# Nuclear Metrology & Applications

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# 2 x 1 hour lectures on Nuclear Metrology at the National Physical Laboratory (NPL)

- Lecture 1:
  - What is nuclear metrology ?
  - History, definition of activity, units, Ci, Bq etc.
  - Nuclear Data ; the longest half-lives.
  - Naturally Occurring Radioactive Materials.

- Lecture 2:
  - Nuclear Metrology, definitions and primary methods.
  - Example $^{223}$Ra standardisation for medical application.
  - Gaseous radionuclide metrology example, CTBTO test;

# Lecture 1

# What is nuclear metrology?

- The measurement of nuclear properties.

  - The collection, evaluation and application of (primary and secondary) nuclear data, including

    - Fundamental decay properties (e.g. $T_{1/2}$,  Q-value...)

    - Nuclear decay /  structure  signatures for decay e.g.  characteristic gamma-ray energies, internal conversion coefficients $\alpha_{tot}$, $\alpha_K$, $\alpha_L$ etc. ; $P\gamma(\%)$ etc;

    - Reaction properties: e.g.  neutron capture / scattering cross-sections, (n,fission) probabilities....

# Lots of applications of nuclear science measurement

- Nuclear power & waste management ($^{134,7}$Cs ; $^{90}$Sr; $^{239,240}$Pu, $^{241}$Am.....)

- Medical therapies & diagnostics ($^{99m}$Tc, $^{18}$F, $^{82}$Sr, $^{177}$Lu $^{149,152,155,161}$Tb, $^{223}$Ra...)

- Naturally Occurring ($^{222}$Rn; $^{226,228}$Ra, $^{40}$K) radioactivity measurements.

- Man-made (nuclear weapons tests) radioactivity evaluations ($^{90}$Sr, $^{137}$Cs, Pu..)

- Chronology / long-term dating ($^{14}$C ; $^{238}$U/$^{206}$Pb ; $^{40}$K/$^{40}$Ar)

- Elemental / isotopic ratio analysis (in-beam techs PIGE ; PNAA..)

  – Cultural heritage – are your paintings, coins, wine etc. fake?

- Nuclear forensics (e.g.,CTBTO weapons test), criticality monitoring

  – Noble gas radioactivity signatures & isotopic ratios e.g. $^{133,135}$Xe and $^{87,88}$Kr

  All these rely on **accurate Nuclear Metrology** and high-quality and **traceable references standards** & relate the absolute activity to the number of atoms

$$A = \lambda N = - ( dN / dt)$$

## Today's lecture:

- Measurements of Radioactivity in the Environment:
    - NORMS ($^{235,8}$U ; $^{232}$Th ; $^{40}$K....)

## Tomorrow's lecture:

- Radionuclide Metrology:
    - Primary and secondary standards.
    - Xe/Kr radioactive gas metrology

## NUCLEAR METROLOGY?

It is important to keep accurate, traceable records of previous (nuclear data) measurements and <u>reference standards</u> for potential future applications and comparisons….

# Some 'old data' and previous standards...
Nuclear Physics Summer School, U. York - *who's who 1989*.



Includes: **Gavin Smith** (Manchester); **Martin Freer** (B'ham); **Sean Freeman** (Manchester / CERN); David Ireland (Glasgow) ; PHR (Surrey / NPL); **Gerda Neyens** (Leuven); John Roberts (IAEA);  Alison Bruce (Brighton); Mike Bentley (York); Peter Butler (Liverpool);

Bangor EPSRC Summer School 1995

PHR, Martin Freer (B'ham), Jim Al-Khalili (Surrey), Alison Bruce (B'ton),
David Ireland (Glasgow), Robert Page (Liverpool), Sean Freeman, David Joss (Liv),

Durham Summer School 1999

Inc. Helen Boston (Liverpool), Kate Jones (Tennessee), Iain Moore (Jyvaskyla), David Joss (Liverpool), Dan Watts (York) + Rodi Herzberg (Liverpool), PHR, Sean, Freeman & David Ireland (Glasgow).
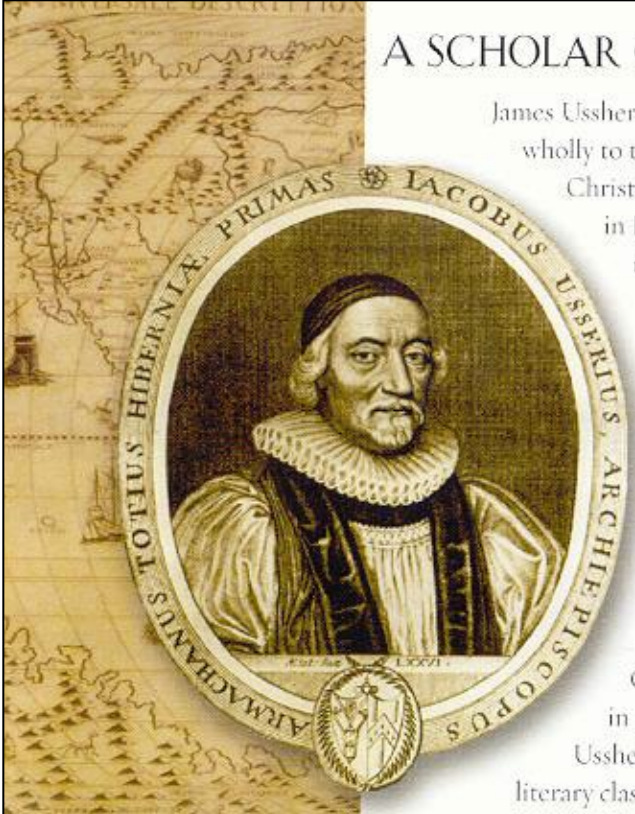
# Radioactivity in the Environment

## (TE)NORM.

- Primordial (long-lived) & cosmogenic sources.
- Decay chains (U, Th & Ac); odd-odd primordials
- How do you measure it ? How much is there?
- NORM activity concentrations using HPGe

## Anthropogenic radioactivity:

- Which fission fragments do you expect, when?
- Fission reactor vs nuclear weapons tests.
- Fukushima, criticality signatures, CTBTO ?

# ASIDE: Evidence based research, using the best the available literature.
# Famous example : **Archbishop Ussher**,



A SCHOLAR OF HONOR

James Ussher devoted himself wholly to the defense of the Christian Faith. Born in Dublin, Ireland in 1581, this highly educated, well-traveled historian devoted years to writing the history of the world from creation to A.D. 70. Originally published in Latin in the 1650s, Ussher's masterpiece is a literary classic.

ANNALES

VETERIS TESTAMENTI,

*à primâ Mundi origine deducti.*

IN PRINCIPIO creavit DEUS Cœlum & Terram. [Genef. I. 1] quod temporis principium (juxta noftram Chronologiam) incidit in noctis illius initium, quæ XXIII. diem Octobris præcessit, in anno Periodi Julianæ 710.

Primo igitur feculi die (Octob. 23. feriâ 1.) cum fupremo Cœlo creavit Deus Angelos : deinde fummo operis fastigio primùm perfecto, ad ima Mundanæ hujus fabricæ fundamenta progreffus miran-

7104004

Bishop James Ussher (1581-1656)

*Annals of the Old Testament, Deduced From the First Origins of the World*

Others also tried similar methods to age the Earth inc. Isaac Newton & Johannes Kepler.

Moral of the Story:

The physical interpretation of the results is limited by the accuracy of the input data (ideally measured) .

| DATE | EVENT | SCRIPTURE | AGE OF EARTH |
|---|---|---|---|
| 4004 BC | Creation | Gen. 1:1–31 | 0 |
| 3874 BC | Seth born when Adam was 130 | Gen. 5:3 | 130 yrs. |
| 3769 BC | Enos born when Seth was 105 | Gen. 5:6 | 235 yrs. |
| 3679 BC | Cainan born when Enos was 90 | Gen. 5:9 | 325 yrs. |
| 3609 BC | Mahalaleel born when Cainan was 70 | Gen. 5:12 | 395 yrs. |
| 3544 BC | Jared born when Mahalaleel was 65 | Gen. 5:15 | 460 yrs. |
| 3382 BC | Enoch born when Jared was 162 | Gen. 5:18 | 622 yrs. |
| 3317 BC | Methuselah born when Enoch was 65 | Gen. 5:21 | 687 yrs. |
| 3130 BC | Lamech born when Methuselah was 187 | Gen. 5:25 | 874 yrs. |
| 2948 BC | Noah born when Lamech was 182 | Gen. 5:28 | 1,056 yrs. |
| 2446 BC | Shem born when Noah was 502 | Gen. 11:10 | 1,558 yrs. |
| 2348 BC | Flood when Noah was 600 | Gen. 7:6 | 1,656 yrs. |
| 2346 BC | Arphaxad born when Shem was 100 | Gen. 11:10 | 1,658 yrs. |
| 2311 BC | Salah born when Arphad was 35 | Gen. 11:12 | 1,693 yrs. |
| 2281 BC | Eber born when Salah was 30 | Gen. 11:14 | 1,723 yrs. |
| 2246 BC | Peleg born when Eber was 34 | Gen. 11:16 | 1,758 yrs. |
| 2217 BC | Reu born when Peleg was 30 | Gen. 11:18 | 1,787 yrs. |
| 2185 BC | Serug born when Reu was 32 | Gen. 11:20 | 1,819 yrs. |
| 2155 BC | Nahor born when Serug was 30 | Gen. 11:22 | 1,849 yrs. |
| 2126 BC | Terah born when Nahor was 29 | Gen. 11:24 | 1,878 yrs. |
| 1996 BC | Abraham born when Terah was 130 | Gen. 11:32; 12:4 | 2,008 yrs. |
| 1921 BC | Abraham enters Canaan at 75 | Gen. 12:4 | 2,083 yrs. |

# How old is the Earth ? A Chronology of Chronologies…

- **<u>Comte de Buffon (1779)</u>** - Believes earth is slowly cooling, from the rate compared to a small globe, estimated ~75,000 years.

- **<u>James Hutton (1795)</u>** - 'The Theory of the Earth (1795)' Geological evolution of the earth's crust, rock strata formed in layers?

- **<u>Lord Kelvin aka William Thompson (1862)</u>** - Earth had formed between 20 and 40 million years, estimated from time to cool and heat assuming heat generated by gravitational contraction and scientific estimates of earth's heat conduction (2nd law of thermodynamics).

- **<u>Rutherford and Soddy (1903)</u>** - Explanation of radioactivity of Uranium (U); earth's internal heat could come from radioactivity. Half-lives for decays could be billions of years.

   Rutherford suggests use of helium ($\alpha$ particles) in rocks to age them.

- **<u>Boltwood (1907)</u>** - ratios of U to Lead to get age of rocks > $10^9$ years.

L. Badash, 'The Age of the Earth Debate' Scientific American, August 1989 p 90ff

I. *A Radio-active Substance emitted from Thorium Compounds. By* E. Rutherford, *M.A., B.Sc., Macdonald Professor of Physics, McGill University, Montreal* *.

IT has been shown by Schmidt† that thorium compounds give out a type of radiation similar in its photographic and electrical actions to uranium and Röntgen radiation. In addition to this ordinary radiation, I have found that thorium compounds continuously emit radio-active particles of some kind, which retain their radio-active powers for several minutes. This "emanation," as it will be termed for shortness, has the power of ionizing the gas in its neighbourhood and of passing through thin layers of metals, and, with great ease, through considerable thicknesses of paper.

In order to make clear the evidence of the existence of a radio-active emanation, an account will first be given of the anomalous behaviour of thorium compounds compared with those of uranium. Thorium oxide has been employed in most of the experiments, as it exhibits the "emanation" property to a greater degree than the other compounds; but what is true for the oxide is also true, but to a less extent, of the other thorium compounds examined, viz., the nitrate, sulphate, acetate, and oxalate.

In a previous paper‡ the author has shown that the radiation

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\* Communicated by Prof J. J. Thomson, F.R.S.
† Wied. *Ann.* May 1898.
‡ Phil. Mag. Jan. 1899, p. 109.
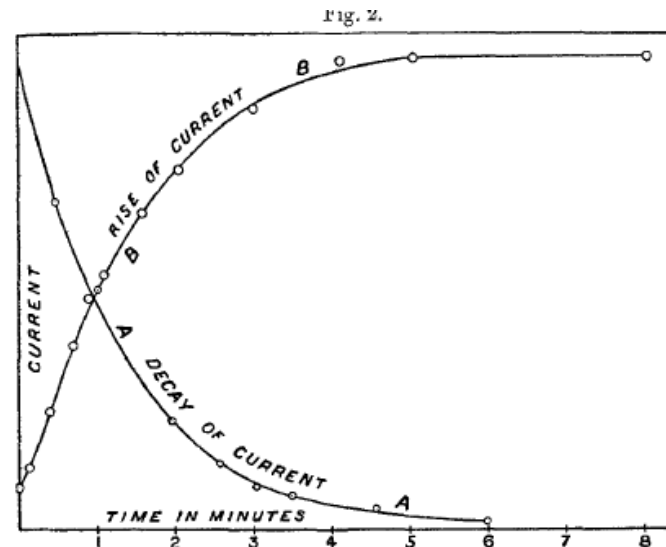
Fig. 2, curve A, shows the relation existing between the current through the gas and the time. The current, just before the flow of air is stopped, is taken as unity. It will be observed that the current through the gas diminishes in a geometrical progression with the time. It can easily be shown,

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The results are expressed in fig. 2, curve B, where the ordinate represents current and the abscissa time. It will be observed that the curve of rise of the current is similar in form to the rise of an electric current in a circuit of constant inductance. The current reaches half its value about one minute after the current of air has stopped,—a result which agrees with the equation given, for $e^{-\lambda t} = \frac{1}{2}$ when $t = 60$ seconds (see Table IV.). At the instant of stopping the current of air the current has a definite value, since most of the ions given off by the emanation, before it is blown out of the cylinders, reach the electrodes.

When the source of the emanation is removed, $q = 0$, and the decay of the number of ions produced by the emanation is given by the equation
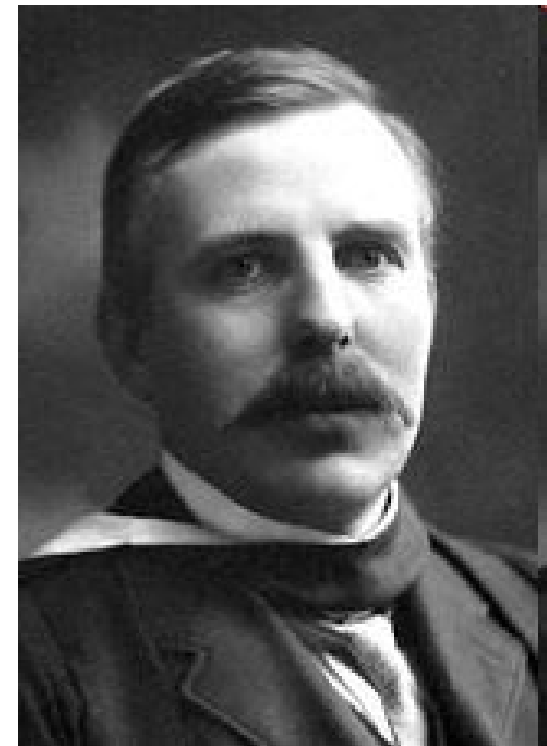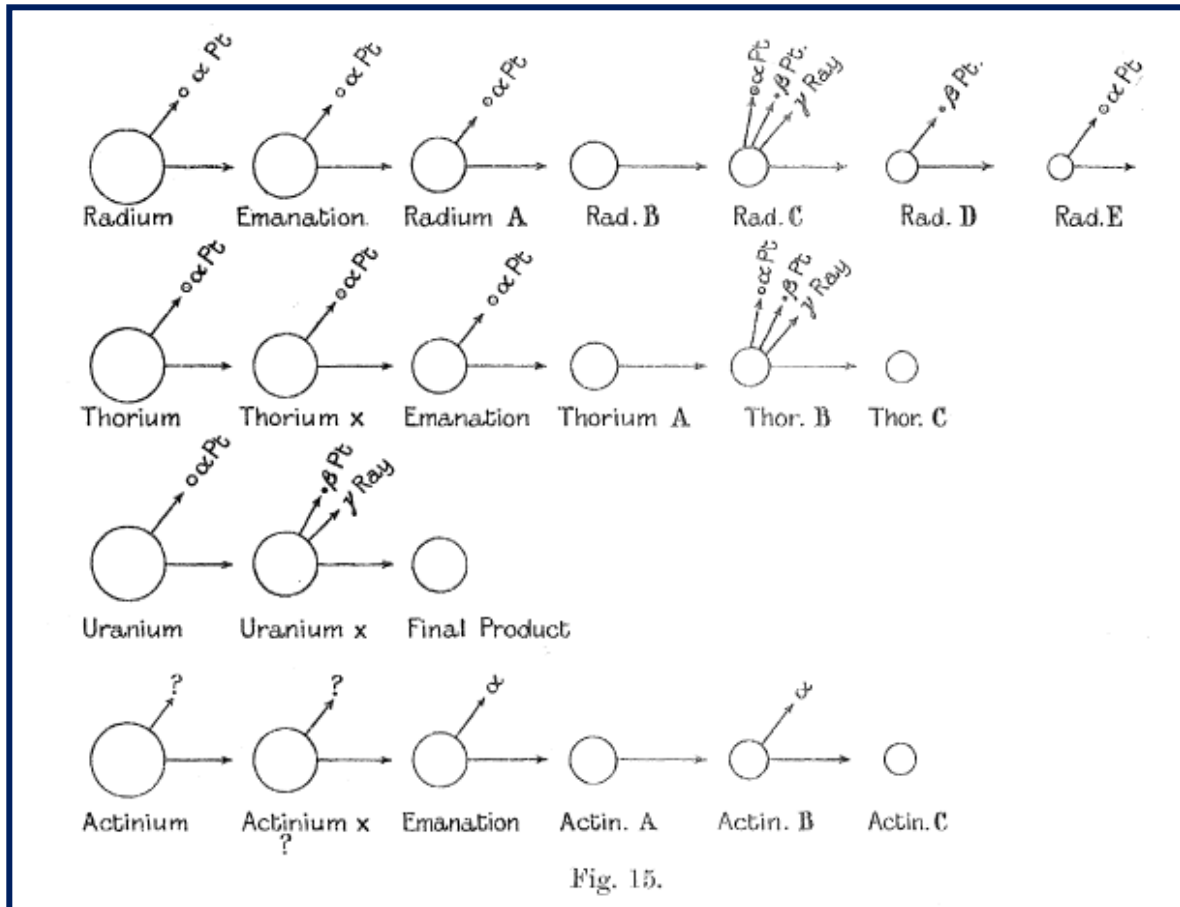
$$\frac{dn}{dt} = -\lambda n.$$

If $n = N$ when $t = 0$, it is easily seen that

$$\frac{n}{N} = e^{-\lambda t},$$

## V. BAKERIAN LECTURE. —*The Succession of Changes in Radioactive Bodies.*

*By Professor* E. RUTHERFORD, *F.R.S., Macdonald Professor of Physics, McGill University, Montreal.*

Fig. 15.

The solution of this equation is of the form

$$Q = n\left(ae^{-\lambda_1 t} + be^{-\lambda_2 t}\right) \quad \cdots \quad \cdots \quad (3).$$

By substitution it is found that $a = \lambda_2/(\lambda_2 - \lambda_1)$.

Since $Q = 0$ when $t = 0$, $b = -\lambda_1(\lambda_2 - \lambda_1)$.
Thus

$$Q = \frac{n\lambda_1}{\lambda_1 - \lambda_2}\left(e^{-\lambda_2 t} - e^{-\lambda_1 t}\right) \quad \cdots \quad \cdots \quad (4).$$

Substituting this value of Q in (2), it can readily be shown that

$$R = n\left(ae^{-\lambda_1 t} + be^{-\lambda_2 t} + ce^{-\lambda_3 t}\right). \quad \cdots \quad \cdots \quad (5),$$

where $a = \dfrac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)(\lambda_1 - \lambda_3)}$, $\quad b = \dfrac{-\lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)(\lambda_2 - \lambda_3)}$, $\quad c = \dfrac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}$.

The variation of the values of P, Q, R with the time $t$ after removal is shown graphically in fig. 8, curves A, B, and C respectively. In order to draw the curves
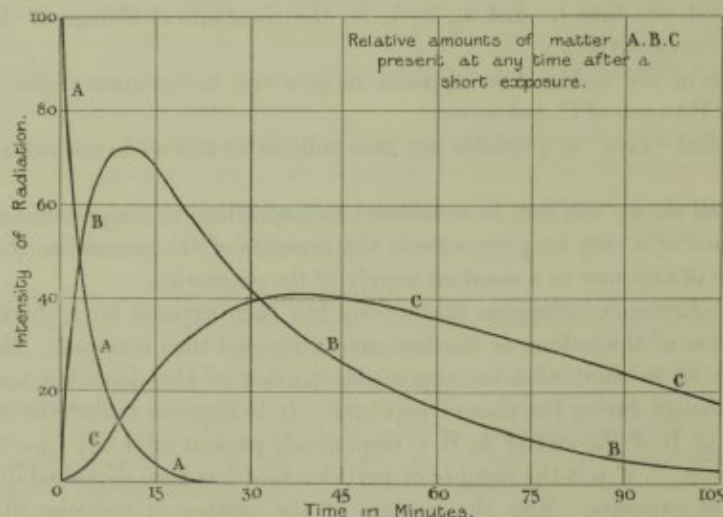


Fig. 8.

for the practical case corresponding to the first three changes in radium A, the values of $\lambda_1$, $\lambda_2$, $\lambda_3$ were taken as $3 \cdot 85 \times 10^{-3}$, $5 \cdot 38 \times 10^{-4}$, $4 \cdot 13 \times 10^{-4}$ respectively, i.e., the times required for each type of matter to be half transformed are about 3, 21, and 28 minutes respectively.
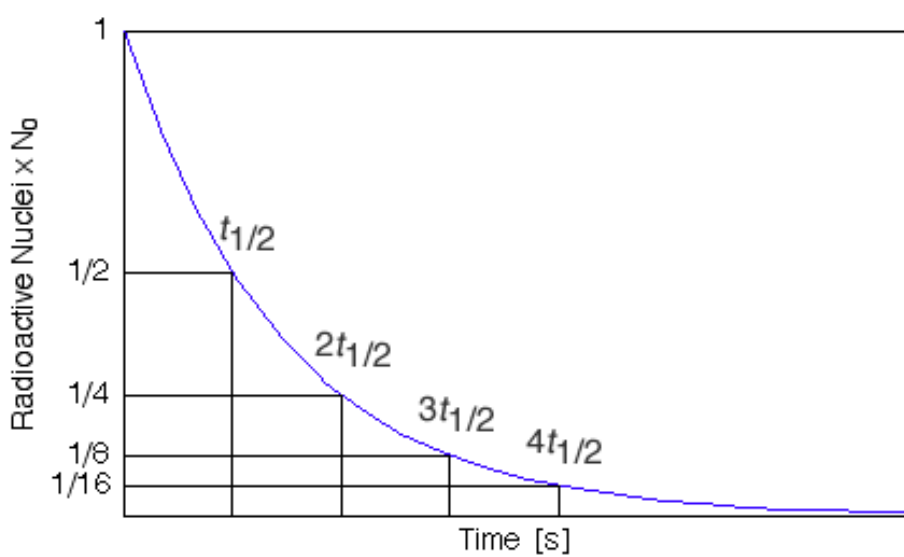
$$A = -dN / dt = \lambda N$$

$$A_{mother}(t) = A_0 \cdot e^{-\lambda \cdot t}$$

$$A_{daughter}(t) = A_0 \cdot \left(1 - e^{-\lambda \cdot t}\right)$$

# What if the 'daughter' nucleus is also radioactive?
# SECULAR EQUILIBRIUM

$$\frac{dN_2}{dt} = -\lambda_2 N_2 + \lambda_1 N_1$$

The solution of this differential equation is:

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^{\,0}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + N_2^{\,0} e^{-\lambda_2 t}$$

and

$$A_2 = \lambda_2 N_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1^{\,0}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)$$



..if $\lambda_2 \gg \lambda_1$ ($T^{1/2}_1 \gg T^{1/2}_2$) then $A_2 \approx A_1$

# THE
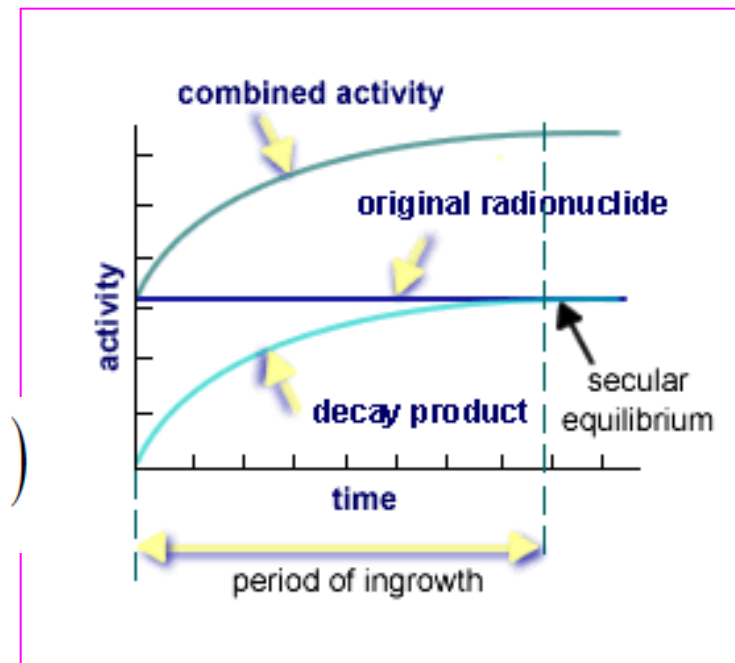
# AMERICAN JOURNAL OF SCIENCE

## [FOURTH SERIES.]

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Art. XXVIII.—*On the Ultimate Disintegration Products of the Radio-active Elements;* by BERTRAM B. BOLTWOOD.

IN a paper by Rutherford and Soddy,* the authors have called attention to the probability that an intimate knowledge of the composition of radio-active minerals will lead to the recognition and identification of the ultimate, stable products formed by the disintegration of the relatively unstable radio-active elements.†

It is an extremely impressive fact that it was from the somewhat meager information available on the occurrence of helium in radio-active minerals, and from the consideration of the data derived from the experiments of one of them on the nature of the expelled alpha particle, that in 1902 the same authors were enabled to make that brilliant prediction of the production of helium‡ which was afterwards confirmed by the experiments of Ramsay and Soddy.

The natural minerals represent chemical systems which are in most instances of extreme antiquity, their original formation having frequently taken place during the earliest geological periods of our planet. With the assistance of the data supplied by geology and mineralogy, it is often possible to assign the origin of a given mineral to some definite geological period and to arrange a series of different individuals roughly in the order

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\* Phil. Mag. (6), v, 576 (1903).

† "In the naturally occurring minerals containing the radio-elements these changes must have been proceeding steadily over very long periods, and, unless they succeed in escaping, the ultimate products should have accumulated in sufficient quantity to be detected, and should therefore appear in nature as the invariable companions of the radio-elements."—*Rutherford and Soddy, loc. cit.*

‡ Phil. Mag. (6), iv, 582.

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Art. VII.—*On the Ultimate Disintegration Products of the Radio-active Elements.  Part II.  The Disintegration Products of Uranium;* by BERTRAM B. BOLTWOOD.

[Contributions from the Sloane Physical Laboratory of Yale University.]

THE general question of the nature of the ultimate disintegration products of the radio-active elements, as indicated by the occurrence of certain chemical elements in the radio-active minerals, has been discussed in an earlier paper,* and it was there pointed out that lead, bismuth and barium might perhaps be included among the possible disintegration products. As more recent experiments† have indicated, however, that actinium is probably an intermediate product between uranium and radium, the number of possible ultimate products has been correspondingly reduced. In addition to this careful examinations have been made of specially selected samples of typical primary uraninites from Branchville, Conn., and Flat Rock, N. C., and of thorianite from Ceylon, which have led to the conclusion that neither bismuth nor barium can be considered as disintegration products in the main line of descent from either uranium or thorium, at least on the basis of the present disintegration theory.

The conditions essential for the identification of the final disintegration products of uranium from a study of the composition of the natural minerals which contain this element would appear to be the following : In unaltered primary minerals of the same species, and of different species from the same locality, that is, in minerals formed at the same time and therefore of equal ages, a constant proportion must exist between the amount of each disintegration product and the

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\* This Journal, xx, 253, 1905.        † Ibid., xxii, 537, 1906.

$$\frac{^{206}\text{Pb}^{*}}{^{238}\text{U}} = e^{\lambda t} - 1$$

# Can calculate the age of rocks (and the earth) via $^{206}$Pb / $^{238}$U ratio: assuming **secular equilibrium**.

$$\frac{^{206}Pb*}{^{238}U} = e^{\lambda t} - 1$$

where $\lambda$ is ln2 $/T_{1/2}$ and $T_{1/2}$ is the decay half-life of $^{238}$U (~4.5x10$^9$ years).

which solves to

$$t = \frac{1}{\lambda}\ln\left(\frac{^{206}Pb*}{^{238}U} + 1\right)$$

Can do the same for $^{235}$U : $^{207}$Pb ratios in the same samples.

# *By 1930, the main 'NORM' decay chains were characterised....*

## THE RADIOACTIVE CONSTANTS AS OF 1930

### REPORT OF THE INTERNATIONAL RADIUM-STANDARDS COMMISSION

By M. Curie, A. Debierne, A. S. Eve, H. Geiger, O. Hahn, S. C. Lind,
St. Meyer, E. Rutherford, and E. Schweidler

### I. INTRODUCTION

FOLLOWING the reorganization of the International Union of Chemistry and of the International Atomic Weights Commission, the need has arisen for the publication of special Tables of the Radioactive Constants.

This responsibility has been assumed by the International Radium Standards Commission chosen in Brussels in 1910, which has expressed its willingness to cooperate with the International Union.

Besides the members, M. Curie, A. Debierne, A. S. Eve, H. Geiger, O. Hahn, S. C. Lind, St. Meyer, E. Rutherford, E. Schweidler, the following have taken part as experts: J. Chadwick, I. Joliot-Curie, K. W. F. Kohlrausch, A. F. Kovarik, L. W. McKeehan, L. Meitner and H. Schlundt, to whom it is desired to express especial obligations.

The following report will be simultaneously published* also in the Physikalische Zeitschrift, in the Journal of the American Chemical Society, Philosophical Magazine, and Journal de Physique et le Radium.

Symbols:

UI, UX$_1$, UX$_2$, UII, Io, Ra, Rn, RaA, RaB, RaC', RaC'', RaD, RaE, RaF = Po, RaG, UY, UZ

Th, MsTh$_1$, MsTh$_2$, RdTh, ThX, Tn, ThA, ThB, ThC', ThC'' ThD, AcU, Pa, Ac, RdAc, AcX, An, AcA, AcB, AcC, AcC', AcC'', AcD,

Pa is for protactinium (not proto-actinium)

Em is the joint symbol for Rn, Tn, and An

| | | $T$ | $\lambda$ | $\tau$ | Literature |
|---|---|---|---|---|---|
| Thorium | Th At. Wt. 232.12 At. No. 90 P. No. 232 | $1.8 \cdot 10^{10}$yr $5.6 \cdot 10^{17}$s | $4.0 \cdot 10^{-11}$yr$^{-1}$ $1.2 \cdot 10^{-18}$s$^{-1}$ | $2.5 \cdot 10^{10}$yr $8.0 \cdot 10^{17}$s | 33 |
| Mesothorium 1 | MsTh$_1$ At. No. 88 P. No. 228 | 6.7yr $2.1 \cdot 10^8$s | 0.103yr$^{-1}$ $3.26 \cdot 10^{-9}$s$^{-1}$ | 9.7yr $3.05 \cdot 10^8$s | |
| Mesothorium 2 | MsTh$_2$ At. No. 89 P. No. 228 | 6.13h $2.21 \cdot 10^4$s | 0.113h$^{-1}$ $3.14 \cdot 10^{-5}$s$^{-1}$ | 8.84h $3.18 \cdot 10^4$s | 34 |
| Radiothorium | RdTh At. No. 90 P. No. 228 | 1.90yr $6.0 \cdot 10^7$s | 0.365yr$^{-1}$ $1.16 \cdot 10^{-8}$s$^{-1}$ | 2.74yr $8.65 \cdot 10^7$s | 35 |
| Thorium X | ThX At. No. 88 P. No. 224 | 3.64d $3.14 \cdot 10^5$s | 0.190d$^{-1}$ $2.20 \cdot 10^{-6}$s$^{-1}$ | 5.25d $4.54 \cdot 10^5$s | |
| Thoron | Tn At. No. 86 P. No. 220 | 54.5s | $1.27 \cdot 10^{-2}$s$^{-1}$ | 78.7s | 36 |
| Thorium A | ThA At. No. 84 P. No. 216 | 0.14s | 4.95s$^{-1}$ | 0.20s | 37 |
| Thorium B | ThB At. No. 82 P. No. 212 | 10.6h $3.82 \cdot 10^4$s | $6.54 \cdot 10^{-2}$h$^{-1}$ $1.82 \cdot 10^{-5}$s$^{-1}$ | 15.3h $5.51 \cdot 10^4$s | |
| Thorium C | ThC At. No. 83 P. No. 212 | 60.5m $3.63 \cdot 10^3$s | $1.15 \cdot 10^{-2}$m$^{-1}$ $1.91 \cdot 10^{-4}$s$^{-1}$ | 87.3m $5.24 \cdot 10^3$s | 38 |
| Thorium C' 65% 65.7% | ThC' At. No. 84 P. No. 212 | $10^{-9}$s (?) $<10^{-6}$s | $10^9$s$^{-1}$ (?) $>10^6$s$^{-1}$ | $10^{-9}$s (?) $<10^{-6}$s | 40 |
| Thorium C'' 35% 34.3% | ThC'' At. No. 81 P. No. 208 | 3.1m 186s | $2.24 \cdot 10^{-1}$m$^{-1}$ $3.73 \cdot 10^{-3}$s$^{-1}$ | 4.47m 286.3s | 39 40 |
| Thorium D Thorium lead Pb208 | ThD At. Wt. 208.016 (?) At. No. 82 P. No. 208 | | | | |

TABLE IV. *Thorium Family*

### IV. NUMBER OF ALPHA-PARTICLES Z EMITTED PER SECOND FROM 1 GRAM RA

**Recommended value.**

Use of the value $3.7 \cdot 10^{10}$ is recommended in accord with reference 9.

# Calculation of the value of 1 Curie (Ci) in Bq (becquerels)

1 g of $^{226}$Ra contains $N_A$ / 226 atoms = 1 Ci of activity

$N_A$ = Avogadro's number ($6.02 \times 10^{23}$ per mol)

i.e. 1g of pure $^{226}$Ra contains $2.66 \times 10^{21}$ atoms

Half-life of $^{226}$Ra (to $^{222}$Rn) is $T_{1/2}$ = 1600(7) years.

