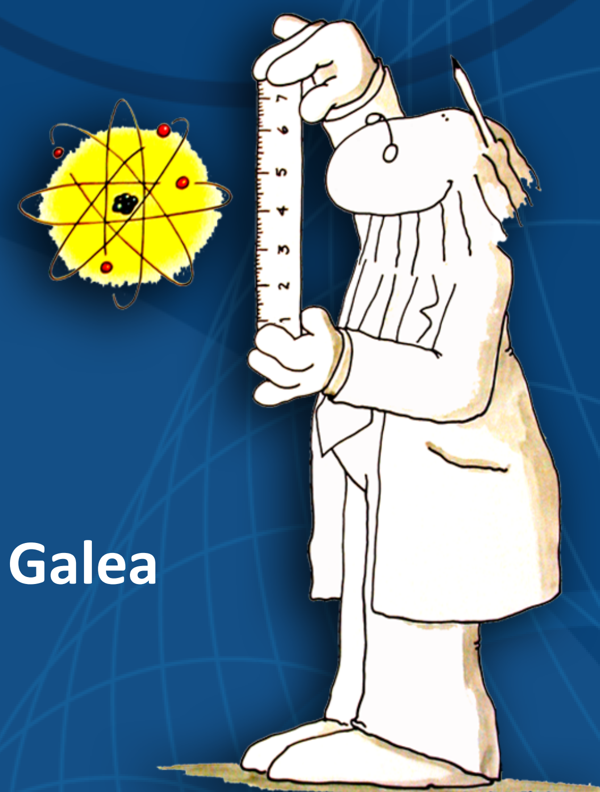
NRC·CNRC

Radionuclide decay scheme modelling in EGSnrc



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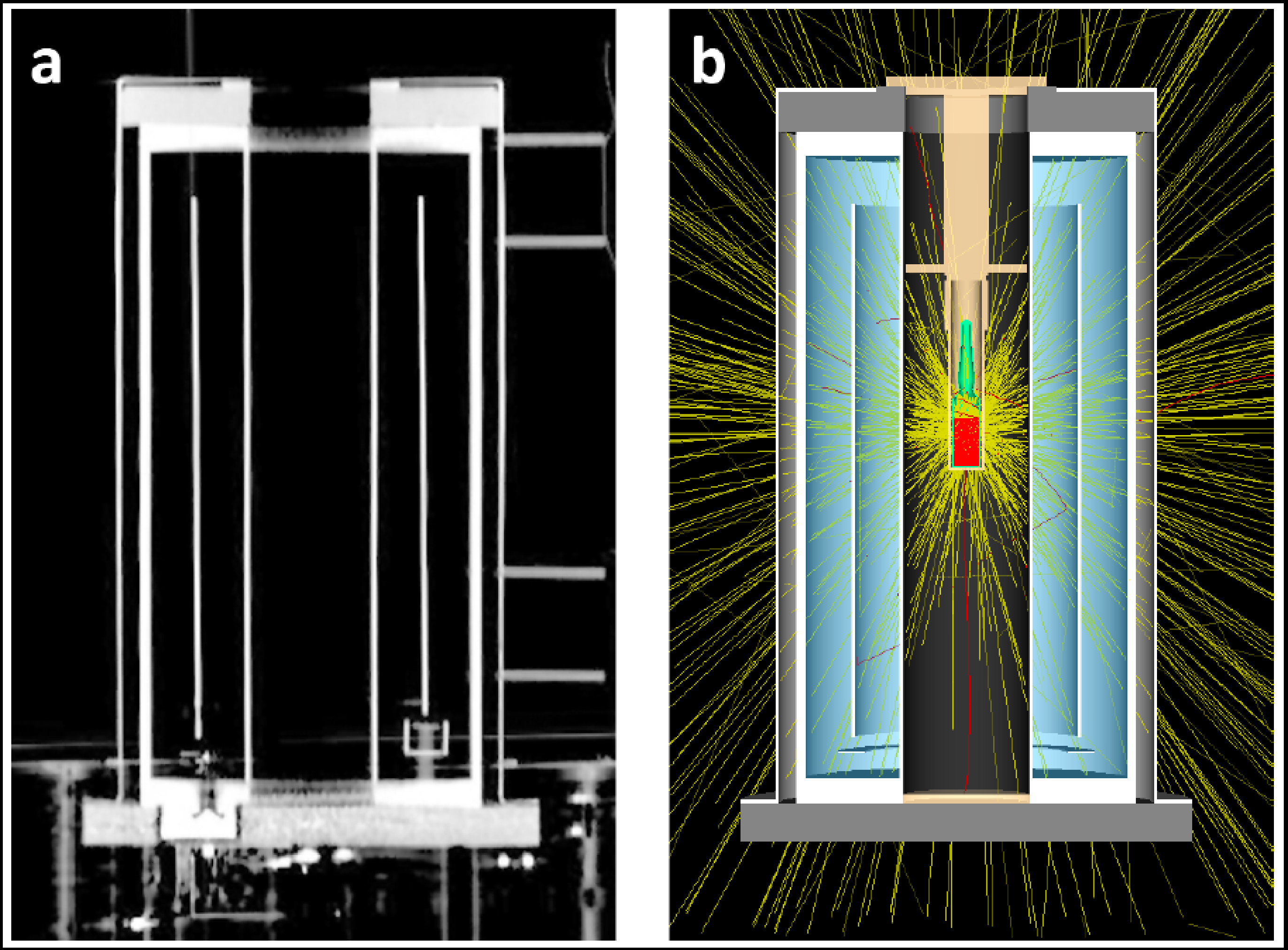
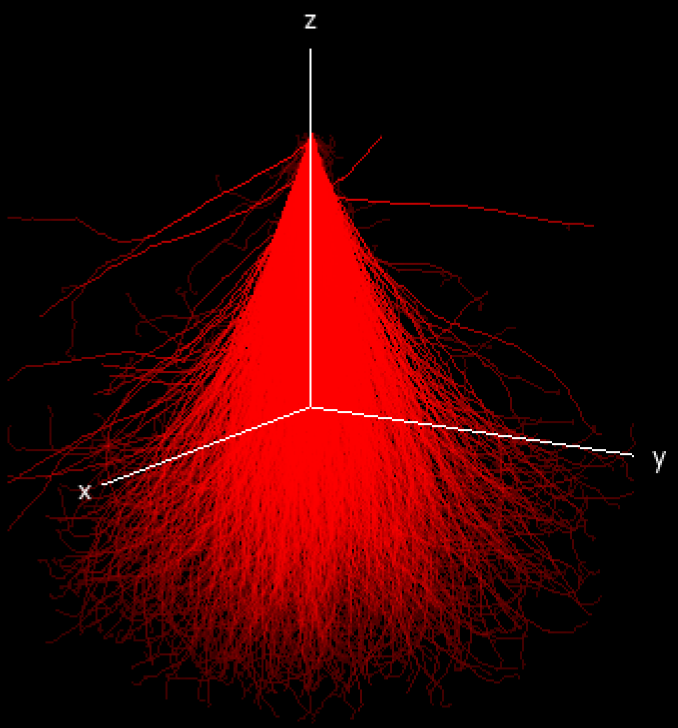
Measurement Science and Standards
National Research Council Canada

Simulation = geometry + source (+...)



