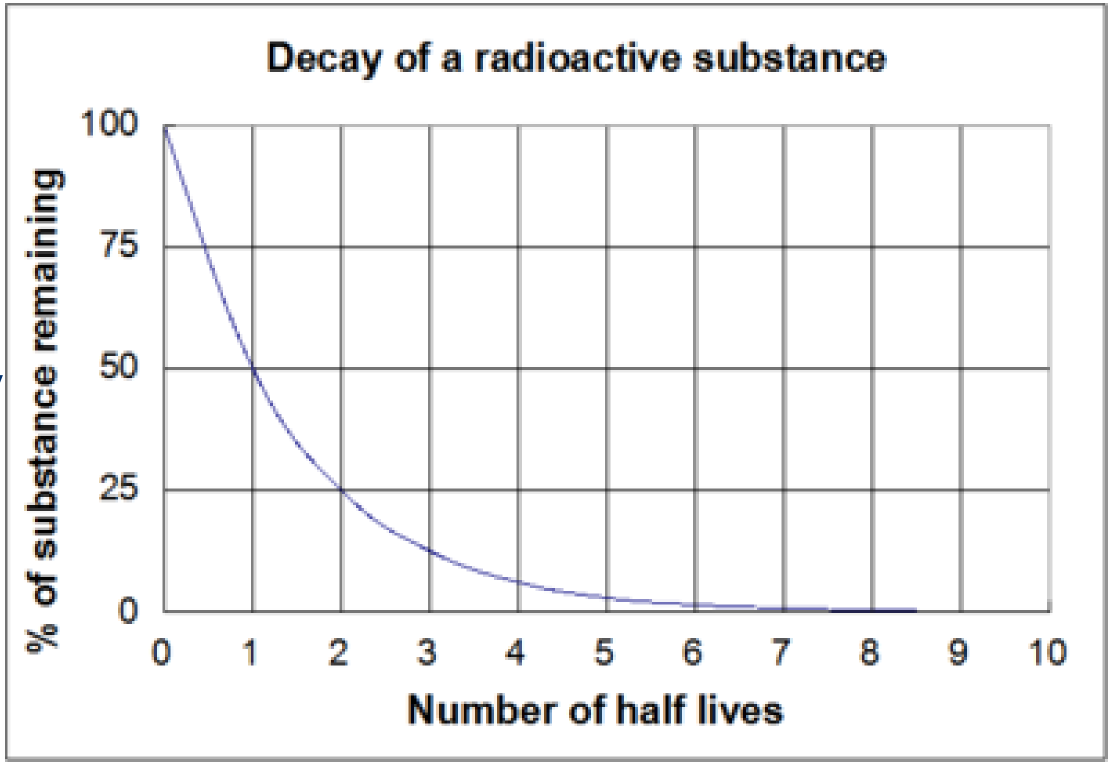
- These all have associated energies and emission probabilities



Radioactive Decay Constants

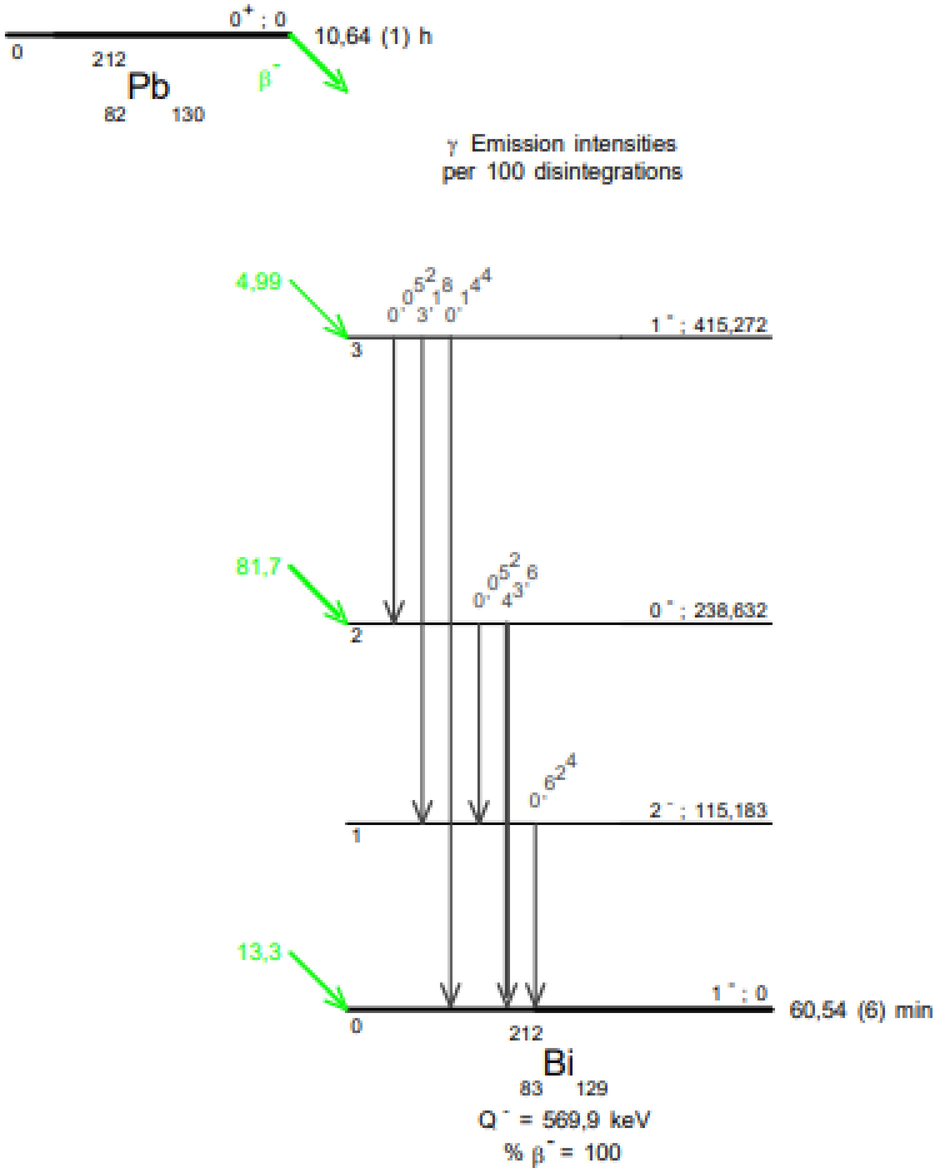
$$T_{1/2} = \frac{ln(2)}{\lambda}$$

- Critical to be able to compare measurements of the same solution at any time and space
- It is impossible to predict when a single atom will decay
- However, for significant numbers of identical atoms the decay rate can be expressed as a decay constant (λ) or half-life ($T_{1/2}$)
- Half-lives of radionuclides have a huge range from near instant to longer than the universe