$T_{1/2}$ = 1600 years = $5.0 \times 10^{10}$ seconds

Activity (1g) = $\lambda.N$ = (ln 2 /$T_{1/2}$(s)) * Numb of atoms in 1 g

A (1g) = ($0.693$ / $5.0 \times 10^{10}$ s) * $2.7 \times 10^{21}$ atoms = $3.7 \times 10^{10}$ s$^{-1}$
        i.e. 1 Ci = $3.7 \times 10^{10}$ Bq

# NORM

**Naturally Occurring Radioactive Materials**

- Two main sub-groups.

  - <u>Primordial</u>, old - here when the earth formed.

    - Singly-decaying nuclei, $^{40}$K, $^{138}$La, $^{176}$Lu, $^{180}$Ta.

    - Chains: $^{232}$Th($^{228}$Ac); $^{235}$U($^{227}$Ac) $^{238}$U($^{226}$Ra)

  - <u>Cosmogenic</u> following cosmic ray interactions)

    - $^{14}$C (via $^{14}$N(n,p)$^{14}$C), $^{7}$Be, $^{26}$Al (spallation)

(Primordial) Radionuclides have **long half-lives** usually due to:

(a) β-decay selection rules / nuclear structure effects and small (allowed) $Q_\beta$ values (e.g. $^{40}$K; $^{138}$La ; $^{176}$Lu ; $^{180}$Ta) and/or

(b) Low $Q_\alpha$ values = long $T_{1/2}$ (Geiger-Nuttall rule) in a decay (e.g., $^{232}$Th)
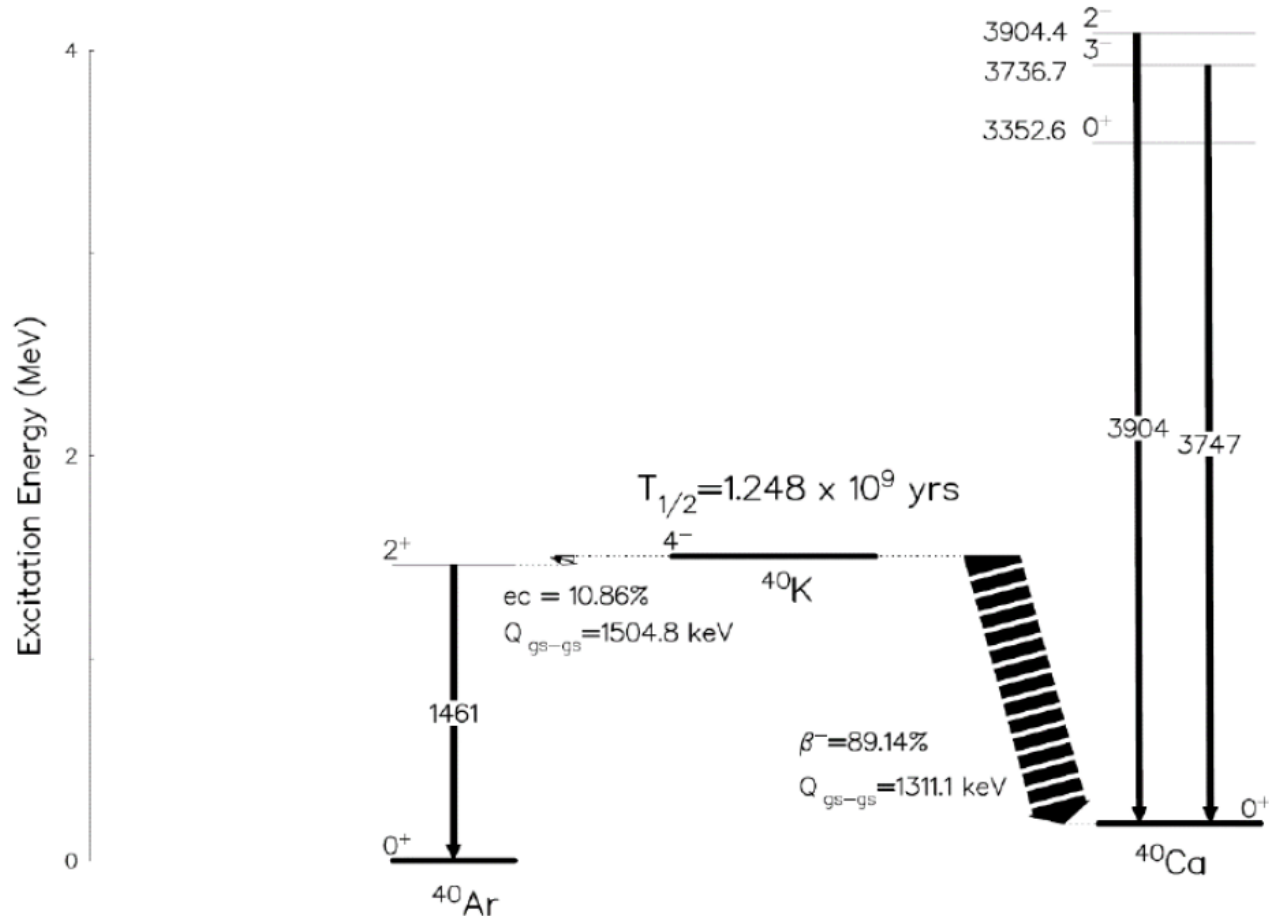
Mass parabolas very useful for understanding NORMS and radioactivity in general....
e.g., Doubly magic nature of $^{40}Ca_{20}$ is responsible for the long half-life of $^{40}K$ due to highly-forbidden $\beta^-$ decay.
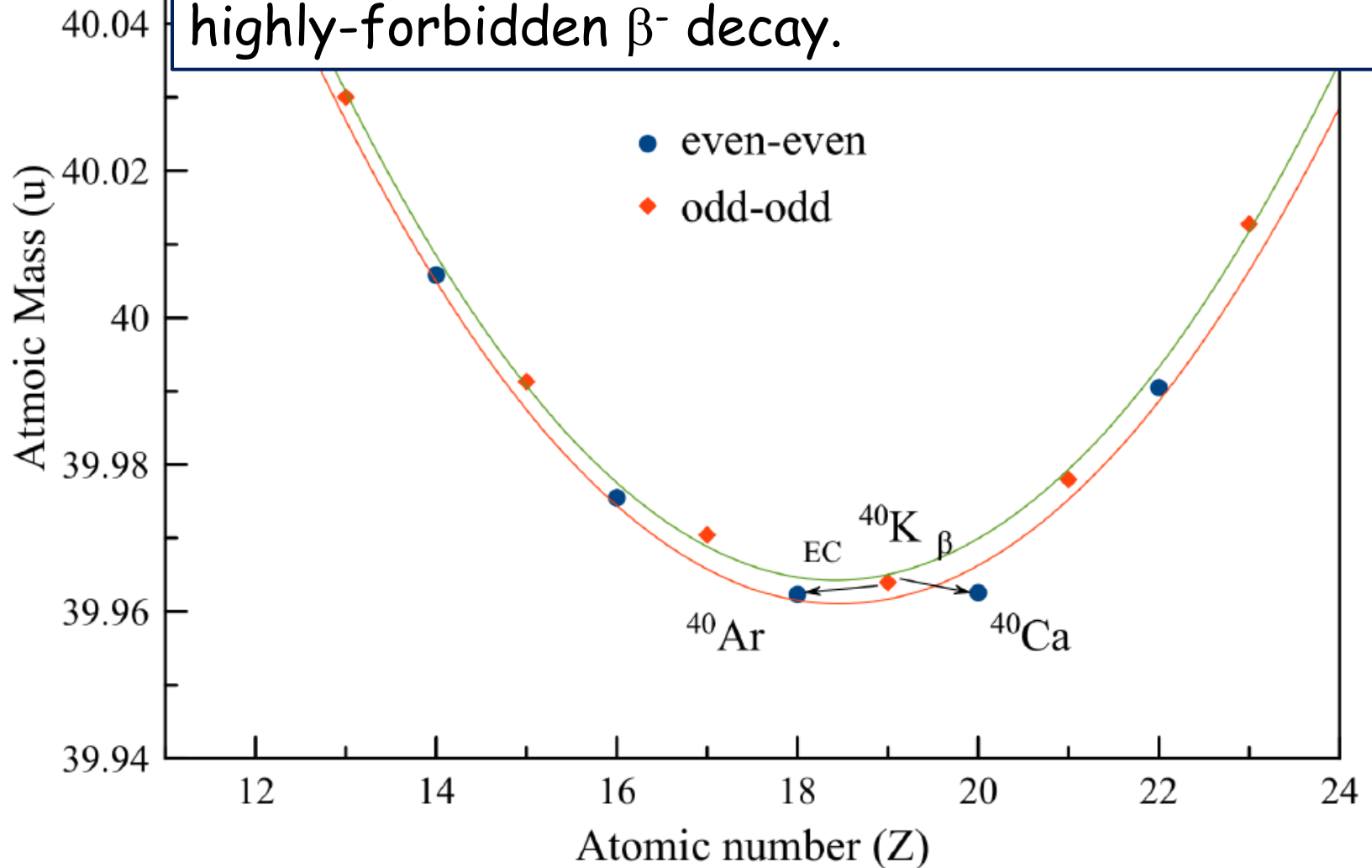
# We can use primordial isotope ratios to age.

- $^{40}K$ decay to $^{40}Ar$ - $T_{1/2}$ for $^{40}K$ is $1.25 \times 10^9$ years.

  - Taking mass ratios using mass spectrographs in rock inclusions, can date the rock using the equation:

$$t = \frac{t_{1/2}}{\ln(2)} \ln\left(1 + \frac{^{40}Ar}{0.109 \cdot ^{40}K}\right)$$

  - The factor 0.109 is due to the 'branching ratio' in the decay of $^{40}K$, i.e., only 10.9% of the time does it decay to $^{40}Ar$.
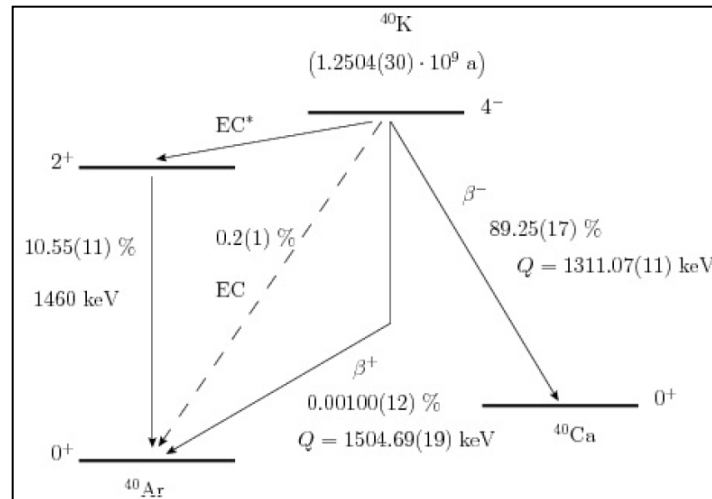
| Nuclide | Half-life (yr) | %Isotopic abundance | Decay mode | Stable disintegration products |
|---|---|---|---|---|
| $^{40}$K | $1.28 \times 10^9$ | 0.0117 | $\beta^+$, EC, $\beta^-$ | $^{40}$Ca, $^{40}$Ar |
| $^{87}$Rb | $4.8 \times 10^{10}$ | 27.83 | $\beta^-$ | $^{87}$Sr |
| $^{113}$Cd | $8 \times 10^{15}$ | 12.2 | $\beta^-$ | $^{113}$In |
| $^{115}$In | $4.4 \times 10^{14}$ | 95.7 | $\beta^-$ | $^{115}$Sn |
| $^{138}$La | $1.02 \times 10^{11}$ | 0.089 | EC, $\beta^-$, $\beta^+$ | $^{138}$Ba, $^{138}$Ce |
| $^{144}$Nd | $2.29 \times 10^{15}$ | 23.8 | $\alpha$ | $^{140}$Ce |
| $^{147}$Sm | $1.06 \times 10^{11}$ | 14.99 | $\alpha$ | $^{143}$Nd |
| $^{148}$Sm | $7 \times 10^{15}$ | 11.24 | $\alpha$ | $^{144}$Nd |
| $^{152}$Gd | $1.08 \times 10^{14}$ | 0.2 | $\alpha$ | $^{148}$Sm |
| $^{176}$Lu | $3.76 \times 10^{10}$ | 2.599 | $\beta^-$ | $^{176}$Hf |
| $^{174}$Hf | $2 \times 10^{15}$ | 0.16 | $\alpha$ | $^{170}$Yb |
| $^{187}$Re | $4.33 \times 10^{10}$ | 62.60 | $\beta^-$ | $^{187}$Os |
| $^{190}$Pt | $6.5 \times 10^{11}$ | 0.0122 | $\alpha$ | $^{186}$Os |

# Cosmogenic radionuclides in the environment

| Nuclide | Half-life | Decay Mode | Particle Energy (MeV) | Decay product | Characteristic $\gamma$-decay energy (keV) |
|---------|-----------|------------|------------------------|---------------|---------------------------------------------|
| $^3$H | 12.32$y$ | $\beta$ | 0.0186 | He | * |
| $^7$Be | 53.22$d$ | EC | 0.478 | Li | 477.595 |
| $^{10}$Be | 1.51$x10^6y$ | $\beta$ | 0.555 | B | * |
| $^{14}$C | 5730$y$ | $\beta$ | 0.1565 | N | * |
| $^{22}$Na | 2.602$y$ | $\beta$ | 0.545 | Ne | 1274.537 |
| $^{26}$Al | 7.17$x10^5y$ | EC | 1.16 | Mg | 1808.65, 1129.67 |
| $^{32}$Si | 153$y$ | $\beta$ | 0.213 | P | * |
| $^{32}$P | 14.26$d$ | $\beta$ | 1.7 | S | * |
| $^{35}$S | 87.51$d$ | $\beta$ | 0.167 | Cl | * |
| $^{36}$Cl | 3.01$x10^5y$ | EC | 0.709 | Ar, S | * |
| $^{39}$Ar | 269$y$ | $\beta$ | 0.565 | K | * |
| $^{53}$Mn | 3.7$x10^6y$ | EC | 0.595 | Cr | * |
| $^{81}$Kr | 2.29$x10^5y$ | EC | 0.28 | Br | 190.46 |

Usually created following spallation with cosmic-ray interactions with the atmosphere.

# Nuclear Metrology is Underpinned by evaluated nuclear decay data.

http://www.nucleide.org/DDEP.htm

https://www.nndc.bnl.gov/nudat2/

Alpha decay (sometimes) leaves the A-4 daughter nucleus in **excited states** which can then decay by characteristic _gamma emission_ (or competing IC).
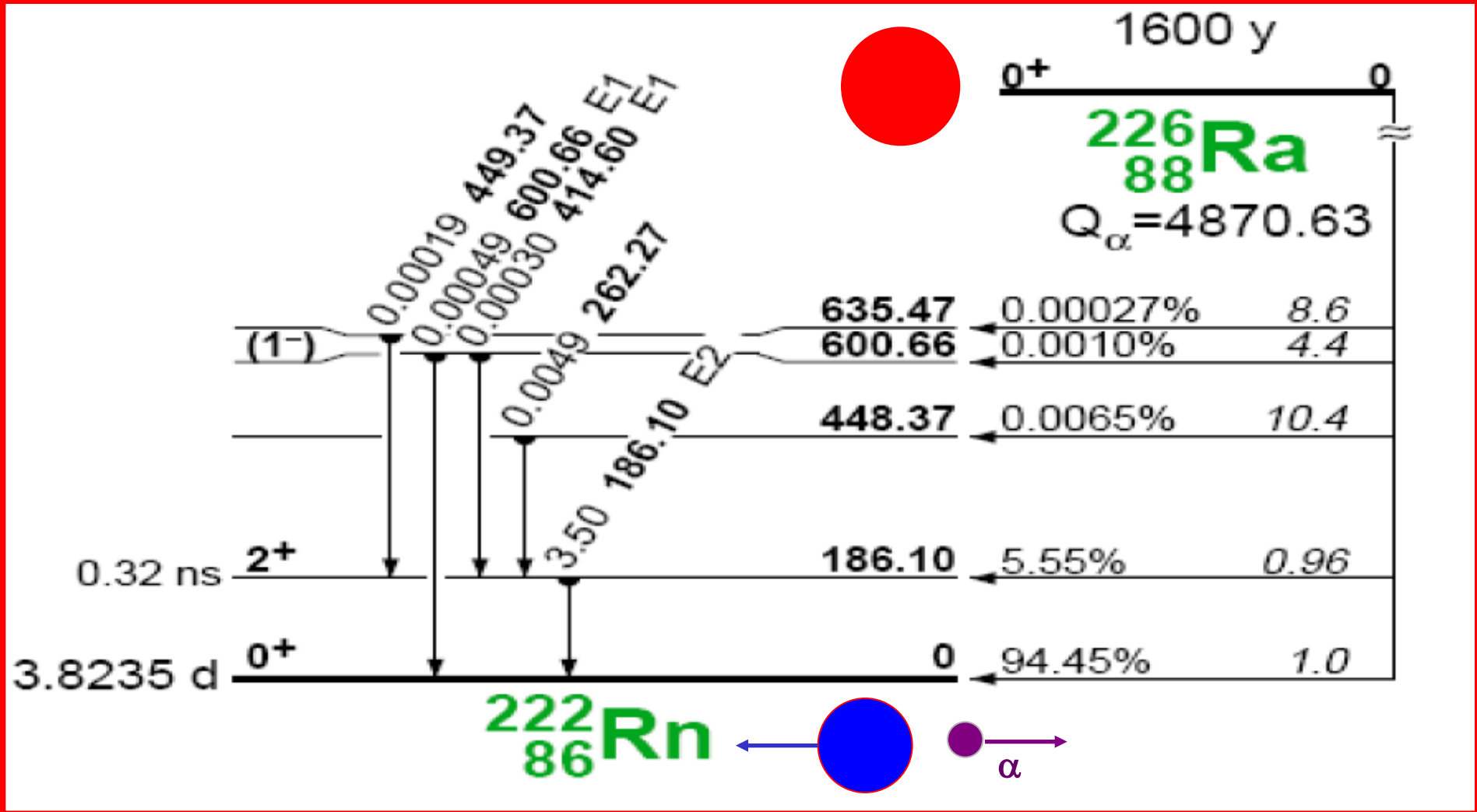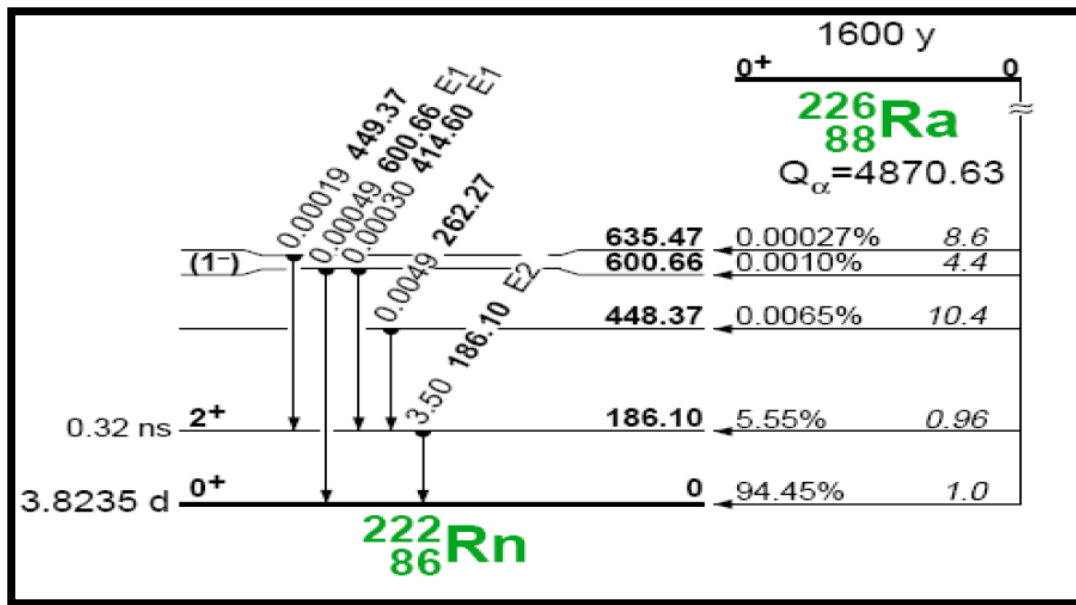
186.1 keV state populated in 5.55% of $^{226}$Ra decays….
Gamma decay competes with internal conversion:
$\alpha_{tot}$ (E2:186 keV) = 0.68

$\alpha = \Gamma_e/\Gamma_\gamma$ ( http://physics.anu.edu.au/nuclear/bricc/ ), then,
$I_{tot}=I_\gamma(1+\alpha)$ = 5.55% → 186.1 keV gamma ray emitted in ~ 3.6% of all $^{226}$Ra decays [3.6% ~ 5.6% / (1+0.68)]

# Underpinned by evaluated nuclear decay data.

Nuclear metrology relies on accurate **Nuclear Decay Data**.

Need to id characteristic signatures of specific decays and understand underlying nuclear (structure) physics.



Introduction: decay schemes (e.g., $^{67}$Cu)

# Underpinned by evaluated nuclear decay data.



http://www.nucleide.org/DDEP.htm

# Primordial Decay Chains

# ($^{235,238}$U, $^{232}$Th)

# Natural decay 'chains'.

Sequences of $\alpha$ and $\beta$ decaying radioisotopes from Uranium (Z=92) or Thorium (Z=90) to Lead (Z=82).

On earth since formation....isotopic ratios (e.g. for $^{235/238}U$) used to age the earth. Longest lived members start the chains:

$T_{1/2}$ ($^{238}U$) ~4.5x10$^9$ yr
$T_{1/2}$ ($^{235}U$) ~7x10$^8$ yr
$T_{1/2}$($^{232}Th$) ~1.4x10$^{10}$ yr

**4n Thorium-232 Decay Series**

Neutron Number (N) vs Proton Number (Z)

Th-232
1.40 × 10$^{10}$ years
100% α decay

Ra-228
5.75 years
100% β⁻ decay

Ac-228
21.72 years
100% β⁻ decay

Th-228
1.92 years
100% α decay

Ra-224
3.63 days
100% α decay

Rn-220
55.6 seconds
100% α decay

Po-216
0.145 seconds
100% α decay

Pb-212
10.64 hours
100% β⁻ decay

Bi-212
61 minutes
64% β⁻ decay / 36% α decay

Tl-208
3.05 minutes
100% β⁻ decay

Po-212
3x10⁻⁷ seconds
100% α decay

Pb-208
STABLE

**4n+2 Uranium-238 Decay Series**

Neutron Number (N) vs Proton Number (Z)

U-238
4.46x10$^9$ years
100% α decay

Th-234
24.1 days
100% β⁻ decay

Pa-234
24.1 days
100% β⁻ decay

U-234
2.46x10$^5$ years
100% α decay

Th-230
7.54x10$^4$ years
100% α decay

Ra-226
1600 years
100% α decay

Rn-222
3.83 days
100% α decay

Po-218
3.1 minutes
99.98% α decay / 0.02% β⁻ decay

At-218
2.0 seconds
100% α decay

Bi-214
19.9 minutes
99.98% β⁻ decay/ 0.02% α decay

Pb-214
26.8 minutes
100% β⁻ decay

Po-214
1.64x10⁻⁴ days
100% α decay

Pb-210
22.2 years
β⁻/ α decay

Tl-210
3.05 minutes

Bi-210
5.01 days
β⁻/ α decay

Po-210
138 days
100% α decay

Hg-206
8.32 minutes

Tl-206
4.2 minutes

Pb-206
STABLE

Figure 2.10 The Thorium ($^{232}$Th) decay series

4n = thorium ($^{232}$Th) chain.

Q.
$T_{1/2}$ ($^{232}$Th) = 1.40(1) x 10$^{10}$ yrs
$T_{1/2}$ ($^{232}$U) = 68.9(4) yrs

Why is $^{232}$Th longer lived?

H. Geiger and J.M. Nuttall (1911) "The ranges of the a particles from various radioactive substances and a relation between range and period of transformation," *Philosophical Magazine*, Series 6, vol. 22, no. 130, pages 613-621.  See also: H. Geiger and J.M. Nuttall (1912) "The ranges of a particles from uranium," *Philosophical Magazine*, Series 6, vol. 23, no. 135, pages 439-445.

| Substance. | Range at 0°. | Initial Velocity. | Transformation Constant. | Half-value Period. |
|---|---|---|---|---|
| Uranium | 2·58 cms. | $1·51 \times 10^9 \frac{cm.}{sec.}$ | $4·6 \times 10^{-18}$ | $5 \times 10^9$ years |
| Ionium | 2·84 ,, | 1·56 ,, ,, | | |
| Radium | 3·13 ,, | 1·61 ,, ,, | $1·1 \times 10^{-11}$ | 2000 years |
| Ra Emanation | 3·94 ,, | 1·74 ,, ,, | $2·085 \times 10^{-6}$ | 3·85 days |
| Radium A | 4·50 ,, | 1·81 ,, ,, | $3·85 \times 10^{3}$ | 3·0 minutes |
| Radium C | 6·57 ,, | 2·06 ,, ,, | | |
| Polonium | 3·58 ,, | 1·68 ,, ,, | $5·60 \times 10^{-8}$ | 143 days |
| Radioactinium | 4·55 ,, | 1·82 ,, ,, | $4·1 \times 10^{-7}$ | 19·5 days |
| Actinium X | 4·17 ,, | 1·77 ,, ,, | $7·6 \times 10^{-7}$ | 10·5 days |
| Act Emanation | 5·40 ,, | 1·93 ,, ,, | $1·8 \times 10^{-1}$ | 3·9 seconds |
| Actinium A | 6·16 ,, | 2·02 ,, ,, | 350 | $\frac{1}{355}$ second |
| Actinium C | 5·12 ,, | 1·90 ,, ,, | $5·4 \times 10^{-3}$ | 2·15 minutes |


Fig. 3.

## LVII. *The Ranges of the α particles from Various Radioactive Substances and a Relation between Range and Period of Transformation.* By H. GEIGER, *Ph.D.*, and J. M. NUTTALL, *B Sc., University of Manchester* *.

IT is well known that the α particles from different radioactive substances are characterized by their ranges, *i. e.* by the distance through which they can travel in air at atmospheric pressure. This was first pointed out by Bragg, and the ranges of a number of products have been determined by him and his co-workers. The method applied by Bragg to determine the ranges is well known. By means of a set of parallel tubes placed directly above the active plate α rays with practically parallel paths were obtained. The ionization produced by these rays was measured at different distances in a shallow ionization vessel, and the distance in air at which the ionization just disappeared was taken as the range of the α particles.

It was shown by Rutherford that at the same distance from the source at which the α particles fail to produce ionization they also lose their power of producing scintillations. The observation of the scintillations at different distances from the source therefore presents another way of determining the ranges of the α particles, and this method has frequently been made use of by Hahn and other observers. It appears, however, that the scintillations method gives somewhat smaller values for the ranges than the ionization method.

Great difficulty has been experienced in the determination of the ranges of the very inactive substances uranium and thorium. In these cases, the methods mentioned above are not applicable. Estimates of the ranges of these products were, however, made by Bragg†, but more accurate values

* Communicated by Prof. E. Rutherford, F.R.S.
† W. H. Bragg, Phil. Mag. xi. p. 754 (1906).

$$\ln \lambda = -a_1 Z E^{-1/2} + a_2,$$

# The Geiger-Nuttall Rule – explains long lived α-decays

There are relatively small variations for **a decay energies** across all measured, heavy cases.

Typical a decay energies lie between 4 and 9 MeV for heavy nuclei.

The corresponding range in radioactive **decay half-lives** is by contrast, ENORMOUS, ranging from > $10^{18}$ years to less than microseconds.

When plotted on a semi-log plot of decay half-life verses $T_\alpha$, a correlation becomes clear.
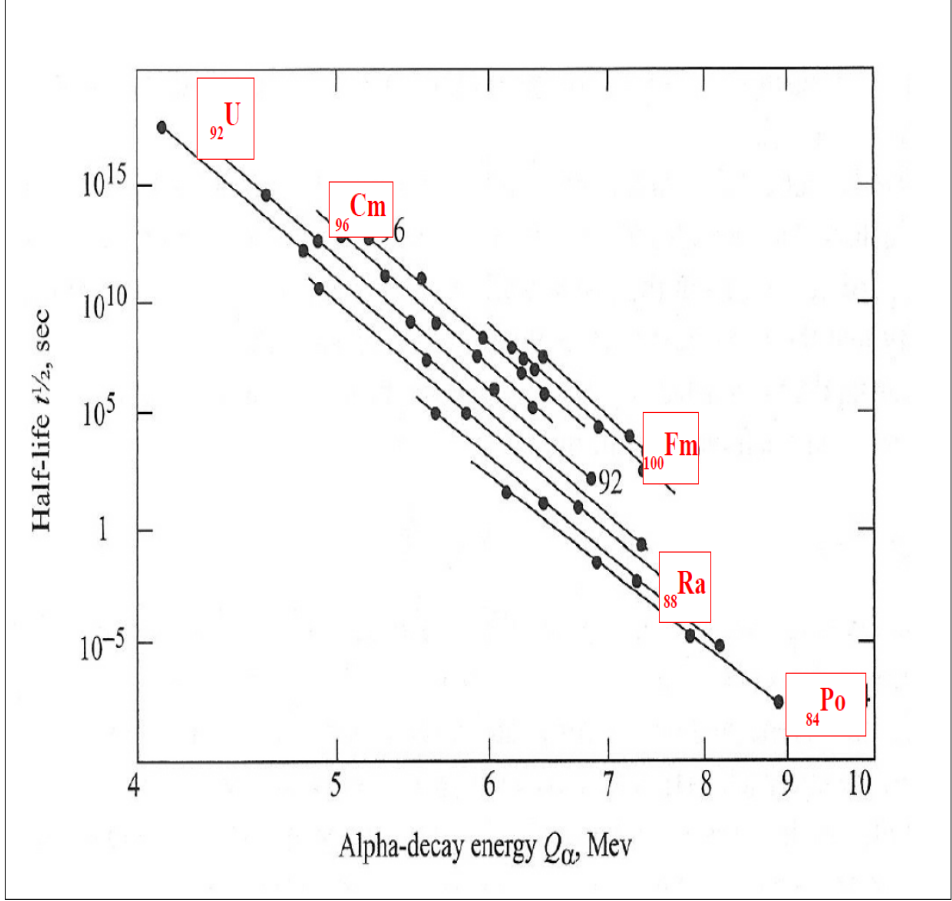


This empirical relation underpins understanding of the α decay process in terms of a <u>NUCLEAR QUANTUM MECHANICAL TUNNELLING PROCESS.</u>

Aside: How do we measure the longest half-lives?

- $^{238}$U: $T_{1/2}$ = 4.5x10$^9$ years

- $^{232}$Th: $T_{1/2}$=1.4x10$^{10}$ years

- $^{40}$K: $T_{1/2}$ = 1.25x10$^9$ years

How do we know ?

Too long to see how long for ½ to decay..

Measure **absolute activity** (A) and number of atoms (N) and use $A = \lambda N$

# Half-life of $^{176}$Lu

G. F. Grinyer,[1,2,*] J. C. Waddington,[2,†] C. E. Svensson,[1] R. A. E. Austin,[2] G. C. Ball,[3] G. Hackman,[3] J. M. O'Meara,[1]
C. Osborne,[3] F. Sarazin,[3] H. C. Scraggs,[3] and H. D. H. Stöver[4]

[1]Department of Physics, University of Guelph, Guelph, Ontario, Canada N1G 2W1
[2]Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada L8S 4K1
[3]TRIUMF, 4004 Wesbrook Mall, Vancouver, British Columbia, Canada V6T 2A3
[4]Department of Chemistry, McMaster University, Hamilton, Ontario, Canada L8S 4K1

The half-life of $^{176}$Lu has been measured using a $\gamma$-$\gamma$ coincidence technique and was found to be $T_{1/2}$ = $(4.08\pm0.03)\times10^{10}$ years. The method employed eliminates most of the uncertainties associated with detector efficiencies, solid angle coverage, internal conversion, self-absorption, angular correlations, and true coincidence summing.
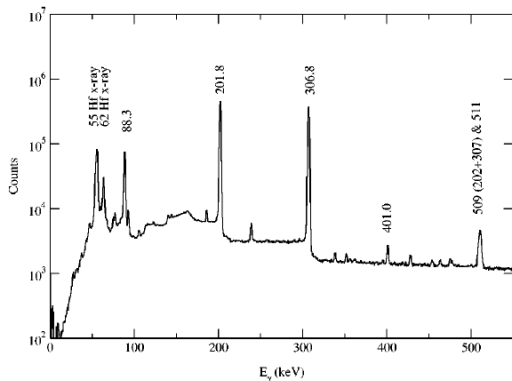


FIG. 2. Singles spectrum from the 48 h experiment showing $^{176}$Lu $\gamma$ rays and associated x rays. Other peaks represent room background $\gamma$ rays that were present during the experiment.
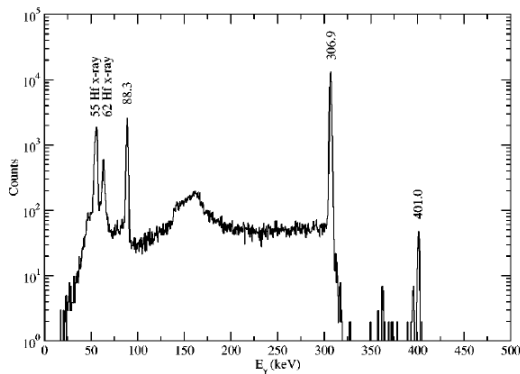


FIG. 3. Coincidence spectrum from the 48 h experiment gated on the 202 keV $\gamma$ ray.



$$\lambda N = \frac{A_1 A_2}{(1-\delta)(1-\sigma)C_{1,2}^{Tot}},$$

TABLE II. Data from the 48 h $^{176}$Lu decay experiment with the $8\pi$ spectrometer.

| Quantity | Description | Result |
|---|---|---|
| $A_1$ | singles rate of the 202 keV $\gamma$ ray | $8.653\pm0.010$ s$^{-1}$ |
| $A_2$ | singles rate of the 307 keV $\gamma$ ray | $9.985\pm0.011$ s$^{-1}$ |
| $C_{1,2}$ | coincidence rate from a gate on the 202 keV $\gamma$ ray | $0.3125\pm0.0014$ s$^{-1}$ |
| $A_{1+2}$ | singles rate of the sum (202+307) photopeak | $0.0209\pm0.0004$ s$^{-1}$ |
| $C_{1,2}^{Tot}$ | total coincidence rate | $0.3334\pm0.0015$ s$^{-1}$ |
| $\sigma$ | angular correlation correction factor | $0.0018\pm0.0004$ |
| $\delta$ | correction factor in Eq. (9) | $0.023\pm0.005$ |
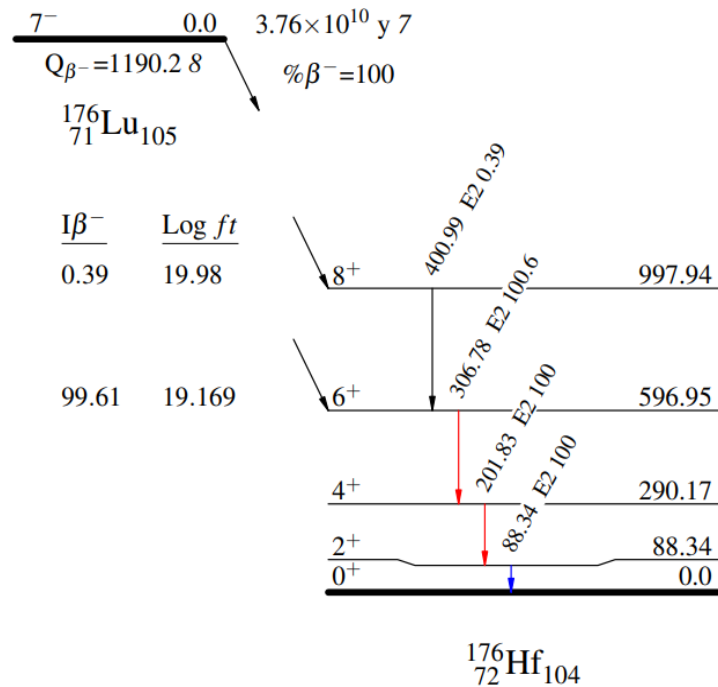
N ($^{176}$Lu) = 4.938(23)x10$^{20}$

by mass and ICP-MS
isotope distribution Analysis.