An accurate particle source is key



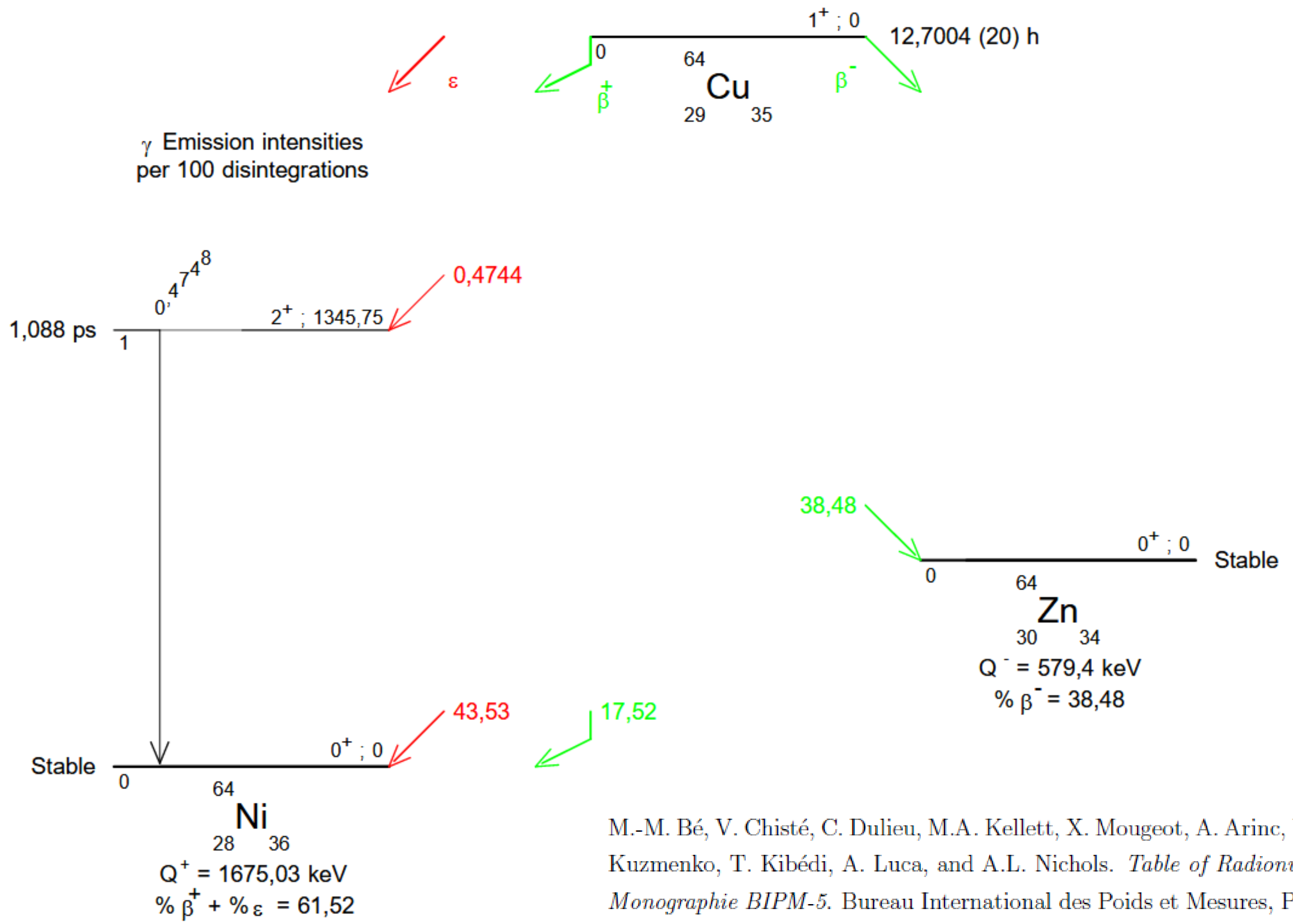
Location

Direction

Energy

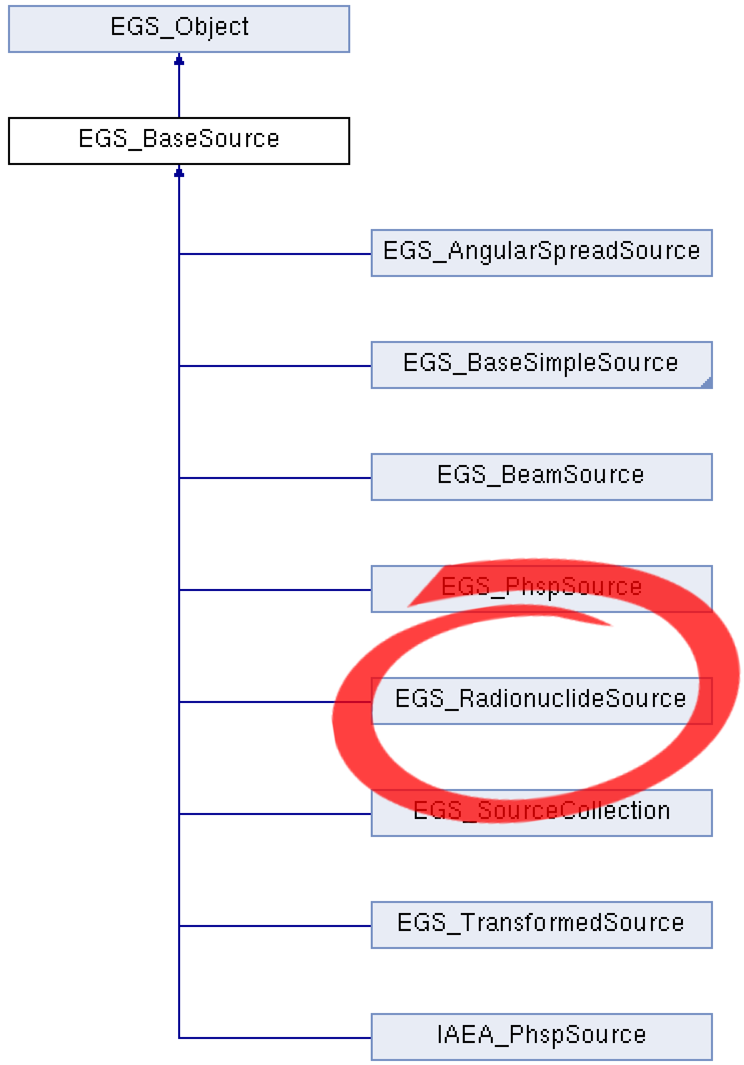
... and more?

Radionuclide decays are complex to model



M.-M. Bé, V. Chisté, C. Dulieu, M.A. Kellett, X. Mougeot, A. Arinc, V.P. Chechev, N.K. Kuzmenko, T. Kibédi, A. Luca, and A.L. Nichols. *Table of Radionuclides*, volume 8 of *Monographie BIPM-5*. Bureau International des Poids et Mesures, Pavillon de Breteuil, F-92310 Sèvres, France, 2016.

Introducing: EGS_RadionuclideSource



Radionuclide data from LNHB

- Data from Laboratoire National Henri Becquerel (LNHB)
  - http://www.nucleide.org/DDEP_WG/DDEPdata.htm

Tables of evaluated data and comments on evaluation
Pages updated by the Laboratoire National Henri Becquerel
All questions about the data must be sent to the authors. See chapter Addresses.

updated: 3rd **March 2017**
newly added: **Pr-142**
recently updated: **Co-57, Xe-133m**
ASCII files updated on: **24/06/2016**
(**221** nuclides in table, sorted by **alphabetical order** / atomic number / mass number / edition date)

(**History of older evaluations**, sorted by **alphabetical order**)

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Vol.	Publication
99	CEA Report - Table de Radionucléides
1	Monographie BIPM-5 - Table of Radionuclides, vol. 1
2	Monographie BIPM-5 - Table of Radionuclides, vol. 2
3	Monographie BIPM-5 - Table of Radionuclides, vol. 3
4	Monographie BIPM-5 - Table of Radionuclides, vol. 4
5	Monographie BIPM-5 - Table of Radionuclides, vol. 5
6	Monographie BIPM-5 - Table of Radionuclides, vol. 6
7	Monographie BIPM-5 - Table of Radionuclides, vol. 7
8	Monographie BIPM-5 - Table of Radionuclides, vol. 8

(*Type of updates: **N** - new evaluation; **1** - update in comments only; **2** - minor update in table; **3** - major update in table)

Nuclide		Tables	Comments	ASCII files			Vol.	UpDate	Type*
				ENSDF	PenNuc	Lara			
Ac-225	^{225}Ac	table	comments	ensdf	pennuc	txt	5	26/08/2009	3
Ac-227	^{227}Ac	table	comments	ensdf	pennuc	txt	4	16/02/2009	2
Ac-228	^{228}Ac	table	comments	ensdf	pennuc	txt	6	22/01/2010	3
Ag-108	^{108}Ag	table	comments	ensdf	pennuc	txt	3	4/09/2006	2

O-15
P-32
P-33

NRC·CNRC

The ENSDF format is widely used

- Evaluated Nuclear Structure Data File (ENSDF)

```
67ZN    67GA EC DECAY (3.2613 D)
...
 67ZN T  Auger electrons and X ray energies and emission intensities:
 67ZN T       {U Energy (keV)}   {U Intensity}  {U Line}
 67ZN T
 67ZN T        8.61587            17.0      6      XKA2
 67ZN T        8.63896            33.0     12      XKA1
...
 67ZN T
 67ZN T        7.21-7.55      |]                   KLL AUGER
 67ZN T        8.31-8.63      |]  60.4     21      KLX AUGER
 67ZN T        9.39-9.65      |]                   KXY AUGER
 67ZN T        0.732-0.997        167.5    21      L AUGER
 67GA  P 0.0          3/2-             3.2613 D  5             1000.8    12
 67ZN  N 1.0          1.0       1        1.0
 67ZN  L 0            5/2-              STABLE
 67ZN  E                        3.3    326.532
 67ZN2 E CK=0.8836   15$CL=0.0989   12$CM=0.0164    4$CN=0.0011    1
 67ZN  L 93.31        1/2-             9.00 US   4
 67ZN  E                        50.5    175.261
 67ZN2 E CK=0.8834   15$CL=0.0991   12$CM=0.0164    4$CN=0.0011    1
 67ZN  G 93.307     1238.1      7E2                        0.854  12
 67ZN2 G KC=0.748    11$LC=0.0922   13$MC=0.01300  19$NC=0.000388  6
 67ZN  L 184.58       3/2-             1.028 NS  14
 67ZN  E                        22.3    275.523
 67ZN2 E CK=0.8832   15$CL=0.0993   12$CM=0.0164    4$CN=0.0011    1
 67ZN  G 91.263     153.09      7M1+E2     0.123   25      0.091   6
```