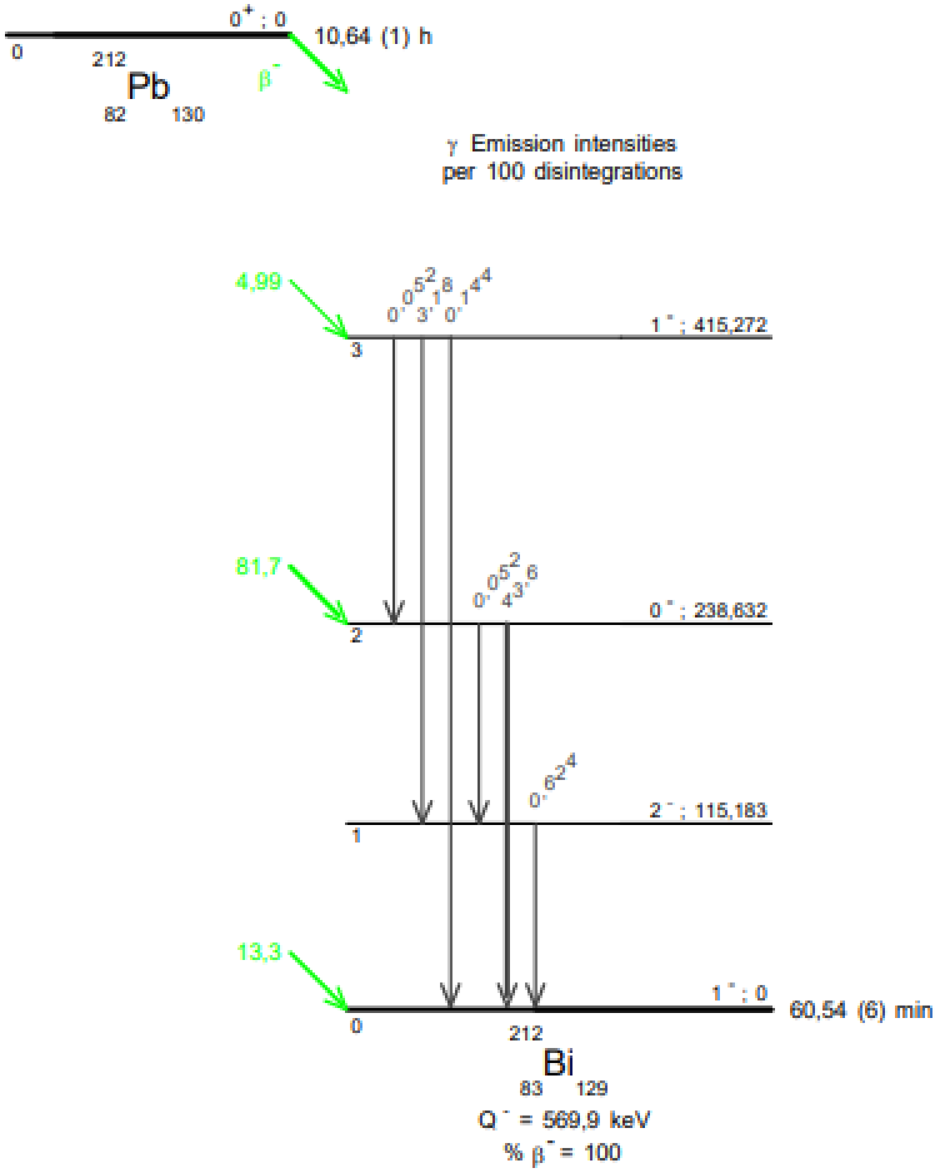
Emission probabilities/intensities

- Along with the half-life, these are probably the most important values for everyday use
- These are the probabilities of the any given emission arising from a single atom decaying
- But it's like flipping a coin….

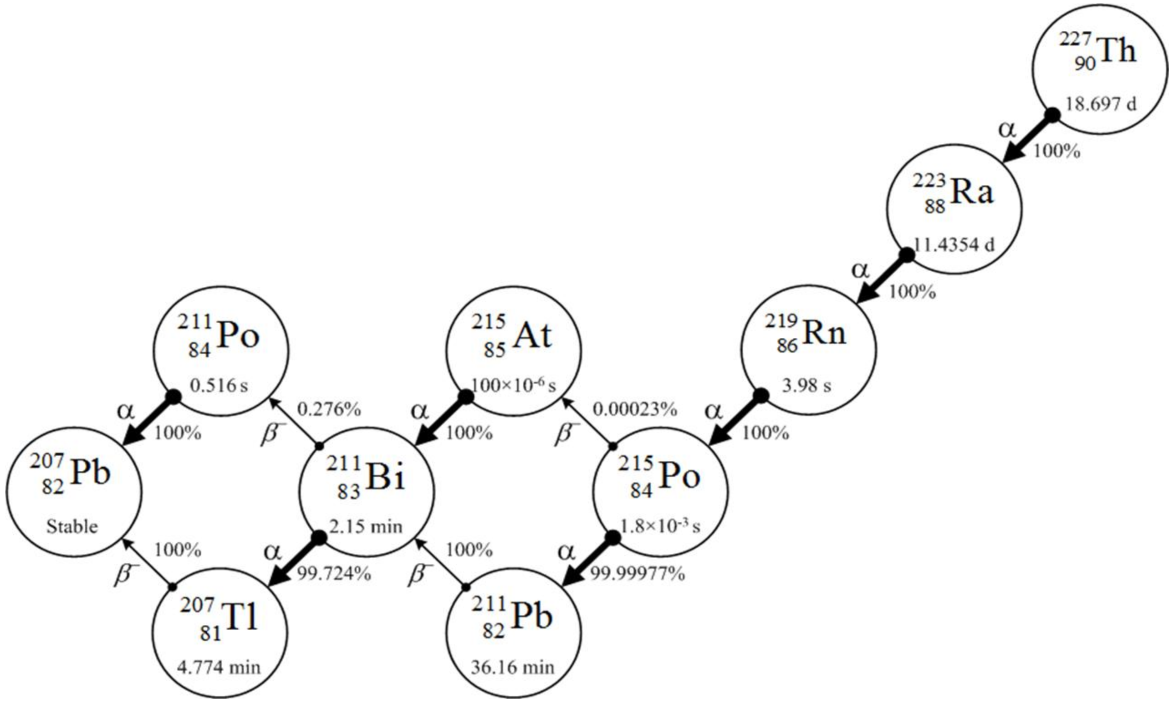


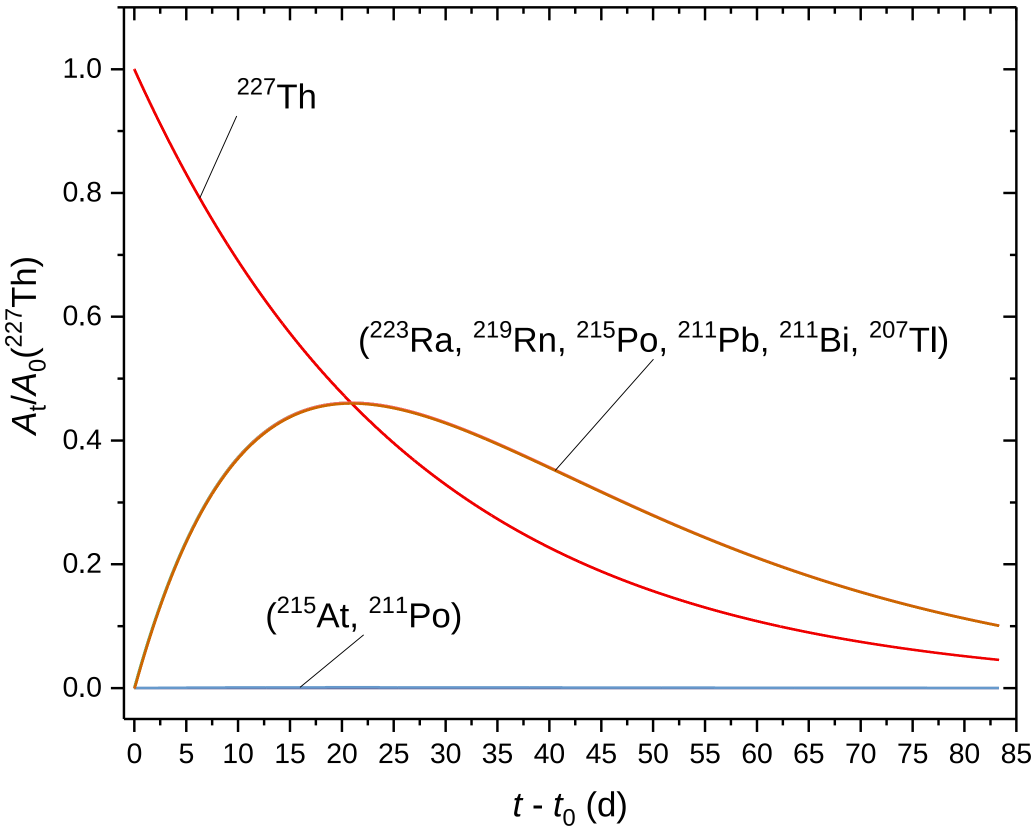
Emission probabilities/intensities

- Along with the half-life, these are probably the most important values for everyday use
- These are the probabilities of the any given emission arising from a single atom decaying
- But it’s like flipping a coin….
- Who measures just one atom decaying?
- We measure lots of decays!



