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Determination of precision nuclear decay data for the decay of ^{153}Gd

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National Physical Laboratory, UK

MEDICIS-Promed Final Conference, 30 April – 4 May 2019, Erice, Sicily

Need for nuclear decay data for medical applications

- MIRD formalism of Dose[1]

$$\bar{D} = 2.13\bar{c}\sum_i n_i \bar{E}_\mathrm{i} \phi_i$$

- Reliant on the absolute emission probabilities of the radiation and the half-life of the isotope.

- Change of these data changes the relevant dose calculations for patient and practitioner.

- Move into quantitative imaging requires high precision nuclear decay data in PET and SPECT.

- NPL combines unique expertise in radiochemical separation techniques (Peter Ivanov) & nuclear spectrometry for radionuclide standardisations & underpinning nuclear decay data measurements.

Nuclear decay data at NPL

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Applied Radiation and Isotopes

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

The half-life of ^{227}Th by direct and indirect measurements

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H I G H L I G H T S

- First direct measurement of ^{227}Th half-life by HPGe γ-ray spectrometry.
- Precision half-life measurement by ionisation chamber.
- New half-life of 18.695 (4) days determined.
- Critical evaluation of published half-lives and recommended value determined.

Applied Radiation and Isotopes 99 (2015) 46–53

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Applied Radiation and Isotopes

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

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

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H I G H L I G H T S

- 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.

Applied Radiation and Isotopes 102 (2015) 15–28

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Applied Radiation and Isotopes

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

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

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H I G H L I G H T S

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

Applied Radiation and Isotopes xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

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

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

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H I G H L I G H T S

- An 11% spread in activities determined from recommended nuclear data observed.
- A negative 3.4% bias observed in the weighted mean activity from recommended nuclear data.
- Absolute emission intensities of 70 γ rays from the α-decay of ^{227}Th determined.
- Normalisation scaling factor reduced by 3.3% from the recommended data.
- Precision improved by an order of magnitude or more over the recommended nuclear data.

EPJ Web of Conferences **146**, 08002 (2017) DOI: 10.1051/epjconf/201714608002

ND2016

Half-life measurement of the medical radioisotope ^{177}Lu produced from the ^{176}Yb(n,γ) reaction

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Gadolinium-153
Motivation for study

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- Gadolinium-153 is used as a line (calibration) source for SPECT.[2]

- Historically was used in bone absorptiometry, bone mineral density and marrow content studies.[3]

- Identified as a potential radionuclide to use for interstitial rotating shield brachytherapy (I-RBST) to replace ^{192}Ir. [4]

- Inconsistencies within the dataset identified as well as SIRIC disagreements.

Further motivation for study

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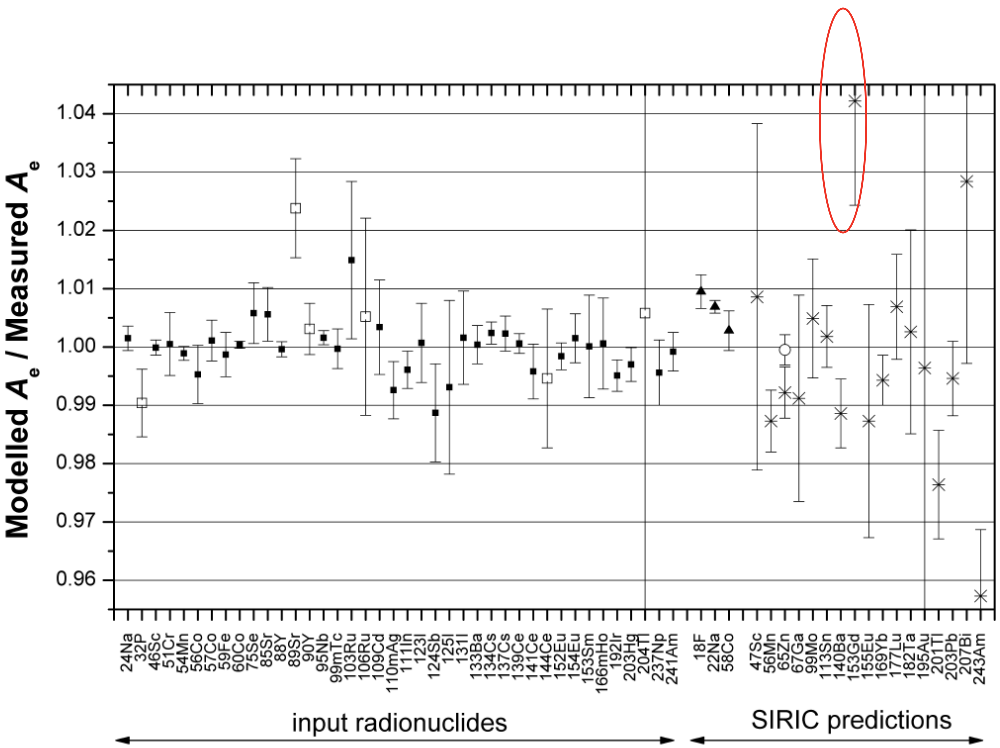
- BIPM-Monographie 7, "*Measurement modelling of the International Reference System (SIR) for gamma emitting radionuclides*" found a **4.3 %** discrepancy between modelled and experimental data in the case ^{153}Gd for the measured activity and the SIRIC predicted value according to a well understood least squares fitting algorithm to the efficiency of the counter. Similar nuclides with similar decay properties were not in disagreement.