# Precision Measurement of Half-Lives and Specific Activities of $^{235}$U and $^{238}$U[†]

A. H. Jaffey, K. F. Flynn, L. E. Glendenin, W. C. Bentley, and A. M. Essling

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(Received 29 April 1971)

New determinations of the half-lives of $^{235}$U and $^{238}$U have been made. Improved techniques have allowed the half-life values to be measured with greater accuracy than has been heretofore achieved. Samples were prepared by molecular plating and counted in a intermediate-geometry $\alpha$-proportional counter with an extremely flat pulse-height plateau. The small amount of residual nonplated uranium was counted in a $2\pi$ counter. Energy analysis with a silicon-junction detector was used to measure the presence of "foreign" activities. For $^{235}$U, the measured specific activity was $(4798.1 \pm 3.3)$ (dis/min)/(mg $^{235}$U), corresponding to a half-life of $(7.0381 \pm 0.0048) \times 10^8$ yr. For $^{238}$U, the specific activity was measured as $(746.19 \pm 0.41)$ (dis/min)/(mg $^{238}$U), corresponding to a half-life of $(4.4683 \pm 0.0024) \times 10^9$ yr. Errors quoted are statistical (standard error of the mean), based upon the observed scatter of the data. This scatter exceeds that expected from counting statistics alone. We believe that systematic errors, if present, will no more than double the quoted errors.

# The Disintegration Constant of Thorium and the Branching Ratio of Thorium C

ALOIS F. KOVARIK AND NORMAN I. ADAMS, JR.

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(Received June 30, 1938)

In age determinations of minerals in which thorium predominates, some inconsistencies have appeared when the value used for the disintegration constant of thorium is that adopted by the International Radium-Standard Commission (1930). This value differs from the original Geiger-Rutherford value by 30 percent. The present research was undertaken to determine the disintegration constant of thorium by the method of counting alpha-particles. Easily weighed sources were used. The material was obtained from a thorite from the same lot that was used by Geiger and Rutherford. The thorite contained only a small amount of uranium and correction for the ionium present was made. Various checks on the counting are given. The half-value period of thorium was found to be $1.39(10)^{10}$ years and the branching ratio, thorium C' to thorium (C+C'), is 0.663. A method of impounding the thoron, yet permitting alpha-particles to escape, is described.

# Half-life $T_{1/2}$ for $^{209}$Bi 2.01(8)x$10^{19}$ years

**Experimental detection of α-particles from the radioactive decay of natural bismuth**

Pierre de Marcillac, Noël Coron, Gérard Dambier, Jacques Leblanc & Jean-Pierre Moalic

Institut d'Astrophysique Spatiale, CNRS & Université Paris Sud, UMR 8617, Bât. 121, 91405 Orsay Cedex, France
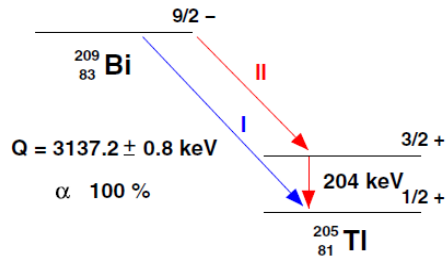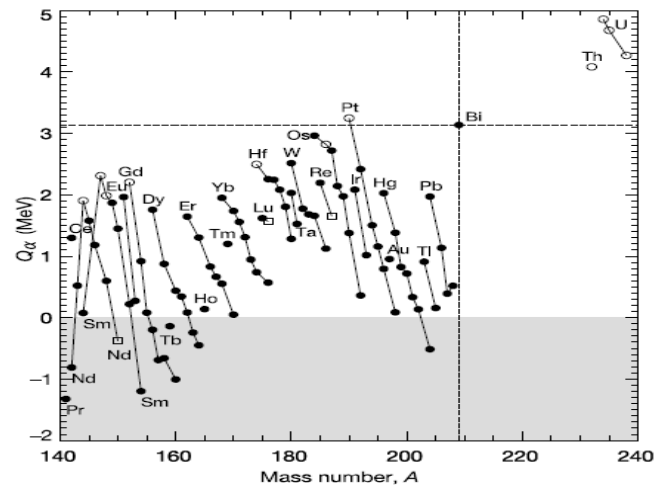
**First Measurement of the Partial Widths of $^{209}$Bi Decay to the Ground and to the First Excited States**

J. W. Beeman,[1] M. Biassoni,[2,3] C. Brofferio,[2,3] C. Bucci,[4] S. Capelli,[2,3] L. Cardani,[5] M. Carrettoni,[2,3] M. Clemenza,[2,3] O. Cremonesi,[3] E. Ferri,[2,3] A. Giachero,[3] L. Gironi,[2,3,*] P. Gorla,[4,6] C. Gotti,[3,7] A. Nucciotti,[2,3] C. Maiano,[2,3] L. Pattavina,[3] M. Pavan,[2,3] G. Pessina,[3] S. Pirro,[3,4] E. Previtali,[2,3] M. Sisti,[2,3] and L. Zanotti[2,3]
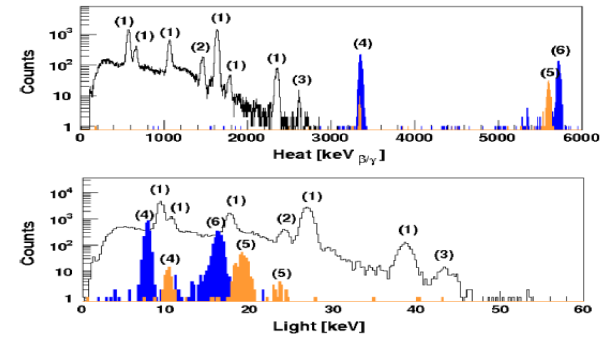


FIG. 3 (color online). Heat (top) and light (bottom) spectra of events belonging to the β/γ band (black histogram) and α band (colored filled histograms). Numbers are used to identify the main lines: (1) is for γ lines of $^{207}$Bi, (2) for those of $^{40}$K, (3) for $^{232}$Th, (4) indicates the α lines of $^{209}$Bi, (5) those of $^{210}$Bi, and (6) that of $^{210}$Po.

Half-life obtained using

$A = -dN/dt = \lambda.N = (\ln 2/T_{1/2}).N$

N = number of $^{209}$Bi atoms in BGO Bolometer
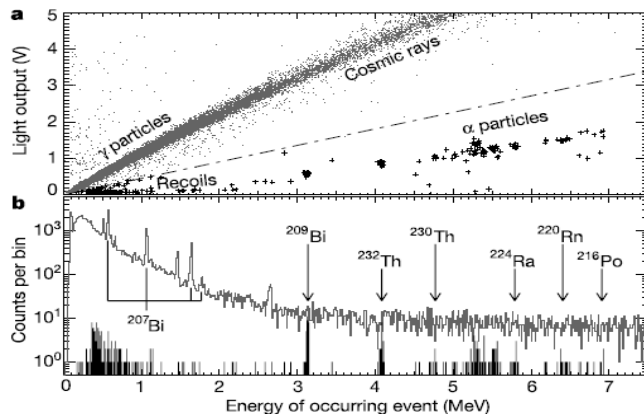
# ASIDE: Relevance of NORMS and decay chains to nuclear (astro)physics?

- $\alpha$ decay of $^{210}$Po decay terminates the s-process

- $^{235,238}$U, $^{232}$Th evidence for the r-neutron process.

- Abundance ratios used to estimate age of earth.

- Possible (pre-weapon) $^{244}$Pu ($T_{1/2}$~80 million years) infers local, recent supernovae / r-process event.

## Measuring the amount of NORM?

We can identify the **decay products** present and determine their <u>activity concentrations</u> ($Ac$) i.e. decays per unit mass of sample by

quantifying the <u>characteristic emission signatures of the decays</u> in the respective decay chains.

# Origins of NORM



Source: IAEA Tecdoc 1712
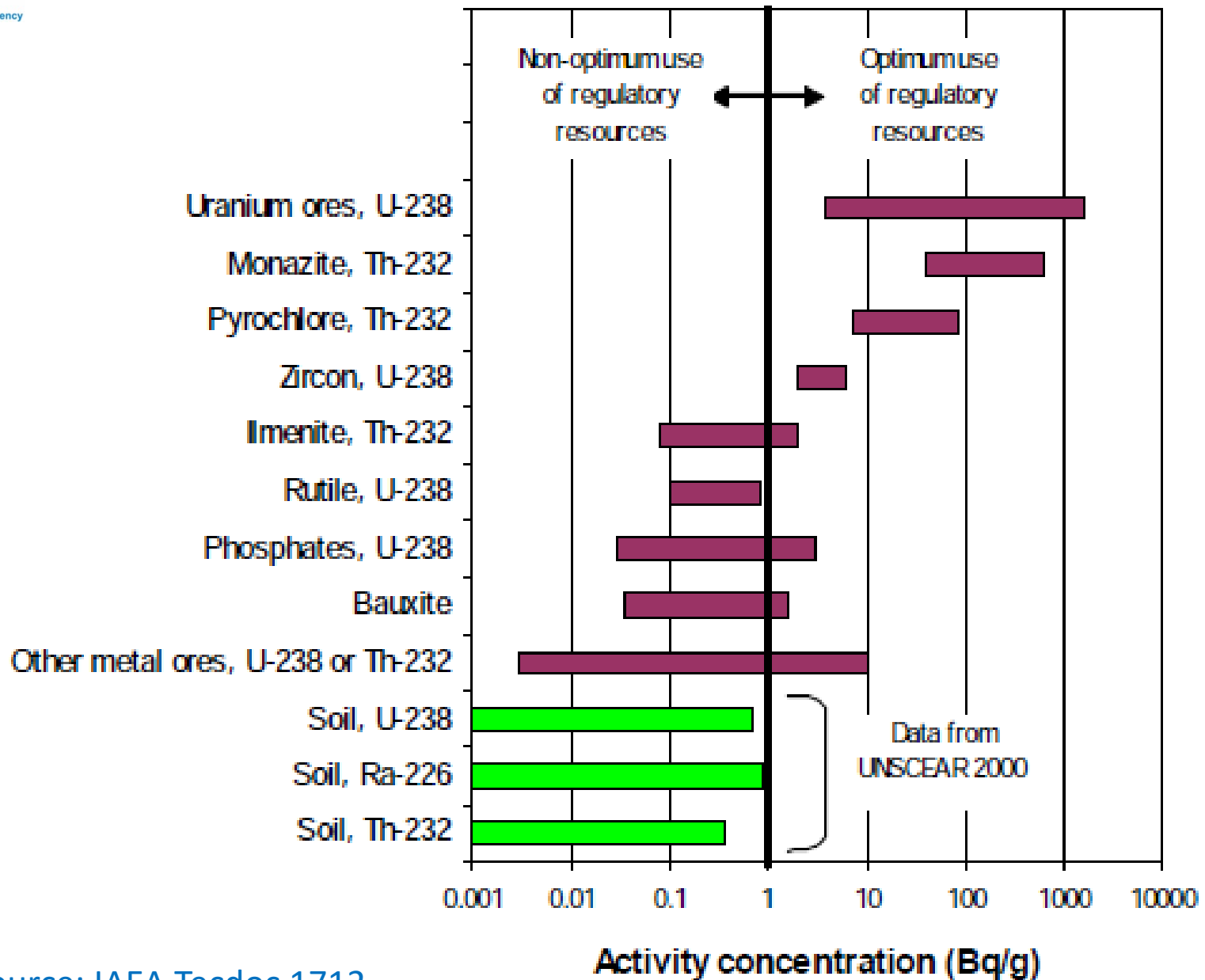
Chemical fractionation of NORM decay chains materials.
Usually result in concentration of Ra and subsequent decay
Products as Ra is water soluble and Rn is a noble gas.
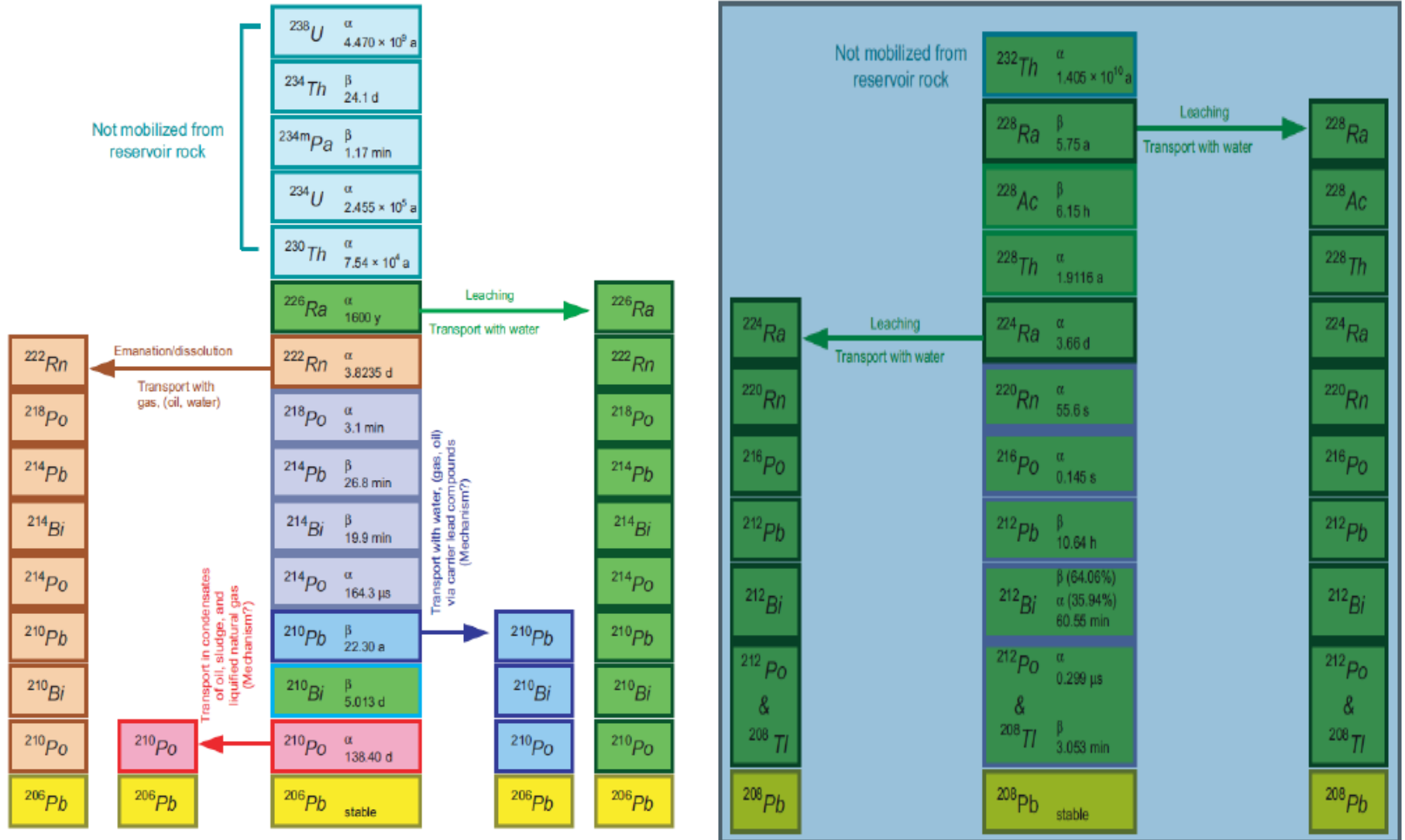
# Thorium ($^{232}$Th) decay chain experimental gamma signatures.
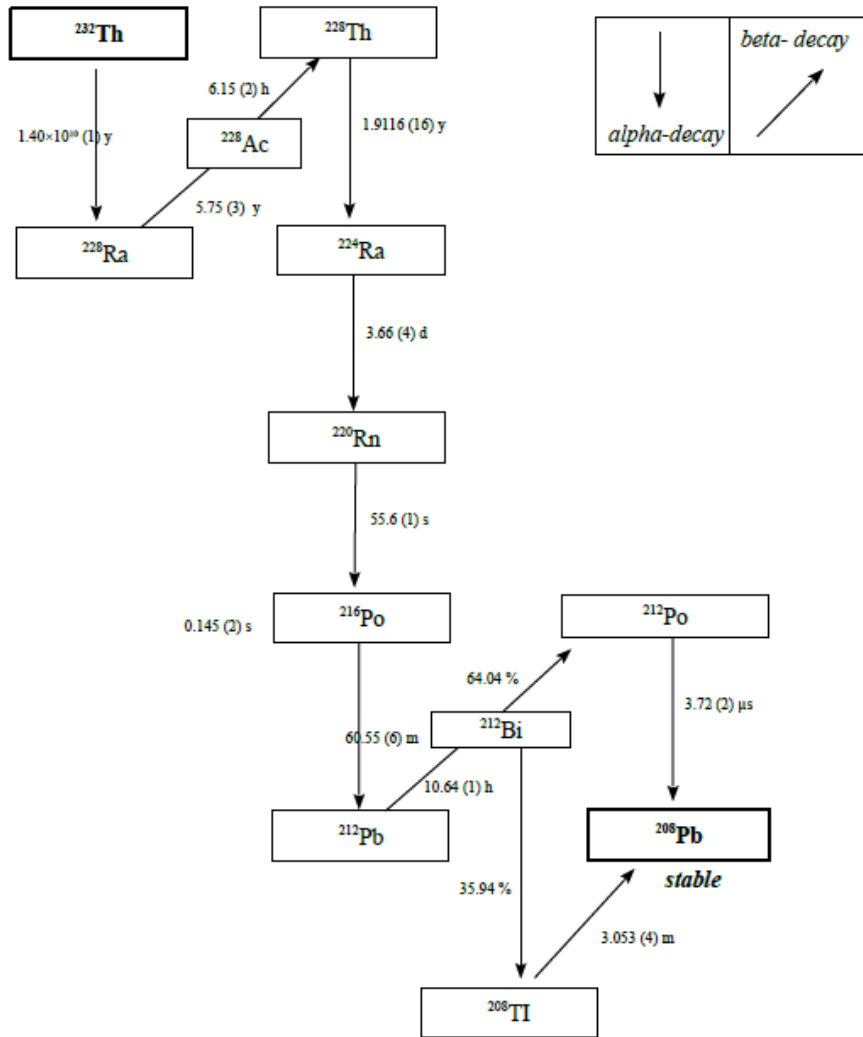


| Nuclide | Half-life | Decay mode | Gamma-ray energy (keV) | Emission probability (%) |
|---|---|---|---|---|
| **Th-232 Series** $^{232}$Th | 1.405 (6)× 10$^{10}$ y | α | 63.81 (2) | 0.263 (13) |
| $^{228}$Ra | 5.75 (3)y | β$^-$ | 13.52 (2) | 1.6 |
| $^{228}$Ac | 6.15 (2)h | β$^-$ | 911.024 (4) | 25.8 (4) |
| | | | 968.971 (17) | 15.8 (3) |
| | | | 338.320 (3) | 11.27 (12) |
| | | | 964.766 (10) | 4.99 (9) |
| | | | 463.004 (6) | 4.40 (7) |
| $^{228}$Th | 1.9116 (16)y | α | 84.373 (3) | 1.22 (2) |
| $^{224}$Ra | 3.66 (4) d | α | 240.986 (6) | 4.10 (5) |
| $^{220}$Rn | 55.6 (1) s | α | 549.76 (4) | 0.114 (17) |
| $^{216}$Po | 0.145 (2) s | α | 804.9 (5) | 0.0019 (3) |
| $^{212}$Pb | 10.64 (1)h | β$^-$ | 238.632 (2) | 43.3 (3) |
| | | | 300.087 (10) | 3.28 (3) |
| $^{212}$Bi | 60.55 (6) m | α, β$^-$ | 727.330 (9) | 6.58 (6) |
| | | | 1620.50 (10) | 1.49 (4) |
| $^{212}$Po | 0.299 (2)μs | α | No γ-ray | - |
| $^{208}$Ti | 3.053 (4) m | β$^-$ | 2614.533 (13) | 35.64 (60 |
| | | | 583.191 (2) | 30.4 (2) |
| | | | 510.77 (10) | 8.13 (2) |
| | | | 860.564 (5) | 4.47 (4) |
| $^{208}$Pb | Stable | - | - | - |

# Uranium ($^{238}$U) decay chain experimental gamma signatures.

$^{238}$U

$^{234}$U

4.468 (3) × 10⁹ y   1.159 (11) m

$^{234m}$Pa

2.455 (6) ×10⁵ y

24.10 (3) d

$^{234}$Th

$^{230}$Th

7.54 (3) x10⁴ y

$^{226}$Ra

1,600 (7) y

$^{222}$Rn

3.82 (3) d

$^{218}$Po       $^{214}$Po       $^{210}$Po

19.9 (4) m   164.3 (20) µs   5.012(5) d

3.098 (12) m    $^{214}$Bi       $^{210}$Bi    138.376 (2) d

26.8 (9) m        22.20 (22) y

$^{214}$Pb       $^{210}$Pb       $^{206}$Pb

stable

beta-decay

alpha-decay

| Nuclide | Half-life | Decay mode | Gamma-ray energy (keV) | Emission probability (%) |
|---|---|---|---|---|
| U-238 series | | | | |
| $^{238}$U | 4.468 (3)× 10⁹y | α | 49.55(6) | 0.063(7) |
| $^{234}$Th | 24.10 (3) d | β⁻ | 63.283 (3) | 4.1 (7) |
| | | | 92.370 (10) | 2.42 (15) |
| | | | 92.793 (10) | 2.39 (15) |
| $^{234m}$Pa | 1.17 (3) m | β⁻ | 1001.03 (3) | 0.837 (10) |
| | | | 766.38 (2) | 0.294 (12) |
| | | | 742.81 (3) | 0.080 (4) |
| $^{234}$U | 2.455 (6)× 10⁵y | α | 53.20 (2) | 0.123 (2) |
| $^{230}$Th | 75380 (30) y | α | 67.672 (2) | 0.373 (21) |
| $^{226}$Ra | 1600 (7) y | α | 186.211 (13) | 3.59 (6) |
| $^{222}$Rn | 3.8235 (3) d | α | 511 (2) | 0.076 |
| $^{218}$Po | 3.10 (1) m | α, β⁻ | No γ-ray | - |
| $^{214}$Pb | 26.8 (9) m | β⁻ | 351.932 (2) | 35.1 (4) |
| | | | 295.224 (2) | 18.2 (2) |
| | | | 241.997 (3) | 7.12 (11) |
| $^{218}$At | 1.6 s | α | No γ-ray | - |
| $^{214}$Bi | 19.9 (4) m | α, β⁻ | 609.312 (7) | 44.6 (5) |
| | | | 1764.494 (14) | 15.1 (2) |
| | | | 1120.287 (10) | 14.7 (2) |
| | | | 1238.110 (12) | 5.78 (8) |
| $^{214}$Po | 164.3 (20) µ s | α | 799.7 (1) | 0.0104 (35) |
| $^{210}$Ti | 1.3 m | β⁻ | No γ-ray | - |
| $^{210}$Pb | 22.3 (2) y | β⁻ | 46.539 (1) | 4.25 (4) |
| $^{210}$Pi | 5.013 (5) d | α, β⁻ | No γ-ray | - |
| $^{210}$Po | 138.376 (2) d | α | 803.10 (5) | 0.00122 (4) |
| $^{206}$Pb | Stable | - | - | - |

# $^{235}$U heads the 4n+3 chain (Actinium)
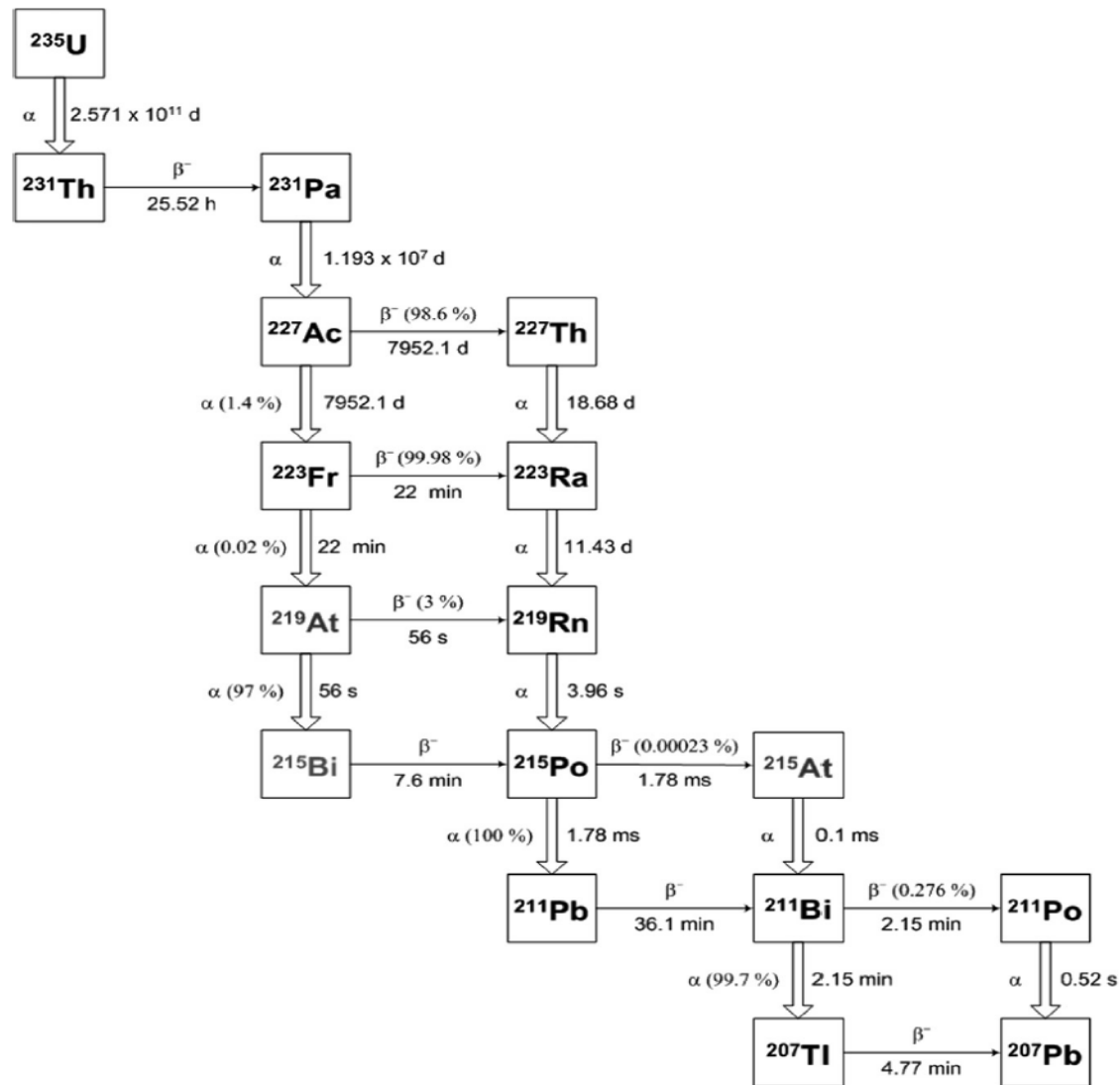
Fig. 1. Decay series of $^{235}$U.

# Discovery of actinium, thorium, protactinium, and uranium isotopes

C. Fry, M. Thoennessen*

National Superconducting Cyclotron Laboratory and Department of Physics and Astronomy, Michigan State University, East Lansing, MI 48824, USA

ARTICLE INFO

ABSTRACT

Thirty-one actinium, thirty-one thorium, twenty-eight protactinium, and twenty-three uranium isotopes have so far been observed; the discovery of these isotopes is described. For each isotope a brief summary of the first refereed publication, including the production and identification method, is presented.

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## $^{227}Ac$

Giesel reported the observation of a new active substance later identified as $^{227}$Ac in the 1902 paper "Ueber Radium und radioactive Stoffe" [22]. A raw sample consisting of barium, strontium, calcium, and small amount of radium was chemical separated and radiation was observed on a zinc sulfide screen.

## $^{227}Th$

A new radioactive substance, later identified as $^{227}$Th, was reported by Hahn in the 1906 paper "A new product of actinium" [67]. Alpha- and beta-ray activities of an actinium solution were measured following chemical separation. "...I have found that a new product is present in actinium which is intermediate between actinium and actinium X, and, from analogy to thorium, will be called for convenience 'radio-actinium.' This product emits $\alpha$ rays, is half-transformed in about twenty days, and is the parent of actinium X". The currently adopted half-life is 18.68(9) days.

[23] F. Giesel, Ber. Deuts. Chem. Ges. 37 (1904) 3963.

[67] O. Hahn, Nature 73 (1906) 559.
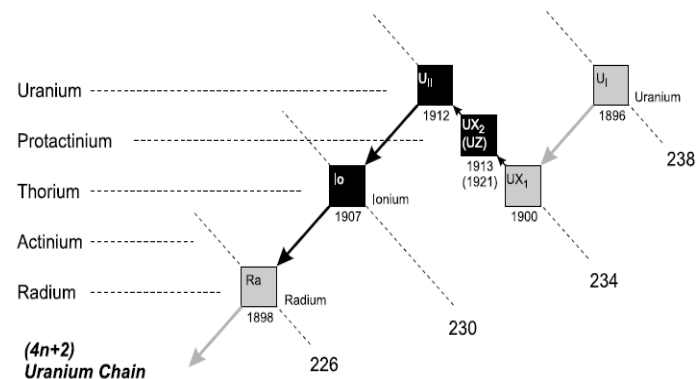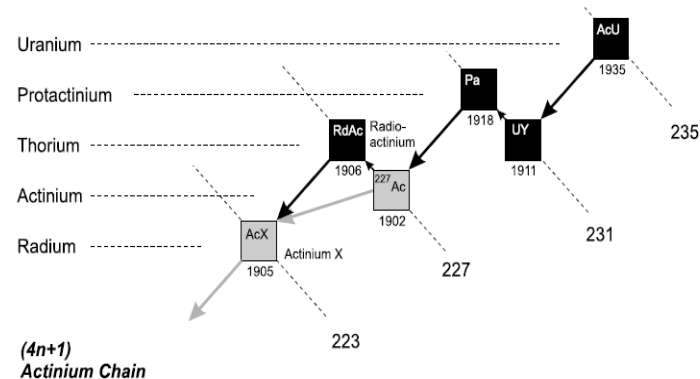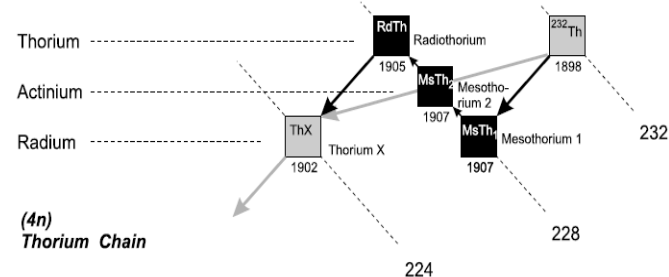
Fig. 1. Original nomenclature used to describe radium, actinium, thorium, protactinium, and uranium isotopes within the three natural occurring radioactive decay series. The gray squares connected by the gray arrows depict the activities labeled by Rutherford in his Bakerian lecture [19]. The black squares correspond to radioactive substances discovered later.

*The $^{235}$U parent of $^{227}$Ac and $^{223}$Ra was identified ~30 years later.....*

$^{235}U$

In the 1935 article "Isotopic constitution of uranium" Dempster reported the discovery of $^{235}$U in natural uranium[131]. Uranium samples were used in a spark source of the Chicago mass spectrograph. "It was found that an exposure of a few seconds was sufficient for the main component at 238 reported by Dr. Aston; but in addition on long exposures a faint companion of atomic weight 235 was also present. With two different uranium electrodes it was observed on eight photographs, and two photographs with the pitchblende electrode also showed the new component". $^{235}$U is of primordial origin with a half-life of $7.01(1) \times 10^8$ year and an abundance of 0.72%. Rutherford had predicted the existence of $^{235}$U in 1929 naming it actino-uranium [132] based on Aston's extrapolation of 231 for the mass of protactinium as the precursor of $^{207}$Pb [133].

[131] A.J. Dempster, Nature 136 (1935) 180.
[132] E. Rutherford, Nature 123 (1929) 313.
[133] F.W. Aston, Nature 123 (1929) 313.