Decay chains of alpha emitters





$$N_1(t) = N_1(0)e^{-\lambda_1 t} \quad (j = 1; 227Th)$$

$$N_j(t) \approx N_1(0)\left(\prod_{i=1}^{j-1} b_{i+1}\lambda_i\right)\sum_{k=1}^{j}\left(\frac{e^{-\lambda_k t}}{\prod_{\substack{i=1\\(i\neq k)}}^{j}(\lambda_i - \lambda_k)}\right)$$

$$A_j(t) = \lambda_j N_j(t)$$

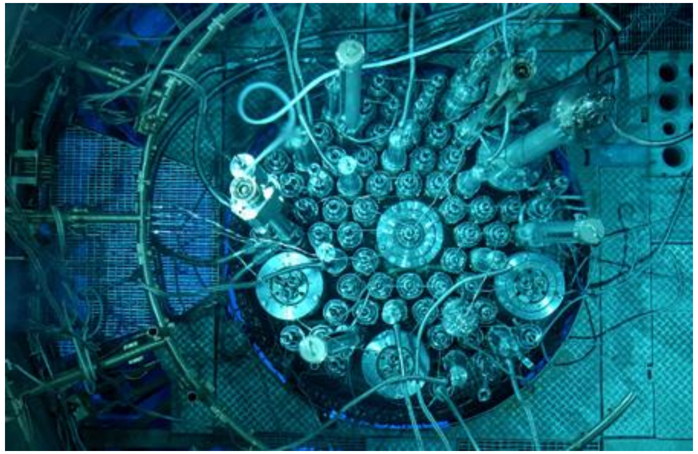
$N_j(t)$ = number of atoms of nuclide j at time t

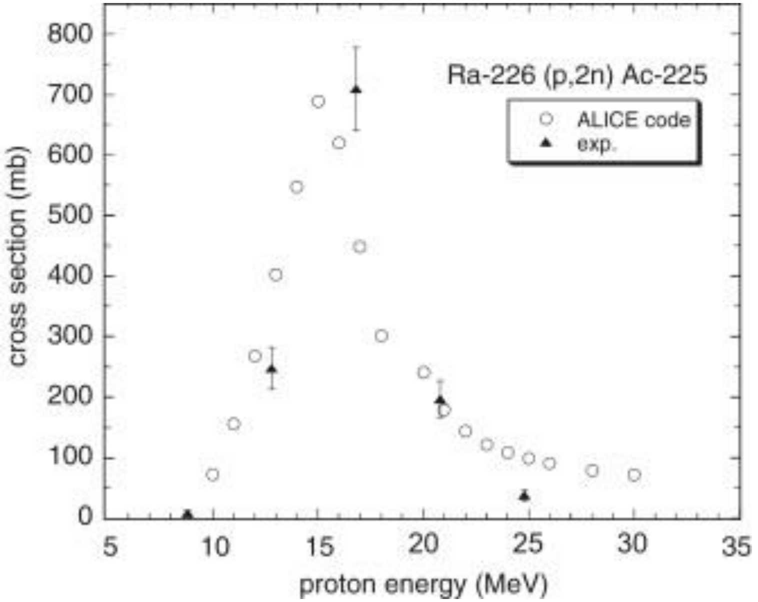
λ_j = decay constant for nuclide j (s^{-1})

$A_j(t)$ = Activity of nuclide j at time t

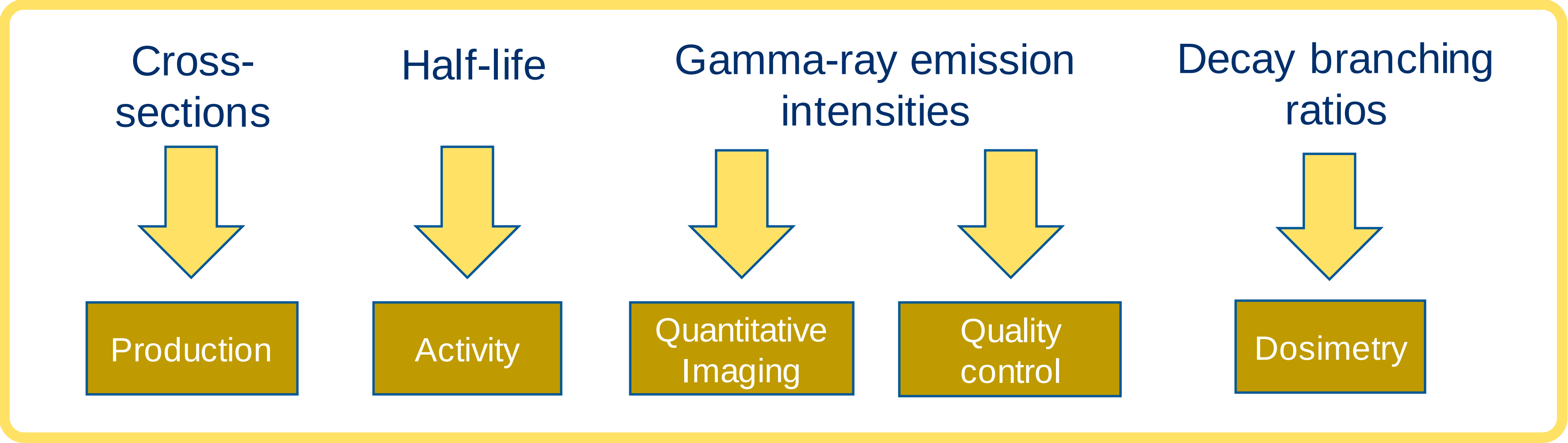
Reaction cross-section data

- Radionuclides can be produced from stable or unstable isotopes by firing protons, neutrons, or photons at a target
- There are different reactions to create the same radionuclide
  - $^{226}Ra(p,2n)^{225}Ac$
  - $^{226}Ra(d,3n)^{225}Ac$
  - $^{226}Ra(\gamma,n)^{225}Ra \rightarrow {}^{225}Ac$
  - $^{232}Th(p,x)^{225}Ra \rightarrow {}^{225}Ac$
- Nuclear cross-section of a nucleus is the probability that a nuclear reaction will occur.
- These can be different at different particle energies



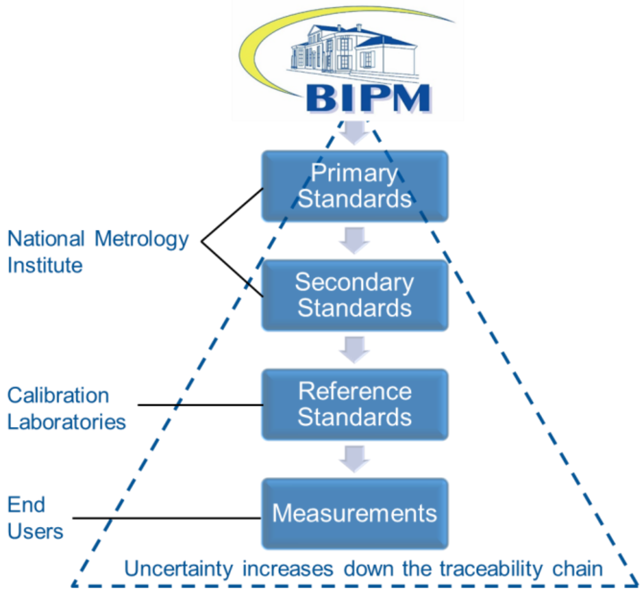


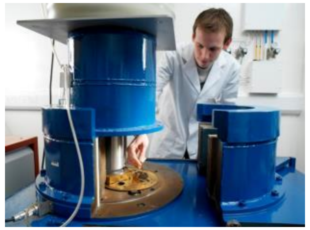
Importance in Nuclear Medicine

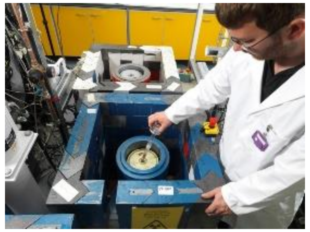


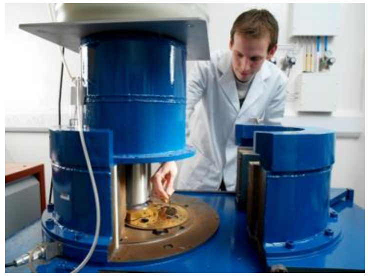
How does metrology underpin decay data

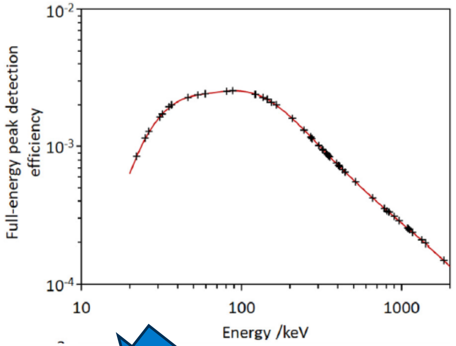
- Decay Data parameters are measurable quantities
- Traceability and standards underpin these measurements
- Primary standards provide significant benefits for determining absolute emission intensities