- It was stated, "***The discrepancy for the ^{153}Gd is probably due to nuclear data… The efficiency curve therefore provides motivation for to investigate the decay data of that radionuclide.***"

Taken from [7]

The electron capture of ^{153}Gd

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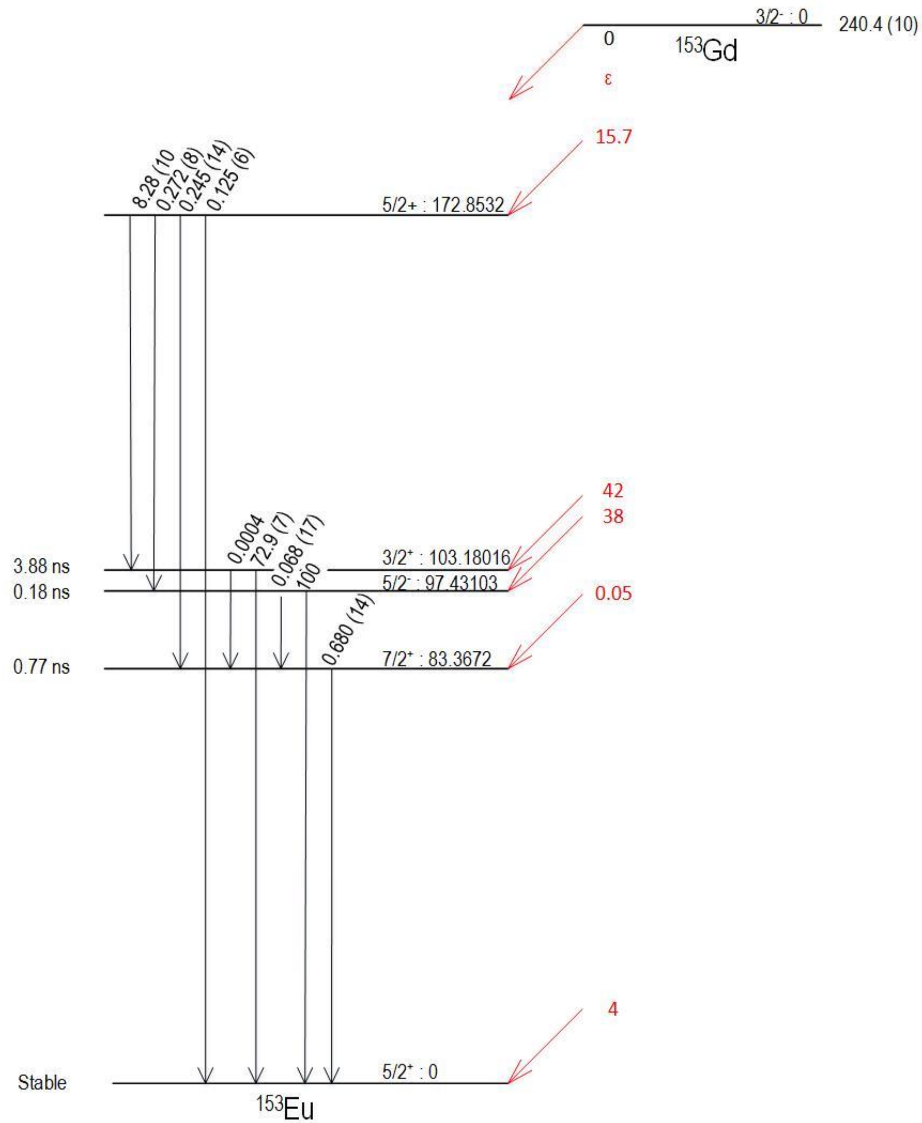
^{153}Gd decays 100% to the A=153 stable isobar ^{153}Eu.

The DDEP [5] evaluation disagrees with recent evaluation by [6], which includes (7/2+) 269.7 keV state and the 7/2- 151.6 keV state.

If these states were populated 54.1, 68.9, 96.8, 118.1 and 166.5 keV emissions would be seen.



Data taken from [5]

Experimental technique

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- Three flame sealed aliquot sources of chemically pure ^{153}Gd were prepared by dispensing ~1g of ^{153}Gd from a 0.5M HCl solution.
- Two HPGe detectors (BART and LOKI) measured the three sources over a period of 5 years, for acquisition times between 86400 s and 250000 s.
- CANBERRA AFT Research amplifier 2025, Analogue-to-Digital Convertor 8715, AIM connected to a computer running CANBERRA GENIE 2000 v2.1c software was used to acquire spectra.
- Different techniques for pile up and dead time corrections implemented for each detector.