NRC·CNRC

Radionuclide production branches

- Disintegration modes
  - β^- decay
  - β^+ decay
  - Electron capture decay
  - α decay → Decay is modelled but α's are discarded
- Gamma transitions
  - ϒ photon emission
  - Conversion electron emission

Atomic relaxation cascades

- Electron rearrangement
  - fluorescent photons, Auger electrons, Coster-Kronig electrons

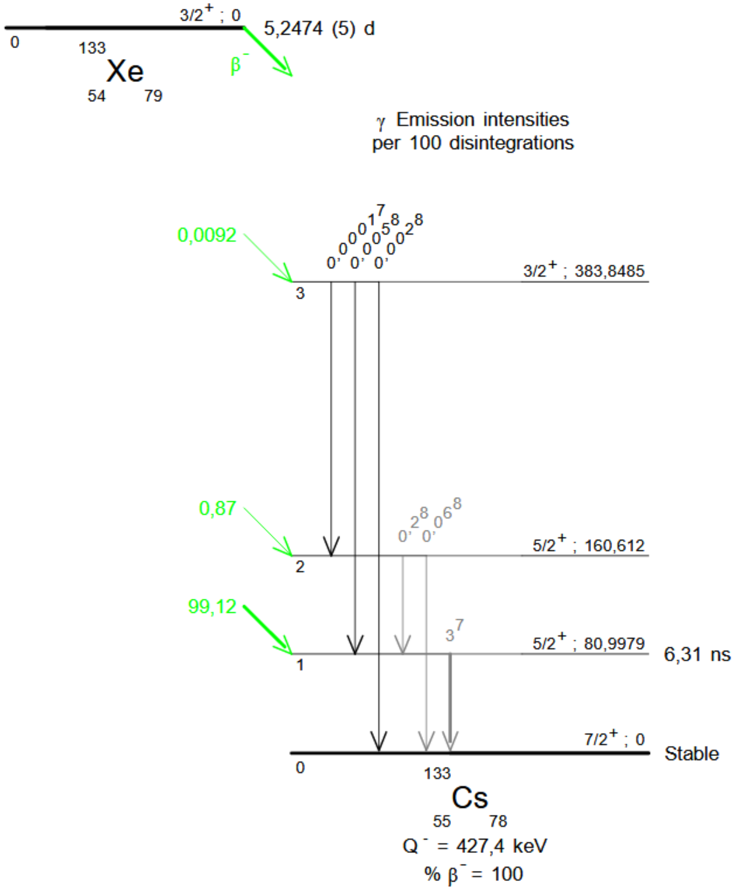
→ Option 1: Statistical model using ENSDF data

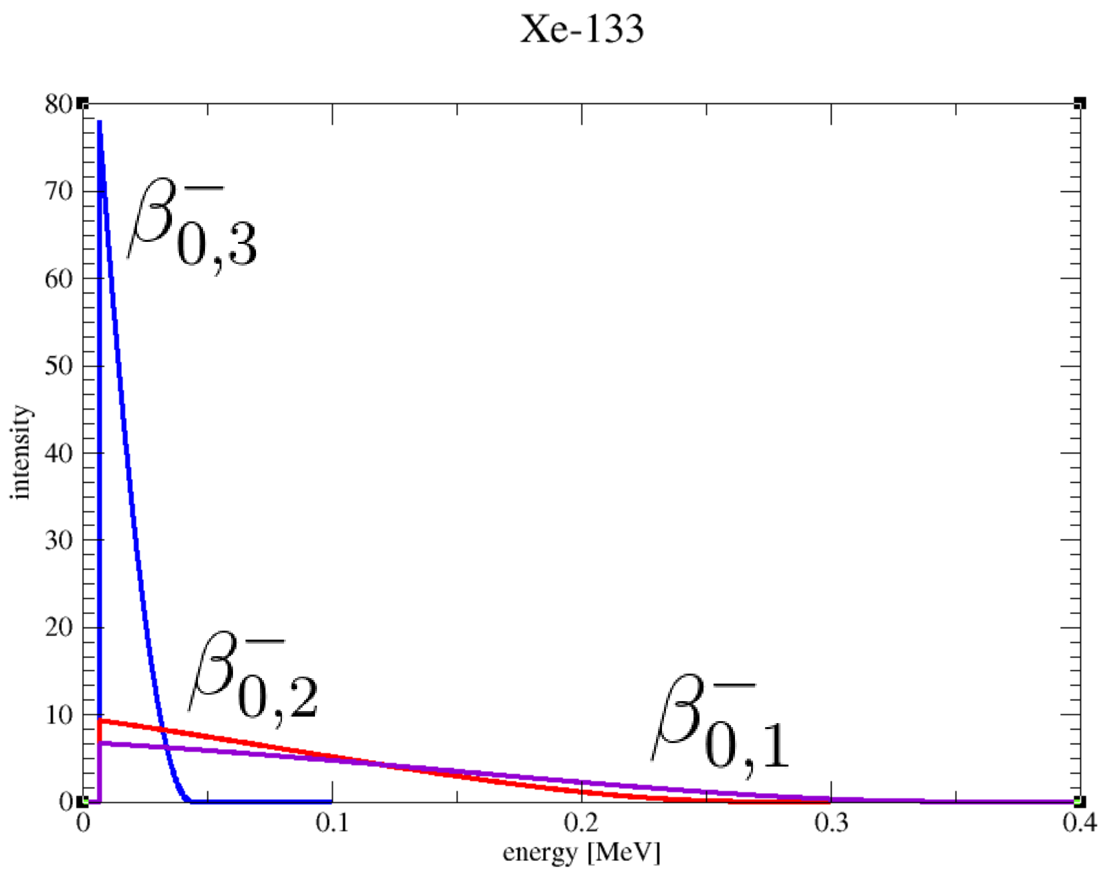
→ Option 2: Sample initial vacancy (correlated with transition)

Simulate entire relaxation cascade

Uses EGSnrc relaxations (EADL database)

Beta energies sampled from Fermi distribution





Coincidence count "realistically"

- All particles are assigned a time of source emission
- No time of flight modelling
- Currently no gamma-gamma directional correlations

```
source->getTime()
```

$$t_{\text{disintegration}} = t_{\text{disintegration-1}} - \ln(1-u)/A$$

$$t_{\text{IT}} = t_{\text{disintegration}} - \frac{t_{\frac{1}{2},\text{IT}} \cdot \ln(1-u)}{\ln(2)}$$

Coincidence count “exactly”

- All emissions & secondaries resulting from the same disintegration return the same “shower index”

```
source->getShowerIndex()
```