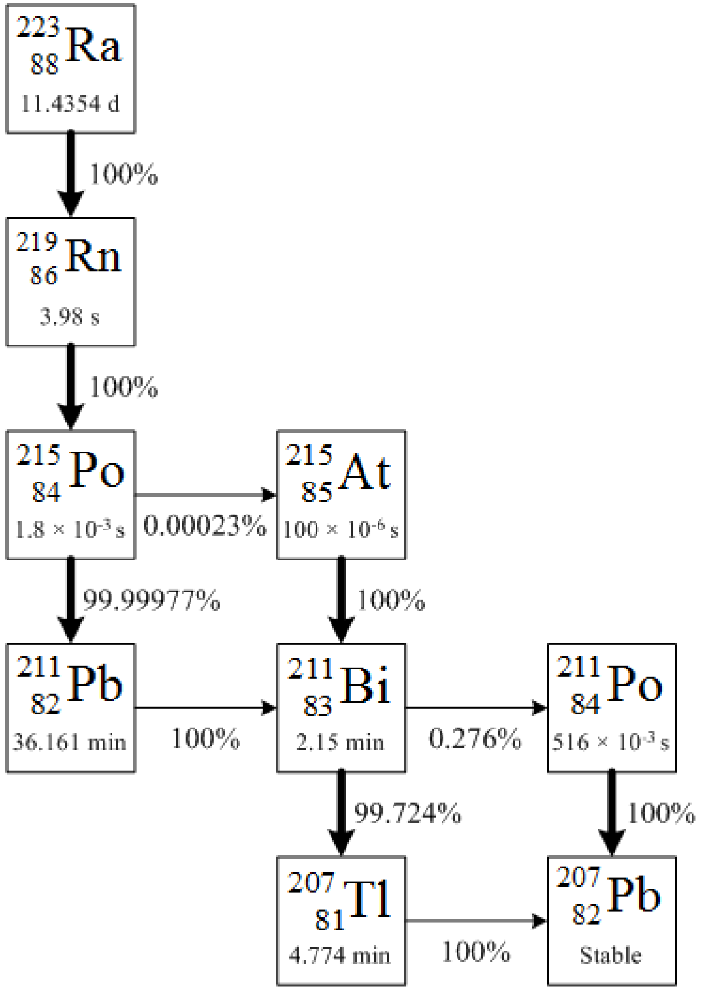


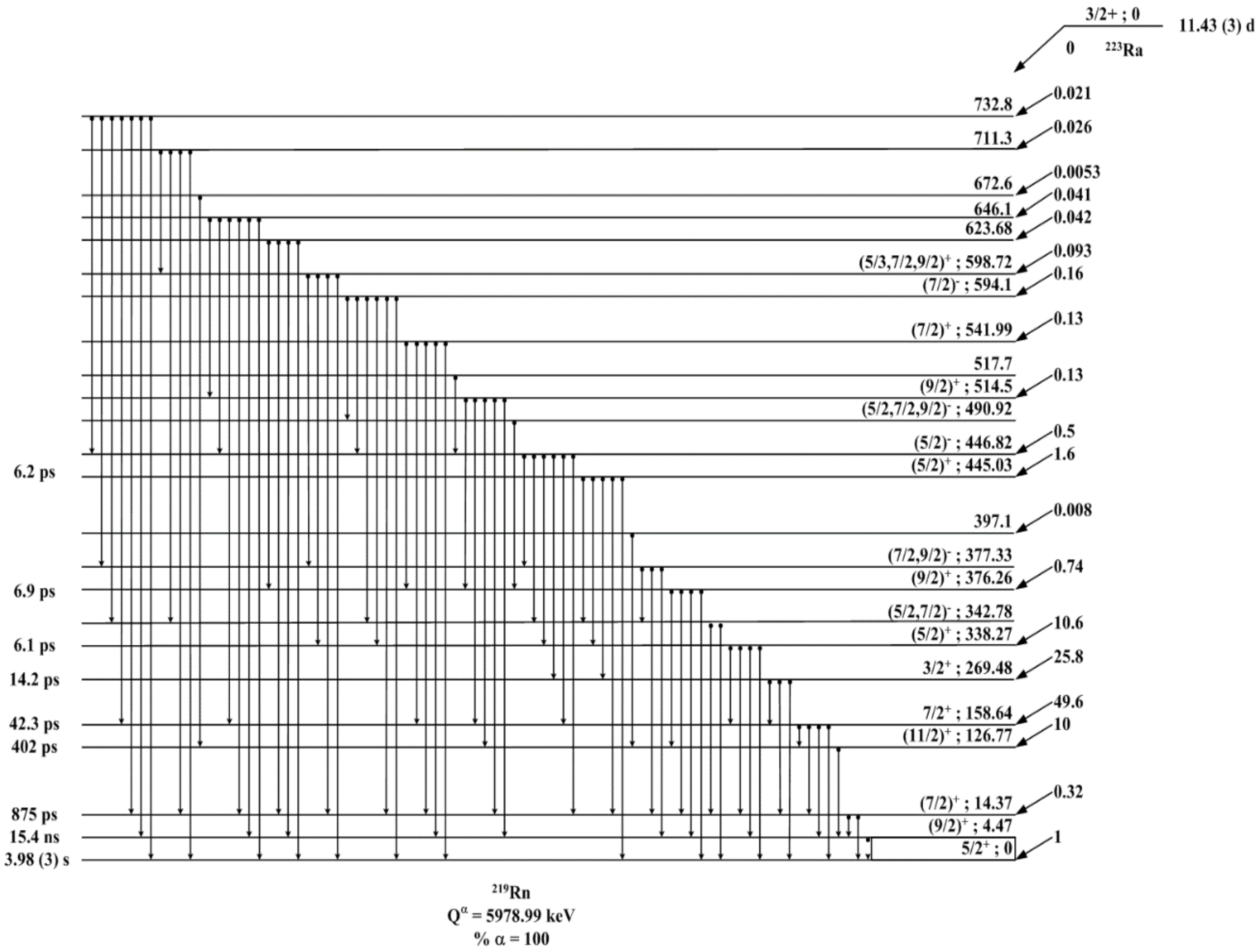
Absolute gamma-ray emission intensities

Examples of ‘recent’ decay data measurements

A Knights Tale (2001)

Ra-223 and decay progeny gamma-ray emission intensities





Ra-223 and decay progeny gamma-ray emission intensities

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- 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?

Applied Radiation and Isotopes 95 (2015) 114–121

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

ELSEVIER

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

CrossMark

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

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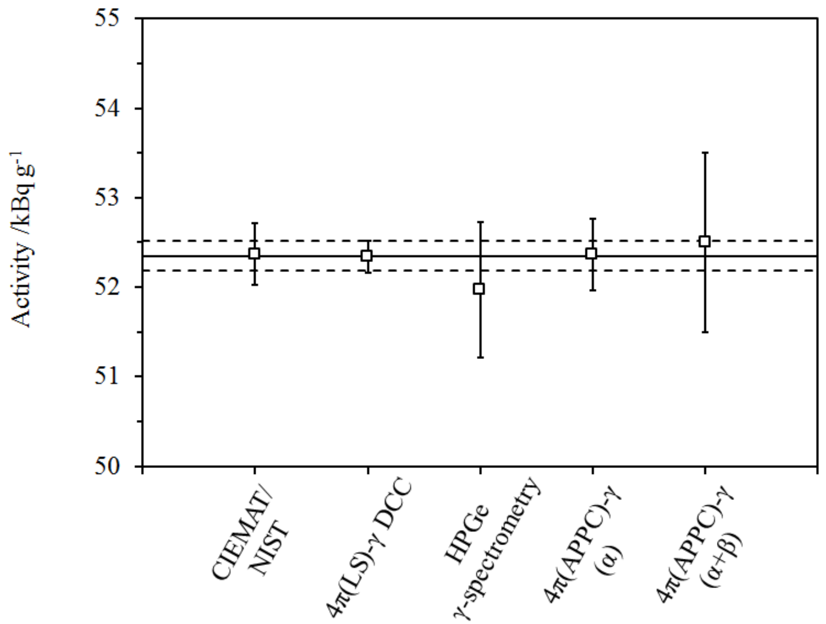
HIGHLIGHTS