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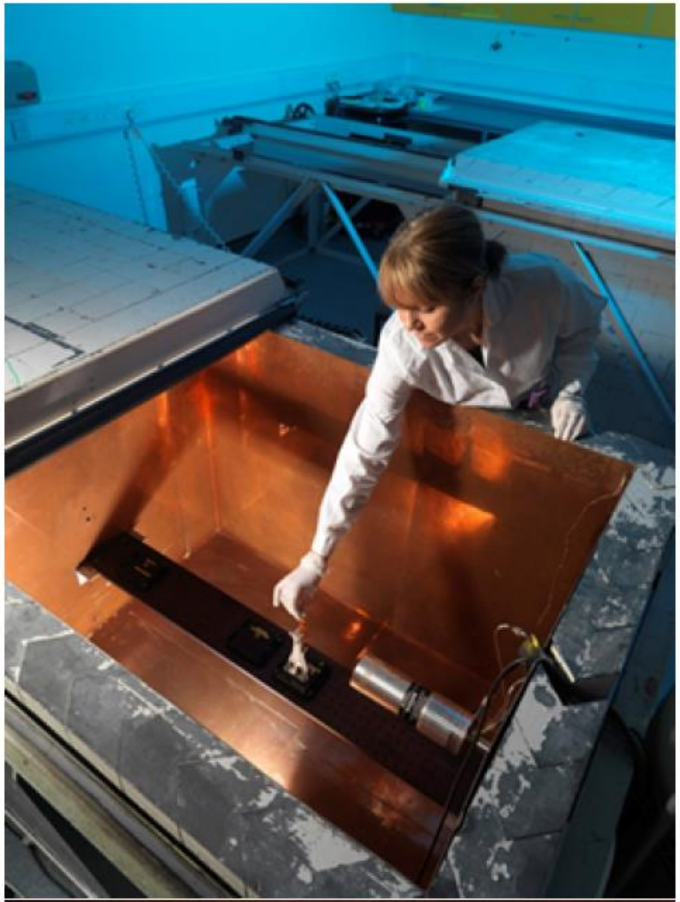
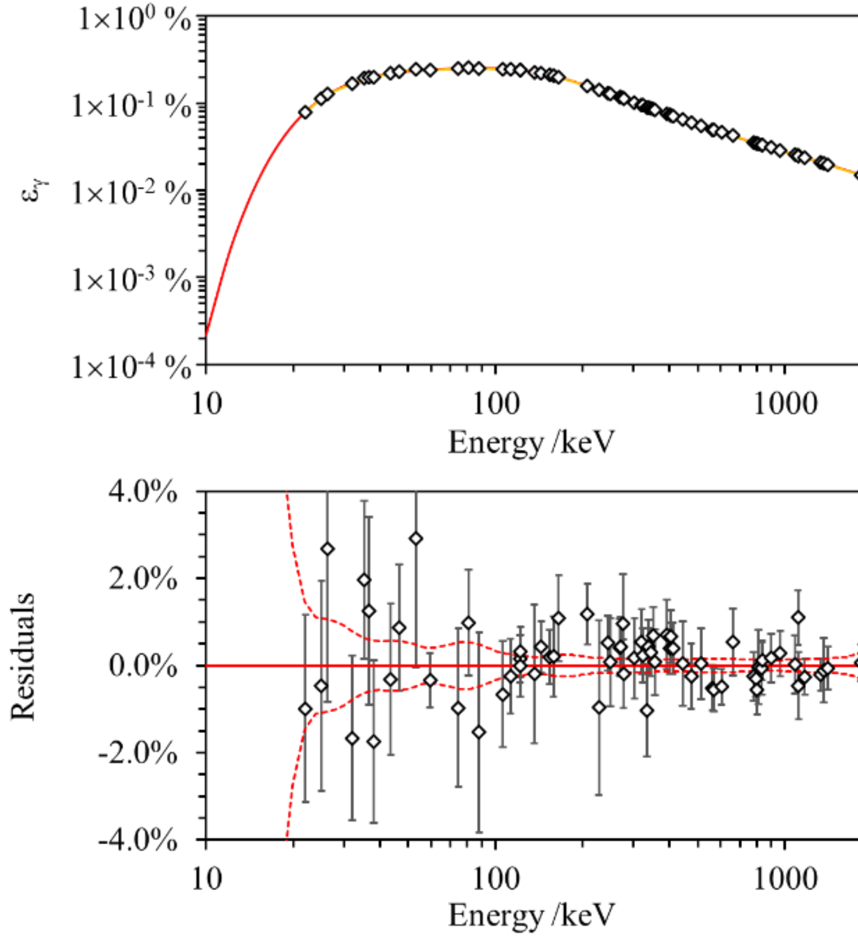
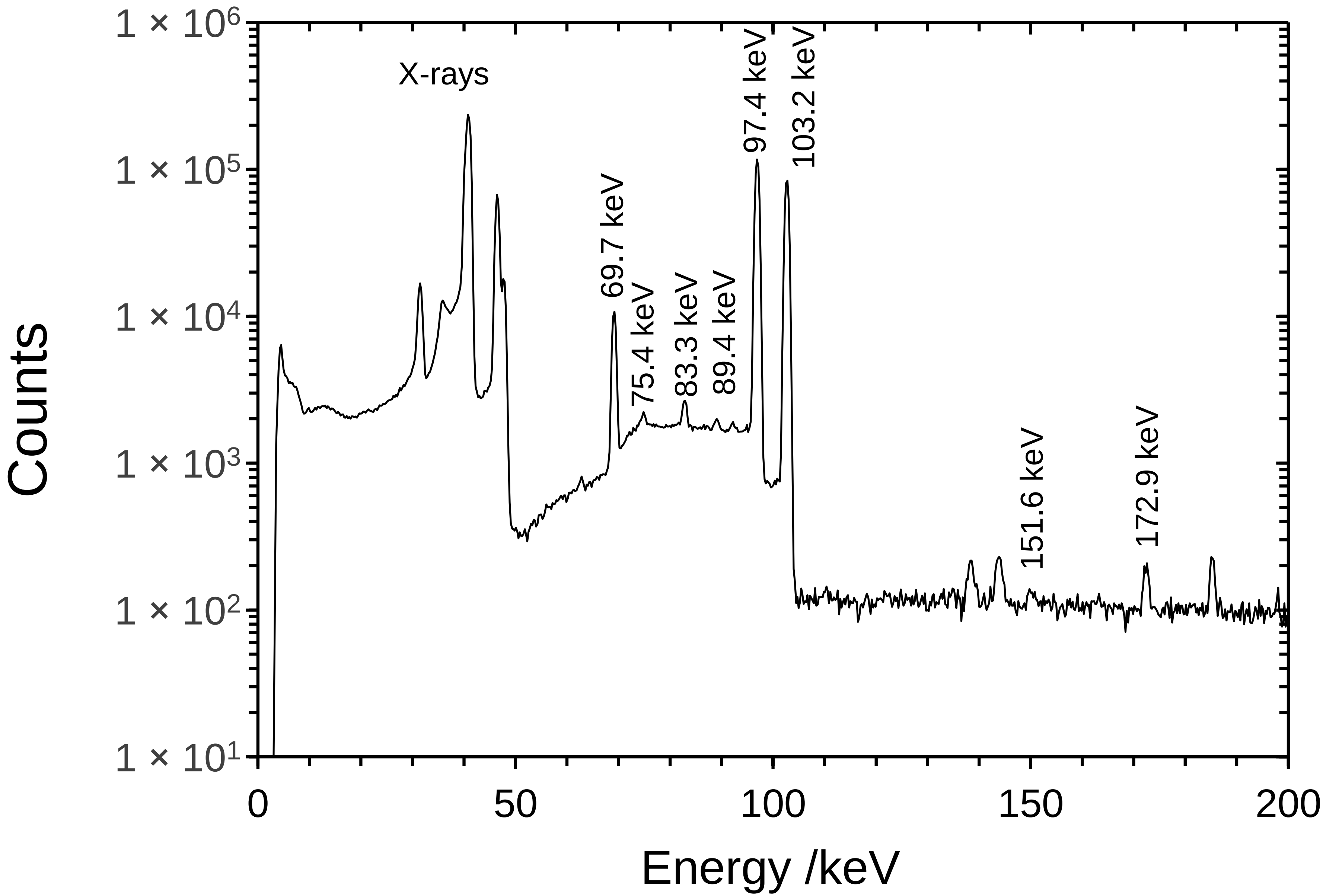