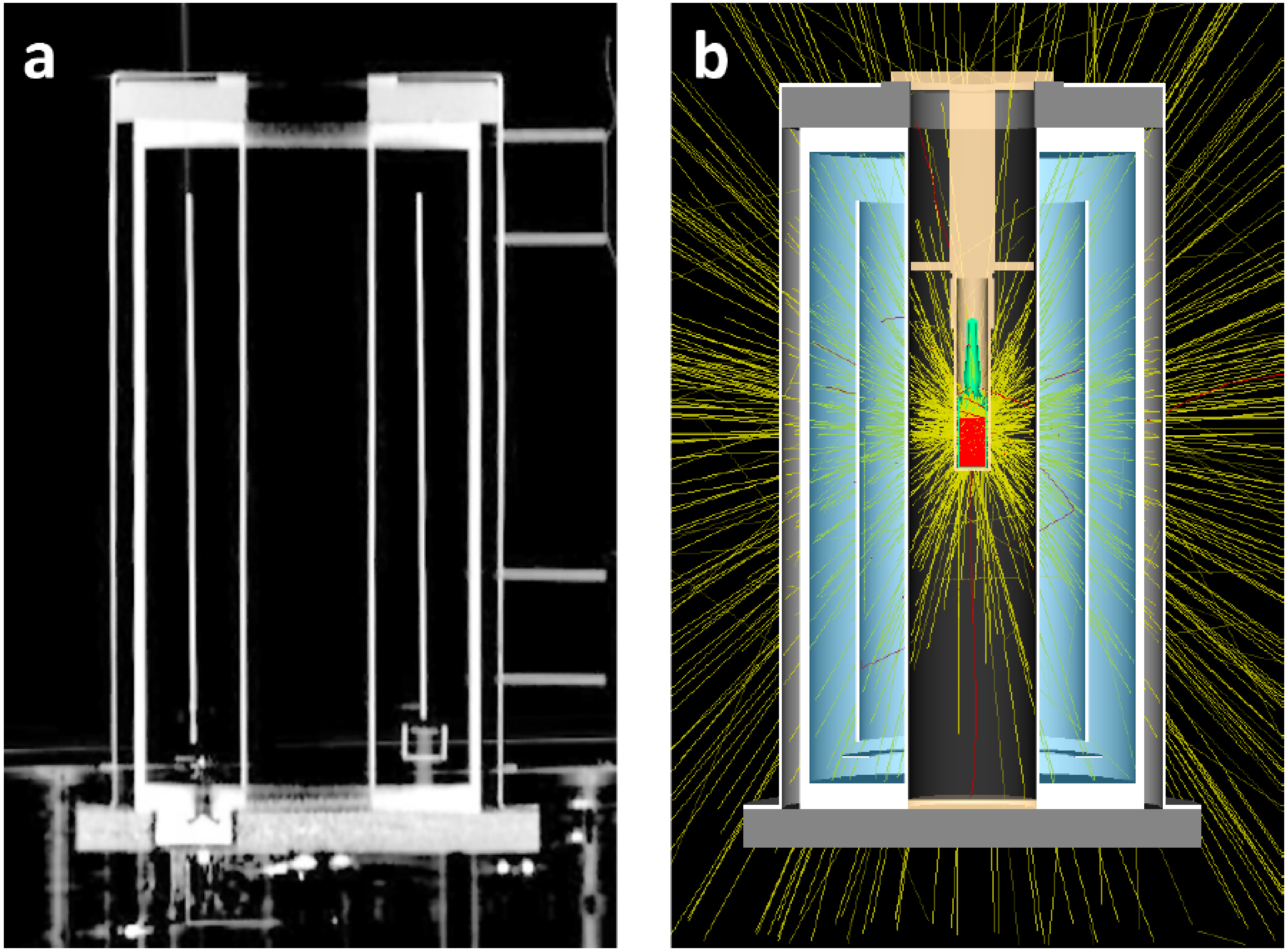
The input file is easy

```
:start source:

    name                    = my_mixture

    library                 = egs_radionuclide_source
    activity                = total activity of mixture, assumed constant

    ... optional arguments ...

    :start shape:
        definition of the source shape
    :stop shape:

    :start spectrum:
        Next slide...
    :stop spectrum:

:stop source:
```

The input file is easy

```
:start source:
    ... (previous) ...

    :start spectrum:

        type              = radionuclide
        nuclide           = name of the nuclide (e.g. Sr-90)

        relative activity = [optional] the relative activity (sampling
                            probability) for this nuclide in a mixture

    :stop spectrum:

    :start spectrum:
        type                  = radionuclide
        nuclide               = next nuclide (e.g. Y-90)
        relative activity     = ...
    :stop spectrum:

:stop source:
```

Calibration coefficients for the Vinten chamber



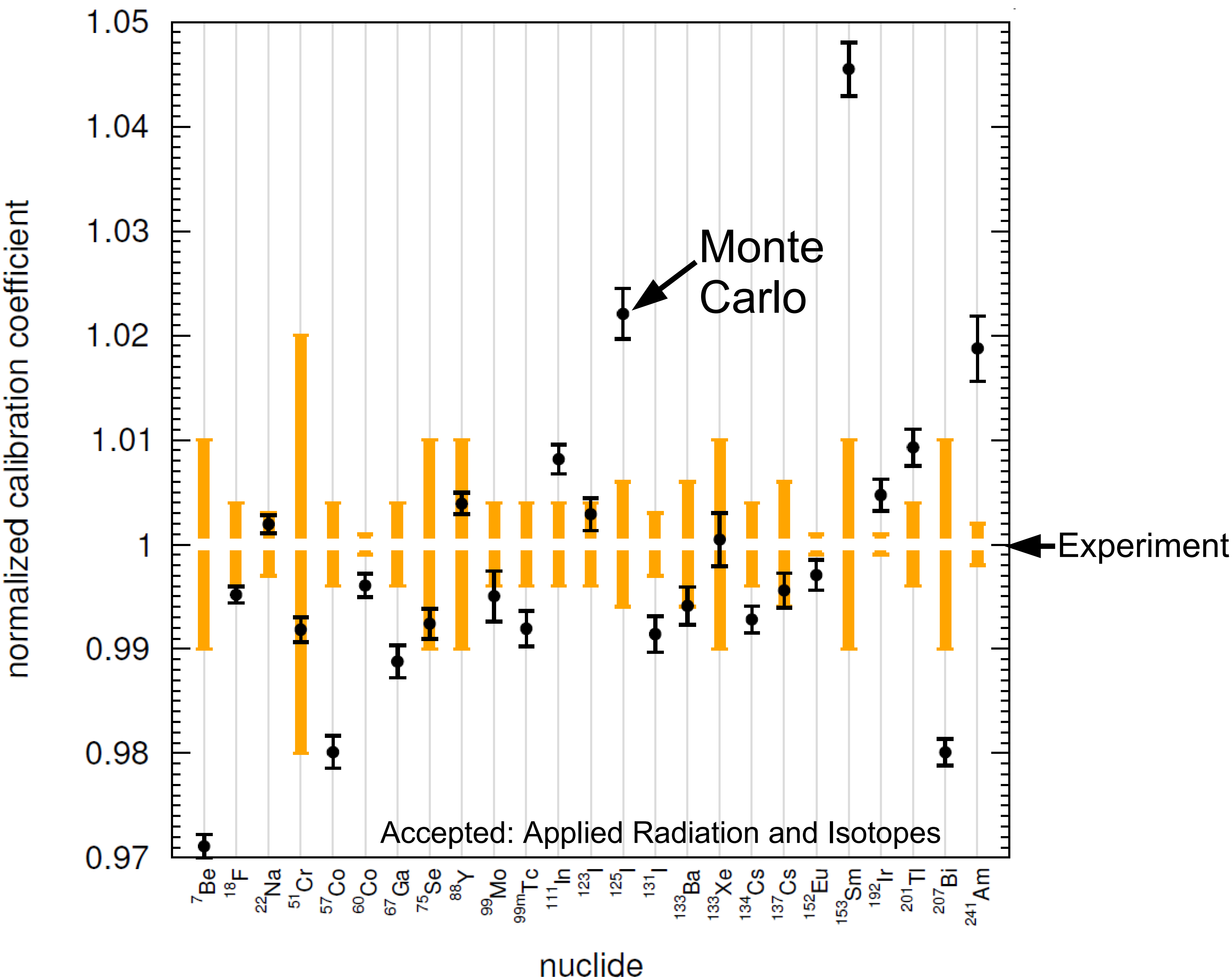
EGSnrc cumulates energy depositions

- EGSnrc reports energy deposited in nitrogen [eV]: E_{g}
- Convert to total charge [C]: $Q = \left(\frac{E_{\mathrm{g}}}{W}\right) e$

$W = 34.8 \pm 0.2\ \mathrm{eV}$ (average energy to create ion pair in nitrogen)

- The charge is deposited for exactly N decays

$$k_{\mathrm{mc}} = \frac{I\ (\mathrm{pA})}{A\ (\mathrm{MBq})} = 10^{18} \cdot \frac{Q}{N} = 10^{18} e\, \frac{(E_{\mathrm{g}}/N)}{W}$$



1.05
1.04
1.03
1.02
1.01
1
0.99
0.98
0.97
normalized calibration coefficient
Monte
Carlo
Experiment
Accepted: Applied Radiation and Isotopes
7Be
18F
22Na
51Cr
57Co
60Co
67Ga
75Se
88Y
99Mo
99mTc
111In
123I
125I
131I
133Ba
133Xe
134Cs
137Cs
152Eu
153Sm
192Ir
201Tl
207Bi
241Am
nuclide

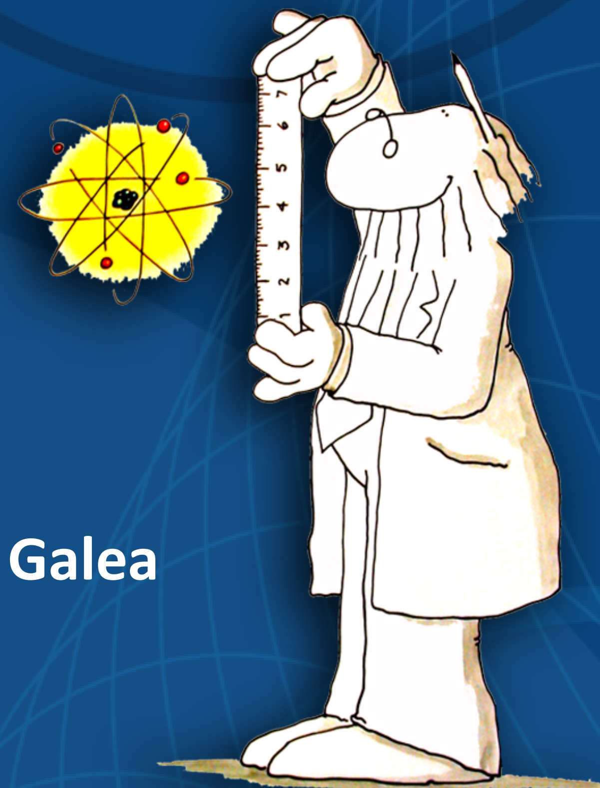
Now we know where to focus

- In the experiment:
  - Radio-impurities?
  - Re-standardization by primary method?
  - Sharpen uncertainties by testing different conditions

- In the model:
  - Pure water was used as the source solution (even for gases!)
  - Refinement of materials, geometries, source modelling etc.