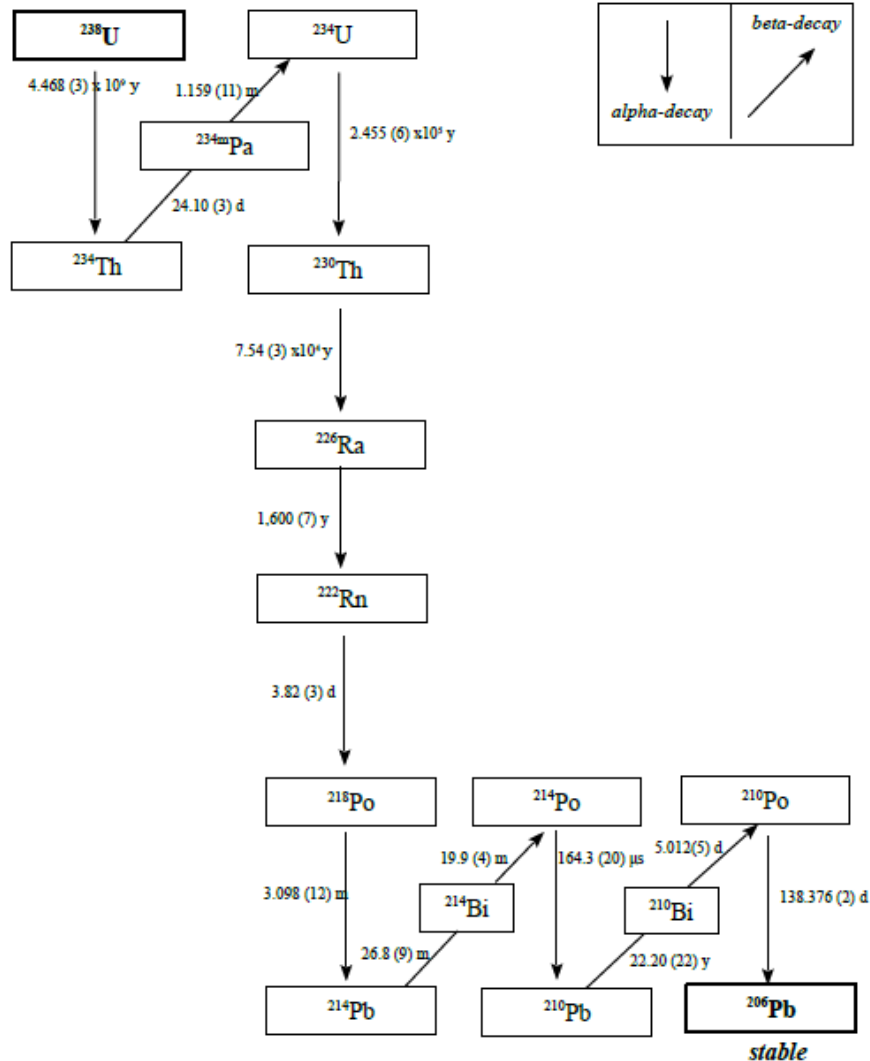# Actinium ($^{235}$U / 227Ac) decay chain experimental gamma signatures.
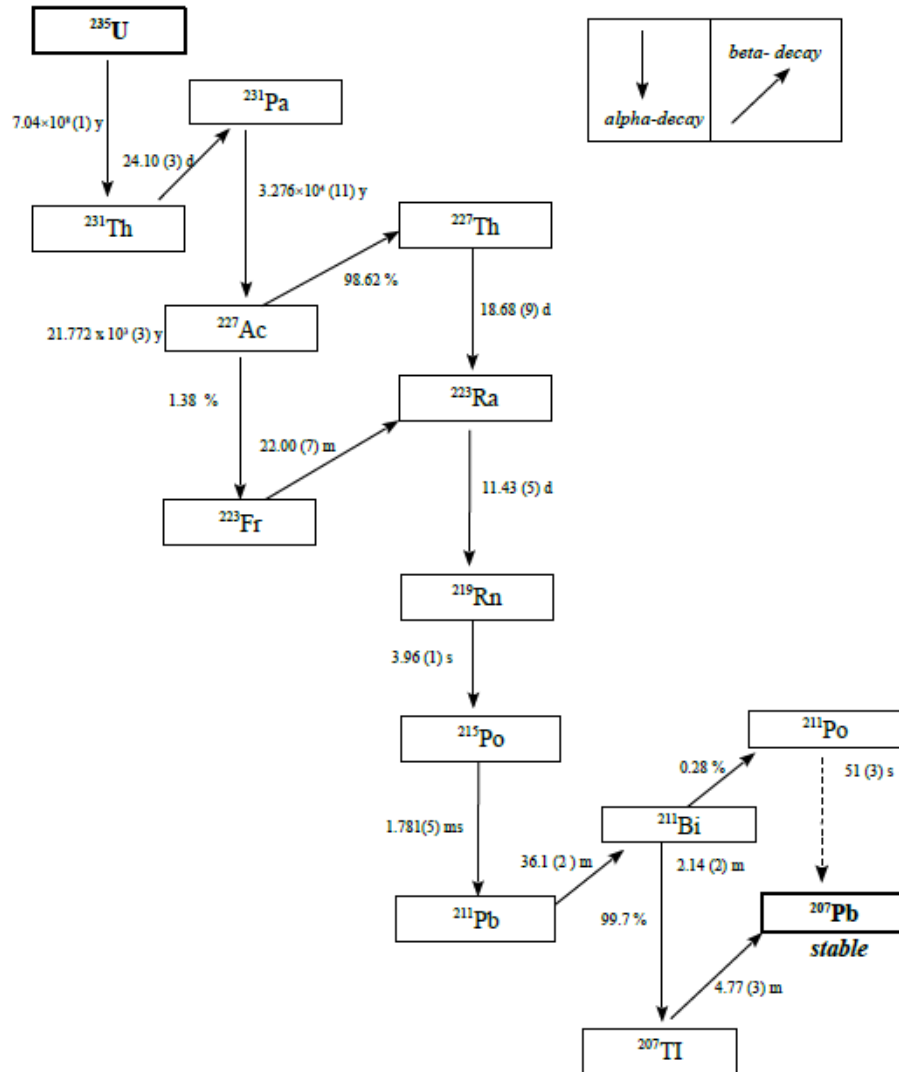


| Nuclide | Half-life | Decay mode | Gamma-ray energy (keV) | Emission probability (%) |
|---|---|---|---|---|
| U-235 series | | | | |
| $^{235}$U | $7.038\ (5) \times 10^8$ y | $\alpha$ | 185.715 (5) | 57.2 (5) |
| | | | 143.764 (2) | 10.96 (8) |
| | | | 163.358 (2) | 5.08 (4) |
| | | | 205.309 (2) | 5.01 (5) |
| $^{231}$Th | 25.52 (1)h | $\beta^-$ | 25.646 (4) | 14.5 (3) |
| | | | 84.216 (3) | 6.6 (3) |
| $^{231}$Pa | $3.276\ (11) \times 10^4$y | $\alpha$ | 27.36 (1) | 10.3 (4) |
| | | | 300.07 (1) | 2.47 (6) |
| | | | 302.65 (2) | 2.87 (27) |
| $^{227}$Ac | 21.773 (3)y | $\alpha, \beta^-$ | 99.6 (1) | 0.0056 (17) |
| $^{227}$Th | 18.72 (2) d | $\alpha$ | 235.971 (20) | 12.3 (13) |
| | | | 50.13 (1) | 8.0 (9) |
| | | | 256.25 (2) | 7.0 (4) |
| | | | 329.851 (20) | 2.69 (27) |
| $^{223}$Ra | 11.435 (4)d | $\alpha$ | 269.459 (10) | 13.7 (3) |
| | | | 154.21 (1) | 5.62 (14) |
| | | | 323.871 (10) | 3.93 (7) |
| | | | 144.232 (10) | 3.22 (7) |
| $^{219}$Rn | 3.96 (1) s | $\alpha$ | 271.23 (1) | 10.8 (3) |
| | | | 401.81 (41) | 6.4 (2) |
| $^{215}$Po | 1.781 (4) ms | $\alpha, \beta^-$ | 438.8 (3) | 0.04 (2) |
| $^{211}$Pb | 36.1 (2)m | $\beta^-$ | 404.853 (10) | 3.78 (50 |
| | | | 832.01 (3) | 3.52 (5) |
| | | | 427.088 (10) | 1.76 (4) |
| $^{211}$Bi | 2.14 (2) m | $\alpha, \beta^-$ | 351.059 (20) | 12.91 (11) |
| Po-211 | 0.516 (3)s | $\alpha$ | 897.80 (5) | 0.561 (11) |
| | | | 569.70 (22) | 0.545 |
| $^{207}$Ti | 4.77 (2)m | $\beta^-$ | 897.80 (5) | 0.260 (9) |
| $^{207}$Pb | Stable | - | - | - |

# How do you measure NORM?
## You could use α spectrometry....

C. Larijani[a,b,*], A.K. Pearce[a], P.H. Regan[a,b], B.C. Russell[a], S.M. Jerome[a], M.T. Crespo[c], P. de Felice[d], G. Lutter[e], F. Maringer[f], M. Mazánová[g]

| Parent Nucleus → Daughter Nucleus | Decay Half-Life | Energy of main α decays and % of decays with this energy. |
|---|---|---|
| $^{210}Po \rightarrow {}^{206}Pb + \alpha$ | 138.4 days | 5.304 MeV (100%) |
| $^{234}U \rightarrow {}^{230}Th + \alpha$ | $2.455 \times 10^5$ years | 4.774 MeV (71%) & 4.722 MeV (28%) |
| $^{235}U \rightarrow {}^{231}Th + \alpha$ | $7.038 \times 10^8$ years | 4.399 MeV (55%) & 4.366 MeV (17%) |
| $^{238}U \rightarrow {}^{234}Th + \alpha$ | $4.468 \times 10^9$ years | 4.198 MeV (79%) & 4.151 MeV (21%) |
| $^{238}Pu \rightarrow {}^{234}U + \alpha$ | 87.7 years | 5.499 MeV (71%) & 5.465 MeV (29%) |
| $^{239}Pu \rightarrow {}^{235}U + \alpha$ | 24,110 years | 5.157 MeV (71%) & 5.144 (17%) |
| $^{240}Pu \rightarrow {}^{236}U + \alpha$ | 6,561 years | 5.168 MeV (73%) & 5.124 MeV (27%) |
| $^{241}Am \rightarrow {}^{237}Np + \alpha$ | 432.6 years | 5.486 MeV (86%) & 5.443 MeV (13%) |

Needs radiochemistry & tracers.
Can be a bit messy, but ok / only ideal for some (non-gamma).
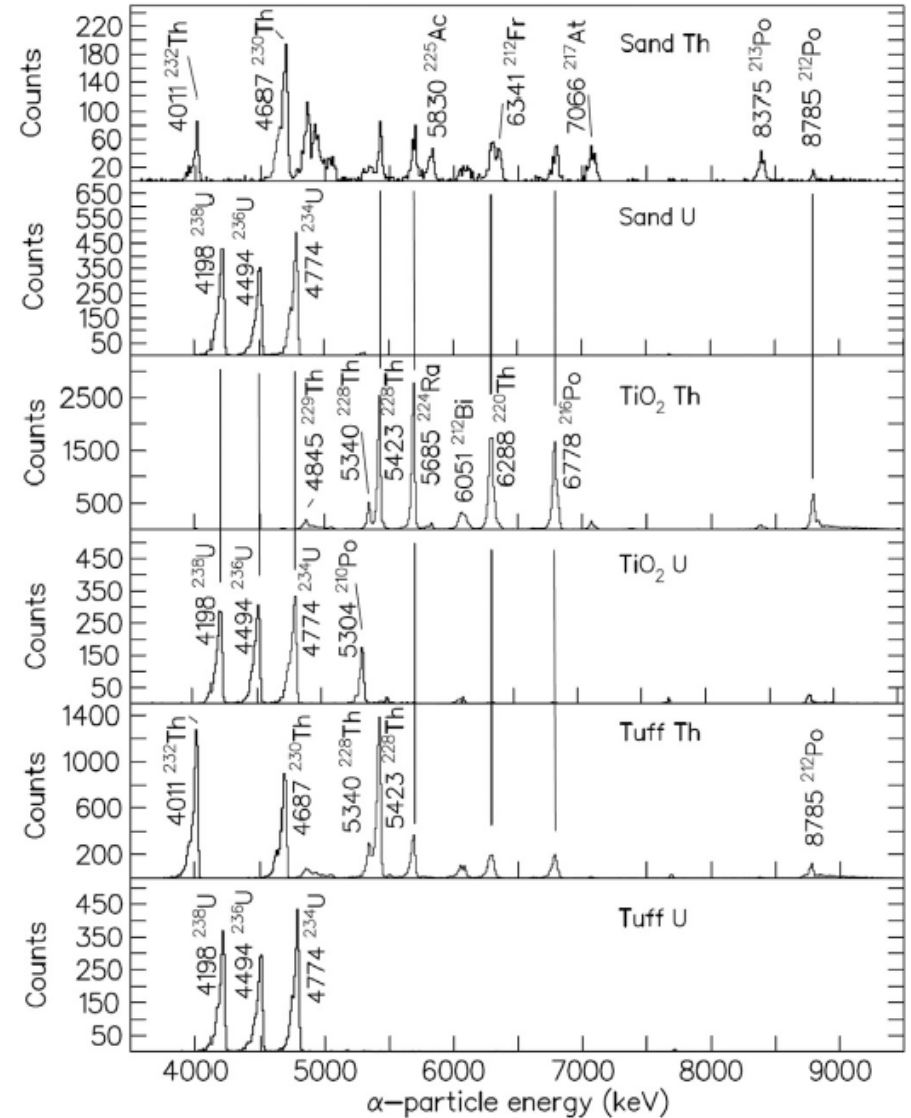
Gamma spectrometry works well.
Calibrate full-energy peak detection
efficiency using same geometry
(Marinelli) standards sources







(b)

Absolute calibration of the system is vital.

Use known sources in similar density matrices.

Calibrate efficiency from ~40 keV to 2614 keV.
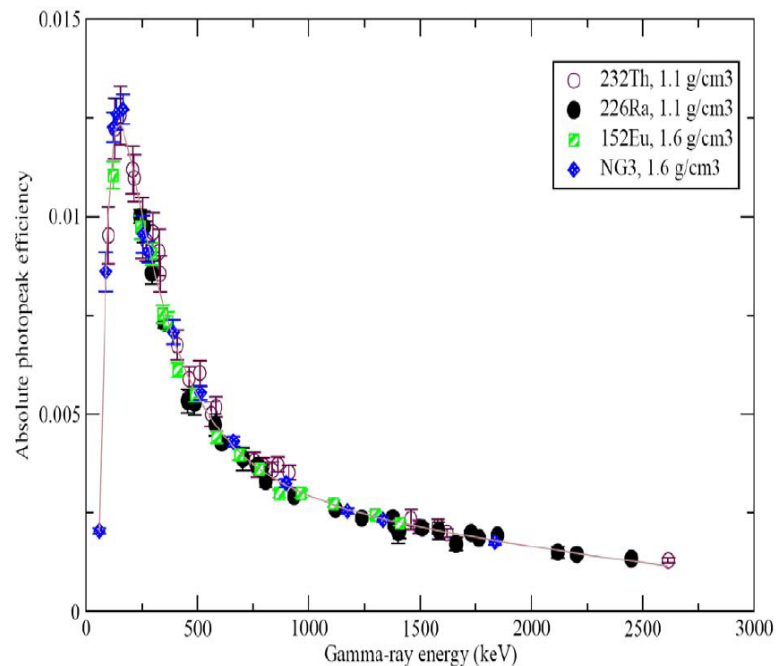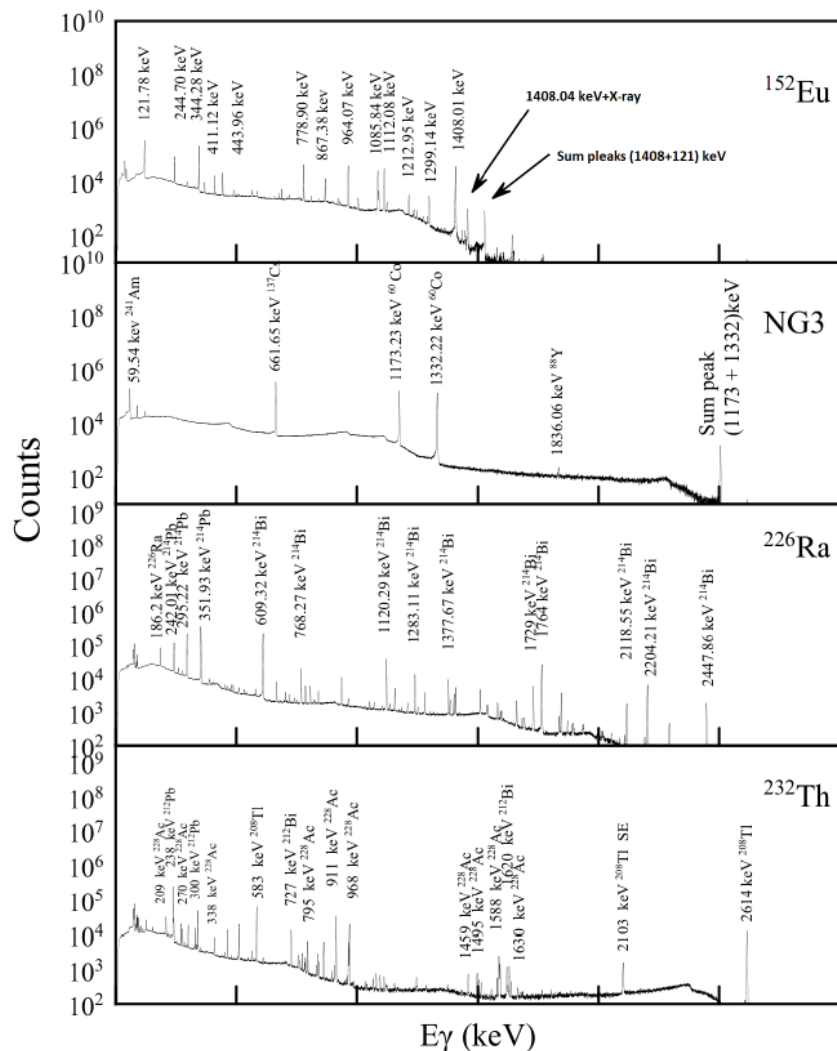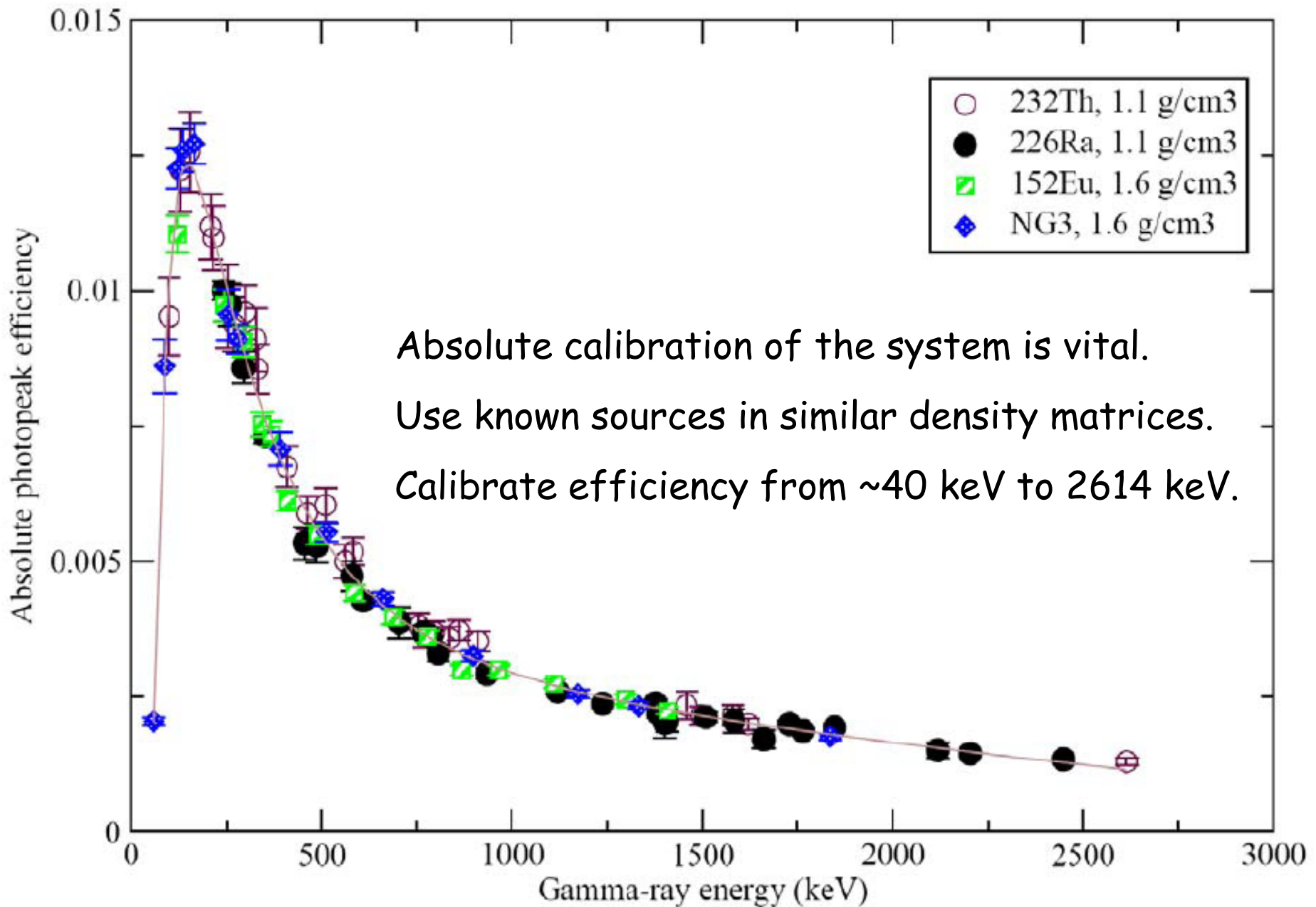
# background (2 days)
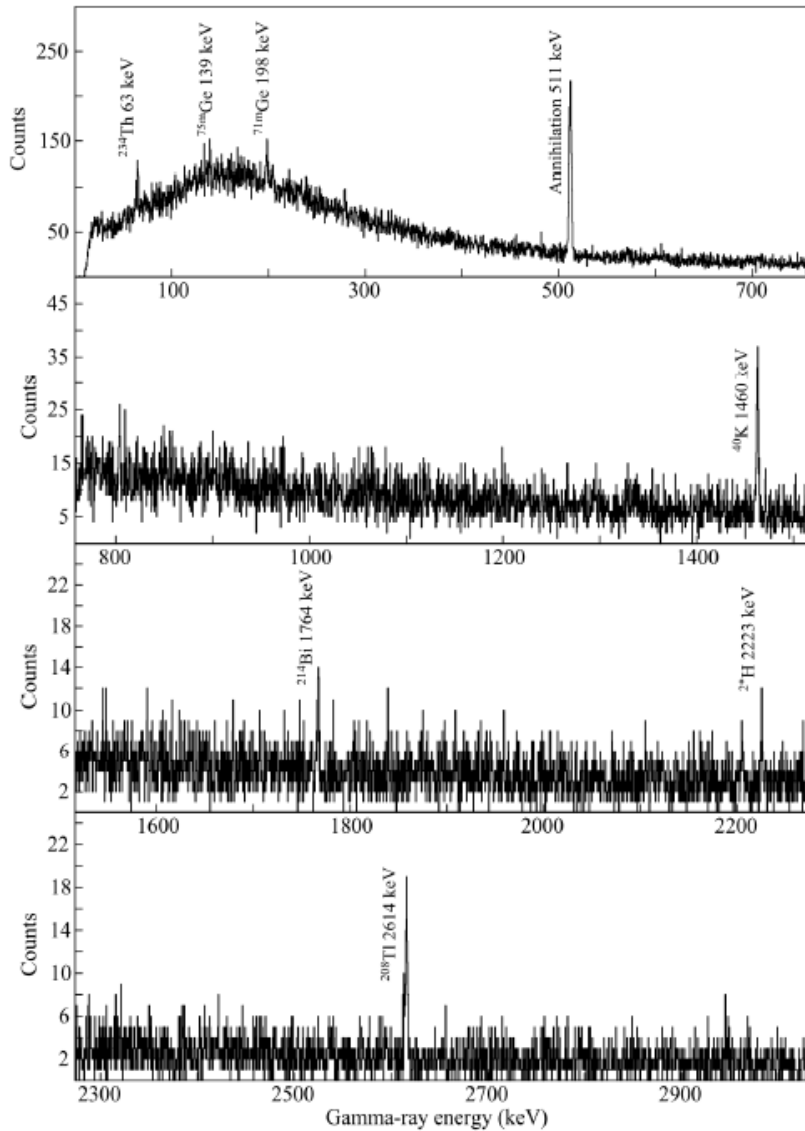
# sand sample (2 days)



Figure 4-4: Background spectra collected from de-ionised water for 172,800s (i.e. 2 days).
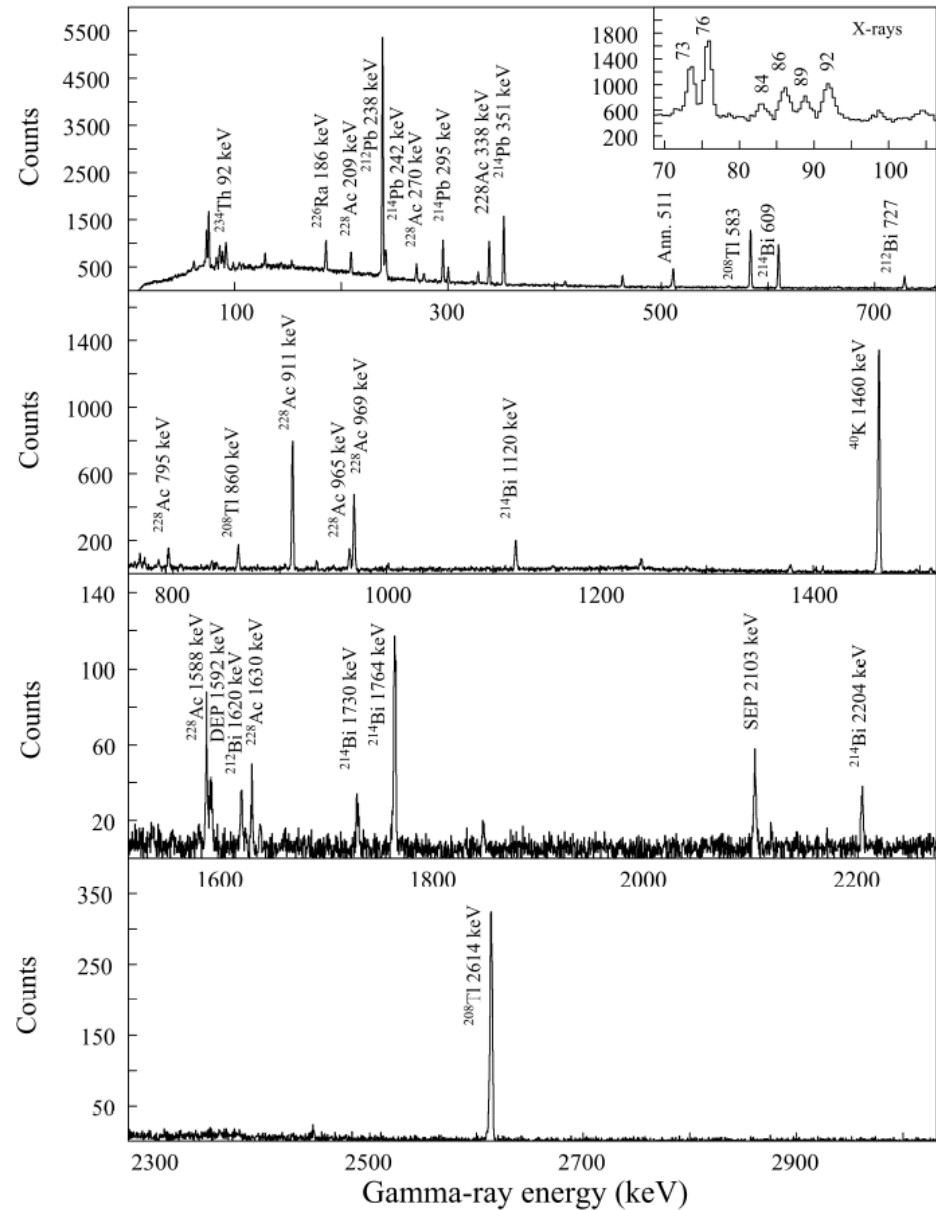
Figure 4-19: Gamma-ray spectrum of the 'Ao Po' beach sand sample subtracted background collected for 172,800s (i.e. 48 hours).

The specific activity in a (soil) sample is then given by:

$$A = \frac{Cnet}{E_f P_\gamma t_s m K}$$

$C_{net}$ = net number of counts (after back sub) in peak
$E_f$ = full-energy peak detection efficiency
$P\gamma$ = gamma-ray emission probability
ts = live time for spectrum collection (in secs)
 m = Mass of sample (in kg)
K – production of correction factors $K_1.K_2.K_3$..etc. for dead time, decay correction since start of measurement; self-attenuation in sample compared to calibration efficiency; random summing; and true coincidence summing etc.

# Determination of $^{238}$U, $^{232}$Th and $^{40}$K activity concentrations in riverbank soil along the Chao Phraya river basin in Thailand

T. Santawamaitre [a], D. Malain [a], H.A. Al-Sulaiti [a], D.A. Bradley [a], M.C. Matthews [b], P.H. Regan [a, c, *]

[a] Centre for Nuclear and Radiation Physics, Department of Physics, University of Surrey, Guildford GU2 7XH, UK
[b] Centre for Environmental Health Engineering, Department of Civil Engineering, University of Surrey, Guildford GU2 7XH, UK
[c] National Physical Laboratory, Teddington, Middlesex, TW11 0LW, UK
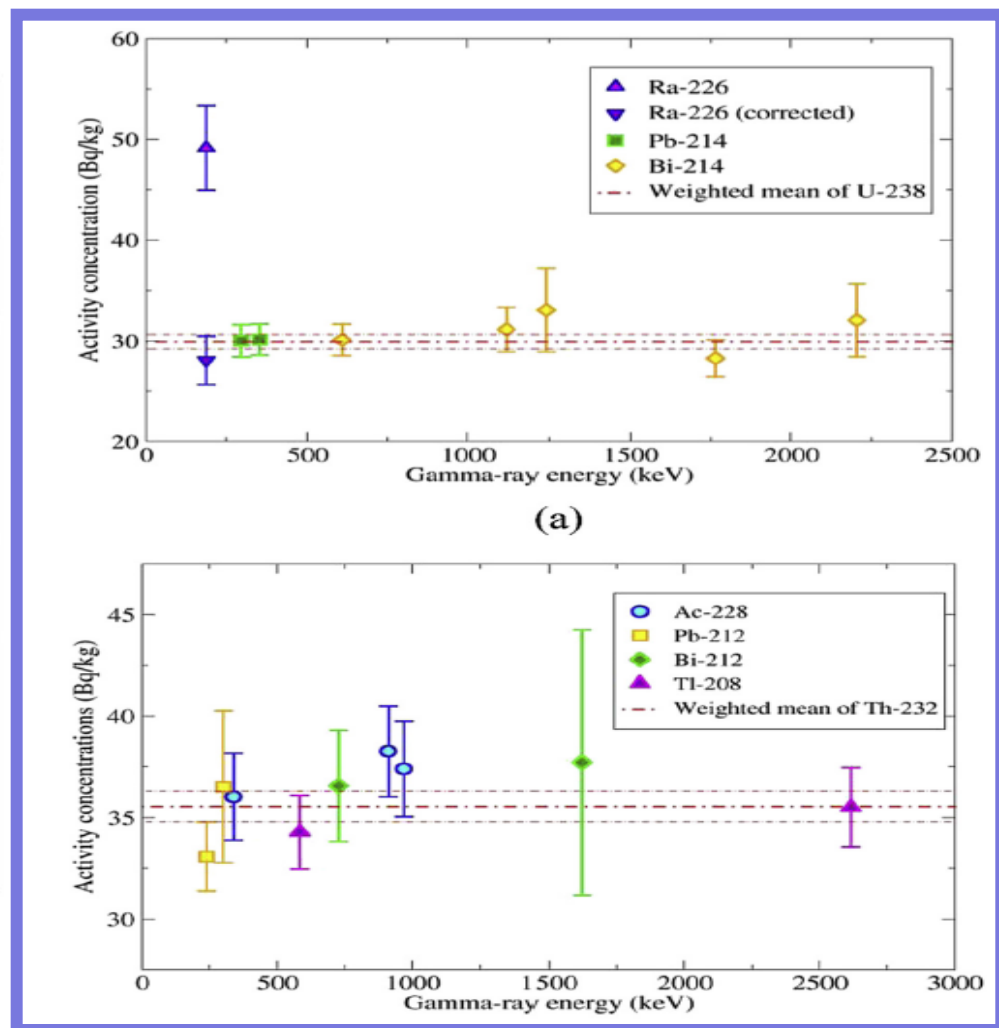
# Beware Energy Doublets....beware ($^{226}$Ra / $^{235}$U for example)
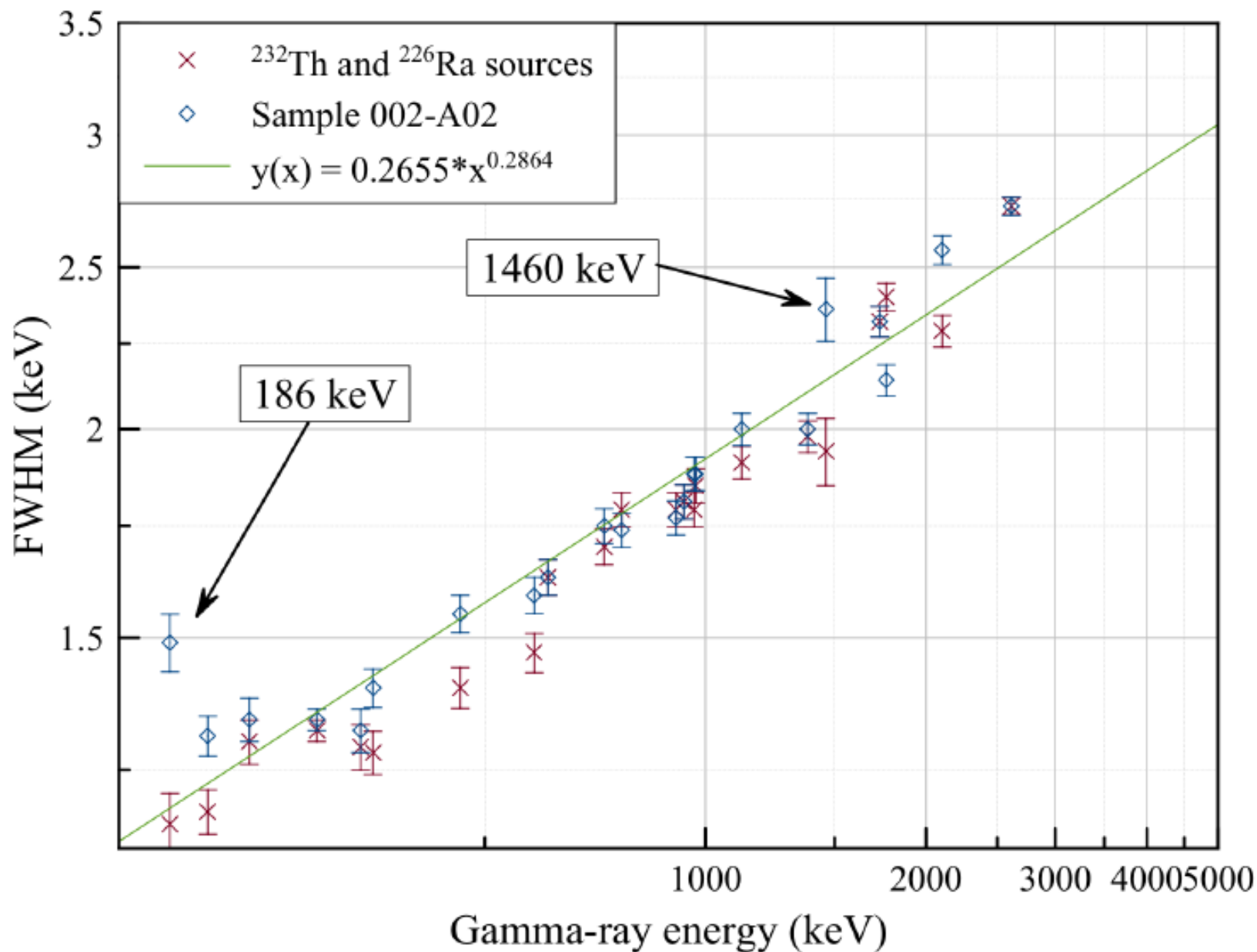
**Table 2.5** Details of the Actinium ($^{235}$U) decay chain [WAH07]