Experimental results – LK16071901



Experimental results

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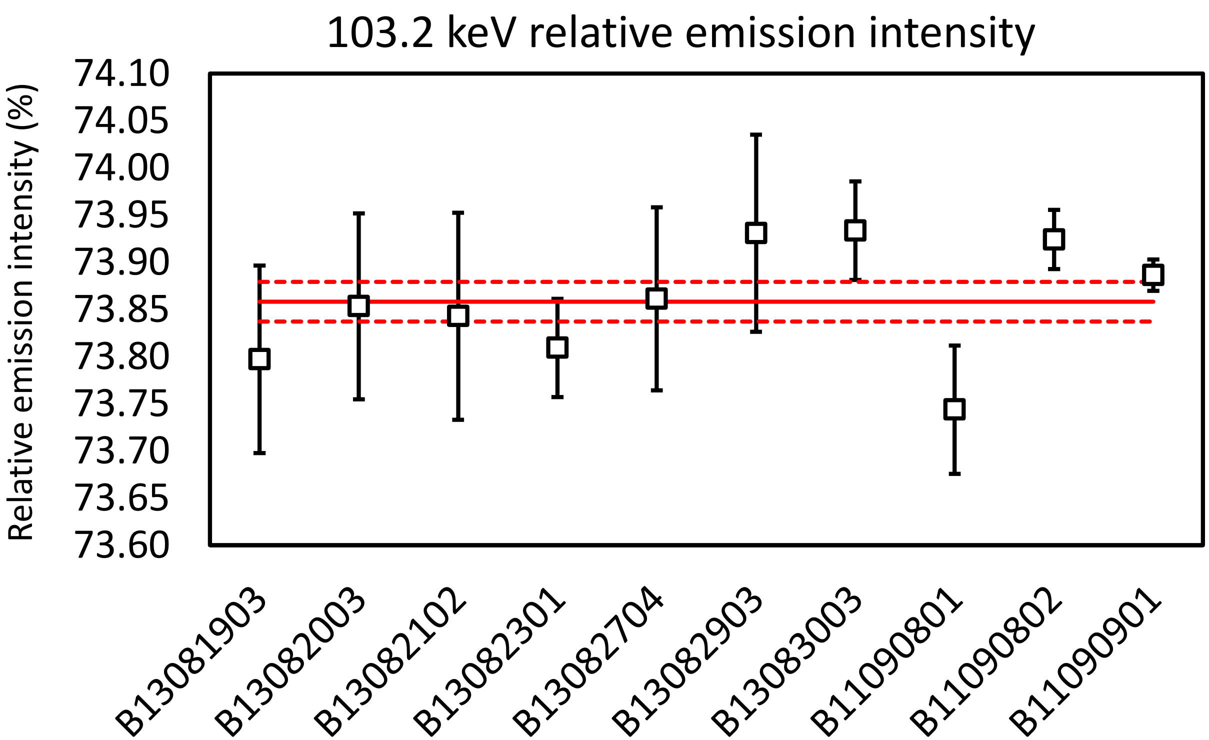

Energy /keV	Mean	χ^2
69.6	7.8455	0.09
89.46	0.3249	0.61
97.43	100.0	
103.18	73.98	0.11
151.62	0.0157	0.27
172.85	0.1312	0.12

Relative intensities – comparison

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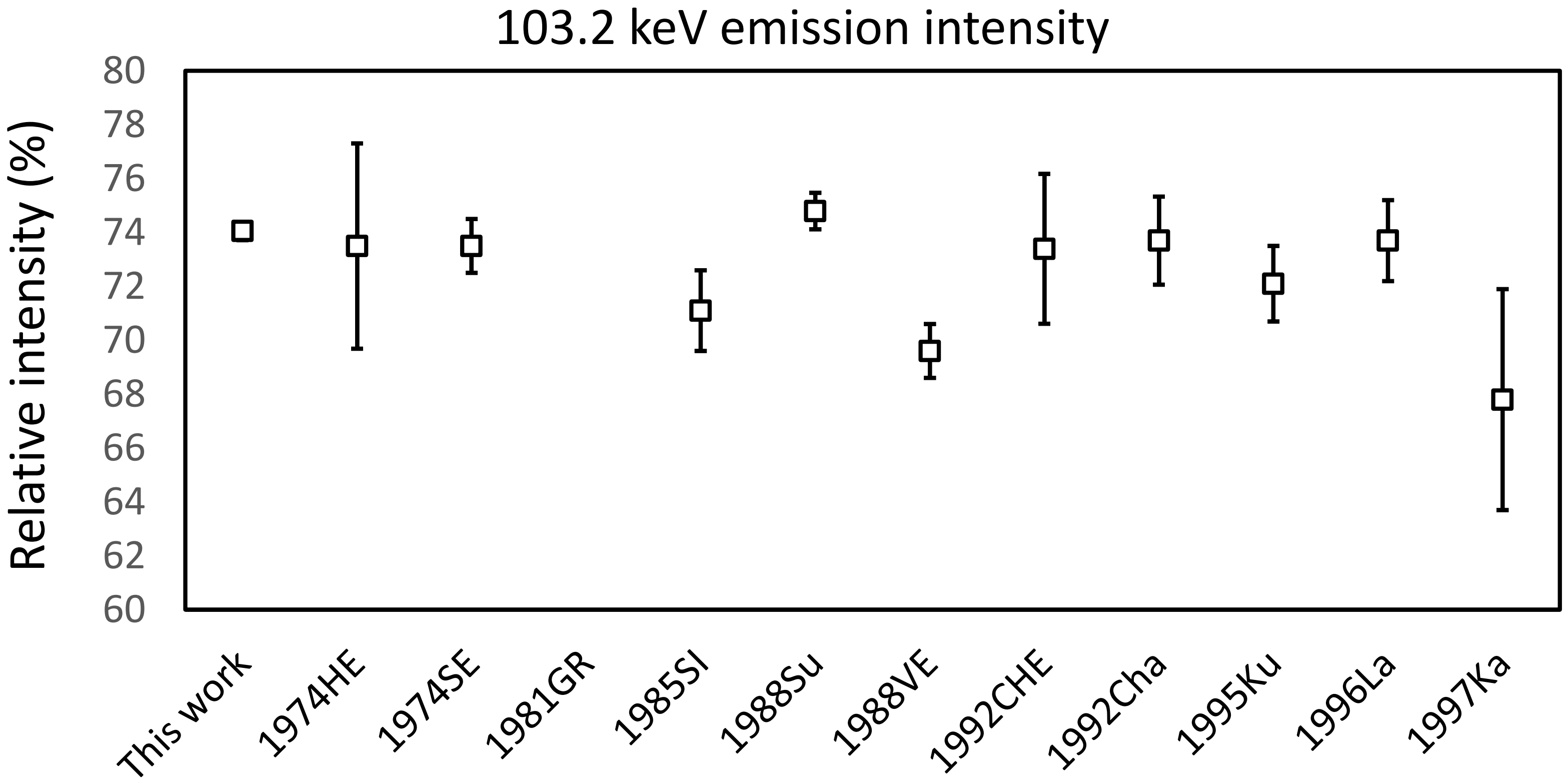
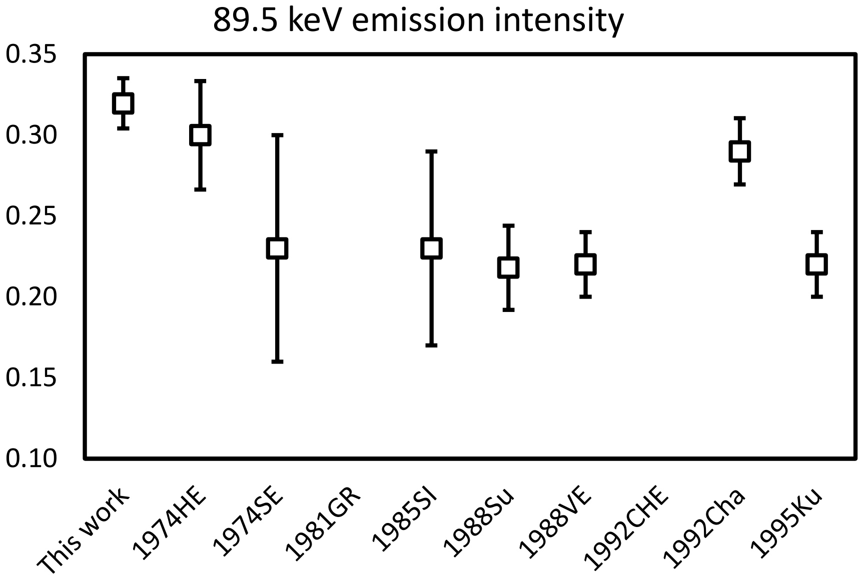
Results from the weighted mean of the two detectors intensity measurements show that there are large ($\sigma > 2$) differences from some of the previous work, however we consistently agree with work done by Chand et al.