Thanks to Patrick Saull for his help with beta spectra

Thanks to LNHB for providing ENSDF data

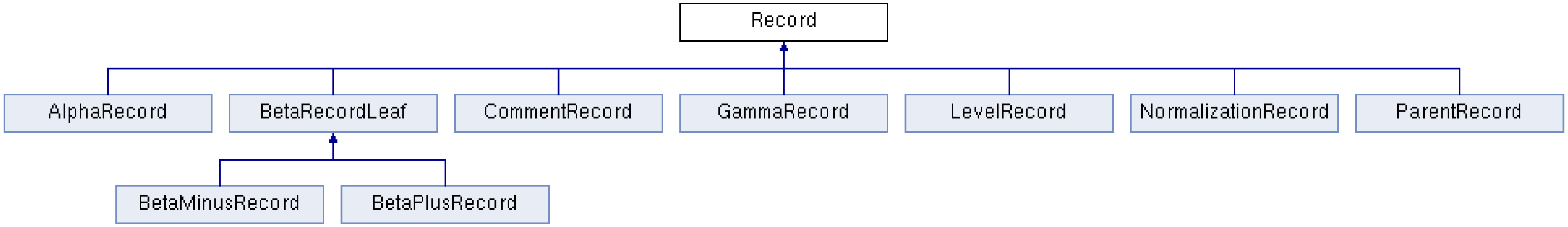


Reid Townson, Frédéric Tessier, Raphael Galea

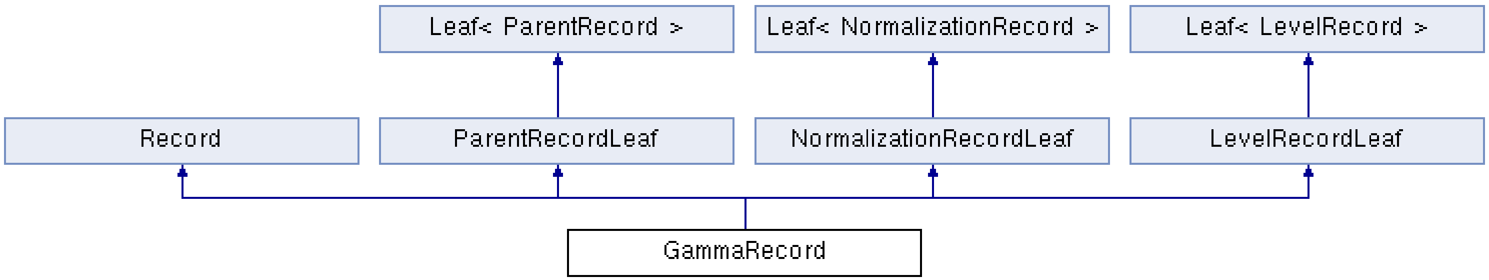
Measurement Science and Standards
National Research Council Canada

ENSDF records converted to c++ objects

- egs++ design is object-oriented



It's a tree-like structure

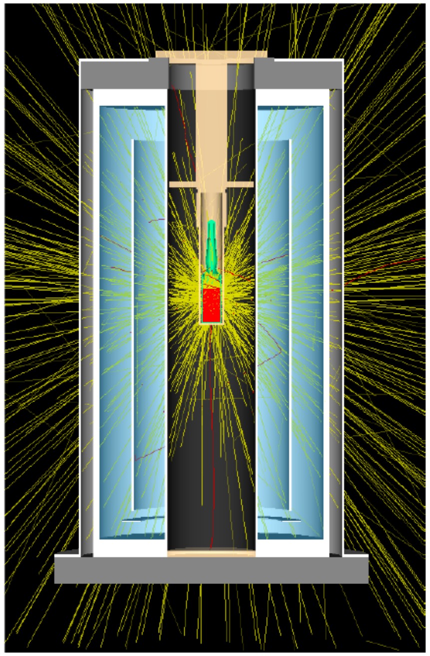


Public Member Functions

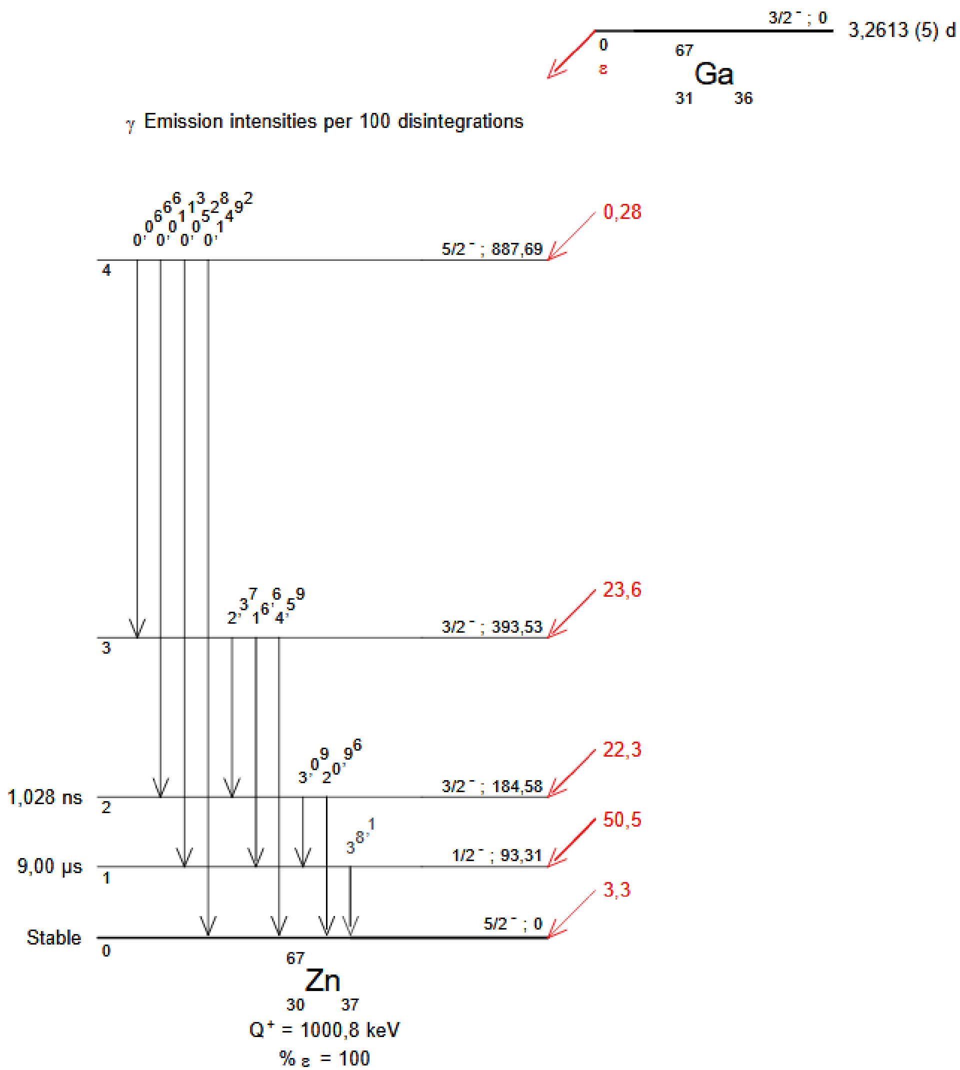
	GammaRecord (vector< string > ensdf, ParentRecord *myParent, NormalizationRecord *myNormalization, LevelRecord *myLevel)
	GammaRecord (GammaRecord *gamma)
double	**getDecayEnergy** () const
double	**getTransitionIntensity** () const
void	**setTransitionIntensity** (double newIntensity)
int	**getCharge** () const
LevelRecord *	**getFinalLevel** () const
void	**setFinalLevel** (LevelRecord *newLevel)
void	**incrNumSampled** ()
EGS_I64	**getNumSampled** () const