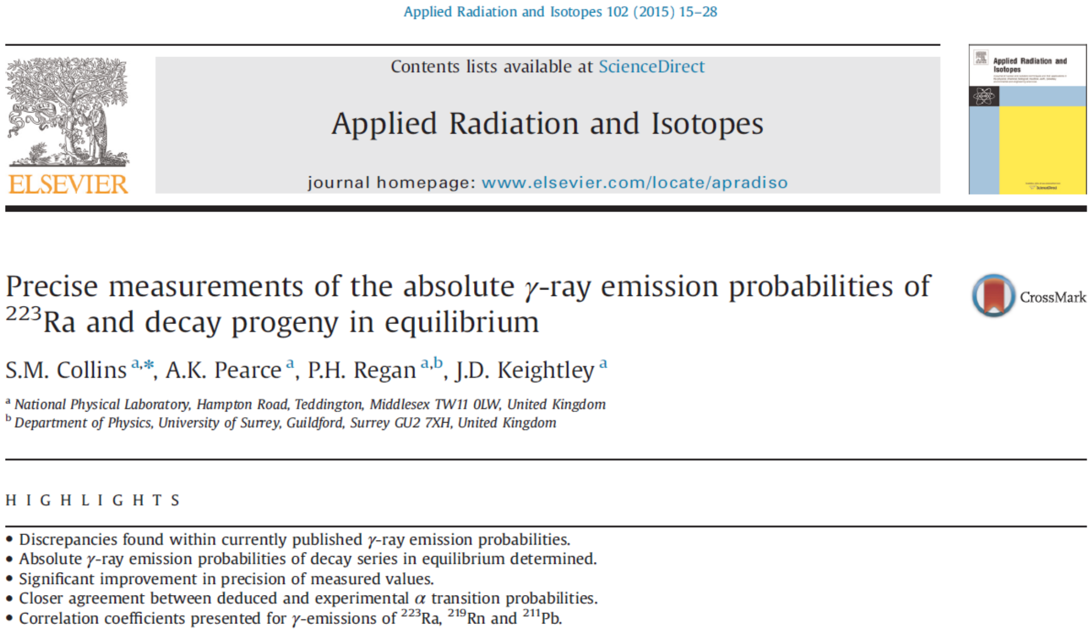
- An aqueous solution of ^{223}Ra chloride was standardised by liquid scintillation counting.
- CIEMAT/NIST efficiency tracing and Digital Coincidence Counting were utilised.
- Calibration factors for a variety of radionuclide calibrators were calculated.
- A discrepancy of around 9% was identified utilising 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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Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

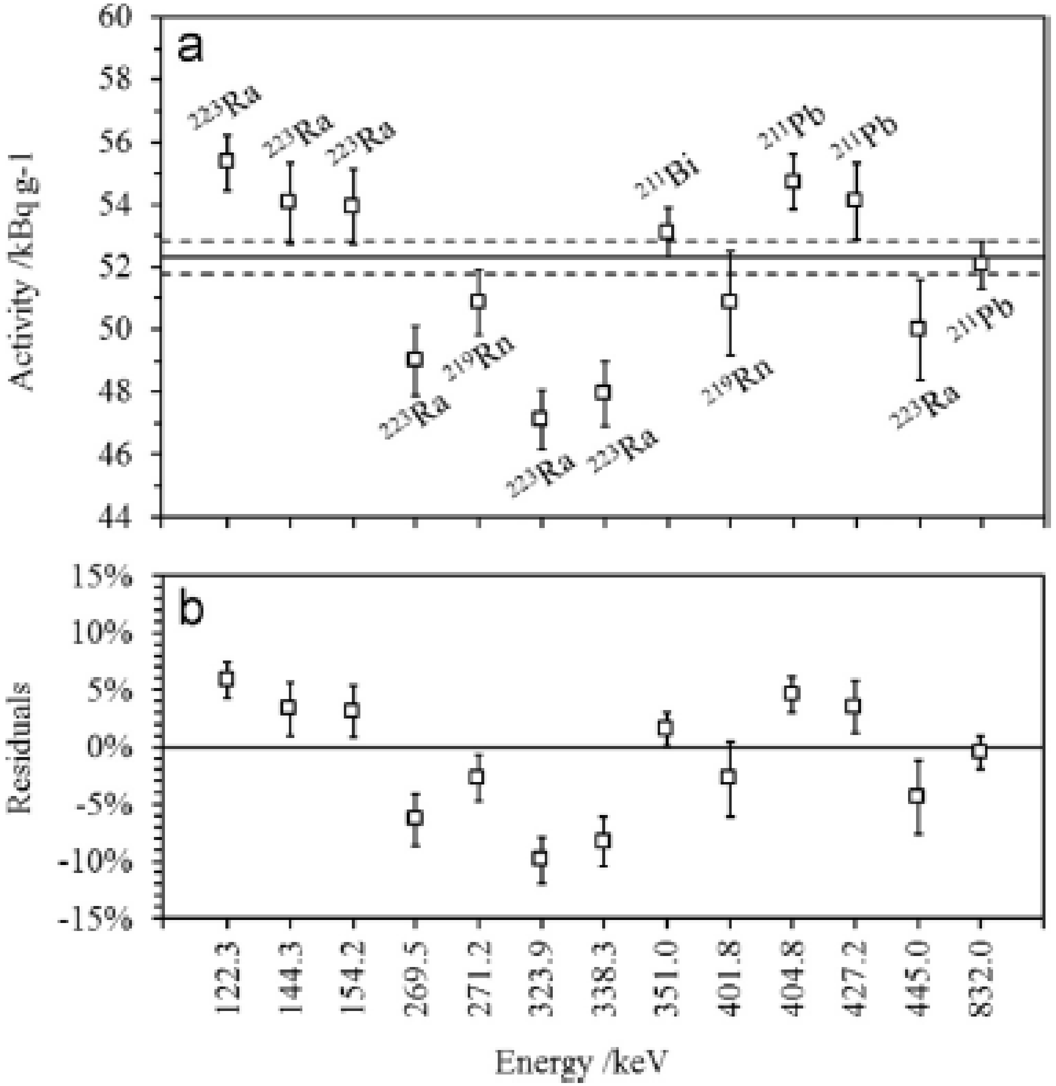
Precise measurements of the absolute γ-ray emission probabilities of ^{223}Ra and decay progeny in equilibrium

S.M. Collins [a,*], A.K. Pearce [a], P.H. Regan [a,b], J.D. Keightley [a]

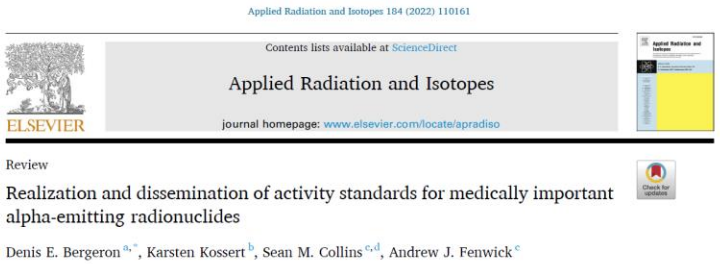
[a] National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, United Kingdom
[b] Department of Physics, University of Surrey, Guildford, Surrey GU2 7XH, United Kingdom

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.



Ra-223 and decay progeny gamma-ray emission intensities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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[c]

Absolute emission intensities

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)	$\chi^2/(\nu\text{-}1)$
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 ^{223}Ra in equilibrium with its progeny. Appl. Radiat. Isot. 101, 15–19.

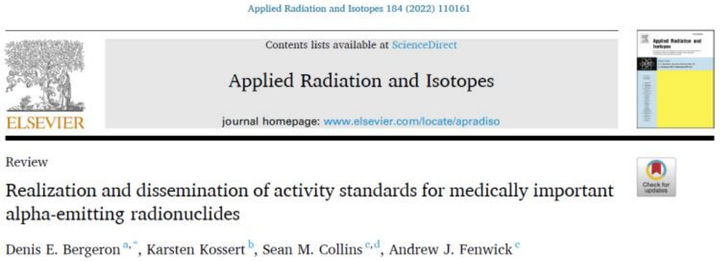
Collins, S.M., Pearce, A.K., Regan, P.H., Keightley, J.D., 2015. Precise measurements of the absolute γ-ray emission probabilities of ^{223}Ra and decay progeny in equilibrium. Appl. Radiat. Isot. 102, 15–18.,

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

Marouli, M., Lutter, G., Pomm´e, 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 ^{227}Ac 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´a, L.V., Poledna, R., de Oliveira, A.E., Iwahara, A., da Cruz, P.A.L., Delgado, J.U., 2021. Standardization of ^{223}Ra by live-time anticoincidence counting and gamma-ray emission determination. Appl. Radiat. Isot. 170, 109559.

Ra-223 and decay progeny gamma-ray emission intensities



Relative emission intensities

Energy /keV	Radionuclide	Pibida et al. (2015) $(I_\gamma/I_{\gamma,154\ \mathrm{keV}})$/100 decays	Collins et al. (2015)	Kossert et al. (2015)	Marouli et al. (2019)	Simões et al. (2021)	$\chi^2/(\nu\text{-}1)$
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 ^{223}Ra in equilibrium with its progeny. Appl. Radiat. Isot. 101, 15–19.