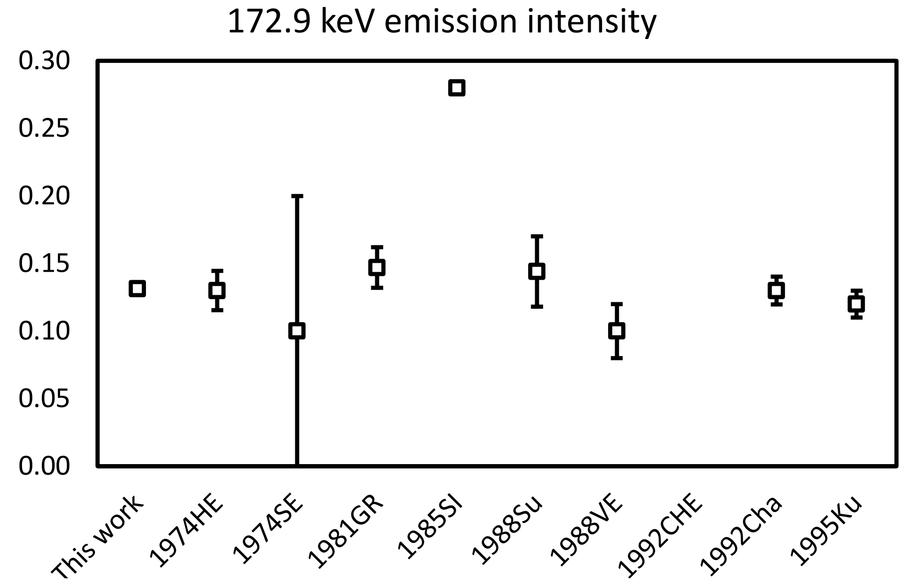


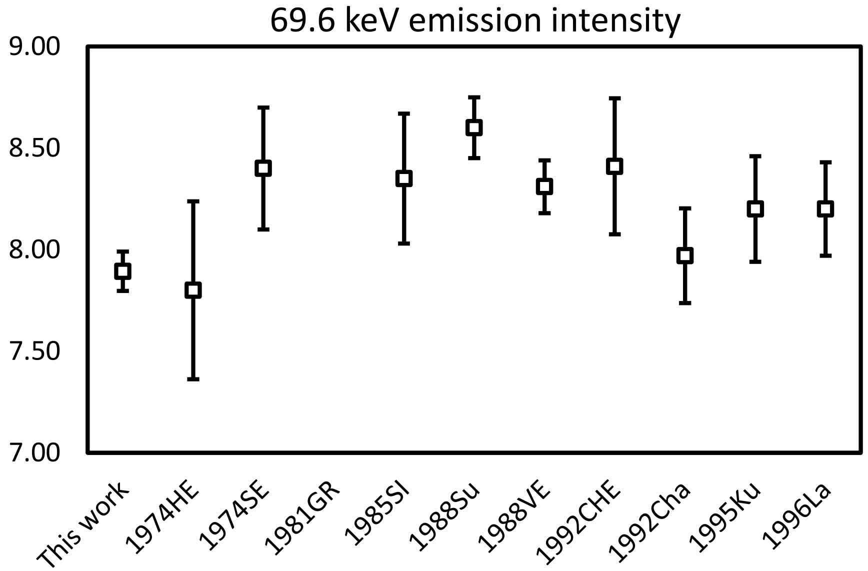
$u(I_{88SU})\% = 0.9\ \%$

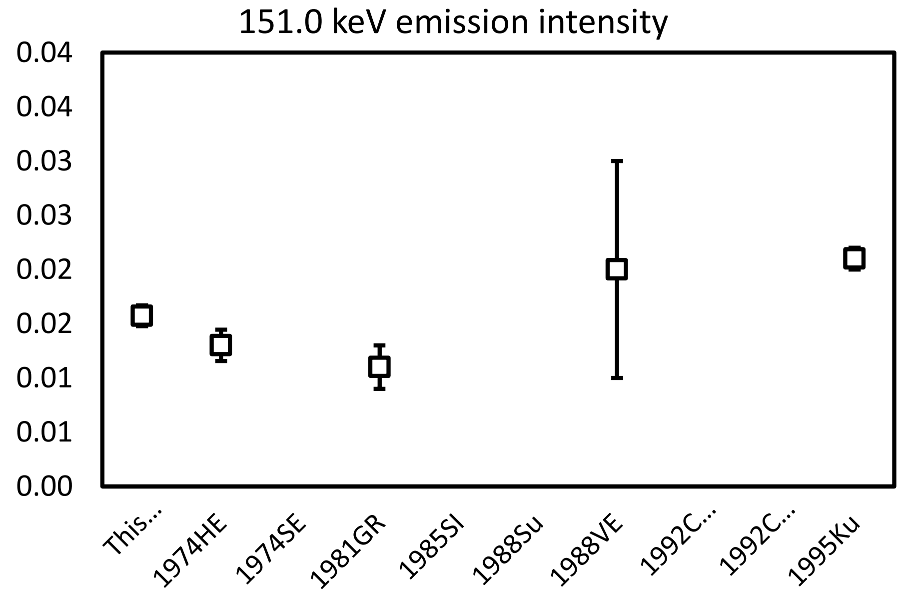
$u(I_{NPl})\ \% = 0.5\ \%$

Relative intensities – comparison









Relative intensity comparison

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Energy	This work	DDEP [5]	Relative difference	Z-score
/keV	$I_\gamma(E)/I_\gamma(97.4)$	$I_\gamma(E)/I_\gamma(97.4)$	(%)	
69.6	7.89 (9)	8.28 (10)	4.71	2.90
89.4	0.320 (16)	0.245 (14)	30.6	-3.52
97.4	100	100	-	-
103.1	74.06 (27)	72.9 (7)	-1.59	-1.55
151.6	0.0157 (10)	0.017 (4)	7.65	0.31
172.8	0.1313 (18)	0.125 (6)	-5.04	-1.00

Uncertainty component	Value /%