| Parent nuclide | Half-life $t_{1/2}$ | Decay Mode (% branch) | Decay Energy (MeV) | % Intensity | Daughter nuclide | γ-emission Energy (keV) | % γ-emission Intensity |
|---|---|---|---|---|---|---|---|
| $^{235}$U | 7.0x10$^8$ y | α (100) | 4.3978 | 55.0 | $^{231}$Th | 185.72 | 57.2 |
| | | | 4.3661 | 17.0 | | 143.76 | 10.96 |
| | | | 4.2147 | 5.7 | | 163.36 | 5.08 |
| $^{231}$Th | 25.52 h | β (100) | 0.2881 | 40.0 | $^{231}$Pa | 25.65 | 14.5 |
| | | | 0.3053 | 33.0 | | 84.22 | 6.6 |
| | | | 0.2060 | 12.8 | | 89.94 | 0.94 |
| $^{231}$Pa | 3.3x10$^4$ y | α (100) | 5.0138 | 25.4 | $^{227}$Ac | 27.36 | 10.3 |
| | | | 4.9513 | 22.8 | | 300.07 | 2.47 |
| | | | 5.0284 | 20.0 | | 302.65 | 2.87 |
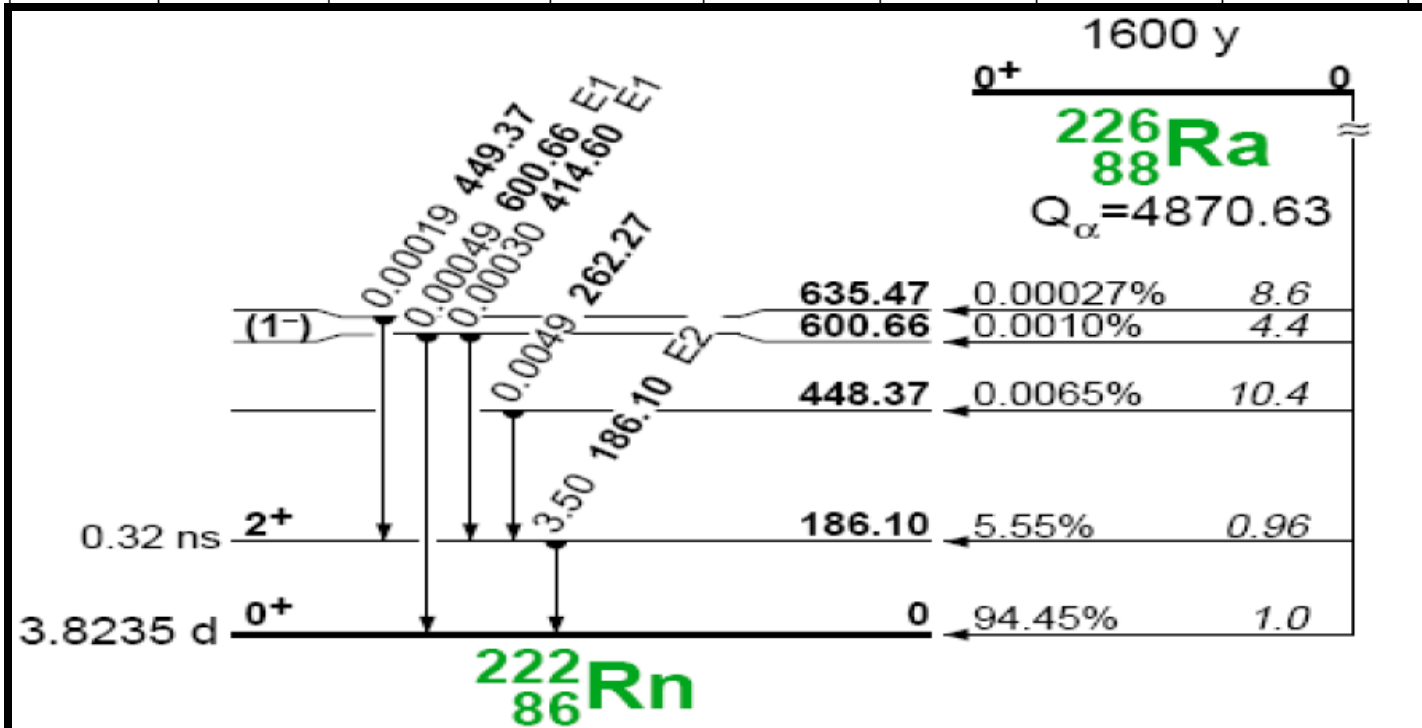
# How do we use the <span style="color:red">Activity Concentration</span> measurements?

The gamma dose rate (D) in units of <u>nGy/hour</u> in the outdoor air can be estimated using dose conversion coeffs...

$$D = 0.462 A_{Ra} + 0.604 A_{Th} + 0.0417 A_{K}$$

$^{226}$Ra     $^{232}$Th     $^{40}$K

A = measured <u>specific activity concentration</u> (Bq/kg)

Gy = unit of radiation dose = 1 Joule / kg  (1 Sv = Wr x 1Gy)

This leads to calculation of effective annual doses from NORM (in mSv/year)  = 0.00123D  assuming an outdoor occupancy of 20%.

[UNSCEAR00] United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), *Sources and effects of ionizing radiation*, Report to General Assembly, with Scientific Annexes, New York, 2000.

# Gamma dose across Europe, from NUPECC 2022



Map of typical annual doses from natural background radiation exposure in Europe.

European Annual Terrestrial Gamma Dose Map (mSv)

- 1.17 - 1.60 (99 %ile - Max)
- 0.80 - 1.17 (95 - 99 %ile)
- 0.72 - 0.80 (90 - 95 %ile)
- 0.61 - 0.72 (75 - 90 %ile)
- 0.50 - 0.61 (50 - 75 %ile)
- 0.39 - 0.50 (25 - 50 %ile)
- 0.33 - 0.39 (15 - 25 %ile)
- 0.30 - 0.33 (10 - 15 %ile)
- 0.26 - 0.30 (5 - 10 %ile)
- 0.14 - 0.26 (Min - 5 %ile)

https://www.nupecc.org/pub/np_lite_eb.pdf
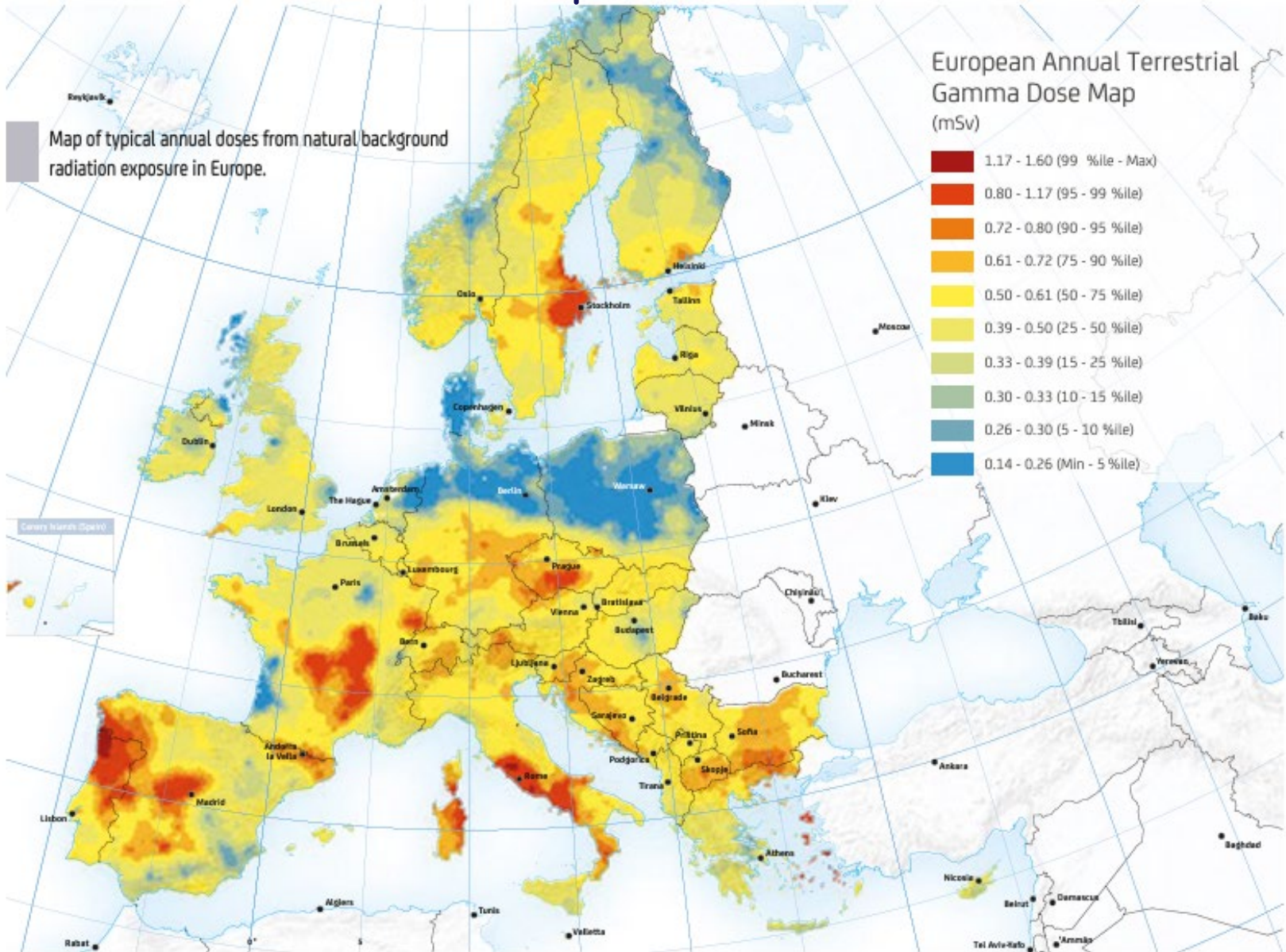
# Comparison of doses from sources of exposure

| Source of exposure | Dose |
|---|---|
| Dental x-ray | 0.005 mSv |
| 100g of Brazil nuts | 0.01 mSv |
| Chest x-ray | 0.014 mSv |
| Transatlantic flight | 0.08 mSv |
| Nuclear power station worker average annual occupational exposure (2010) | 0.18 mSv |
| UK annual average radon dose | 1.3 mSv |
| CT scan of the head | 1.4 mSv |
| UK average annual radiation dose | 2.7 mSv |
| USA average annual radiation dose | 6.2 mSv |
| CT scan of the chest | 6.6 mSv |
| Average annual radon dose to people in Cornwall | 7.8 mSv |
| CT scan of the whole spine | 10 mSv |
| Annual exposure limit for nuclear industry employees | 20 mSv |

# Lecture 2:

# Standards, traceability and two examples..

- Primary and Secondary standards

    - Why and how? Links to Nuclear Data.

    - Radiopharmaceutical standards

        - $^{223}$Ra XOFIGO©, an example

    - Radioactive gas releases

        - CTBTO tests (Xe) and reactor criticality(Kr)

# Links between primary standards of activity & underpinning Nuclear Data

- Primary standards needed are needed to calibrate measurement systems.

- These can then be used for measuring **absolute $\gamma$-ray emission intensities per decay**, $P_\gamma(\%)$ .

- These are needed for:
  - medical radiopharmaceutical dose evaluations;
  - nuclear security (e.g., CTBT verification, radioxenon)
  - nuclear waste assay (e.g, Np, Pu, Am, Cs isotopes);
  - environmental assay (NORMs);
  - nuclear forensics (e.g., $^{134,137}$Cs and U isotope ratios);
  - nuclear structure / nuclear (astro)physics research.

## Nuclear Physics News International

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FEATURING:
NUSTAR · Photonuclear Reactions
The AGATA Campaigns at GSI and GANIL

# Radionuclide Metrology and Standards in Nuclear Physics

The development of radionuclide standards for metrology has underpinned nuclear physics since its inception [1]. The current frontier of radionuclide metrology relies on developments in radiation detection and signal processing combined with accurate nuclear decay data evaluations [2] and contributes to a myriad of scientific disciplines. Radionuclide metrology represents a crucial part of the scientific jigsaw that enables societal benefits from nuclear physics research.

Measurements that are traceable to internationally accepted primary standards can give the public confidence in the characterization of civilian nuclear waste materials such as $^{90}Sr$, $^{134,135,137}Cs$, $^{237}Np$, $^{239,240}Pu$, and $^{241}Am$ and measurements of naturally occurring radioactive materials (NORMs) such as $^{3}H$, $^{7}Be$, $^{14}C$, $^{210}Po$, $^{210}Pb$, $^{214}Bi$, $^{214}Pb$, $^{222}Rn$, $^{223}Ra$, $^{226}Ra$, $^{228}Ac$, and $^{234,235,238}U$. Other applications include assay of Technologically Enhanced NORM with potential radiological impact on workers in the oil and mineral production industries, and the use of radiopharmaceutical isotopes such as $^{18}F$, $^{82}Rb/^{82}Sr$, $^{89}Zr$, $^{99m}Tc$, $^{124,131}I$, $^{211}At$, $^{223}Ra$, and $^{227}Th$ for diagnostic imaging and therapy. This article explains the concept of international traceability and how accurate radiation standards are determined for different radioactive decay modes.

## Primary Radioactivity Standards

National Measurement Institutes (NMIs) are responsible for the development and upkeep of primary measurement standards. Primary standards are used to calibrate instruments and/or to certify reference materials; these can then be distributed to other laboratories and used to calibrate their own instruments in an uninterrupted chain of calibrations to the final end-user. All measurements are essentially ratios back to these primary standards. NMIs cross-check their primary standards against sources from other countries through international comparison exercises co-ordinated by the Bureau International des Poids et Mesures (BIPM).

The first primary standard of radioactivity was based on radium. The inaugural Radium Standards Committee was held in Brussels in 1910 and chaired by Lord Rutherford at which 1 curie (Ci) was defined as the amount of radon in equilibrium with 1 g of radium [1]. The radioactivity measurement system based on radium standards became outdated following developments in accelerator technology, which led to an increased range of artificially created radionuclides. In 1950, the curie was redefined as $3.7 \times 10^{10}$ disintegrations per second and, in 1975, the 15th Conférence Générale des Poids et Mesures adopted the becquerel (Bq), which is equal to one inverse second for the SI unit of activity [3, 4].

In 1958, The International Committee for Weights and Measures of the BIPM created the Comité Consultatif des Rayonnements Ionisants (CCRI). The CCRI is responsible for organizing international comparisons, enabling NMIs to cross-check their primary standards. The International Reference System, implemented in 1975, is based on a pressurized well-type ionization chamber based at BIPM [4]. This is a permanent, stable measurement instrument tool that is available to NMIs to compare primary standards of gamma emitters.

A primary standard of radioactivity allows the number of decays from a source in a finite time period to be determined using a technique that does not itself need calibration. Since the activity of each radionuclide species depends on unique decay properties, different experimental techniques are needed for the primary standardizations of individual radioisotopes. The particular technique depends on the radioactive decay mode(s), half-life, decay scheme of the daughter nucleus and branching ratios for competing decay modes. Most modern radioactivity standards are aqueous solutions, quantified by their activity per unit mass on a given reference date [3, 4]. The main methodologies used for primary radionuclide standardization are discussed below.

### High-Geometry Methods

Perhaps the simplest method is to count the number of photons or particles emitted by a source into the full $4\pi$ steradians of solid angle [3]. The perfect $4\pi$ detector does not exist and the "non-detection" probability must be accounted for, either by examining the rate of coincidences between multiple detectors or using Monte-Carlo particle transport simulation codes.

For a complex decay scheme of excited states populated in the daughter nucleus, the $4\pi\gamma$ counting technique can be exploited using either a single well-type NaI(Tl) or two NaI(Tl) detectors sandwiching the source. A higher number of coincident cascade gammas emitted per decay leads to reduced uncertainties from non-detec-

When you measure something to obtain a **calibration standard**, precision is important.

**If the standard is inaccurate, everything based on it will also be inaccurate.**

<u>**Ionisation chambers**</u> are frequently used to determine dose for radiopharmaceuticals.

Accurate calibration is critical, as is access to accurate calibration standards.

The dose delivered must fall within a narrow region for the benefits to the patient to outweigh the harm.

The consequences of a miscalibrated ionisation chamber in a hospital could be serious.
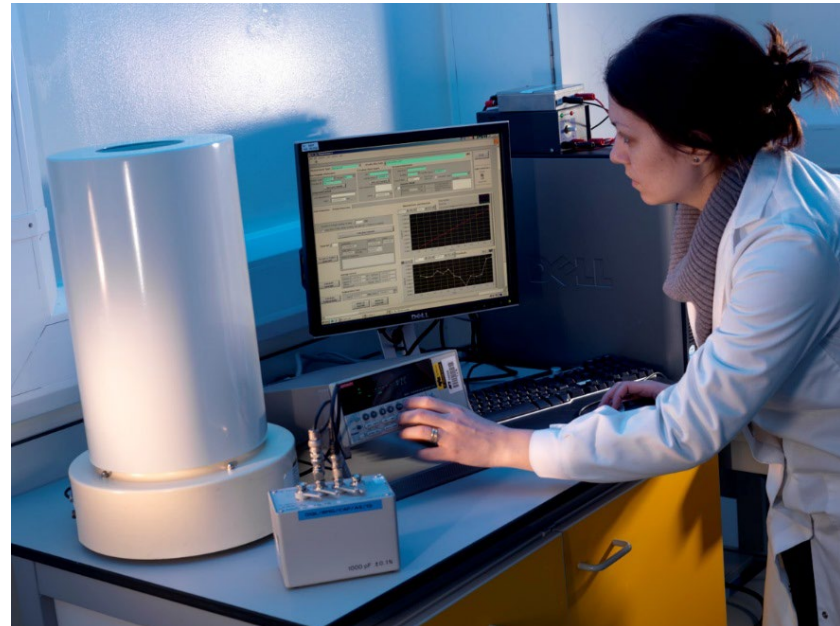
# What is a "primary standard"?

From the International Vocabulary of Metrology:

"A measurement standard established using a **primary reference measurement procedure**"

A **primary measurement procedure** is:

"A reference measurement procedure used to obtain a measurement result **without relation to a measurement standard** for a quantity **of the same kind**"

# Dissemination of a standard

All measurements that are not primary are based off a disseminated standard, thus the precision and accuracy of the standard is vital!

1. The uncertainty of the standard can not be improved upon.

2. If the standard is inaccurate the whole train of measurements after will be.
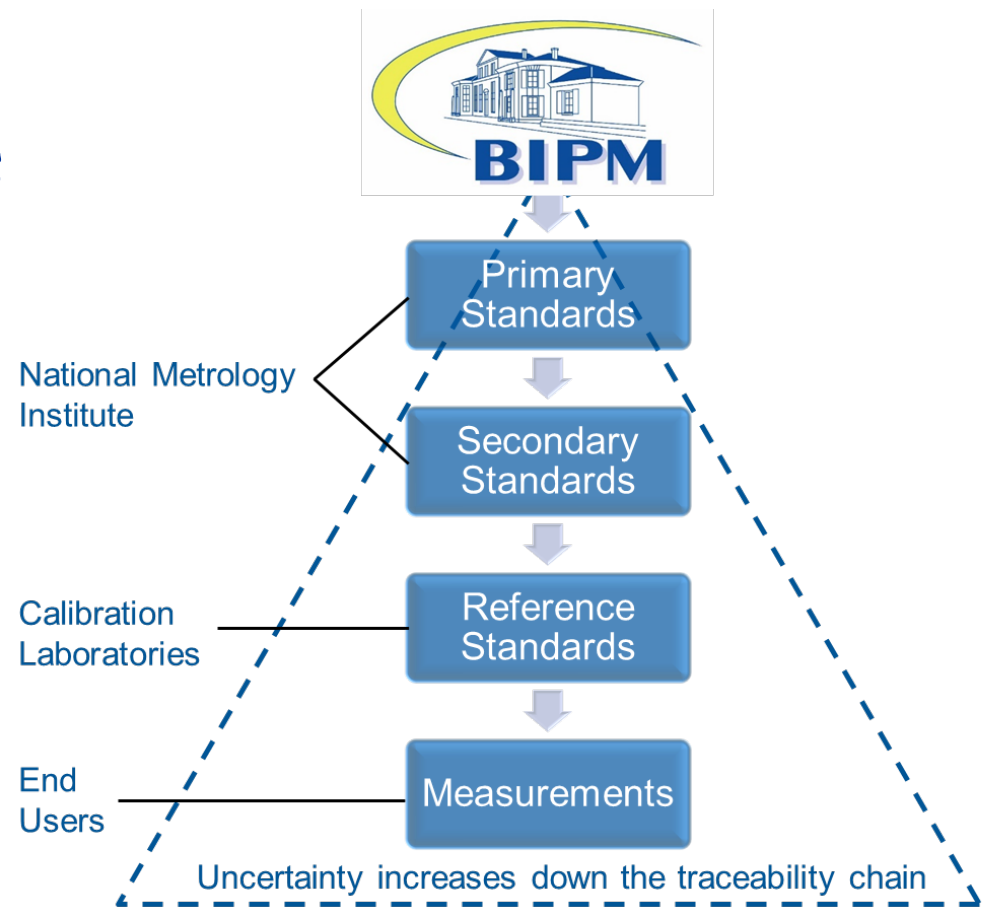
# Metrology chain
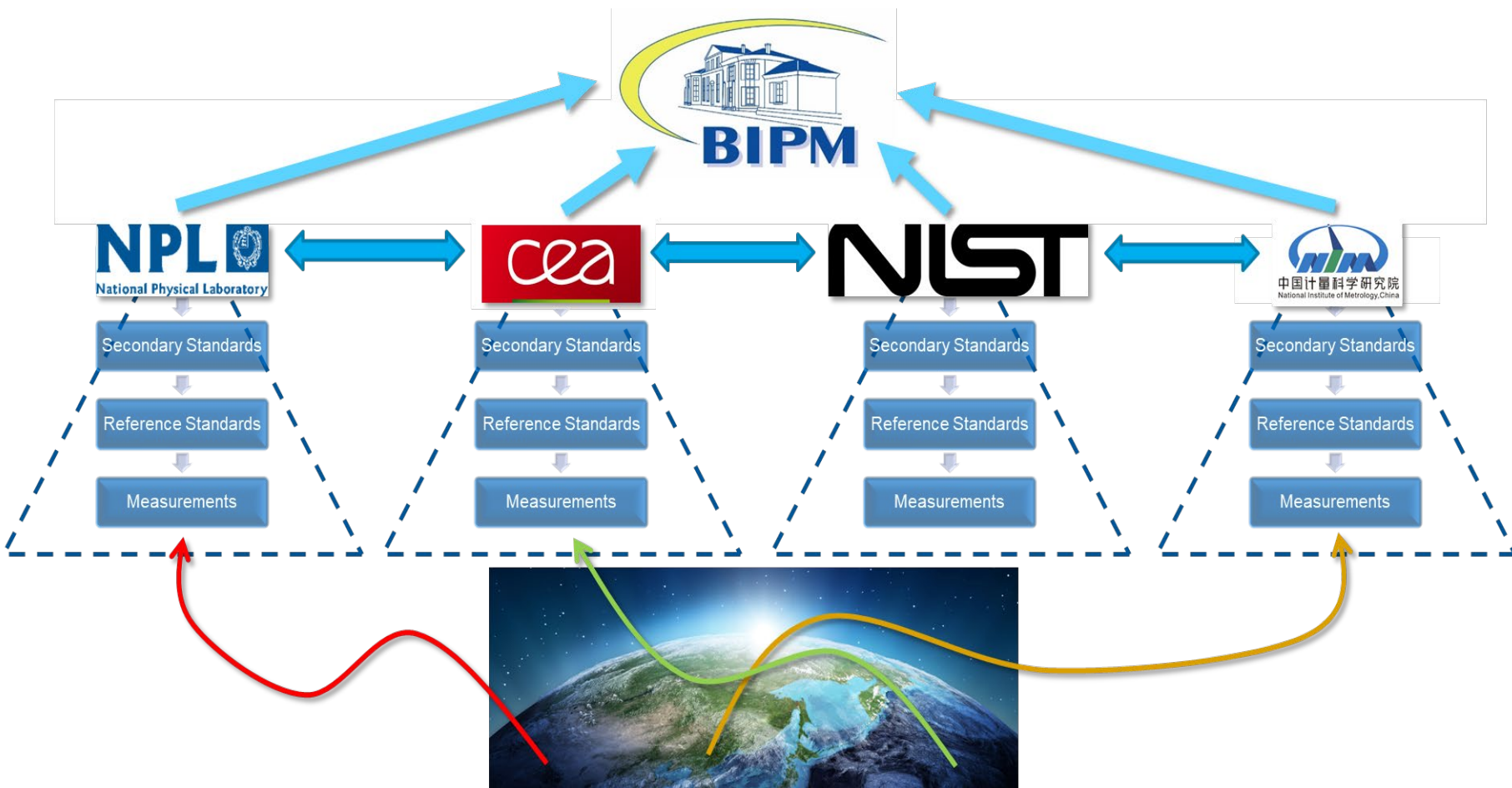
The absolute primary standardisation of activity is a key mission of National Measurement Institutes such as NPL (UK), NIST (USA),

They provide unbroken traceability to the becquerel (Bq) = $s^{-1}$ .

NPL has maintained the traceability of radioactive sources to the Bq in the UK for over 100 years





For Use of Staff.

· CERTIFICATE ·
—— OF ——
EXAMINATION
OF

Radium Bromide preparation

for the Edinburgh Royal Infirmary.

The specimen was contained in a platinum tube about 10 mm. long and 3 mm. diameter, numbered 4485. The preparation has been compared with the British Radium Standard which has been certified by the International Committee after comparison with the International Standard at Sèvres.

The effective gamma ray activity of the preparation in its enclosure is equivalent to

5.2 mgms. of metallic radium in equilibrium.

The accuracy of this result is not claimed to exceed $\pm 1 \%$ . No allowance has been made for absorption of the gamma rays in the containing tube and in the salt itself.

Assuming the atomic weights O = 16, H = 1.008, Br. = 79.92, Ra = 226.4, the above result is equivalent to

9.7 mgms. of Ra Br$_2$ 2H$_2$O

N.B.— The standardisation has been carried out by gamma ray methods. Without special tests this does not distinguish between pure radium preparations and those containing mesothorium or radiothorium.

December 20th,1913.

191

Ra. 1. 5.

Reference

Director.

# Nuclear Metrology Group



NPL radium certificate signed
by Lord Earnest Rutherford



NPL radium bench 1933

COPY.

COMMISSION INTERNATIONALE DES ETALONS DE RADIUM.

CERTIFICATE.

Specimen No. 3 of Radium is prepared as chloride from pitch-blende of St. Joachimstal and is consequently practically free from Mesothorium.

... milligrammes of salt.

... tube (Thuringian

(signed)

Für die Weiner Messung        Pour les Mesures        E. RUTHERFORD.
STEFAN MEYER.                  faites a Paris          Pres. of the Int.
                               M. CURIE.               Ctte. for Radium
                                                       Standard,
                                                       (June 2, 1913).

# Radioactivity standards

**Solid**

Historically, the primary standard was a physical lump of radium, which was chemically purified and weighed

- 1 g $^{226}$Ra = 1 Curie

Standards for calibrating equipment (gamma spectrometers, surface contamination monitors) are often still solid sources, but these are secondary standards cross-calibrated against primary standards.



The first radium standard held by NPL was prepared by Mesrs. Curie, Meyer and Rutherford.

From "Radiation Science at the National Physical Laboratory, 1912-1955", E. E. Smith.

# Other forms

**Gaseous**

Some of the most important radionuclides can be encountered in gaseous form

- $^{222}$Rn – naturally occurring
- Xenon isotopes – fission products
- $^{18}$F, $^{11}$C – medical applications

Gas standards - quantified as activity per unit volume - are often needed

**Liquid**

Almost all modern standards of radioactivity are aqueous solutions which are quantified in terms of activity per unit mass

- The mass of active material is incredibly small -  working with solutions allows for easier handling

- Mass measurements are more reliable than measurements of volume.

# Absolute primary standardisation of activity

- A 'primary' technique has the capability to measure in parallel:
  - Disintegration rate (Activity of sample = A(t) = $\lambda N$ = $N/\tau$)
  - Absolute detection efficiency.

- 'No previous knowledge of nuclear data is required'…but:
  - Caveat 1: Some knowledge of the decay scheme is useful.
  - Caveat 2: The half-life of the radionuclide is needed.

# Links between primary standards of activity & underpinning **Nuclear Data**

- Primary standards needed are needed to calibrate measurement systems.

- These can then be used for measuring <u>**absolute $\gamma$-ray emission intensities per decay**</u>, $P_\gamma$(%) .

- These are needed for:
  - medical radiopharmaceutical dose evaluations;
  - nuclear security (e.g., CTBT verification, radioxenon)
  - nuclear waste assay (e.g, Np, Pu, Am, Cs isotopes);
  - environmental assay (NORMs);
  - nuclear forensics (e.g., $^{134,137}$Cs and U isotope ratios);
  - nuclear structure / nuclear (astro)physics research.

# Primary standardisation techniques
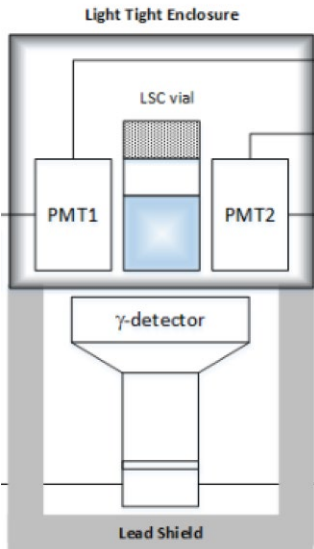


4π(LS)-γ coincidence counting
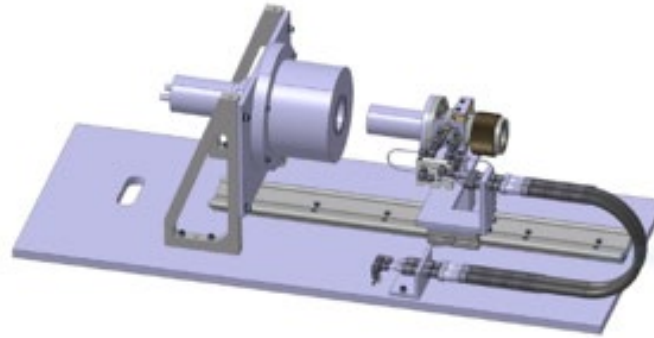


4πα/β-γ coincidence counting
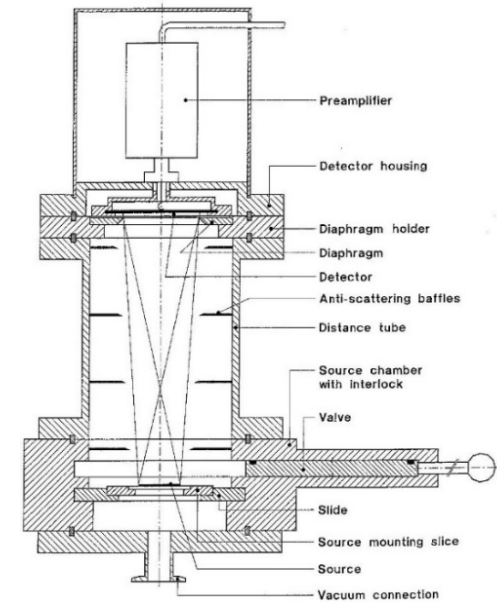


Defined Solid Angle α-counting

Defined by capability to measure simultaneously:
(1) The disintegration rate
(2) The detection efficiency



Triple-to-Double Coincidence Ratio counting

γ-γ coincidence counting
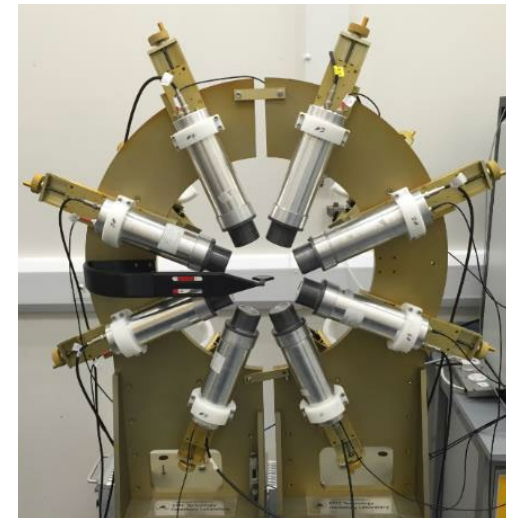
# Defined solid angle alpha counting at NPL

Arzu Arinc *, Michael J. Parfitt, John D. Keightley, Alan Wilson

*National Physical Laboratory, Hampton Road, Teddington, TW11 0LW United Kingdom*

H I G H L I G H T S

- Design of the new defined solid angle counter at NPL.
- Discussion on efforts to ensure reproducible dimensions.
- Solid angle calculation (and uncertainty) is performed by Monte Carlo simulation.
- Report on first DSA measurements at NPL for [241]Am and [243]Am.