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Ra-223 and decay progeny gamma-ray emission intensities

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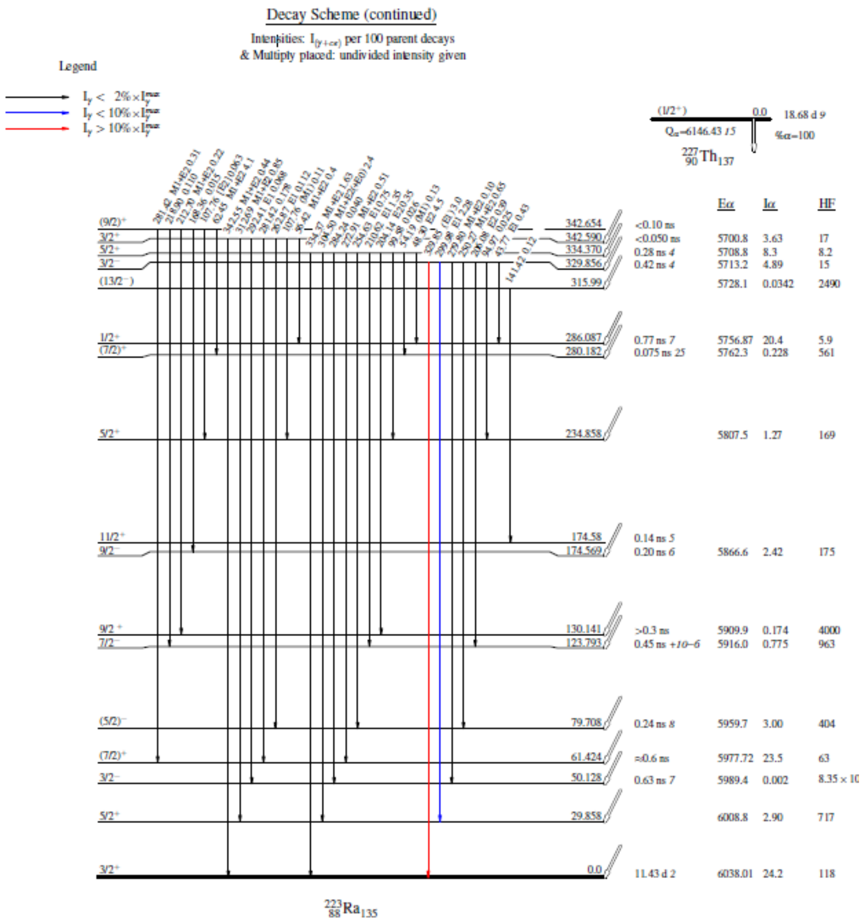
Review

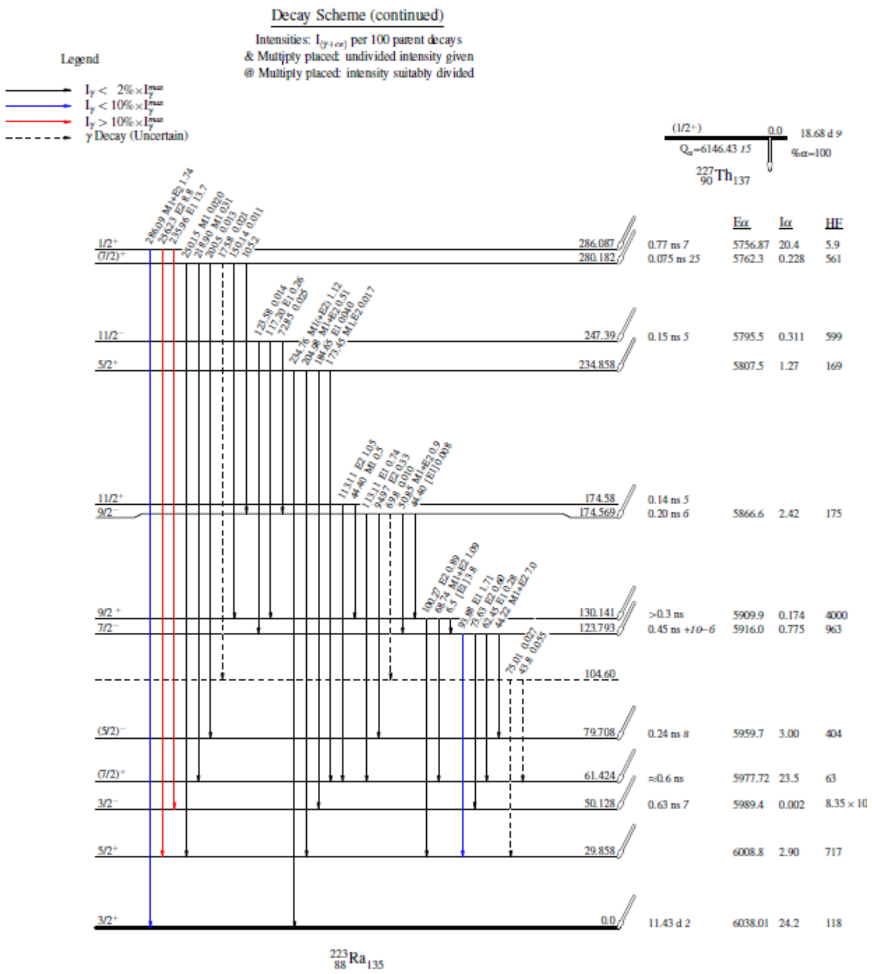
Realization and dissemination of activity standards for medically important alpha-emitting radionuclides

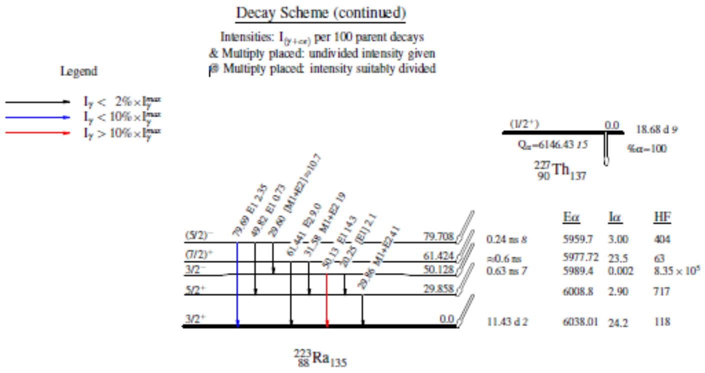
Denis E. Bergeron[a,*], Karsten Kossert[b], Sean M. Collins[c,d], Andrew J. Fenwick[c]

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

Th-227 gamma-ray emission intensities

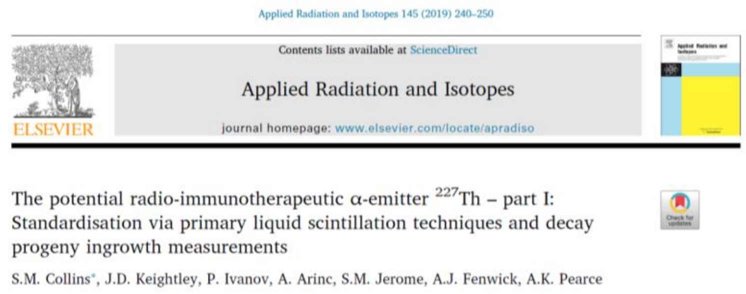






- Calculated absolute gamma-ray emissions lacked precision
  - > 8% uncertainty due to scaling factor
- Previous data published was either in the 1960s or from a private communication in 1992

Th-227 gamma-ray emission intensities


Applied Radiation and Isotopes 145 (2019) 240–250

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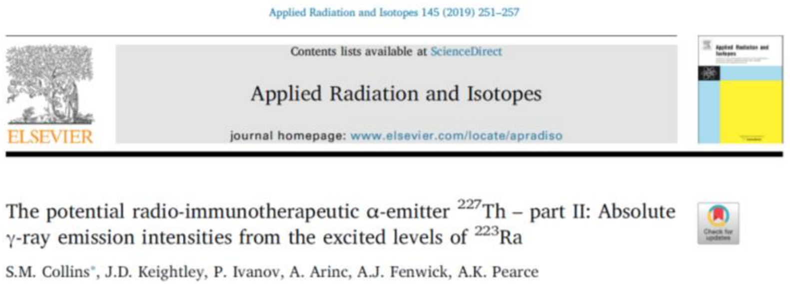
The potential radio-immunotherapeutic α-emitter ^{227}Th – part I: Standardisation via primary liquid scintillation techniques and decay progeny ingrowth measurements