Simulations provide experimental refinement

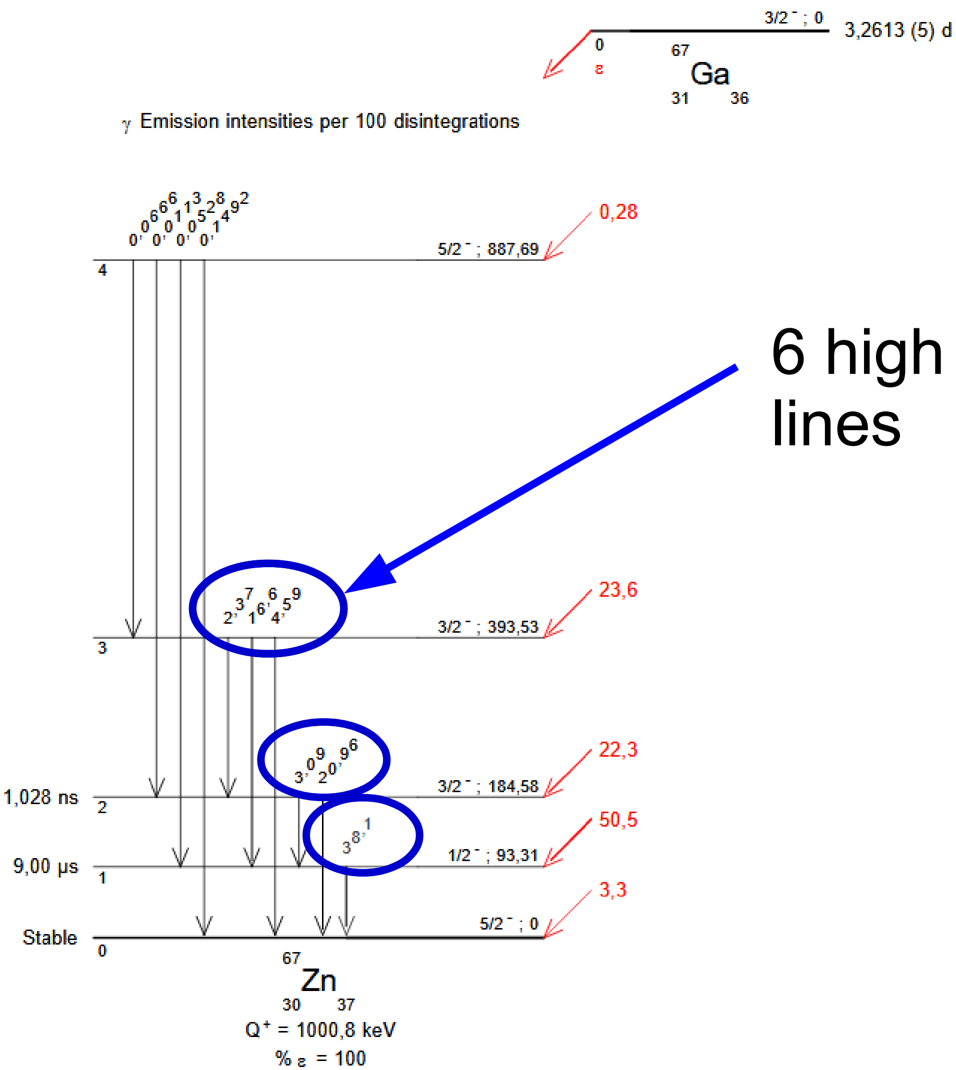
- An EGSnrc model of your detector allows you to:
  - Validate experiments
  - Predict detector response for unknown isotopes
  - Refine experimental uncertainty budget
    - Test geometrical variations
    - Test manufacturing tolerances
    - Test radioimpurity effects



Calculating calibration factors: an example

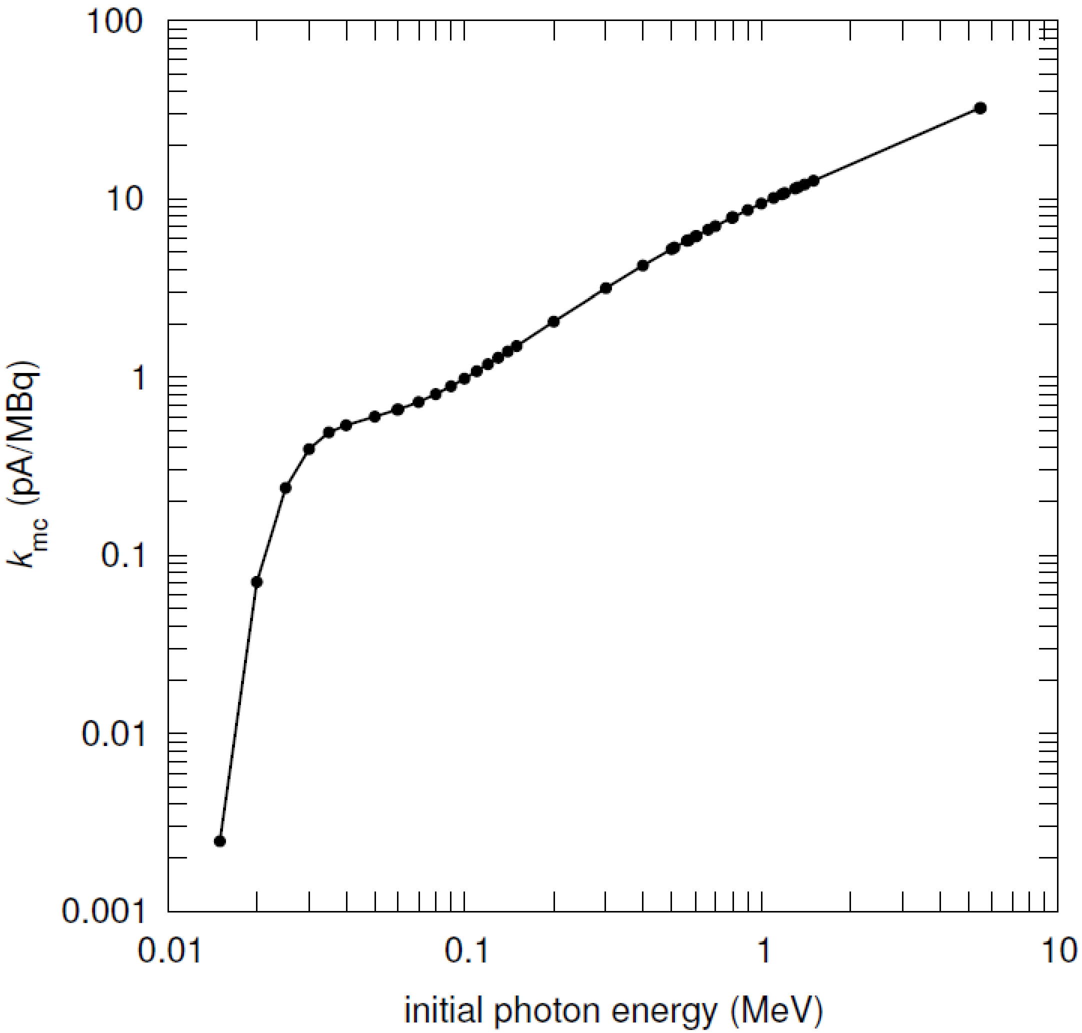


Let's try this the "old way"