FEP detection efficiency	0.38
Absolute activity	0.49
Peak fitting	0.15
Self-absorption	0.013
True coincidence summing	0.10
Gravimetric	0.02
Geometric reproducibility	0.016
Dead-time and pulse pile-up	0.18
Combined uncertainty	**0.67**

Idealised example of the 4πβ–γ coincidence method

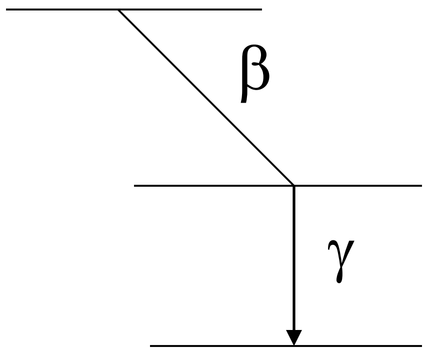
β–γ decay (100% fed single cascade)

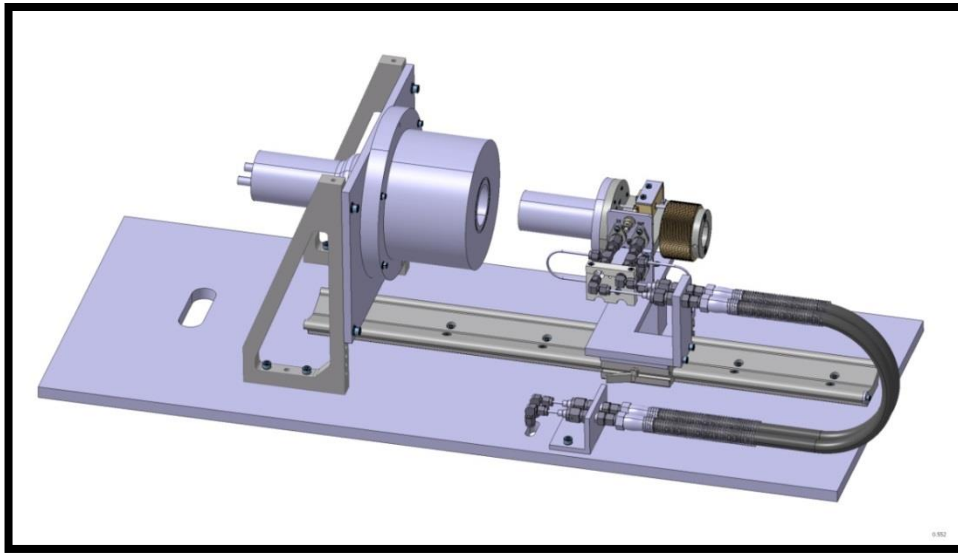
Count Rates

$N_\beta = N_o \varepsilon_\beta$

$N_\gamma = N_o \varepsilon_\gamma$

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





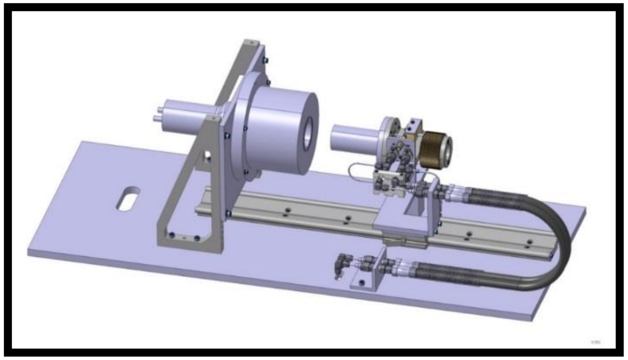
Efficiencies: $\varepsilon_\beta = N_c / N_\gamma$