Metrologia **52** (2015) S73
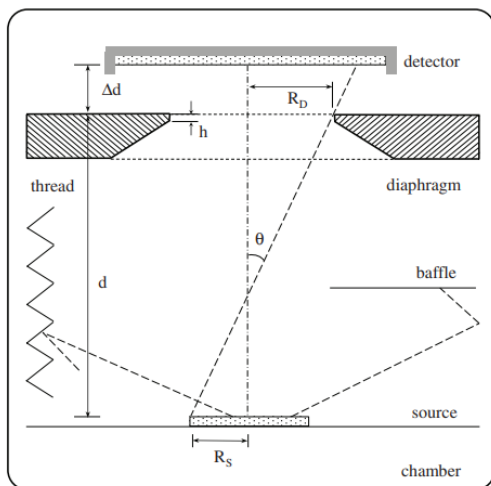


**Figure 1.** Schematic representation of characteristic geometrical parameters.
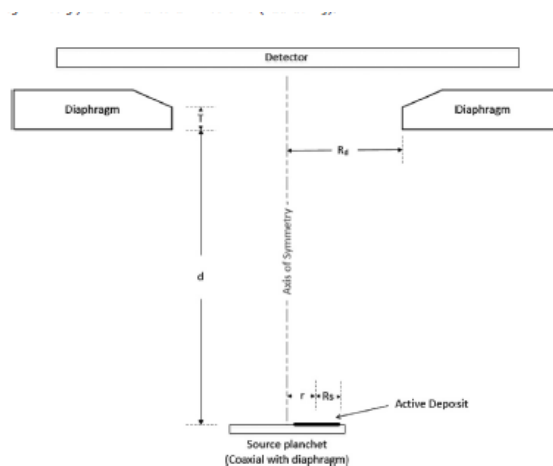


**Fig. 6.** A simplified schematic for 'realistic' geometric configurations.
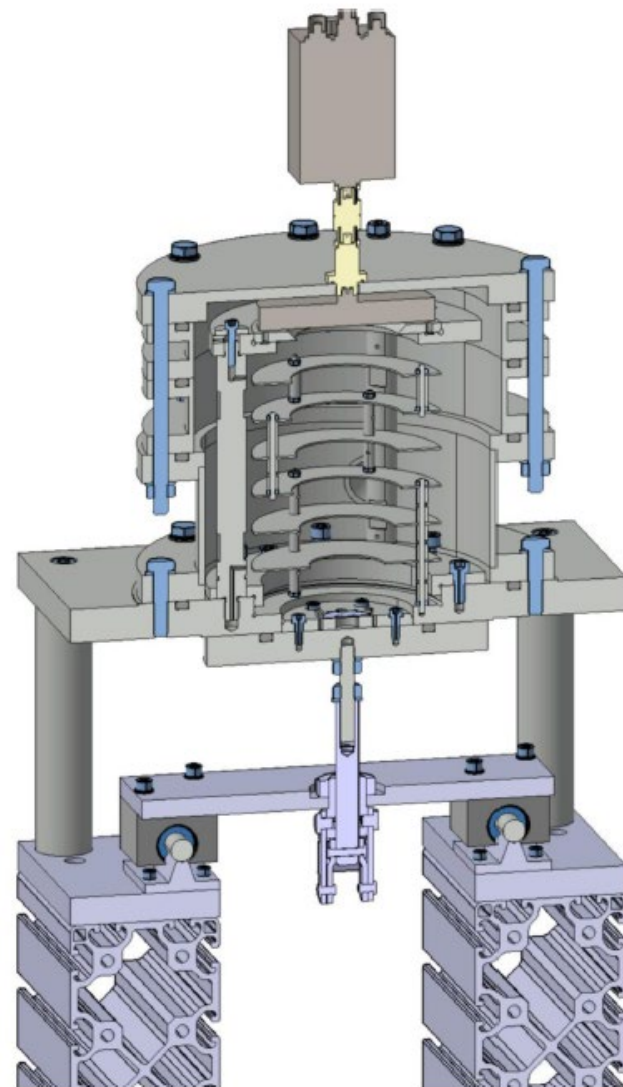


**Fig. 2.** CAD representation of the DSA counting system showing the retractable positioning platform with the source holder, the anti-scattering baffles, diaphragm, detector, preamplifier and the outer shell of the chamber.

# Idealised example of the $4\pi\beta-\gamma$ coincidence method

## $\beta-\gamma$ decay (100% fed single cascade)

*Count Rates*

$N_\beta = N_o\ \varepsilon_\beta$

$N_\gamma = N_o\ \varepsilon_\gamma$

$N_c = N_o\ \varepsilon_\beta\ \varepsilon_\gamma$

$\beta$

$\gamma$
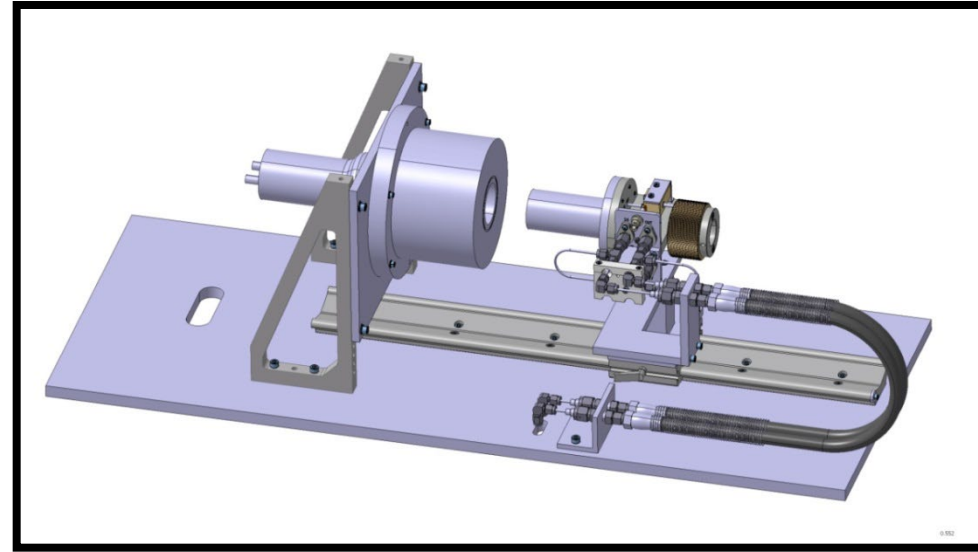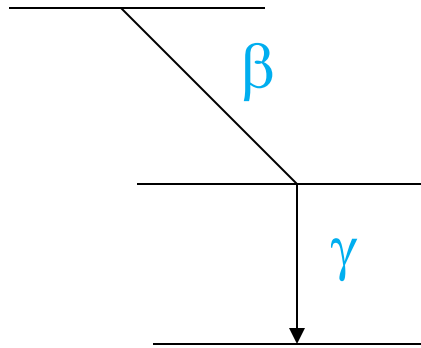


*Efficiencies:* $\quad \varepsilon_\beta = N_c/N_\gamma$

$\varepsilon_\gamma = N_c/N_\beta$

$$Activity : N_o = \frac{N_\beta N_\gamma}{N_c}$$

# Assumptions ?

**(1) The radionuclide under study must produce β and γ emissions in coincidence**

For example: $^{137}Cs$ is a pure beta decay, and has one radioactive decay product $^{137m}Ba$ ($t_{1/2}$ = 2.55 min), which decays by an isomeric transition to $^{137}Ba$ (stable).



$^{137}Cs$   7/2+

$Q_\beta = 1175.63\ (17)\ keV$

$P_\beta = 100\ \%$

~95 %

~5 %

11/2–
2.55 min

662 keV

3/2+

$^{137}Ba$
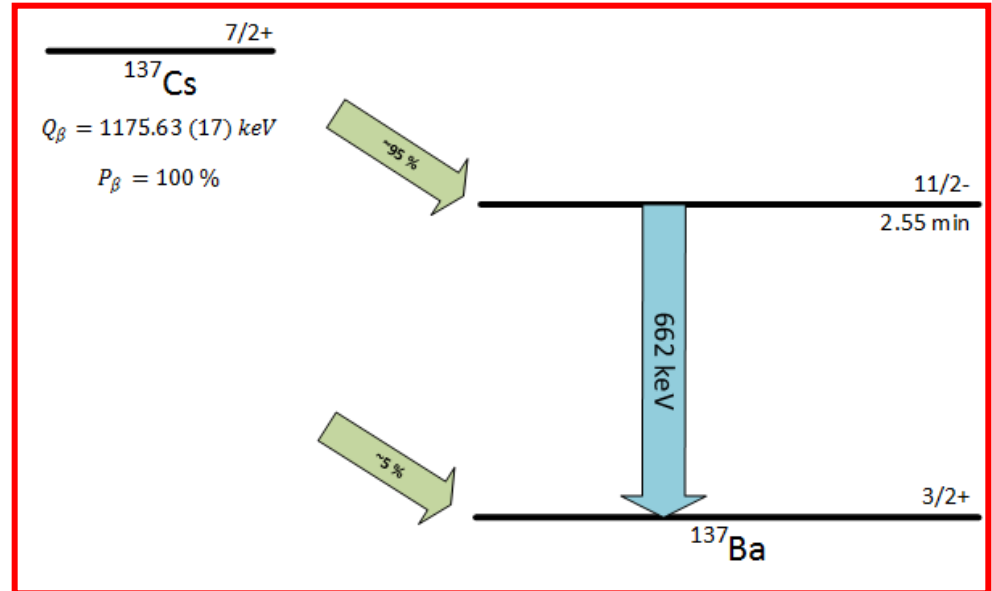
- β and γ emissions are not truly simultaneous;
- The detection and counting processes take a finite and variable amount of time.
- The decays must occur within a given "resolving time".
- The resolving time must be << the mean time between decays otherwise the number of "random coincidences" will be too great.
- Likewise the level lifetimes must be << the chosen resolving time.

# More Assumptions / Corrections ?

- **(2) The radionuclide under study must produce β and γ emissions in coincidence**

    - Need to account for:
        - Any angular distributions/correlations between coincident radiations (ideally measure over $4\pi$ solid angle – e.g. using liquid scintillation counter ).

        - Beta detector sensitive to gammas and vice versa…correct for energy dependent detection efficiency.

        - Account for additional decay branches (e.g. via internal conversion followed by electrons (from ICE) or X-rays.

# Beta sensitivity to gamma emissions

What happens if $\varepsilon_\beta = 1$?

$$\frac{n_\beta\, n_\gamma}{n_C} = A \frac{\left[1 + (0)\left(\frac{\varepsilon_{\beta,\gamma}}{1+\alpha} + \frac{\alpha\varepsilon_{CE}}{1+\alpha}\right)\right]}{1} = A$$

So if we can vary $\varepsilon_\beta$ and estimate what $\frac{n_\beta n_\gamma}{n_C}$ would be at $\varepsilon_\beta = 1$, we can still determine the activity, without the need for additional correction terms.

# Liquid scintillation counting

Samples are mixed with a liquid scintillation cocktail, which produces pulses of light in response to ionising radiation.

The samples are loaded into a liquid scintillation counter which records the number of pulses counted per minute.

This can be related back to the activity of the sample.

Advantages:

- High detection efficiency
- Very low background
- Low energy threshold

# Primary standard of radioactivity standard for $^{60}$Co using γ-γ energy coincidences with NANA

Investigation of γ-γ coincidence counting using the National Nuclear Array (NANA) as a primary standard

S.M. Collins[a,b,*], R. Shearman[a,b], J.D. Keightley[a], P.H. Regan[a,b]

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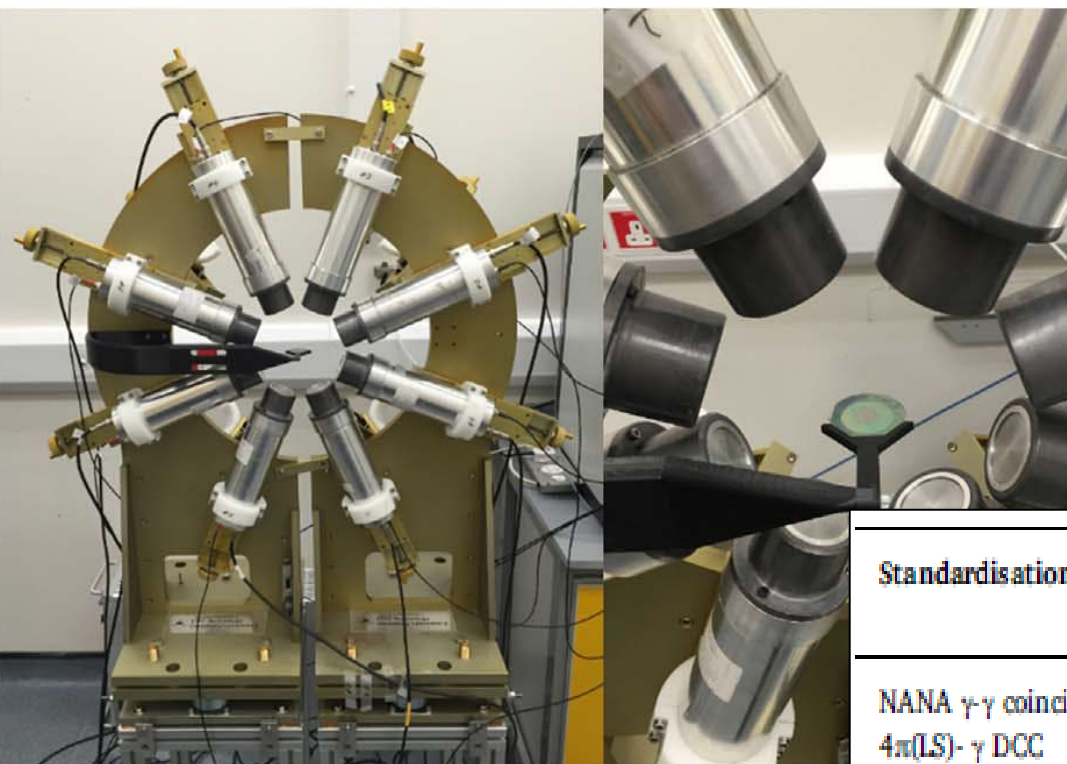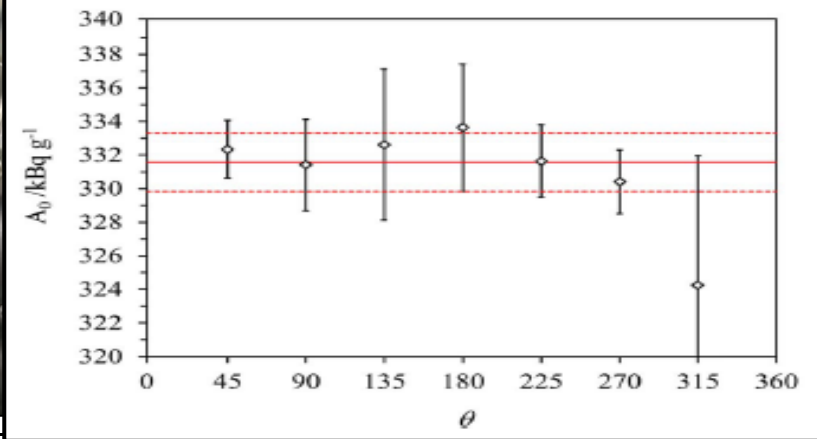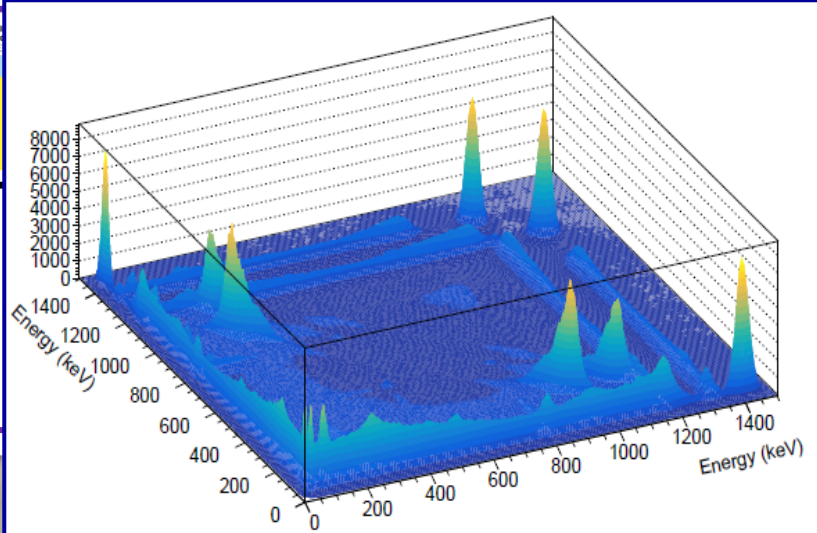




| Standardisation technique | $A_0$ /kBq g$^{-1}$ | u ($A_0$) /kBq g$^{-1}$ |
|---|---|---|
| NANA γ-γ coincidence counting | 330.8 | ± 1.0 |
| 4π(LS)- γ DCC | 330.92 | ± 0.86 |

# Transfer of Primary Standards of Activity to Secondary Standard

Fidelis : traceable link to NPL's Primary Standards.

Primary Standard (MBq/g)

↓

Secondary Standard Ionisation Chamber (pA/g)

↓

Calibration Factor (pA/MBq)
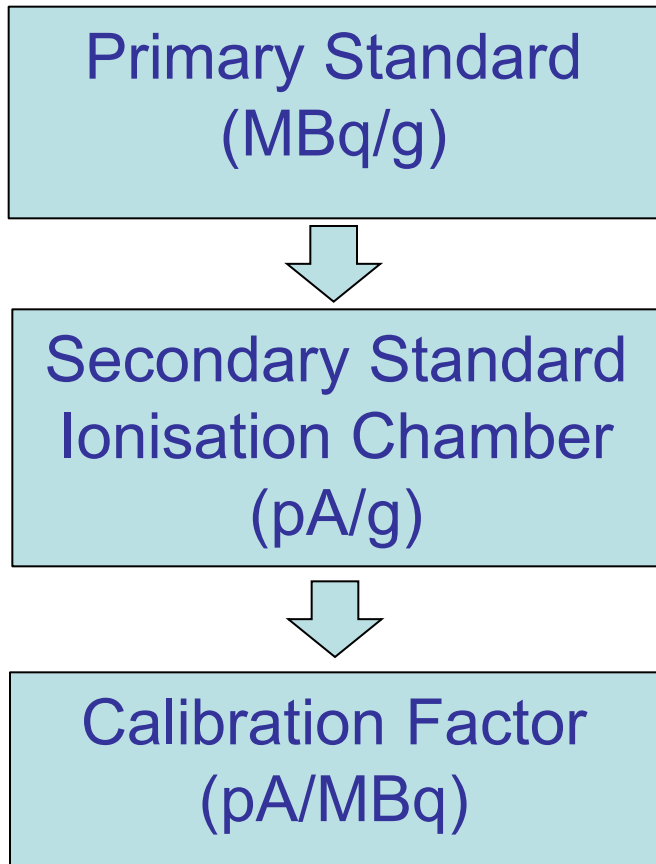
# Ionisation chambers

The ionisation chamber is a form of **gas ionisation detector**.

In a re-entrant chamber, samples to be measured are inserted in to a well at the top of the device.

The current produced by the device $I$ is measured and converted via a predetermined calibration factor $f$ to an activity $A$.

Calibration factors are radionuclide-specific, and also depend on the type and geometry of the sample.



Pulse height (charge collected)

ion chamber region

proportional region

Geiger region

Applied voltage
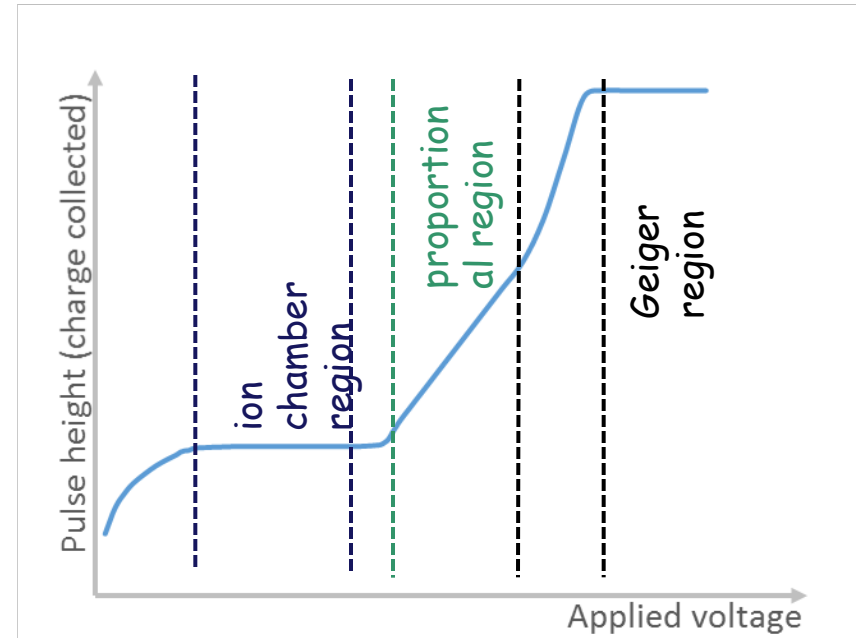


commercial radionuclide calibrator

$I = f A$

# Transfer instruments

The primary techniques described are often time-consuming and challenging, particularly when there is nothing to compare with.

Most standardisation labs such as NPL maintain a suite of transfer instruments, such as **ionisation chambers**, which are calibrated with material standardised by primary techniques.

These are easier and quicker to operate and allow standards to be disseminated easily.



The BIPM in Paris also maintain an ionisation chamber, which is used to compare results from different laboratories across the years.
This is known as the International Reference System (SIR)

# Ionisation chambers

The simplicity of the ionisation chamber is its key advantage.

The manufacturing process is reproducible, so calibration factors measured on one master chamber can be applied to replicate chambers.

For similar reasons, the instrument can be extremely stable.

The response varies very little over decades.

Factors measured 20-30 years ago (or more) can still be used.

# Ionisation chambers

**Monte Carlo simulation of an IC**

From R. Townson, F. Tessier and R. Galea EGSnrc calculation of activity calibration factors for the Vinten ionisation chamber

*Applied Radiation and Isotopes* ([Volume 134](), April 2018, Pages 100-104)
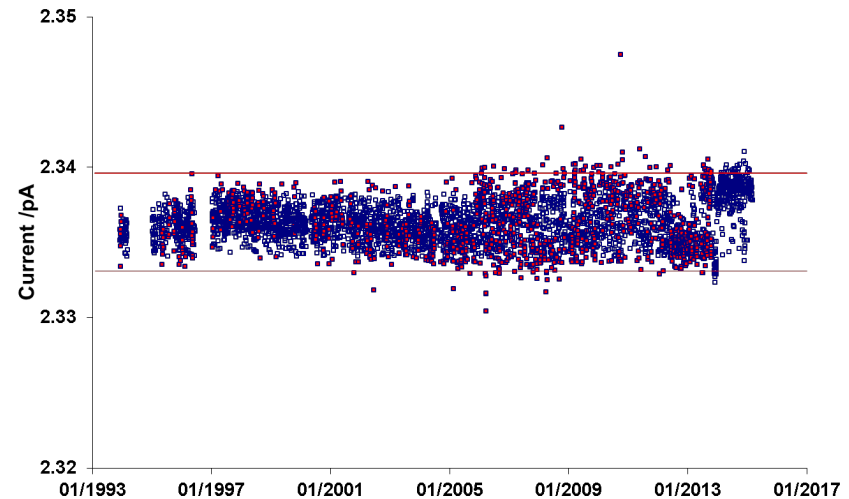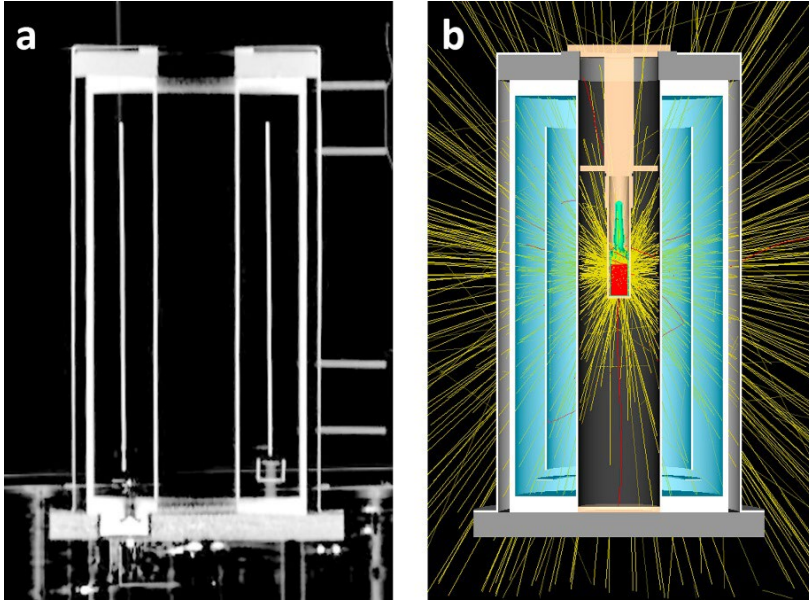
Ionisation chambers are not perfect.

To contain the pressurised gas, they have thick metal walls which will absorb most $\beta^-$ particles and low-energy gamma or X-ray emissions

A lot of interactions are needed to produce a measurable current – therefore they are only useful at higher activities

**They are also not selective – so are of little use when multiple radionuclides are present without extra work**

# Impurities – ionisation chambers

Response to $X$ (cal. factor $f_X$) in presence of $Y$ ($f_Y$) is given by:

$$I = f_X A_X + f_Y A_Y$$

$f_Y$ is not known unless $Y$ has been previously standardised.

A calibration curve can be derived, either by nonlinear fitting or by **Monte Carlo** simulation.

This can be used to derive a calibration factor for impurities.

If the gamma emissions of a particular radionuclide $Y$ form a set $\boldsymbol{I}$ with energies $E_{\gamma i}$ and emission probabilities $P_{\gamma i}$, then:
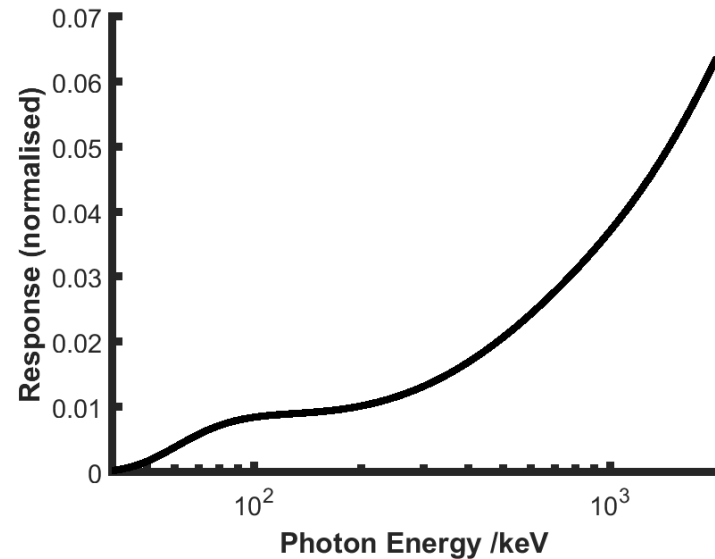


$$f_Y = \sum_{i \in \boldsymbol{I}} P_{\gamma_i}\, g(E_{\gamma,i}) \qquad I = f\,A$$

Where $g(E)$ is a function which describes the ionisation chamber response as a function of energy.

# High Resolution Gamma Spectrometry

A high purity germanium detector can also be used to prepare standards.

The activity calculated depends on:

• Having a precise detector calibration

• Knowing precisely the gamma emissions per decay

Germanium detectors can measure any sample type, and are selective, so can measure samples with more than one radionuclide present.
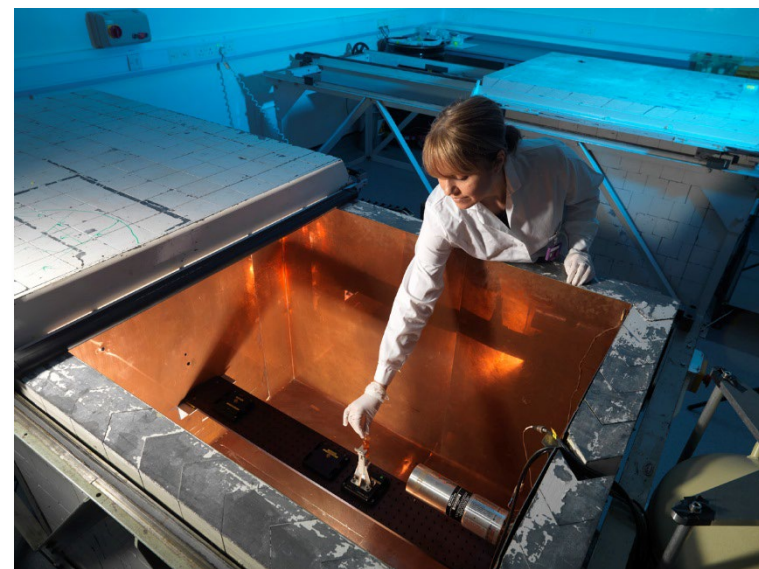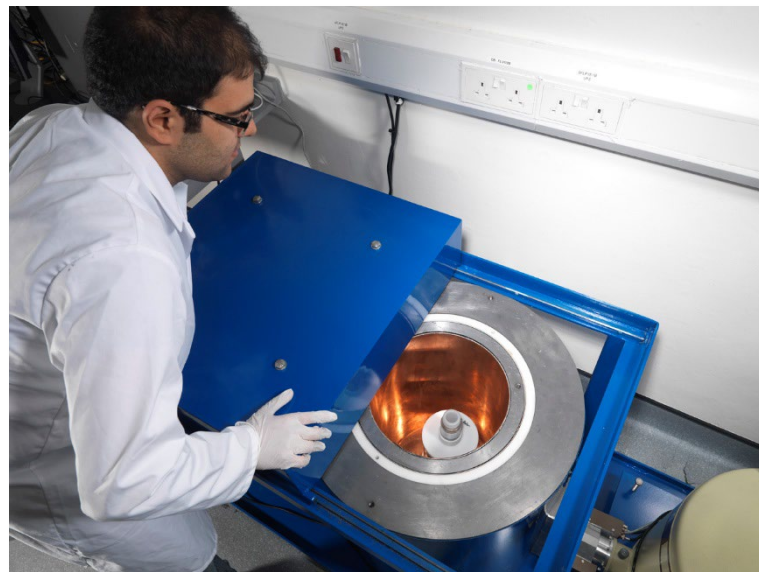
# NPL HPGe γ-ray spectrometers

- HPGe detectors, 'BART' and 'LORAX'.
- Located in 10 cm thick Pb walled 'coffins'
  - Cd/Cu grading to reduce Pb X-rays

- Samples held in place using engineered holders and kinematic mounting systems
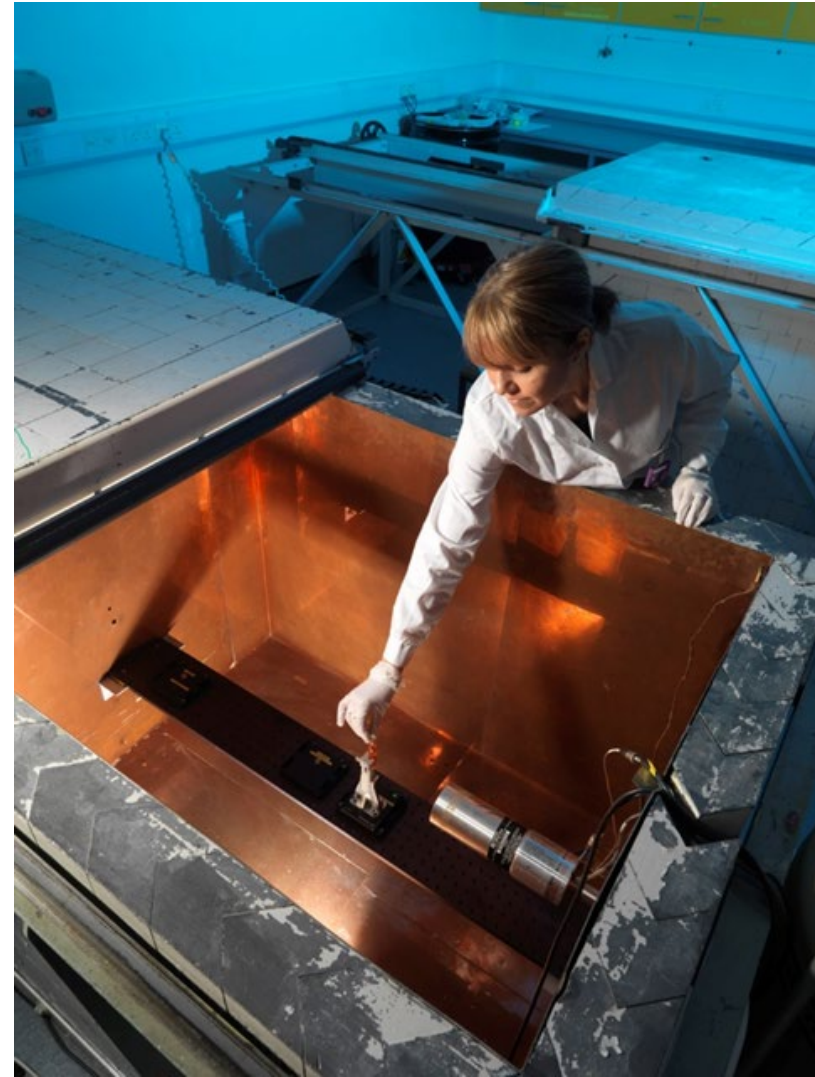
# Full-energy peak efficiency curves
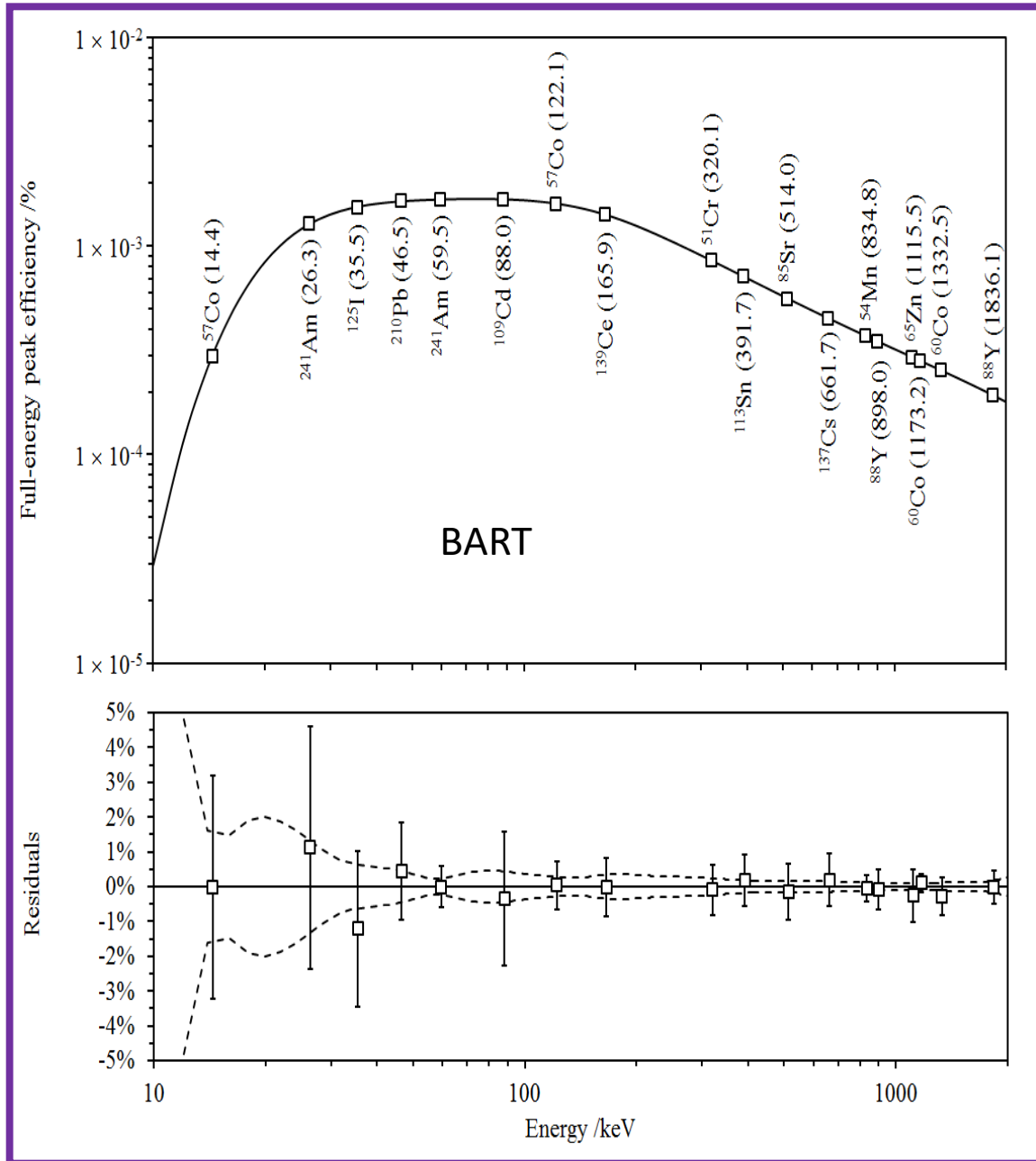
A 'high impact' example:

The standardisation of $^{223}$Ra.