S.M. Collins*, J.D. Keightley, P. Ivanov, A. Arinc, S.M. Jerome, A.J. Fenwick, A.K. Pearce


Applied Radiation and Isotopes 145 (2019) 251–257

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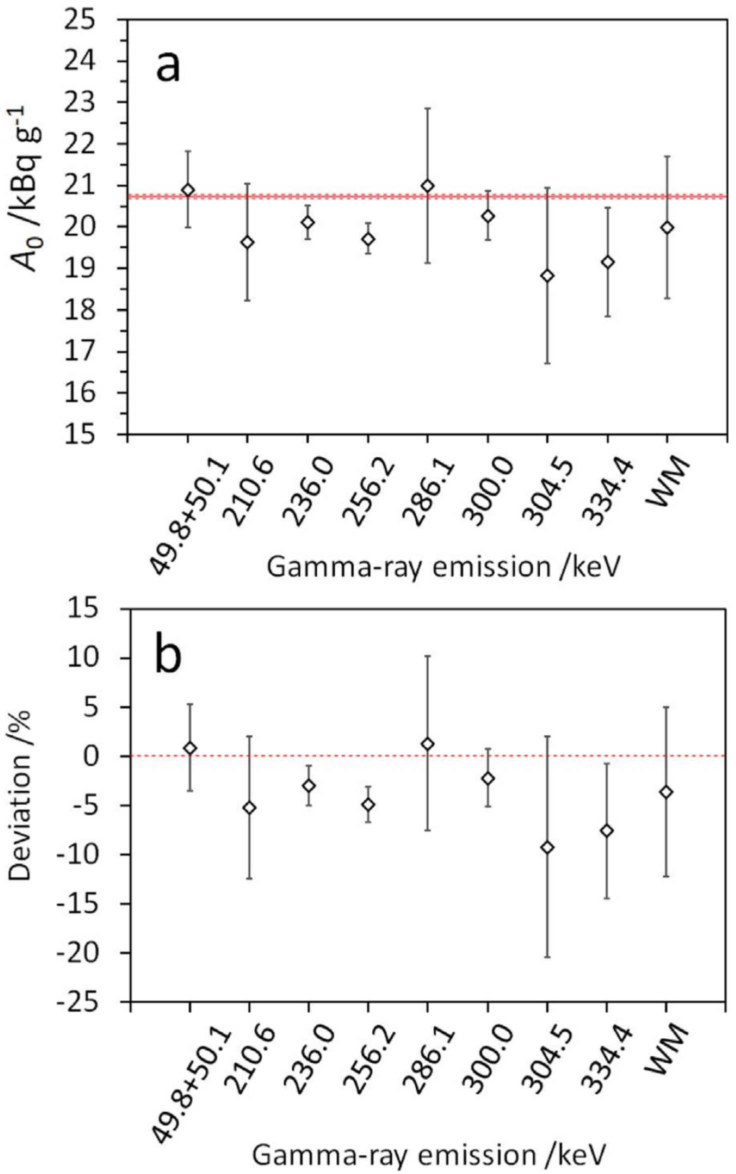
The potential radio-immunotherapeutic α-emitter ^{227}Th – part II: Absolute γ-ray emission intensities from the excited levels of ^{223}Ra

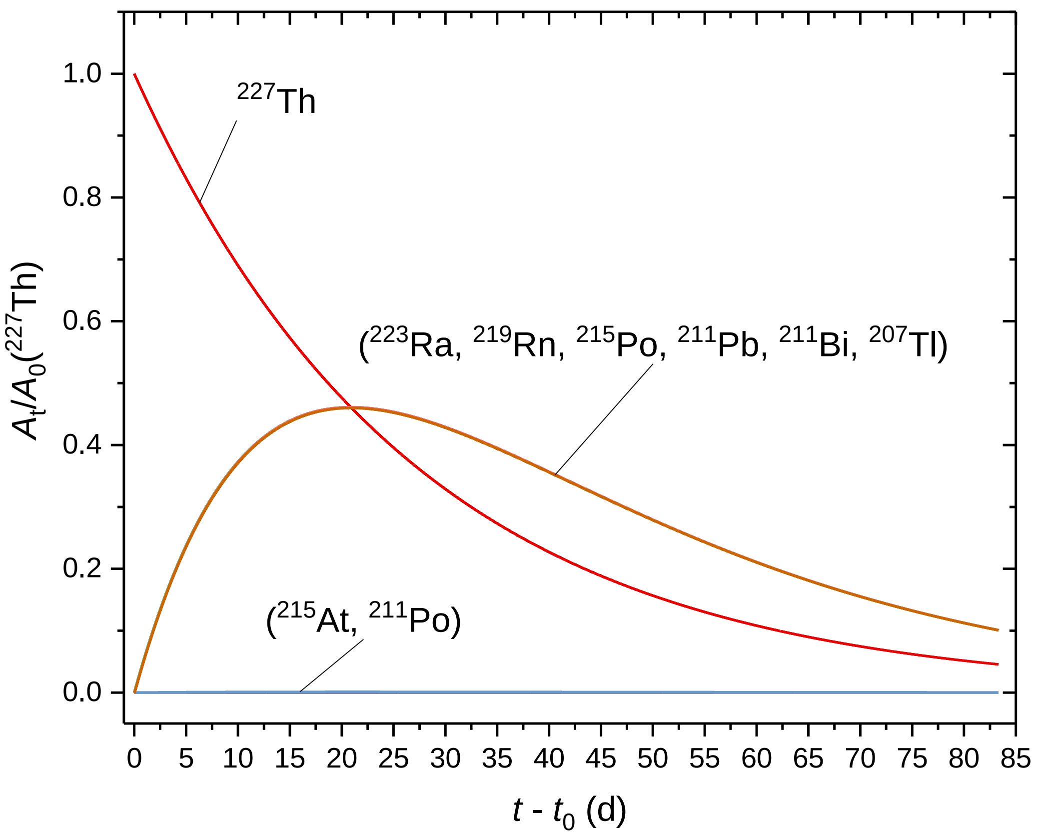
S.M. Collins*, J.D. Keightley, P. Ivanov, A. Arinc, A.J. Fenwick, A.K. Pearce