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

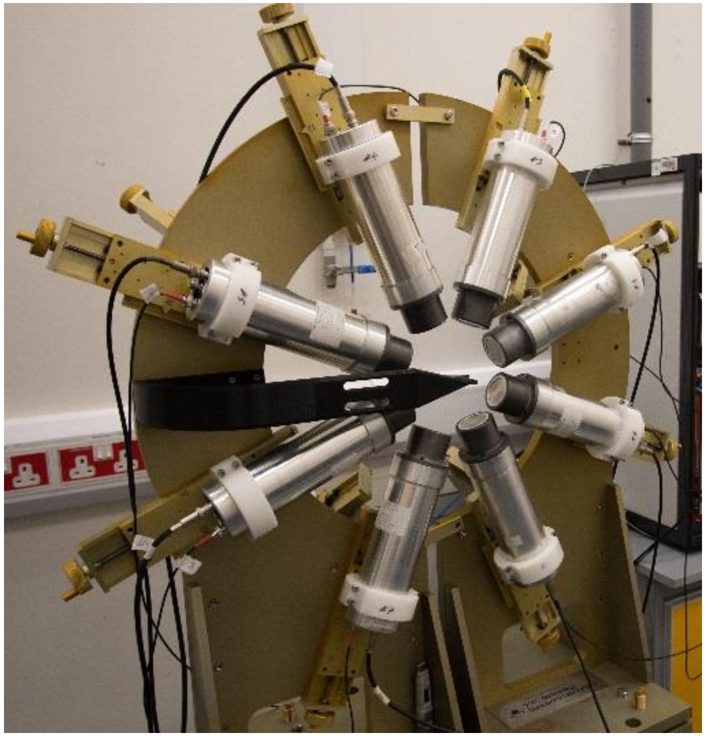
$$Activity : N_o = \frac{N_\beta N_\gamma}{N_c}$$

Several ways to derive absolute activity at NPL

4πβ-γ

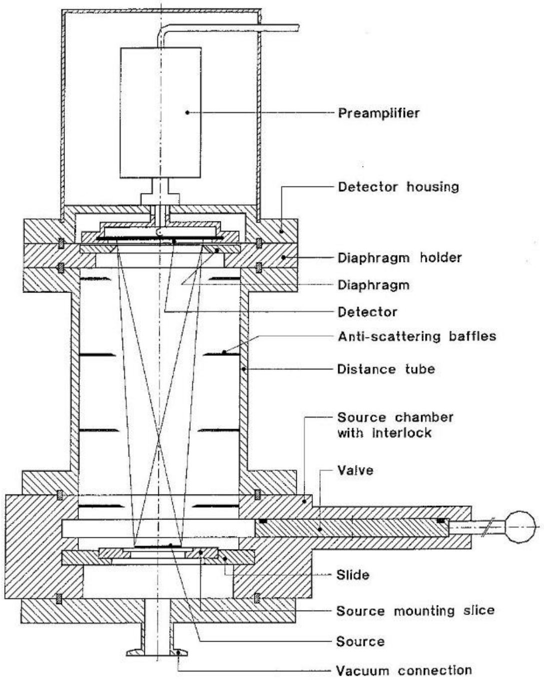


Different needs met by each!



NANA γ-γ counting

TDCR



DSA

Absolute intensity

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4π(LS)-γ DCC calculation of the source reported specific activity of **512.5 (25)** kBq g^{-1}@ 19/7/2011 12:00 UTC

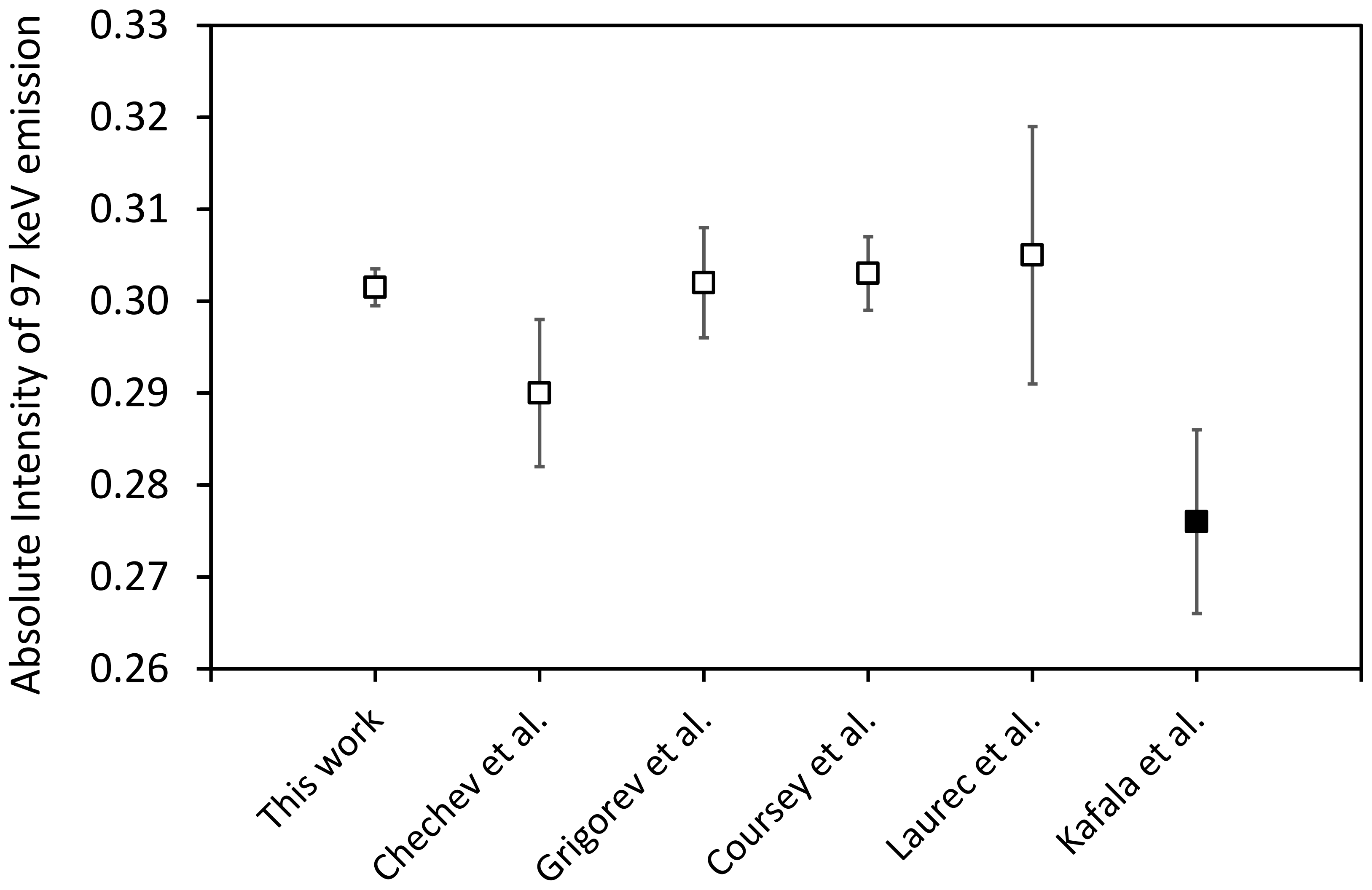
Using this value it is possible to calculate the absolute activity value for the 97.4 keV emission, according to the adopted value of the half life of **240.4 (10) days.**

By taking into account the well known efficiency and the density corrections required for the liquid source and the uncertainty in the mass dispensed, the rate across measurements were calculated and the (decay corrected) DCC value for specific activity used to find the absolute intensity.