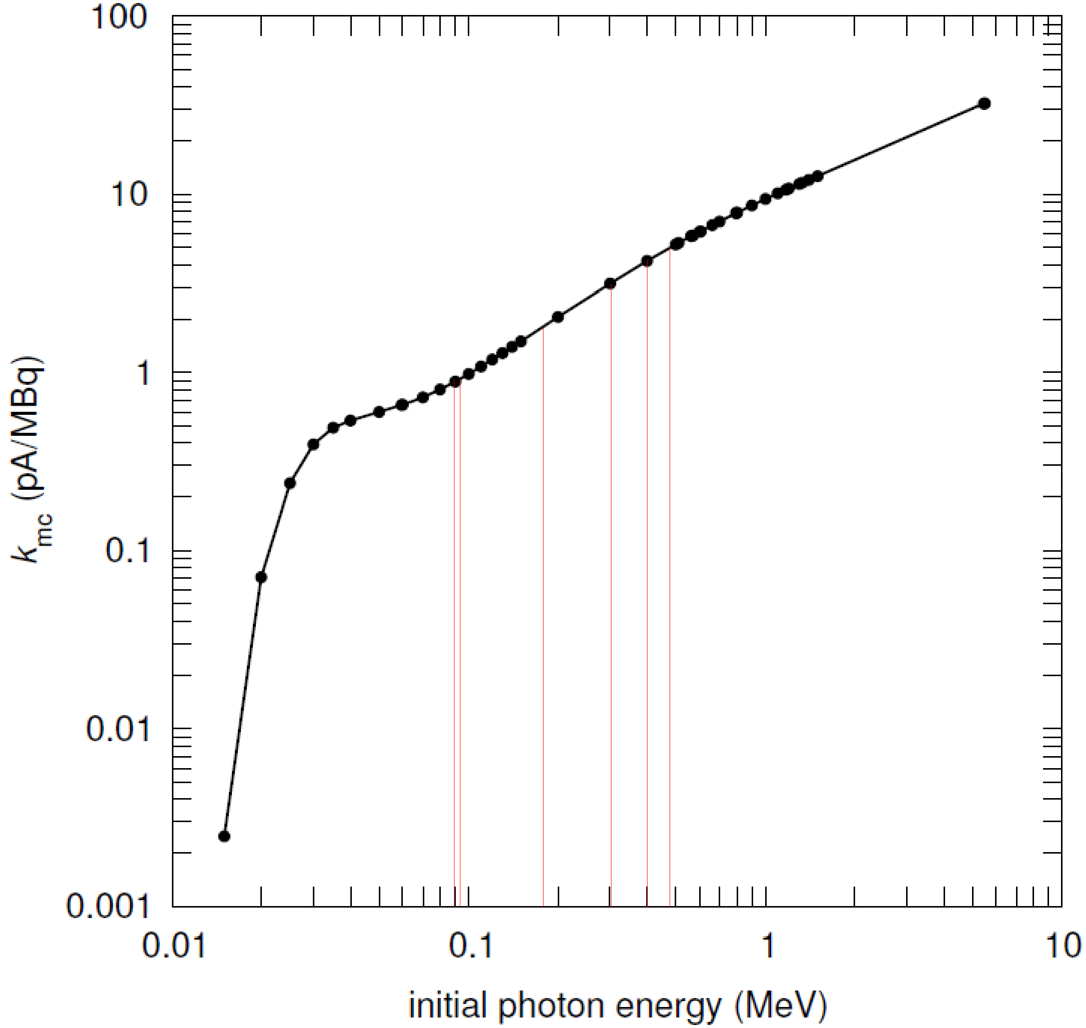
6 high intensity photon lines

Use a series of monoenergetic simulations



Interpolate response

$k_1=0.899$
$k_2=0.917$
$k_3=1.877$
$k_4=2.146$
$k_5=3.142$
$k_6=4.140$



Perform weighted sum using relative intensities

$k_1=0.899$ $P_1=3.09$

$k_2=0.917$ $P_2=38.1$

$k_3=1.877$ $P_3=20.96$ → $k_{hand} = 1.533$

$k_4=2.146$ $P_4=2.37$

$k_5=3.142$ $P_5=16.6$

$k_6=4.140$ $P_6=4.59$

Perform weighted sum using relative intensities

$k_1=0.899$	$P_1=3.09$
$k_2=0.917$	$P_2=38.1$
$k_3=1.877$	$P_3=20.96$
$k_4=2.146$	$P_4=2.37$
$k_5=3.142$	$P_5=16.6$
$k_6=4.140$	$P_6=4.59$