# Nuclear Medicine: Xofigo™


Xofigo®
radium Ra 223 dichloride
INJECTION

- First $\alpha$ emitting radionuclide approved by the US FDA and licensed in the EC from Nov. 2013 - $^{223}RaCl_2$ solution.

- Targeted palliative treatment of bone metastases from late stage castration resistant prostate cancer
  - Extends patient life ~ average 3 months

- Under investigation for bone metastases from breast & ovarian cancer.

- Now used in >3,000 clinics worldwide; supplied through Bayer (formerly Algetha)



Range of α-particle
(short range – 2 to 10 cell diameters[2])

Optimize radiation dose to bone mets; minimize dose to bone marrow (minimize toxicity)

Bone marrow

Tumor

Range of β-particle
(long range – 10 to 1000 cell diameters[2])

Radionuclide

Bone

Bone Mineral
(Hydroxyapatite)

References: 1. Henriksen G, et al. *Cancer Res*. 2002;62:3120–3125. 2. Brechbiel MW. *Dalton Trans*. 2007;43:4918-4928.

**$^{223}$Ra ground state, 3/2$^+$**

**E$_\alpha$=5716(4) keV**

**E$_\alpha$=5607(4) keV**

FIG. 4. Energy level scheme of $^{219}$Rn up to 733 keV resulting from the present study. Alpha energies in keV, their intensities and hindrance factors (HF's) populating the levels are shown to the right and are taken from magnetic spectrograph measurements [2,19]. Transitions are shown as vertical lines with their energies in keV. Spins, parities, and configurations are shown in the simplified level scheme of Fig. 5.

# Interesting aside: (rare) Heavy-particle decay

- Possible to calculate Q values for emission of heavier nuclei
  - Is energetically possible for a large range of heavy nuclei to emit other light nuclei.

- $^{14}C$ emission observed from $^{222,223}Ra$
  - emission probability is much smaller than alpha decay

- Simple barrier penetration estimate can be attributed to very small probability to preform $^{14}C$ residue inside heavy nucleus



$0+$   38.0 s

$^{222}_{88}Ra$   $\alpha$

$^{14}_{6}C$

$3.5 \times 10^{-6}\%$

$Q_\alpha 6681$

$3/2+$   11.435 d

$^{223}_{88}Ra$   $\alpha$

$^{14}_{6}C$

$6.4 \times 10^{-8}\%$

$Q_\alpha 5979.31$
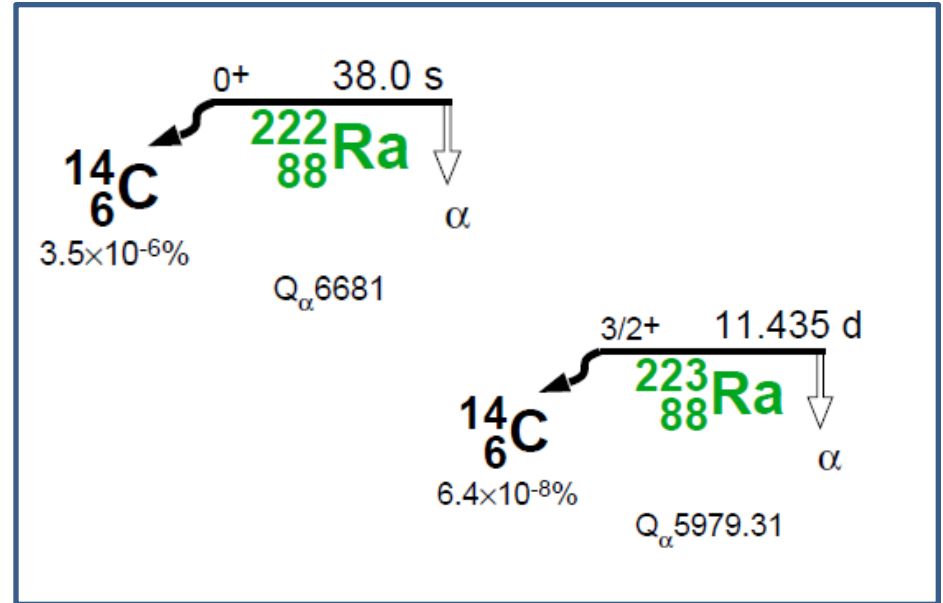
The exotic nuclear decay of $^{223}Ra$ by emission of $^{14}C$ nuclei has been investigated by use of an intense radioactive $^{227}Ac$ source and a magnetic spectrometer with a large solid angle. After a run of 5 d, a group of eleven events was observed at the expected location of $^{14}C$ in a $\Delta E$-$E$ telescope calibrated with a $^{14}C$ beam. A branching ratio of $(5.5 \pm 2.0) \times 10^{-10}$ was measured for the emission of $^{14}C$ nuclei relative to $\alpha$ particles from $^{223}Ra$ in agreement with the previously reported ratio of $(8.5 \pm 2.5) \times 10^{-10}$.

# Direct measurement of the half-life of $^{223}$Ra

CrossMark

S.M. Collins [a,*], A.K. Pearce [a], K.M. Ferreira [a], A.J. Fenwick [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

- Direct measurement of the $^{223}$Ra half-life using an ionisation chamber.
- New measured half-life of 11.4358 (28) days.
- Result consistent with the most precise published value.
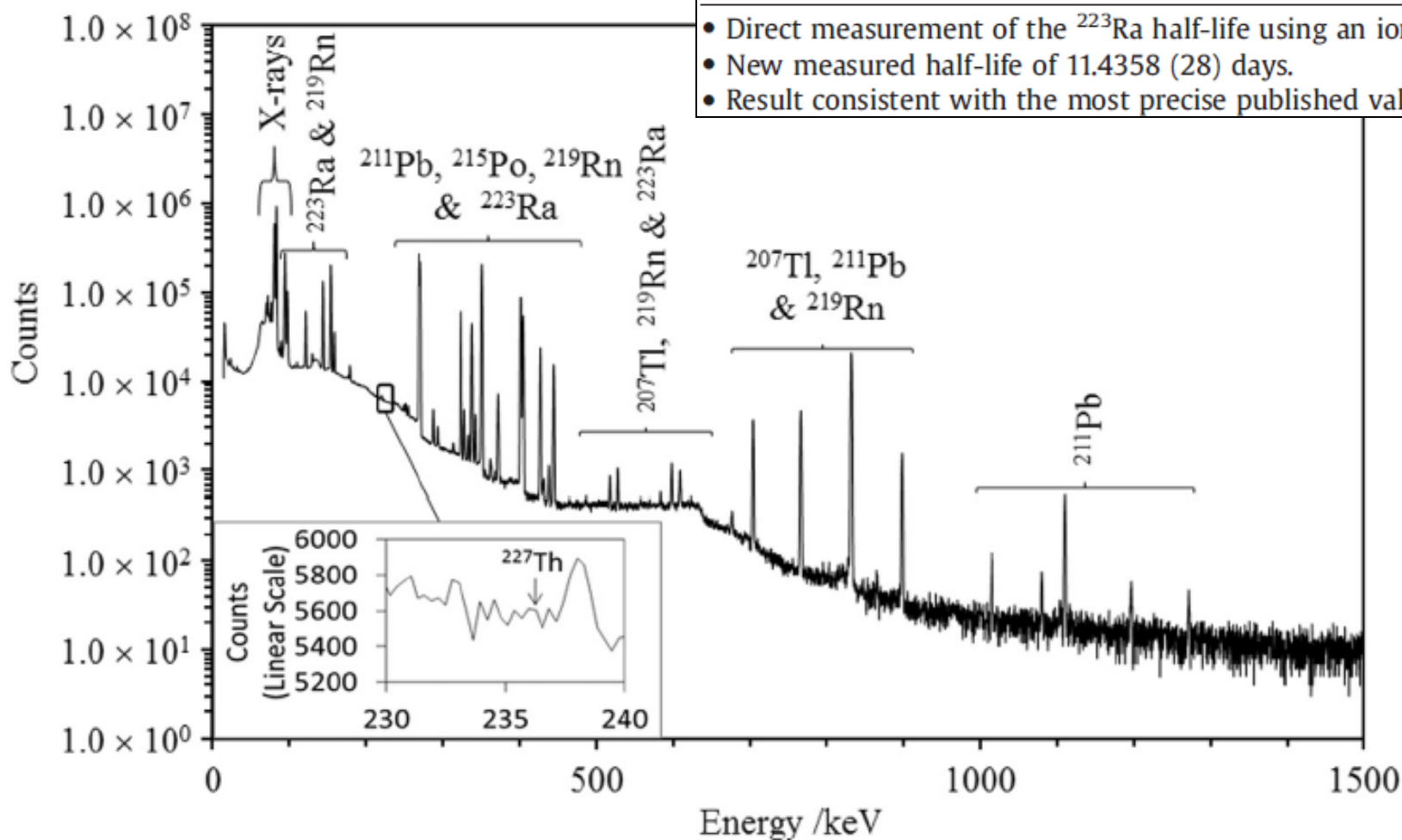
# Direct measurement of the half-life of $^{223}$Ra

S.M. Collins [a,*], A.K. Pearce [a], K.M. Ferreira [a], A.J. Fenwick [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
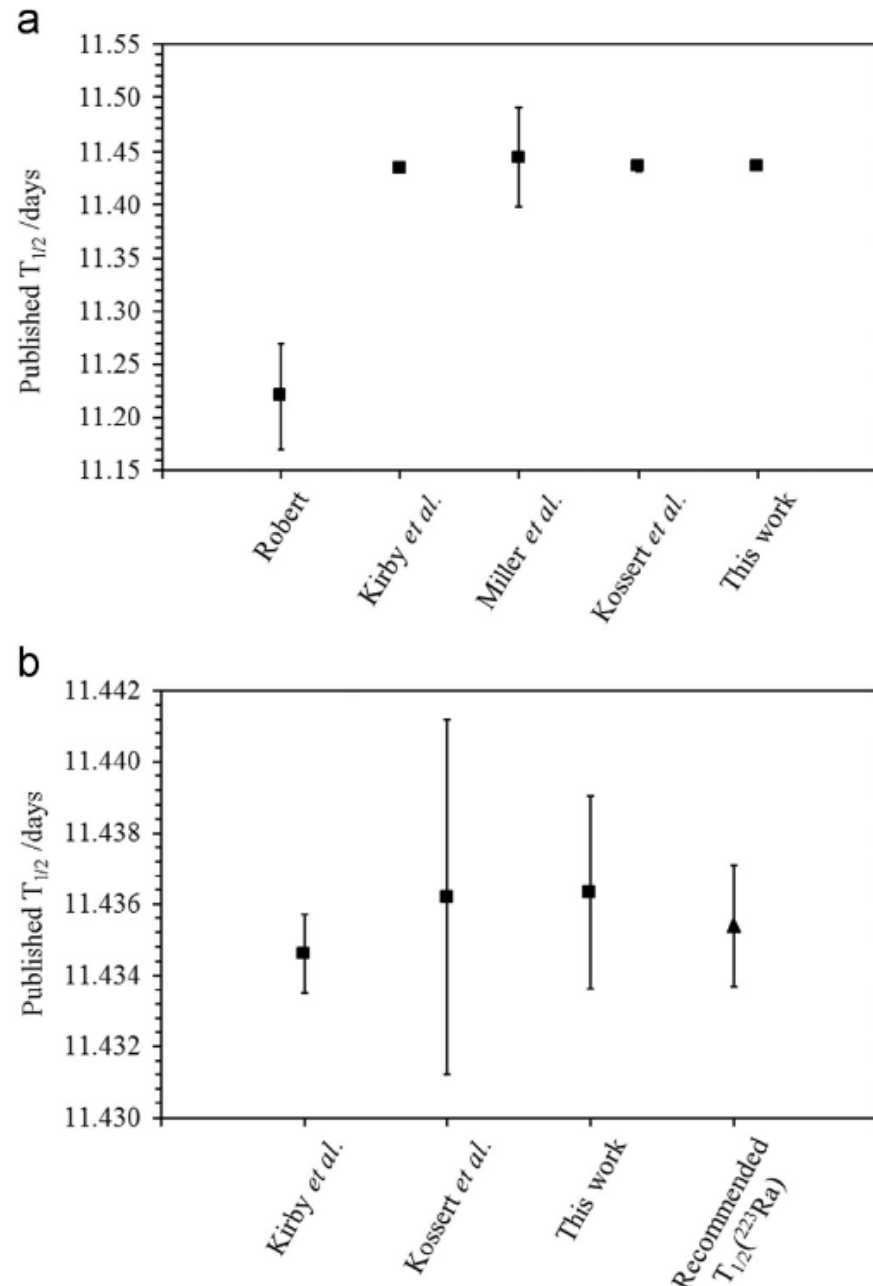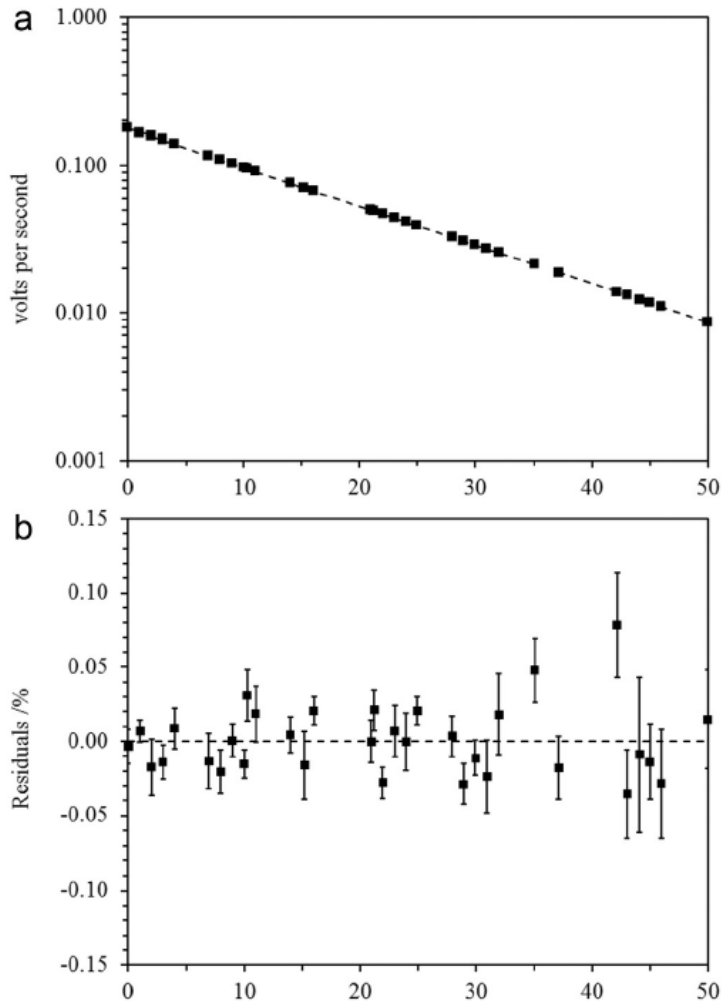
# A problem…absolute <sup>223</sup>Ra standardisation at NPL



9% discrepancy from NIST

- Different techniques showed excellent agreement….
    - Including the γ-spectrometry.

- So why do need new measurements of the γ-ray emission probabilities?
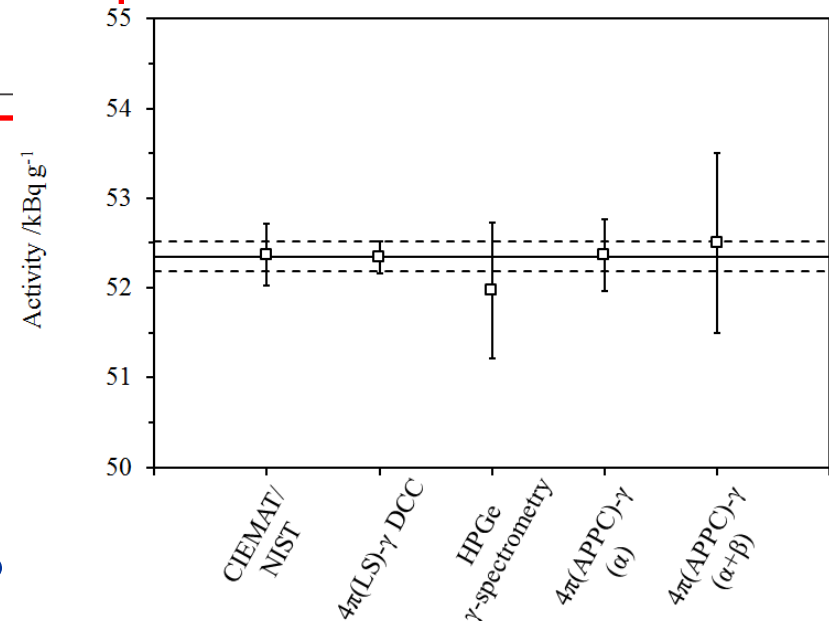
# Large discrepancies in measurements with published γ-ray emission probabilities of $^{223}$Ra.
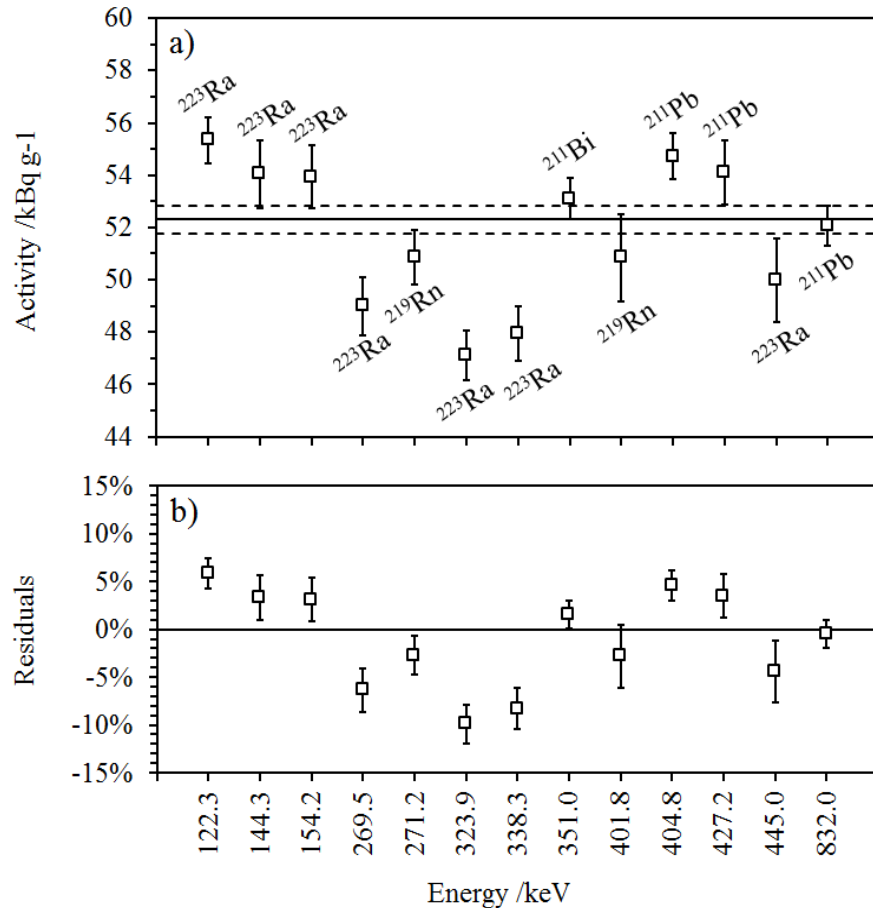


- The main γ-ray emissions from $^{223}$Ra are show using (previous) nuclear data taken from DDEP.

- An 18% range in the deduced activity using different gamma transitions was determined. **This is not good enough.**

- However, the weighted mean gives the 'right' result.

- Previous results 'suspicious' due to the large spread.

# Uncertainty budget of the $^{223}$Ra 269.5 keV γ-ray emission probability

| Uncertainty component | Relative uncertainty ($k = 1$) |
|---|---|
| Standard uncertainty of the weighted mean (includes the full-energy peak efficiency) | 0.24 % |
| Activity concentration | 0.33 % |
| Dead Time & Pile-up correction | 0.050 % |
| Radioactive decay correction | 0.010 % |
| Geometric reproducibility | 0.10 % |
| Gravimetric | 0.10 % |
| Detector stability | 0.20 % |
| Peak fitting | 0.10 % |
| True coincidence summing | 0.10 % |
| Total uncertainty | 0.50 % |

# Most up to date, accurate data on $^{223}$Ra decay.

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

CrossMark

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

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**Table 4**
Absolute γ-ray emission probabilities per 100 decays of $^{223}$Ra and decay progeny in equilibrium.

| Energy (keV) | Source | $I_\gamma$ (%) | Energy (keV) | Source | $I_\gamma$ (%) | Energy (keV) | Source | $I_\gamma$ (%) | Energy (keV) | Source | $I_\gamma$ (%) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 103.9(5) | $^{223}$Ra | 0.0119(6) | 323.9(6) | $^{223}$Ra | 3.655(18) | 438.8(6) | $^{215}$Po | 0.0533(7) | 675.4(6) | $^{211}$Pb | 0.0058(6) |
| 106.7(4) | $^{223}$Ra | 0.0213(11) | 328.4(6) | $^{223}$Ra, $^{211}$Po, $^{207}$Tl | 0.2021(16) | 445.0(6) | $^{223}$Ra | 1.218(6) | 676.9(6) | $^{219}$Rn | 0.0184(5) |
| 110.8(5) | $^{223}$Ra | 0.0512(10) | 333.9(6) | $^{223}$Ra | 0.0756(6) | 462.8(6) | $^{219}$Rn | 0.0011(5) | 704.6(7) | $^{211}$Pb | 0.498(3) |
| 122.3(5) | $^{223}$Ra | 1.312(6) | 338.3(6) | $^{223}$Ra | 2.605(13) | 487.3(5) | $^{223}$Ra | 0.0083(3) | 707.8(7) | $^{219}$Rn | 0.0034(4) |
| 130.6(5) | $^{219}$Rn | 0.1478(10) | 342.9(6) | $^{223}$Ra, $^{211}$Pb | 0.1958(21) | 500.2(6) | $^{223}$Ra | 0.0013(5) | 711.4(7) | $^{223}$Ra | 0.0037(3) |
| 144.3(5) | $^{223}$Ra | 3.481(16) | 351.1(6) | $^{211}$Bi | 13.17(7) | 504.1(6) | $^{211}$Pb | 0.0022(4) | 727.4(7) | $^{223}$Ra | 0.0024(7) |
| 154.2(5) | $^{223}$Ra | 6.02(3) | 355.5(6) | $^{223}$Ra | 0.0124(15) | 517.6(6) | $^{219}$Rn | 0.0453(5) | 766.4(7) | $^{211}$Pb | 0.685(4) |
| 158.7(5) | $^{223}$Ra | 0.749(4) | 361.7(6) | $^{211}$Pb | 0.0341(7) | 522.6(6) | $^{223}$Ra | 0.0021(6) | 831.9(7) | $^{211}$Pb | 3.448(16) |
| 175.6(5) | $^{223}$Ra | 0.01578(10) | 363.0(6) | $^{223}$Ra | 0.0192(9) | 527.6(6) | $^{223}$Ra | 0.0659(8) | 835.6(7) | $^{219}$Rn | 0.00364(19) |
| 177.4(5) | $^{223}$Ra | 0.0426(8) | 368.4(6) | $^{223}$Ra | 0.0134(4) | 531.4(6) | $^{223}$Ra | 0.0028(9) | 865.8(6) | $^{211}$Pb | 0.00540(21) |
| 179.7(5) | $^{223}$Ra | 0.1613(10) | 371.7(6) | $^{223}$Ra | 0.435(3) | 537.5(6) | $^{223}$Ra | 0.0033(6) | 891.3(7) | $^{219}$Rn | 0.00107(20) |
| 221.4(5) | $^{223}$Ra | 0.0304(10) | 372.9(6) | $^{223}$Ra | 0.1133(13) | 542.1(6) | $^{223}$Ra | 0.0026(6) | 897.8(7) | $^{211}$Po, $^{207}$Tl | 0.2725(15) |
| 224.0(5) | $^{219}$Rn | 0.0056(14) | 376.2(6) | $^{223}$Ra | 0.0056(4) | 545.9(6) | $^{223}$Ra | 0.0028(6) | 1014.7(7) | $^{211}$Pb | 0.0171(4) |
| 249.4(5) | $^{223}$Ra | 0.0375(9) | 383.3(5) | $^{223}$Ra | 0.0023(6) | 555.9(5) | $^{219}$Rn | 0.0026(7) | 1074.5(7) | $^{219}$Rn | 0.00044(12) |
| 251.9(5) | $^{223}$Ra | 0.0640(11) | 386.3(5) | $^{223}$Ra, $^{219}$Rn | 0.0052(7) | 564.4(5) | $^{219}$Rn | 0.0035(4) | 1080.1(7) | $^{211}$Pb | 0.01228(21) |
| 255.1(5) | $^{223}$Ra | 0.0499(13) | 390.1(5) | $^{223}$Ra | 0.0053(7) | 569.6(7) | $^{211}$Po, $^{207}$Tl | 0.0043(5) | 1103.3(8) | $^{211}$Pb | 0.00380(12) |
| 269.5(6) | $^{223}$Ra | 13.37(7) | 401.8(6) | $^{219}$Rn | 6.57(3) | 573.7(7) | $^{223}$Ra | 0.0029(13) | 1109.5(8) | $^{211}$Pb | 0.1113(7) |
| 271.3(6) | $^{219}$Rn | 10.75(6) | 404.8(6) | $^{211}$Pb, $^{215}$At | 4.011(19) | 598.6(7) | $^{223}$Ra | 0.0867(12) | 1196.2(8) | $^{211}$Pb | 0.01052(17) |
| 288.2(6) | $^{223}$Ra | 0.1498(16) | 427.1(6) | $^{211}$Pb | 1.890(9) | 609.3(7) | $^{223}$Ra, $^{219}$Rn, $^{211}$Pb | 0.0543(7) | 1234.3(8) | $^{211}$Pb | 0.00092(8) |
| 293.6(5) | $^{219}$Rn | 0.0688(7) | 430.4(6) | $^{223}$Ra, $^{211}$Pb | 0.0206(19) | 619.8(6) | $^{219}$Rn | 0.0056(12) | 1270.7(8) | $^{211}$Pb | 0.00624(19) |
| 313.7(6) | $^{211}$Pb | 0.0276(5) | 432.4(6) | $^{223}$Ra | 0.0297(14) | 623.4(5) | $^{223}$Ra | 0.0082(8) | | | |

# Comparison of main γ-ray emission probability of $^{223}$Ra and decay progeny to DDEP recommended value.

| Radionuclide | Energy /keV | I$_\gamma$(This work) % | I$_\gamma$(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 NPL & (new) NIST and PTB Data.

| Radionuclide | Energy /keV | $I_\gamma$(NPL) % | $I_\gamma$(NIST) % | $I_\gamma$(PTB) % | $\chi^2/(n-1)$ |
|---|---|---|---|---|---|
| $^{223}$Ra | 122.3 | 1.312 (6) | 1.30 (1) | 1.304 (12) | 0.3 |
| $^{223}$Ra | 144.3 | 3.481 (16) | 3.51 (3) | 3.469 (20) | 0.3 |
| $^{223}$Ra | 154.2 | 6.02 (3) | 6.08 (6) | 6.03 (5) | 0.2 |
| $^{223}$Ra | 269.5 | 13.37 (7) | 13.24 (12) | 13.16 (15) | 0.5 |
| $^{223}$Ra | 323.9 | 3.655 (18) | 3.63 (2) | 3.661 (21) | 0.3 |
| $^{223}$Ra | 338.3 | 2.605 (13) | 2.59 (2) | 2.614 (13) | 0.3 |
| $^{223}$Ra | 445.0 | 1.218 (6) | 1.217 (8) | 1.222 (6) | 0.1 |
| $^{219}$Rn | 271.2 | 10.75 (6) | 10.69 (10) | 10.87 (12) | 0.3 |
| $^{219}$Rn | 401.8 | 6.57 (3) | 6.56 (4) | 6.62 (4) | 0.3 |
| $^{211}$Pb | 404.8 | 4.011 (19) | 4.01 (3) | 4.05 (5) | 0.1 |
| $^{211}$Pb | 427.2 | 1.890 (9) | 1.89 (1) | 1.912 (10) | 0.8 |
| $^{211}$Pb | 832.0 | 3.448 (16) | 3.48 (3) | 3.430 (17) | 0.5 |
| $^{211}$Bi | 351.0 | 13.17 (7) | 13.11 (9) | 13.24 (6) | 0.4 |
| No. of γ-rays reported | | 83 | 15 | 43 | |

- Now, good agreement between the three NMIs.