The absolute intensity for the 97.4 keV emission has been calculated to **0.3015 (20)**

Energy	Current work (NPL)
/keV	Absolute I_γ (%)
69.6	2.38 (3)
89.4	0.096 (5)
97.4	30.15 (20)
103.1	22.33 (17)
151.6	0.00049 (6)
172.8	0.0396 (6)

Absolute intensity comparison

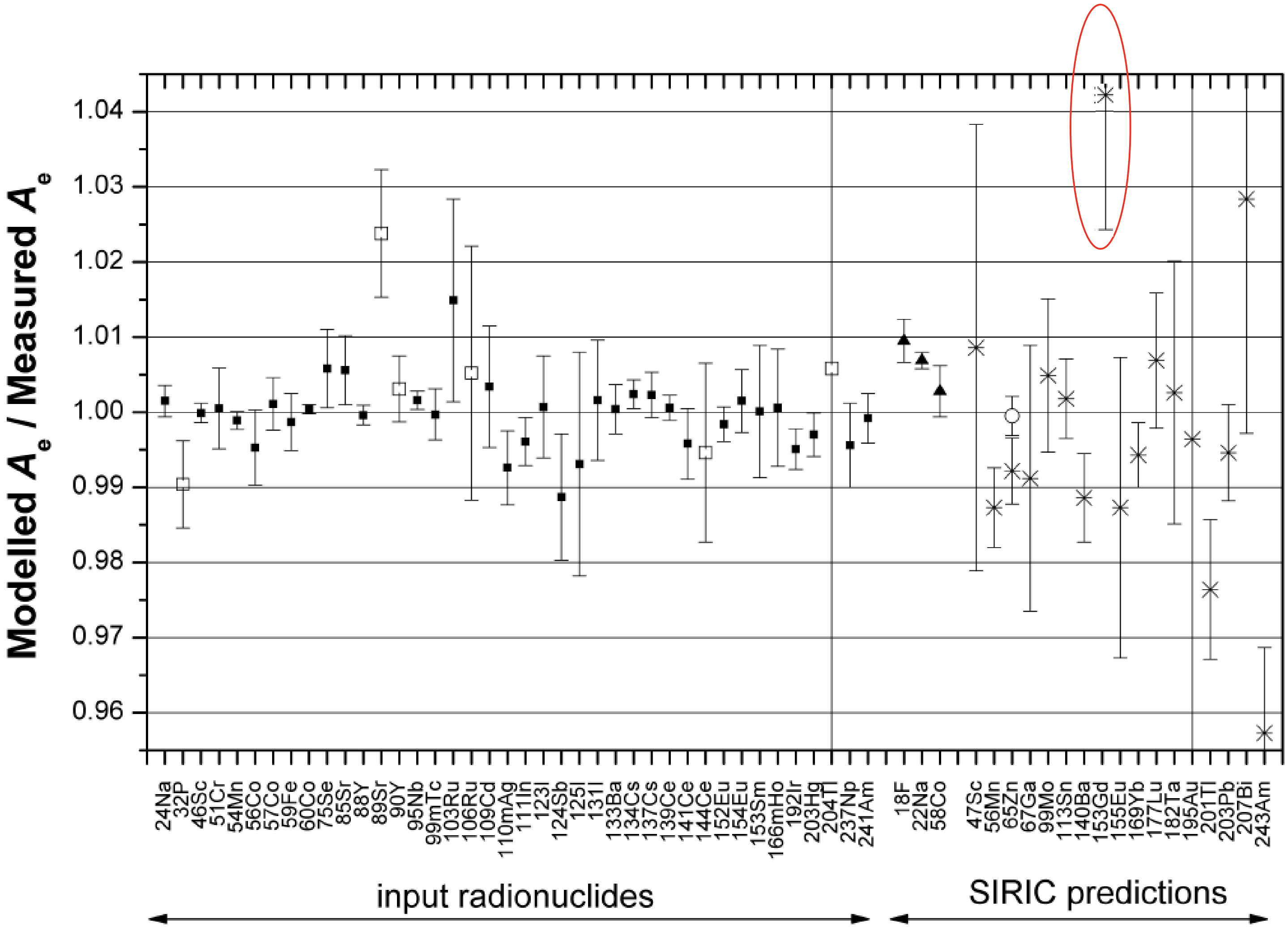


SIRIC comparison

- The adopted evaluated value for the absolute intensity of the 97.4 keV emission is 0.29 (8). The SIRIC discrepancy is **4.3 %**
- The absolute intensity of the 97.4 keV emission from this work (0.3015 (20)) is **4 %** higher than the evaluated value.
- The data in this work has been used to compute a new modelled activity.

Modelled: 368 (19) kBq
KCRV: 368 (17) kBq
Delta: 0.18 %

SIRIC comparison



Comparison with Tb

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Nuclide	NSF	NSF u (%)	Half-life	Half-life u (%)	Notes
Gd-153	0.3015(20)	0.66	240.4 d (10)	0.41	
Tb-149	0.0088 (3)	3.4	4.118 h (25)	0.6	No measurement set after 1970
Tb-152	0.635 (6)	0.95	17.5 h (1)	0.57	Only two measurements
Tb-155	0.0251(13)	5.2	5.32 d (6)	1.1	One measurement in 1976 in evaluation primarily
Tb-161	0.102 (5)	4.9	6.89 d (2)	0.29	

Conclusions

- Measurements of the observed emissions have been determined with greater precision than previous studies.
- No firm evidence that the 269 keV level in ^{153}Eu is populated during this decay (no 96.8, 118.1 or 166.5 keV gamma ray identified)
- Absolute intensity disagrees with current evaluated absolute emission probability of the 97.4 keV γ ray.
- Absolute intensity we have reported agrees with a number of previous measurements and explains SIRIC anomaly.
- Previously well-studied ^{153}Gd nuclear decay data has been enhanced.

Thank you

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