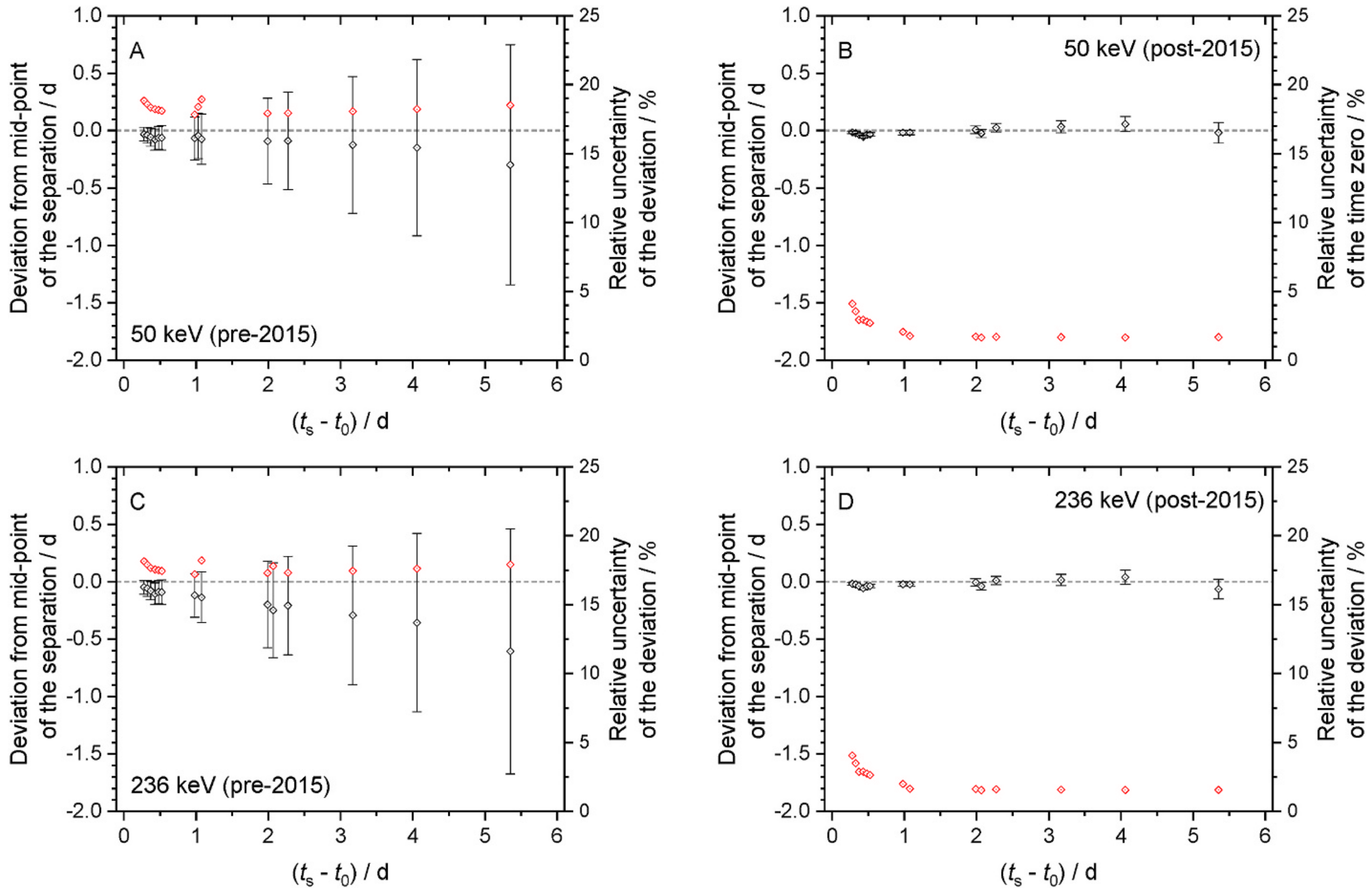


Energy/keV	This work	Abdul-Hadi et al. (1992)	Hesselink et al.(1972)	Briançon et al.(1969)	$\chi^2/(n\text{-}1)$
49.8	71.20 (46)	3.5 (5)	1.7 (13)	4.6 (14)	1.2
50.1		63 (2)	75.7 (52)	65 (3)	
113.1	6.435 (25)	6.6 (3)	4.70 (63)	5.50 (74)	1.8
117.2	1.604 (15)	1.7 (1)	1.50 (31)	1.38 (18)	0.9
204.2	1.525 (14)	1.7 (2)	2.0 (4)	1.6 (4)	0.9
204.9	1.211 (13)	1.2 (2)	1.5 (3)	1.2 (3)	0.6
206.1	1.907 (17)	1.9 (2)	2.32 (41)	1.70 (42)	0.6
210.6	9.497 (35)	9.4 (3)	11.00 (91)	8.5 (11)	1.1
234.8	3.542 (73)	3.4 (3)	5.0 (10)	3.10 (65)	1.0
236.0	100.00 (36)	100 (-)	100 (-)	100 (-)	–
254.6	5.756 (21)	5.3 (3)	7.9 (10)	3.90 (86)	1.9
256.2	53.26 (19)	54 (1)	55.0 (46)	57.0 (55)	2.8
272.9	4.057 (15)	3.9 (2)	4.30 (62)	3.90 (68)	0.5
281.4	1.388 (15)	1.4 (1)	1.30 (30)	1.30 (32)	0.2
286.1	14.063 (51)	15.0 (1)	14.30 (90)	12.3 (11)	4.9
296.5	3.660 (27)	3.3 (3)	3.40 (62)	3.70 (58)	0.7
300.0	17.258 (44)	17.3 (5)	18.8 (15)	16.9 (22)	0.6
304.5	8.343 (22)	8.6 (5)	12.0 (11)	7.7 (10)	2.0
312.7	4.055 (23)	4.0 (3)	4.50 (92)	3.90 (68)	0.3
314.9	3.812 (11)	3.7 (3)	4.70 (92)	3.60 (58)	0.6
329.9	21.615 (55)	21.7 (5)	25.2 (17)	21.5 (26)	1.2
334.4	8.099 (35)	8.2 (3)	10.00 (99)	8.5 (13)	1.1
342.6	3.324 (13)	3.4 (1)	1.70 (41)	3.20 (65)	2.4

- Order of magnitude improvement in precision

Effect on time zero determination





Ac-225 half-life

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- Previously a half-life of 10.0(1) d from a measurement in 1950

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Measurement of the ^{225}Ac half-life

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HIGHLIGHTS

- Measured ^{225}Ac half-life by six methods.
- The result is $T_{1/2}(^{225}\text{Ac})=9.920\ (3)$ d.
- ^{225}Ac and its daughter ^{213}Bi are important for targeted alpha therapy.

References	$T_{1/2}$ (d)	Comments
2012PO14	9.9200 (36)	Minimum measured uncertainty
2013BO	9.9120 (63)	Expanded uncertainty
2020KO06	9.9180 (35)	Minimum measured uncertainty
2023BR08	9.9150 (63)	
2024GA01	9.914 (6)	Minimum measured uncertainty
Recommended	**9.9172 (21)**	Weighted mean, internal uncertainty

Status and needs of radionuclides being investigated for targeted alpha-particle therapy

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DE GRUYTER

Radiochim. Acta 2022; 110(6–9): 609–644

Contribution to "Diamond Jubilee of RCA"

Alan L. Nichols*

Status of the decay data for medical radionuclides: existing and potential diagnostic γ emitters, diagnostic β^+ emitters and therapeutic radioisotopes

https://doi.org/10.1515/ract-2022-0004
Received January 6, 2022; accepted April 10, 2022;
published online May 18, 2022

Abstract: Recommended half-lives and specific well-defined emission energies and absolute emission probabilities are important input parameters that should be well-defined to assist in ensuring the diagnostic and therapeutic [...]
adequate quantification of the required decay data (i.e., dose calculations include half-lives, energies and emission probabilities of α, β^+, various electron particles, γ and X-rays, along with other related parameters). Specific radionuclides possess decay characteristics that have been found to be appropriate or potentially suitable in the diagnosis or radiotherapeutic treatment of tumours, and the various nuclear

27 EU
PRISMAP
Medical Radionuclides

Deliverable D11.1

Nuclear data for day-1 radionuclides

https://www.degruyter.com/document/doi/10.1515/ract-2022-0004/html

https://zenodo.org/records/8247129

Status and needs of radionuclides being investigated for targeted alpha-particle therapy

Tb-149	▪ Precision measurements of the half-lives of Tb-149 and its decay progenies (Eu-145 and Gd-149) are needed. ▪ New studies are required to improve the precision of the alpha decay branching ratio. ▪ There is a requirement for new studies of the gamma-ray emission intensities to confirm the accuracy of the single study and to improve the precision. ▪ There is also a requirement to improve the gamma-ray emission intensities of the decay progenies.
At-211	▪ New half-life determinations with complete uncertainty evaluation are required.
Ac-225	▪ Extensive gamma-ray emission intensity studies and γ-γ coincidence studies are recommended are required.

^{227}Th: Extensive and significant α decay directly to the ground (24.2% α), third (23.5% α), thirteen (20.4% α) and sixteenth (8.3% α) excited states of ^{223}Ra, as well as depopulation by 235.96-, 256.23- and 329.85-keV γ rays and 220 lower-intensity γ transitions from 6.5 to 1025 keV – *extensive and complex decay scheme that includes as many as 223 γ rays of which the placement of thirty-six are in doubt, while at least a further forty-nine γ rays remain unplaced; more extensive γ singles and γ–γ coincidence studies merited*

Importance of Decay Data Evaluations

- Evaluated datasets provide the decay data that most use on a routine basis
- There are several databases of evaluated decay data
  - NNDC
  - Decay Data Evaluation Project (DDEP)
- However, there can be a significant lag between publication of new data and its inclusion in new evaluated datasets
- There are not enough evaluators and too many radionuclides

National Nuclear Data Center

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

Laboratoire National LNHB Henri Becquerel

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

Current Challenges for Decay Data

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RECOGNITION	TECHNICAL
SKILLED PEOPLE	ROBUSTNESS & VALIDITY OF DATA

Future outlook

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- There are new programs that have built in metrology to measure decay data for medical radionuclides

- NuPECC Long Range plan (in progress) has identified decay data, metrology, and needs for societal applications

PRISMAP Medical Radionuclides

EURAMET

ALPHAMET

NuPECC

Department for Energy Security & Net Zero

UK Medical Radionuclide Innovation Programme (MRIP)

Conclusion

- Decay data is a broad term covering a large field
- They are measurable quantities
- A lot of the data is old and does not benefit from traceability or metrology
- It has an impact across the radiopharmaceutical pathway

- There is still plenty to do but significant improvements have been made in the last 10 years
  - State-of-the-art techniques
  - Metrology and traceability
  - Robust uncertainties
- Decay data is seeing an increase in interest and the need for better confidence in the data
- There are new programmes underway that identify the need for metrology in decay data

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