# CTBTO

- Co-located with UN and IAEA in Vienna

- Organisation created to support implementation and verification of international Comprehensive Test Ban Treaty

- Aim to detect any nuclear explosion conducted on Earth – in the underground, underwater or in the atmosphere

- International Monitoring System (IMS) has been established to coordinate monitoring and sharing of data

- IMS comprised of 321 stations and 16 laboratories across the globe, with sensors for:

  - Seismic (underground), hydroacoustic (undersea), infrasound (atmosphere) and radionuclide (particulates and radioxenon in the atmosphere)

Decay chain (A = 131):

$^{131}$In (0.28 s, I: 0.04%, C: 0.04%) — 98% → $^{131}$Sn (56 s, I: 1.4%, C: 1.4%) — 100% → $^{131}$Sb (23 m, I: 1.5%, C: 2.9%)
- 6.5% → $^{131m}$Te (33 h, I: 0.22%, C: 0.43%)
- 93% → $^{131}$Te (25 m, I: 0.08%, C: 2.87%)

$^{131m}$Te — 75% → $^{131}$I; 26% → $^{131}$Te
$^{131}$Te — 100% → $^{131}$I
$^{131}$I (8.0 d, I: <0.01%, C: 3.2%)
- 1.1% → $^{131m}$Xe (12 d, I: <0.01%, C: 0.05%)
- 99% → $^{131}$Xe (stable)
$^{131m}$Xe — 100% → $^{131}$Xe

Decay chain (A = 133):

$^{133}$In (0.18 s, I: < 0.01%, C: < 0.01%) — 15% → $^{133}$Sn (1.5 s, I: 0.14%, C: 0.14%) — 100% → $^{133}$Sb (2.3 m, I: 1.5%, C: 1.7%)
- 29% → $^{133m}$Te (55 m, I: 2.1%, C: 2.8%)
- 71% → $^{133}$Te (13 m, I: 2.6%, C: 3.9%)

$^{133m}$Te — 83% → $^{133}$I; 17% → $^{133}$Te
$^{133}$Te — 100% → $^{133}$I
$^{133}$I (21 h, I: 0.4%, C: 6.7%)
- 2.9% → $^{133m}$Xe (2.2 d, I: <0.01%, C: 0.19%)
- 97% → $^{133}$Xe (5.2 d, I: <0.01%, C: 6.7%)
$^{133m}$Xe — 100% → $^{133}$Xe

Decay chain (A = 135):

$^{135}$In (0.13 s, << 0.001%) — 100% → $^{135}$Sn (0.42 s, I: < 0.01%, C: <0.01%) — 69% → $^{135}$Sb (1.7 s, I: 0.25%, C: 0.25%) — 82% → $^{135}$Te (19 s, I: 2.5%, C: 2.7%) — 100% → $^{135}$I (6.6 h, I: 3.6%, C: 6.3%)
- 16% → $^{135m}$Xe (15 m, I: 0.19%, C: 1.1%)
- 84% → $^{135}$Xe (9.1 h, I: 0.12%, C: 6.6%)
$^{135m}$Xe — 100% → $^{135}$Xe

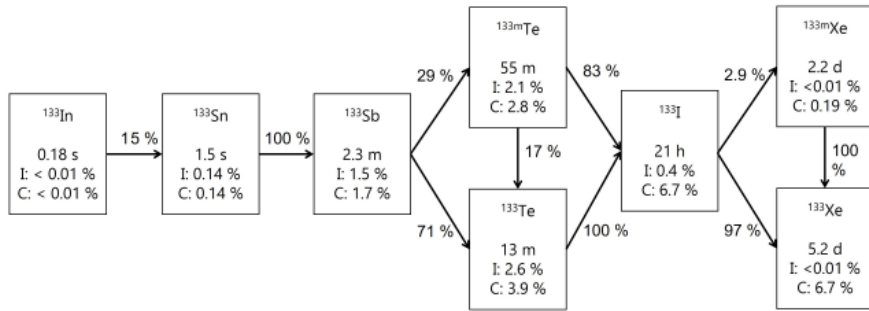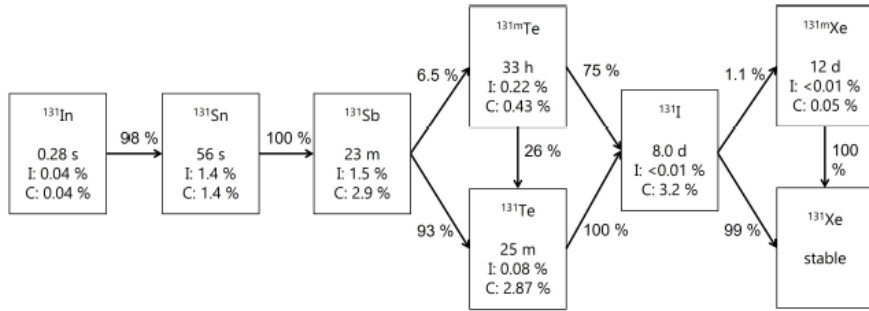Legend: Xe-133, Xe-135, Xe-133m, Xe-135m, Xe-131m

FIGURE 2.12: Simulated activity of five radioxenon isotopes relative to $^{133}$Xe maximum activity, using $^{235}$U thermal neutron-induced fission yields for 10 days. $^{133}$Xe: black dashed; $^{135}$Xe: black solid; $^{133m}$Xe: orange; $^{131m}$Xe: green; $^{135m}$Xe: blue dotted. The activities here are calculated based on no fractionation from the parent nuclei. $^{135m}$Xe is included however it is much shorter-lived ($t_{1/2}$=15.29(5) min).

| Nuclide | Half-life | Independent Yield (%) | Cumulative Yield (%) | t($Y_{max}$) |
|---|---|---|---|---|
| $^{131m}$Xe | 11.9 d | 3.480E-7 | 0.0405 | 15 d |
| $^{133m}$Xe | 2.2 d | 1.890E-3 | 0.1890 | 2 d |
| $^{133}$Xe | 5.2 d | 6.660E-4 | 6.7000 | 3 d |
| $^{135}$Xe | 9.1 h | 7.850E-2 | 6.5400 | 11 h |

BLE 2.3: Fission yield data and associated NuDIC results for $^{235}$U thermal fission om England and Rider [17]). The maximum yield is calculated and the number of s of ingrowth/decay required to reach it. t($Y_{max}$) is the time at which the maximum eld is reached following ingrowth of parent fission product nuclei, post-detonation.
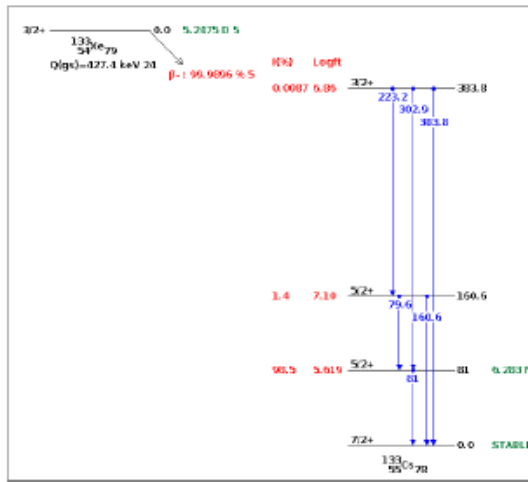
(A) Nuclear decay scheme for $^{133}$Xe $\beta^-$–decay to $^{133}$Cs
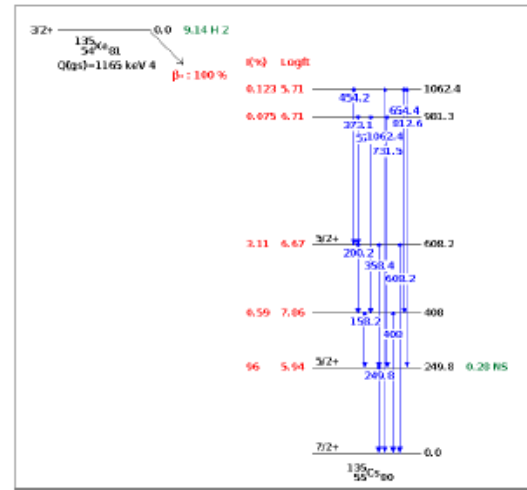
(B) Nuclear decay scheme for $^{135}$Xe $\beta^-$–decay to $^{135}$Cs
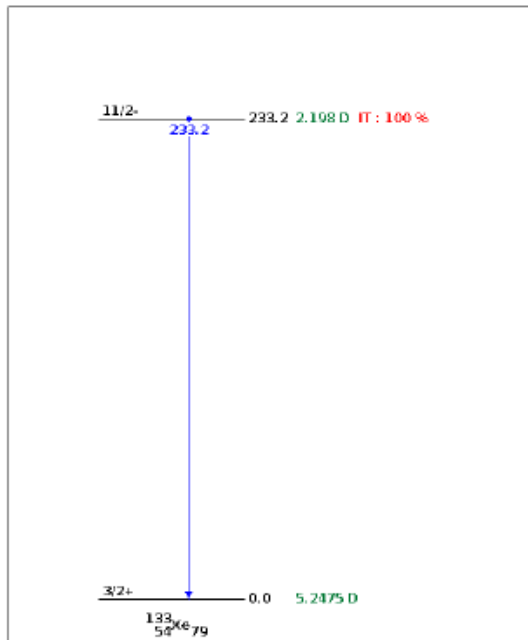
(C) Nuclear decay scheme for $^{133m}$Xe internal conversion decay to $^{133}$Xe
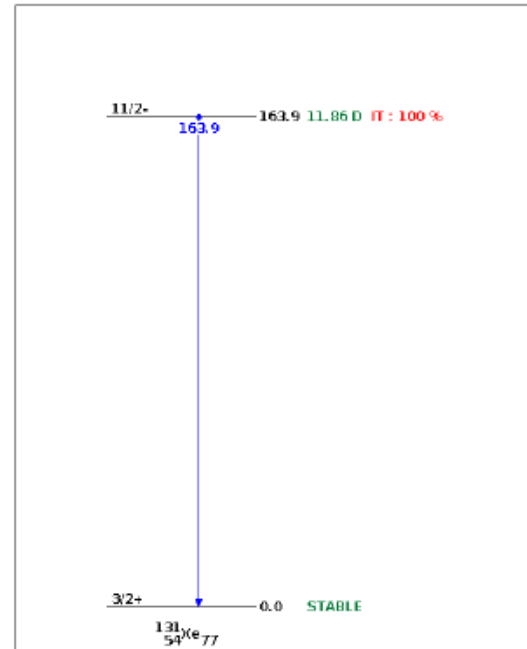
(D) Nuclear decay scheme for $^{131m}$Xe internal conversion decay to $^{131}$Xe

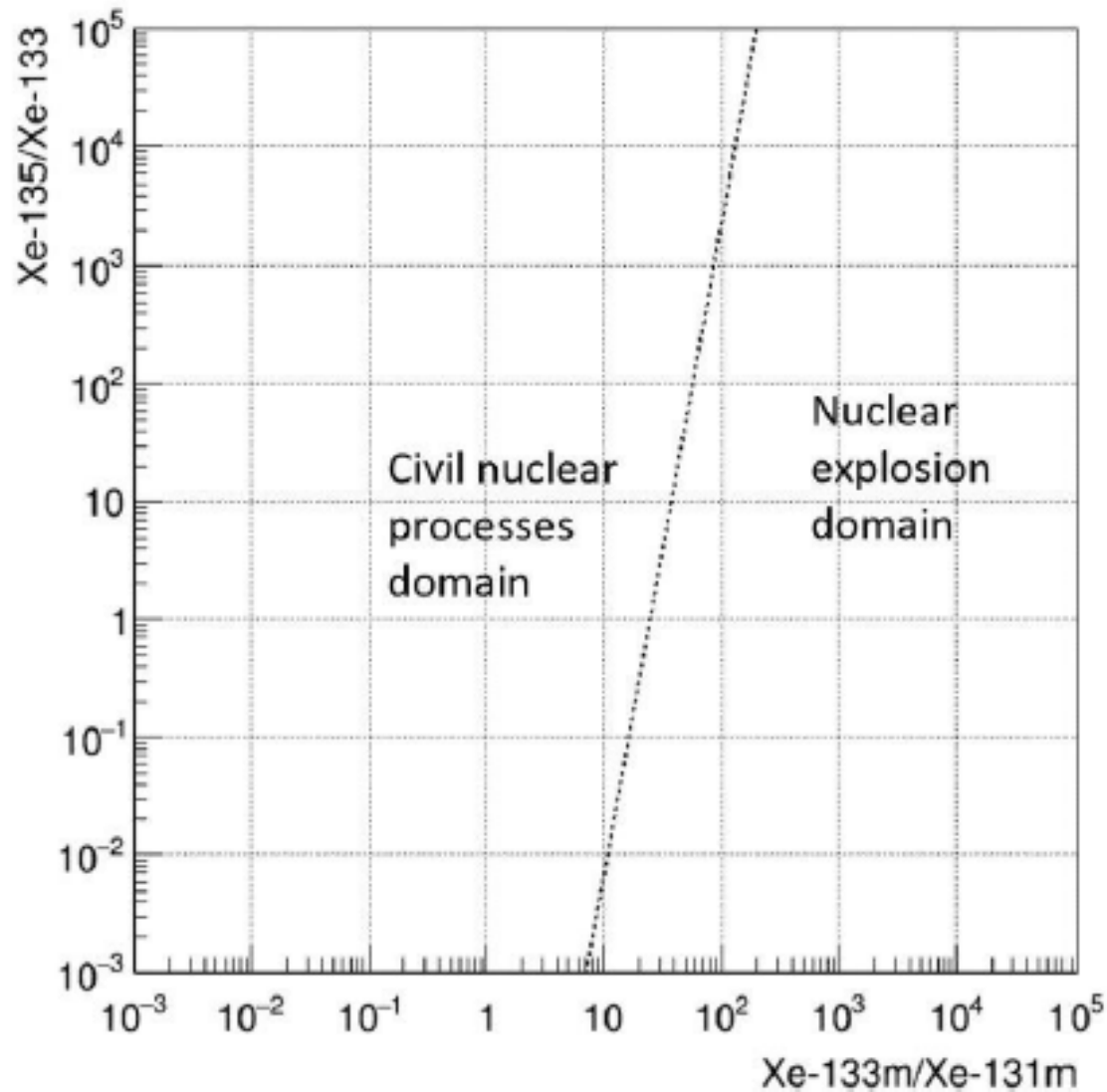FIGURE 2.14: Radioxenon isotopic ratios plot showing the ratios that are consistent with civil nuclear facilities (left) and ratios that are consistent with a nuclear explosion (right). Figure from Goodwin *et al.* [40]

| Parent Radionuclide | Electron Emission Type | Electron Emission Energy (keV) | Photon Emission Type | Photon Energy (keV) | Photons per 100 decays |
|---|---|---|---|---|---|
| $^{135}$Xe | $\beta^-$ | max. 910(5) | $\gamma$ | 249.794(15) | 89.6(16) |
| | | | Cs X K$_{\beta2}$ | 35.9003 | 0.19(1) |
| | | | Cs X K$_{\beta1}$ | 35.1045 | 0.80(4) |
| | | | Cs X K$_{\alpha1}$ | 30.9731 | 2.69(13) |
| | | | Cs X K$_{\alpha2}$ | 30.6254 | 1.45(7) |
| $^{133}$Xe | $\beta^-$ | max. 346.4(24) | $\gamma$ | 80.9979(11) | 37.0(3) |
| | | | Cs X K$_{\beta2}$ | 35.822 | 1.78(6) |
| | | | Cs X K$_{\beta1}$ | 34.9873 | 7.31(15) |
| | | | Cs X K$_{\alpha1}$ | 30.9731 | 25.0(5) |
| | | | Cs X K$_{\alpha2}$ | 30.6254 | 13.54(24) |
| $^{133m}$Xe | c.e. | 198.655(15) | Xe X K$_\beta$ | 33.56 - 34.55 | 10.55(22) |
| | | | Xe X K$_\alpha$ | 29.459, 29.779 | 45.30(81) |
| $^{131m}$Xe | c.e. | 129.366(8) | Xe X K$_\beta$ | 33.56 - 34.55 | 10.27(23) |
| | | | Xe X K$_\alpha$ | 29.459, 29.779 | 44.20(82) |

# Can irradiate U targets for standardised noble gaseous radioactive (Kr, Xe) sources
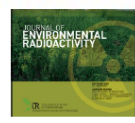
## Production and measurement of fission product noble gases

Matthew A. Goodwin [a,b,*], Steven J. Bell [c], Richard Britton [d], Ashley V. Davies [a], Marc Abilama [c], Sean M. Collins [b,c], Robert Shearman [c], Patrick H. Regan [b,c]

[a] AWE Aldermaston, Reading, Berkshire, RG7 4PR, UK
[b] Department of Physics, University of Surrey, Guildford, GU2 7XH, UK
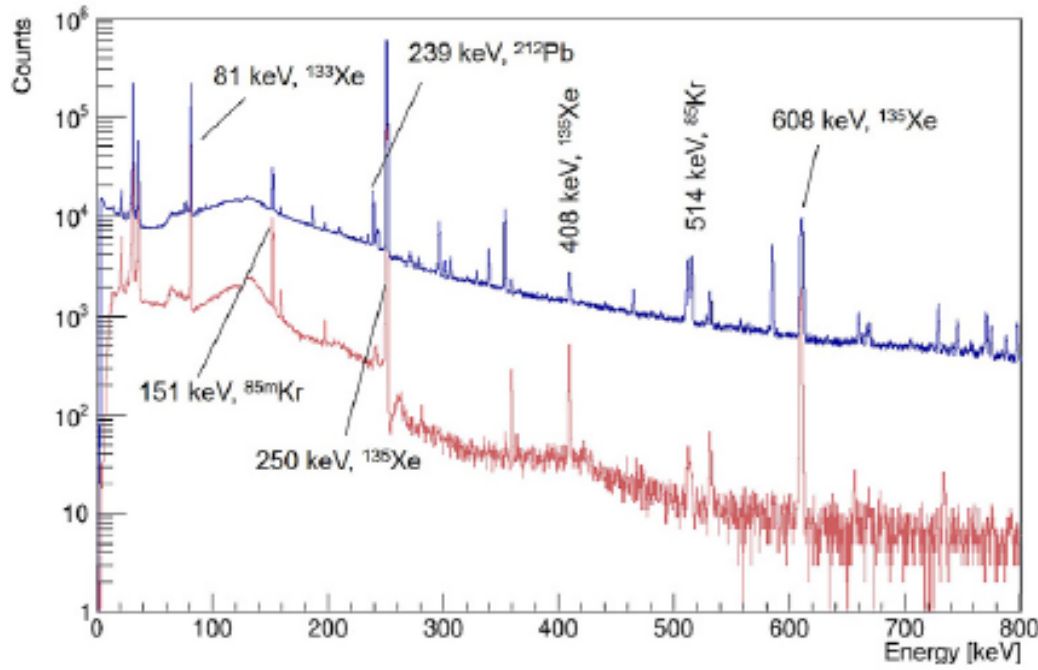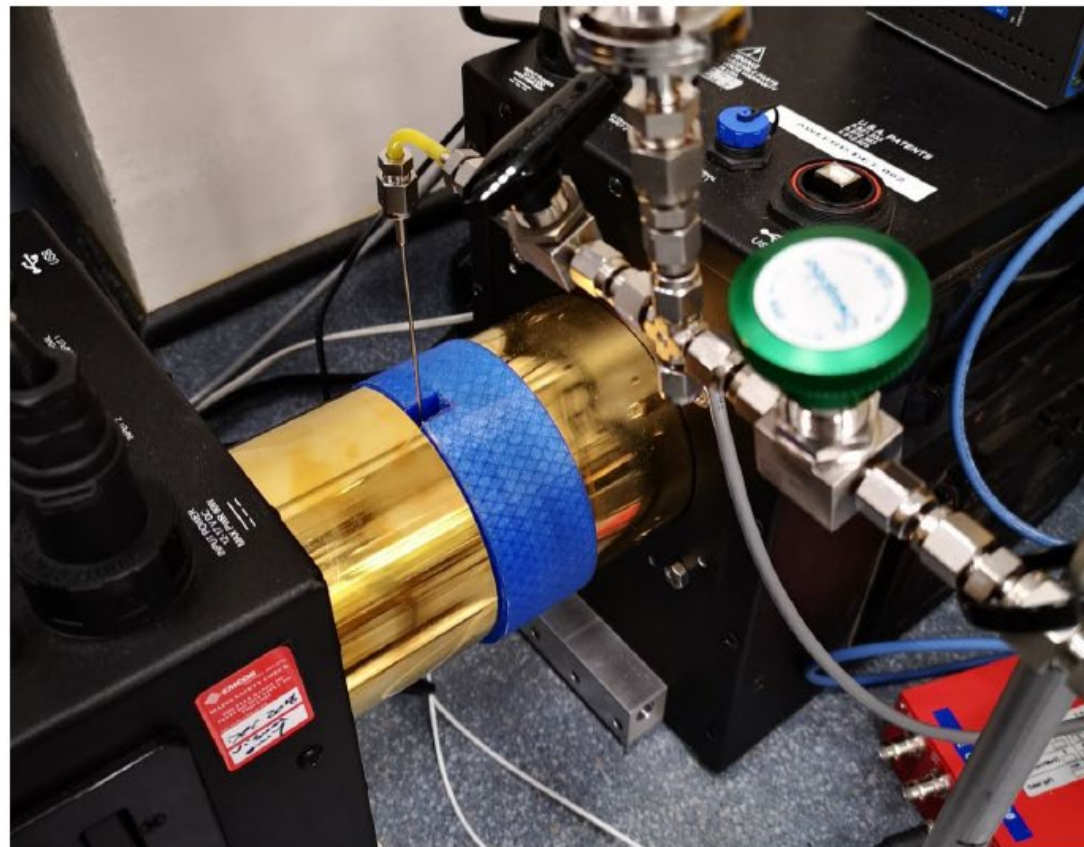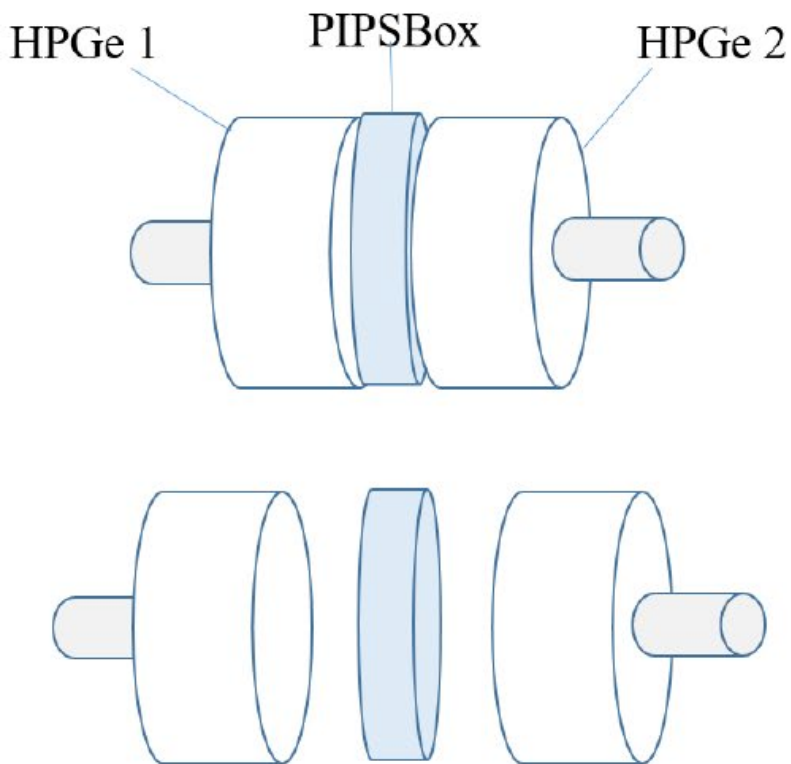[c] National Physical Laboratory, Teddington, Middlesex, TW11 0LW, UK

**Table 3**

Nuclide identification from a peak search of the $\gamma$-ray spectrum from Extraction 1 using the full acquisition (t = 170,731 s).† X-rays from Xe/Cs minus Ge fluoresence (escape peak). The relative $\gamma$-ray intensity is the full energy peak integral divided by the simulated $\gamma$-ray detection efficiency, decay-corrected to the start of the acquisition relative to the 250 keV $^{135}$Xe peak (see Eq. (1)).

| Fitted Energy (keV) | Parent Nucleus | Signature Type | $\gamma$-ray emission probability (%) | Rel. $\gamma$-ray Intensity (RGI) ($\times$ 1000) | Comment |
|---|---|---|---|---|---|
| 20.4 | Xe/Cs† | $e^-$-X | – | – | Multiplet |
| 30.7 | Xe/Cs X $K_\alpha$ | $e^-$-X | – | – | Multiplet |
| 35.3 | Xe/Cs X $K_\beta$ | $e^-$-X | – | – | Multiplet |
| 80.9 | $^{133}$Xe | $\beta - \gamma$ | 37.3(4) | 47.4(7.1) | 81.0 + 79.6 keV |
| 122.8 | $^{88}$Kr | $\beta - \gamma$ | 0.20(1) | 0.807(12) | |
| 129.1 | $^{85m}$Kr | $\beta - \gamma$ | 0.30(8) | 4.73(71) | |
| 151.4 | $^{85m}$Kr | $\beta - \gamma$ | 75.2(8) | 38.1(5.7) | |
| 158.6 | $^{135}$Xe | $\beta - \gamma$ | 0.29(1) | 2.40(36) | |
| 196.6 | $^{88}$Kr | $\beta - \gamma$ | 26(1) | 4.42(67) | |
| 233.4 | $^{133m}$Xe | $\gamma$ | 10.1(2) | 0.65(98) | |
| 240.6 | $^{88}$Kr | $\beta - \gamma$ | 0.25(1) | 0.78(12) | |
| 250.2 | $^{135}$Xe | $\beta - \gamma$ | 90.0(3) | 1000(7) | |
| 305.1 | $^{85m}$Kr | $\beta - \gamma$ | 14.0(4) | 5.32(80) | $^{85m}$Kr > $^{85}$Kr |
| 358.5 | $^{135}$Xe | $\beta - \gamma$ | 0.22(1) | 1.94(29) | |
| 390.0 | $^{88}$Kr | $\beta - \gamma$ | 0.64(5) | 0.87(14) | |
| 407.7 | $^{135}$Xe | $\beta - \gamma$ | 0.36(2) | 5.83(88) | |
| 438.9 | $^{88}$Rb | $\beta - \gamma$ | 0.015(4) | 1.35(21) | |
| 451.1 | $^{85m}$Kr | $\beta - \gamma$ | 0.011(4) | 0.88(14) | |
| 454.4 | $^{135}$Xe | $\beta - \gamma$ | 0.004(1) | 0.55(9) | |
| 514.3 | $^{85}$Kr | $\gamma$ | 0.43(1) | 4.76(72) | $^{85}$Kr > $^{85}$Rb |
| 526.4 | $^{135m}$Xe | $\gamma$ | 80.6(6) | 35.5(5.4) | |
| 530.3 | $^{133}$I | $\beta - \gamma$ | 87(2) | 2.81(42) | |
| 608.3 | $^{135}$Xe | $\beta - \gamma$ | 2.9(1) | 81.4(1.2) | $^{214}$Bi Interference |
| 731.9 | $^{135}$Xe | $\beta - \gamma$ | 0.055(4) | 0.39(6) | |
| 834.9 | $^{88}$Kr | $\beta - \gamma$ | 13(2) | 6.75(10) | |
| 898.2 | $^{88}$Rb | $\beta - \gamma$ | 14.4(2) | 1.73(27) | |
| 1836.5 | $^{88}$Rb | $\beta - \gamma$ | 22.8(1) | 5.07(78) | |



Gamma-ray spectrum: Counts vs Energy [keV], with labelled peaks: 81 keV $^{133}$Xe; 239 keV $^{212}$Pb; 151 keV $^{85m}$Kr; 250 keV $^{135}$Xe; 408 keV $^{135}$Xe; 514 keV $^{85}$Kr; 608 keV $^{135}$Xe.

# β-β-γ-γ

Goodwin et al, 2019. Developing a laboratory-based beta-gamma coincidence detection system: Progress so far. SNT 2019, Vienna.

# β-β-γ-γ
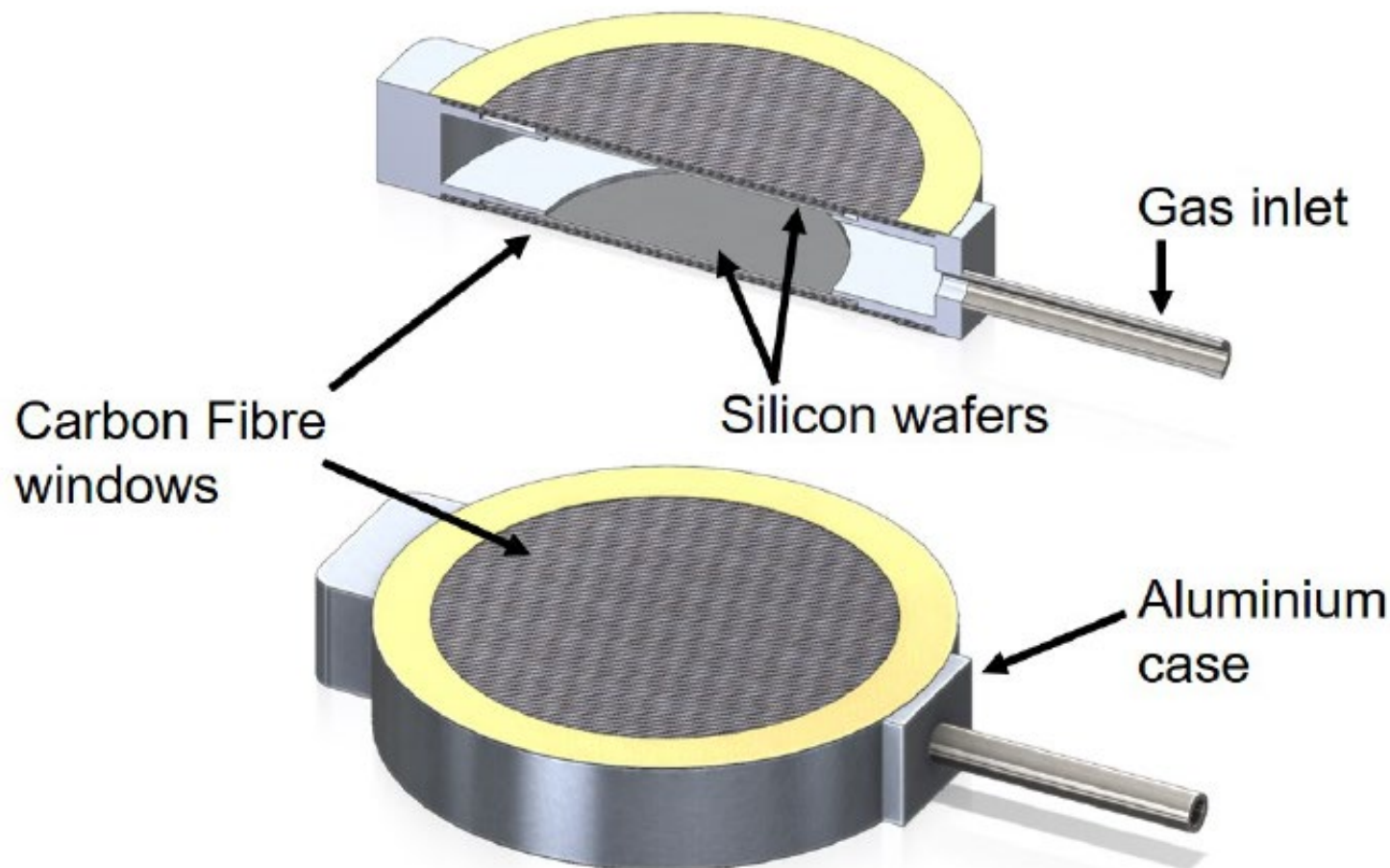


Goodwin et al, 2019. A high-resolution β-γ coincidence spectrometry system for radioxenon measurements. NIMA, 978, 164452.
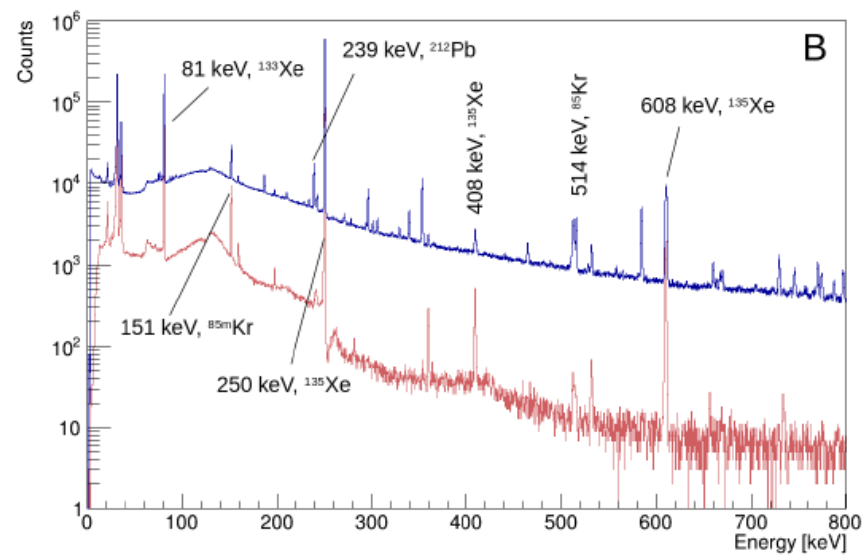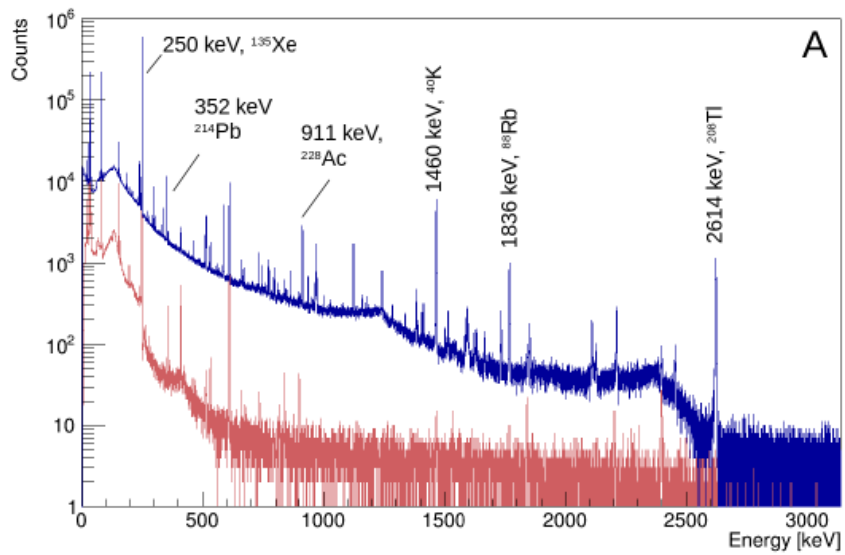https://doi.org/10.1016/j.nima.2020.164452

FIGURE 6.5: γ-spectra from the first extraction of gas (22 hours after irradiation end). Live time: 170,731 s. Blue: Singles γ-spectrum, red: β-gated γ-spectrum. Photon energy range 0-3 MeV (A) and 0-800 keV (B).
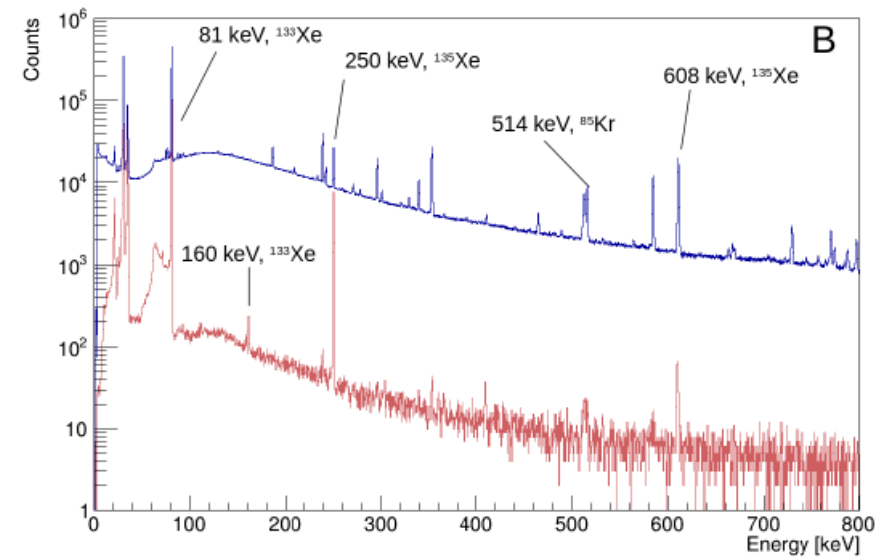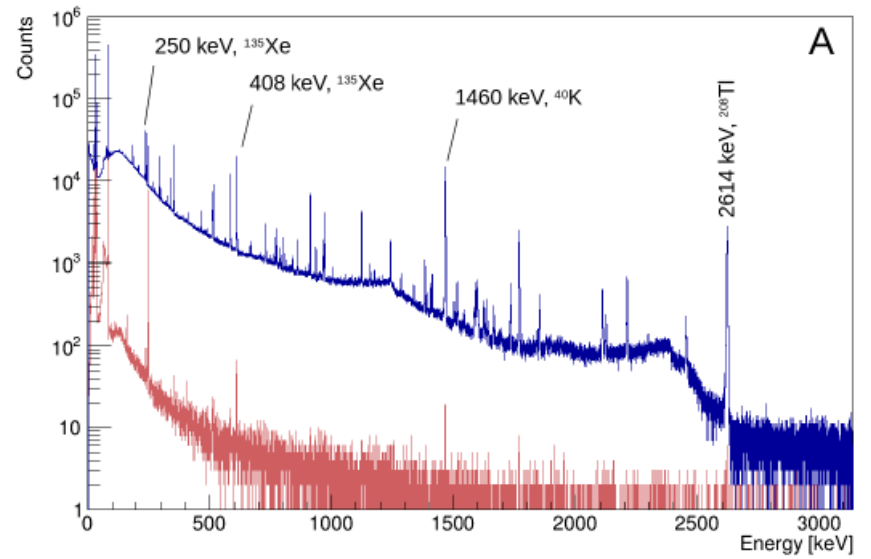
FIGURE 6.6: γ-spectra from the second extraction of gas (70 hours after irradiation end). Live time: 416,000 s. Blue: Singles γ-spectrum, red: β-gated γ-spectrum. Photon energy range 0-3 MeV (A) and 0-800 keV (B).

**A**
$^{135}$Xe $\beta^-$ +
250 keV γ-ray

$^{85m}$Kr $\beta^-$ +
151 keV γ-ray

$^{133}$Xe $\beta^-$ +
81 keV γ-ray

**B**
$^{135}$Xe $\beta^-$ +
250 keV γ-ray

$^{133}$Xe $\beta^-$ +
81 keV γ-ray

**C**
199 keV $e^-$ + 34
keV Xe X(K$_\beta$)

220 keV $e^-$ +
31 keV Cs X(K$_\alpha$)

199 keV $e^-$ +
30 keV Xe X(K$_\alpha$)

$^{133}$Xe & $^{135}$Xe $\beta^-$ +
31 keV Cs X(K$_\alpha$)

$^{133}$Xe & $^{135}$Xe $\beta^-$ +
21 keV Cs X(K$_\alpha$) [escape]

**D**
199 keV $e^-$ + 34
keV Xe X(K$_\beta$)

199 keV $e^-$ +
29.8 keV X (K$_\alpha$)

$^{133}$Xe & $^{135}$Xe $\beta^-$ +
21 keV Cs X(K$_\alpha$) [escape]