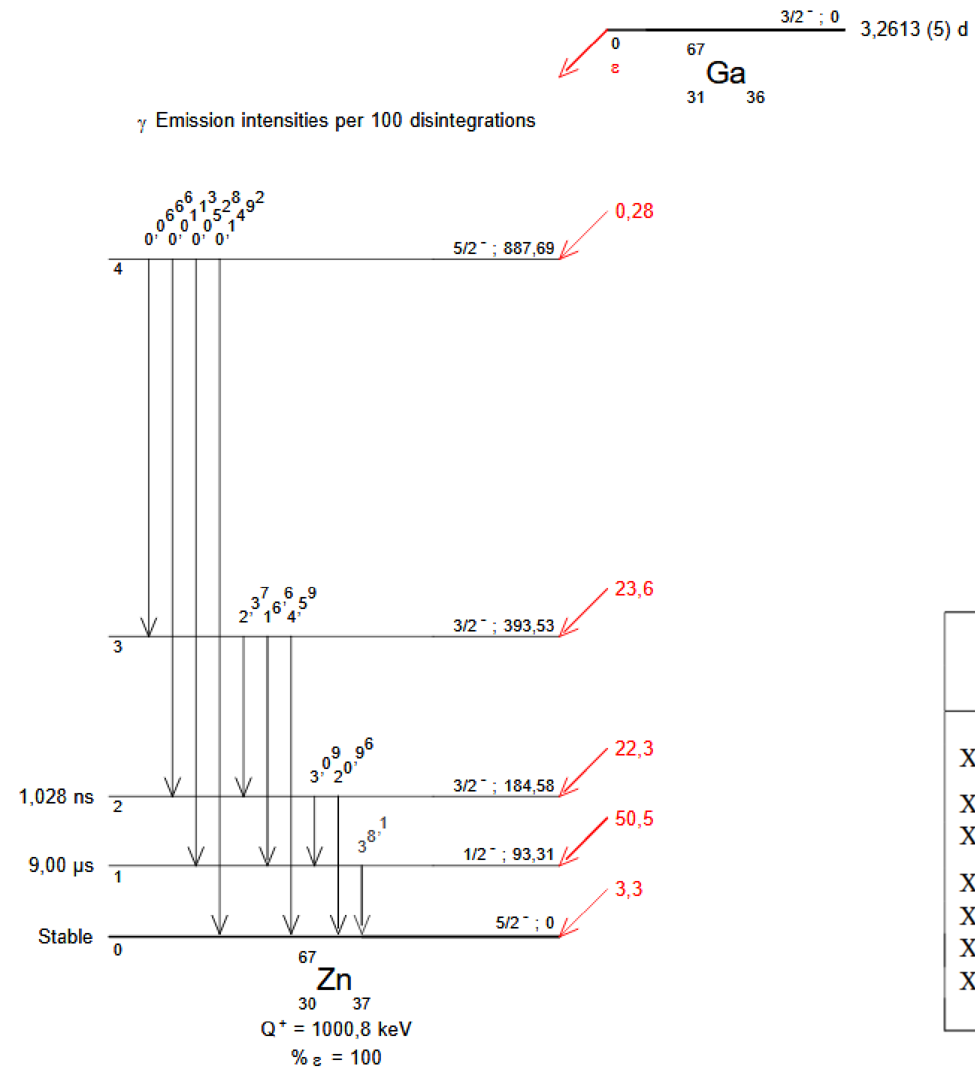
→

$k_{hand} = 1.533$

$k_{exp} = 1.583$

-3.1%

The radionuclide source models a bit more

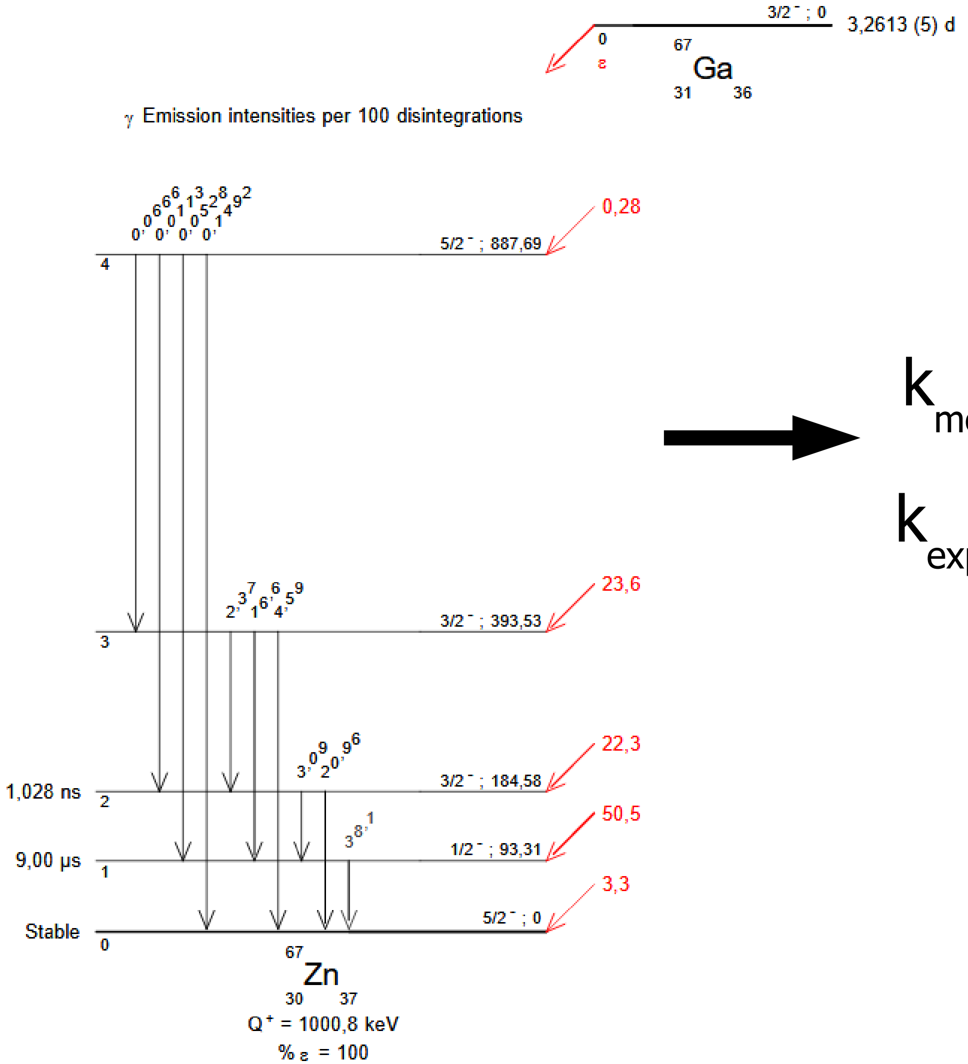


		Energy keV			Electrons per 100 disint.
e_{AL}	(Zn)	0,732	-	0,997	167,5 (21)
e_{AK}	(Zn)				60,4 (21)
	KLL	7,21	-	7,55	}
	KLX	8,31	-	8,63	}
	KXY	9,39	-	9,65	}
$ec_{2,1\ K}$	(Zn)	81,604		(15)	0,250 (16)
$ec_{1,0\ K}$	(Zn)	83,651		(5)	28,4 (7)
$ec_{1,0\ L}$	(Zn)	92,116	-	93,290	3,55 (9)
$ec_{1,0\ M}$	(Zn)	93,174	-	93,302	0,522 (13)
$ec_{2,0\ K}$	(Zn)	174,918		(17)	0,316 (40)
$ec_{3,1\ K}$	(Zn)	290,558		(10)	0,060 (3)

		Energy keV		Photons per 100 disint.	
XL	(Zn)	0,8836 — 1,1861		1,75 (5)	
$XK\alpha_2$	(Zn)	8,61587		17,0 (6)	} $K\alpha$
$XK\alpha_1$	(Zn)	8,63896		33,0 (12)	}
$XK\beta_1$	(Zn)	9,5721	}	7,08 (26)	$K\beta_1'$
$XK\beta_5''$	(Zn)	9,6499	}		
$XK\beta_2$	(Zn)	9,6581	}		
$XK\beta_4$	(Zn)		}		$K\beta_2'$

M.-M. Bé, V. Chisté, C. Dulieu, M.A. Kellett, X. Mougeot, A. Arinc, V.P. Chechev, N.K. Kuzmenko, T. Kibédi, A. Luca, and A.L. Nichols. *Table of Radionuclides*, volume 8 of *Monographie BIPM-5*. Bureau International des Poids et Mesures, Pavillon de Breteuil, F-92310 Sèvres, France, 2016.

Closer agreement!



$$k_{mc} = 1.5547$$

$$k_{exp} = 1.583$$

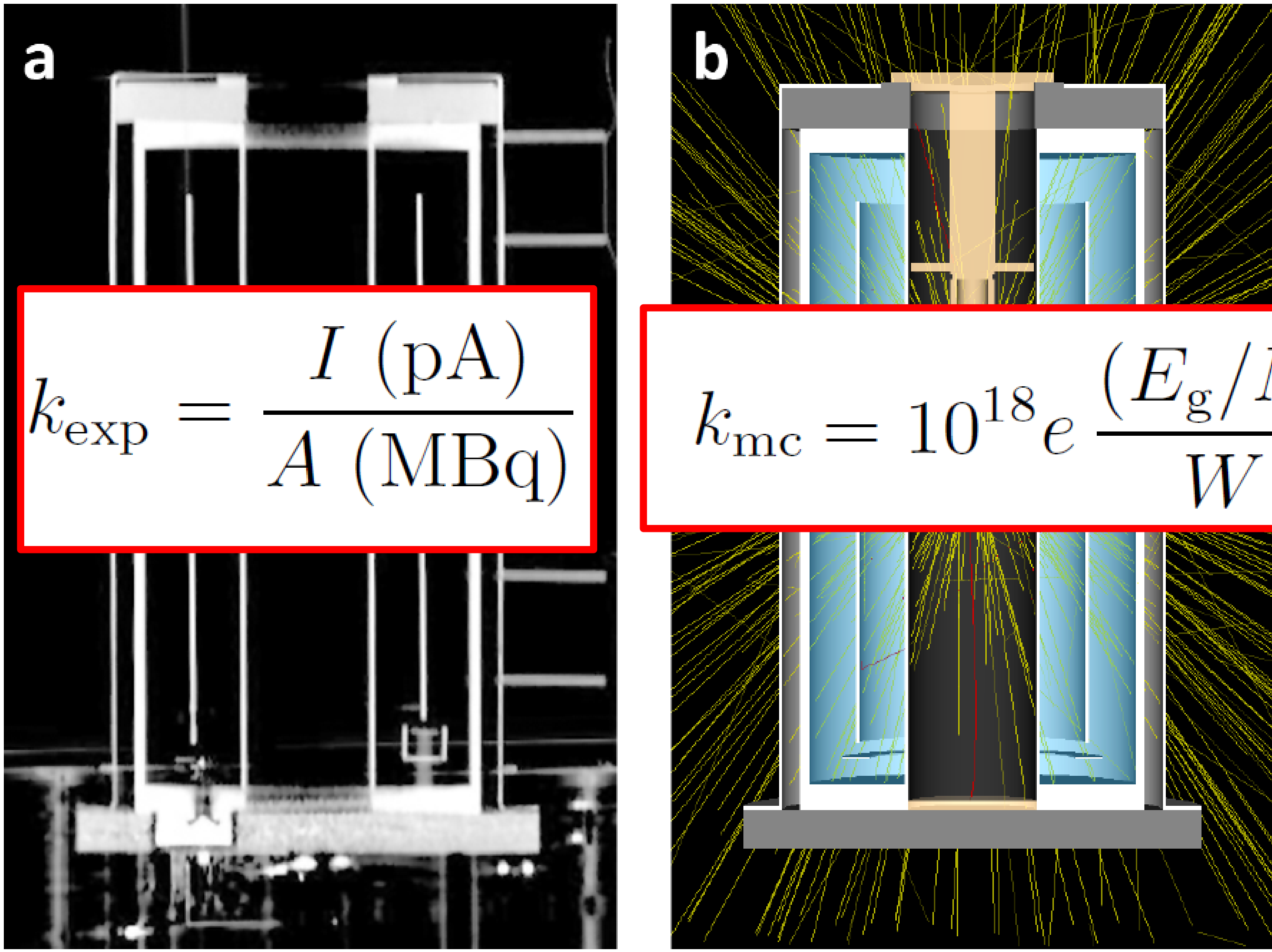
-1.8%

(factor of 1.7 closer)

Simulations provide answers

- With an accurate EGSnrc model at our disposal, we can now look at the questions:
    - How does the uncertainty on a parameter affect measurement?
    - What is the calibration factor for a radionuclide not previously measured?
    - What is the calibration factor for a non-standard geometry?
    - What is the effect of radioimpurities?

Simulations can produce an absolute result



$$k_{\rm exp} = \frac{I\ ({\rm pA})}{A\ ({\rm MBq})}$$

$$k_{\rm mc} = 10^{18} e \, \frac{(E_{\rm g}/N)}{W}$$

There was a problem with the detector model

- Initially, the modelled detector response was systematically low
    - An energy-dependent difference (~7%)

- This indicates a physical discrepancy:
    - Material properties (density, composition)?
    - Geometrical (wall thicknesses)?

We increased the gas pressure

- Varying within manufacturer tolerances could not account
- There was no tolerance on the nitrogen pressure (nominal 1MPa)
    - Increasing the pressure ~7% worked (chi-squared optimized)

- Therefore, our model **predicts** a 7% higher pressure

Turns out it's corroborated

- Strikingly, a previous group also found a 7.2% higher pressure by simulations of a similar chamber using PENELOPE

A De Vismes and MN Amiot. Towards absolute activity measurements by ionisation chambers using the penelope monte-carlo code. *Applied radiation and isotopes*, 59(4):267–272, 2003.

After a few minutes on the cluster...

Radionuclide	k_{mc} (pA/MBq)	Statistical uncertainty	k_{exp} (pA/MBq)	Measurement uncertainty	Percent difference
^{7}Be	0.5195	0.1%	0.535	1%[a]	−2.89%
^{18}F	10.2901	0.1%	10.34	0.3%	−0.48%
^{22}Na	20.8103	0.1%	20.77	0.3%	0.19%
^{51}Cr	0.3326	0.1%	0.3353	2%	−0.82%
^{57}Co	1.2006	0.2%	1.225	0.4%	−1.99%
^{60}Co	22.1523	0.1%	22.24	0.1%	−0.39%
^{67}Ga	1.5653	0.2%	1.583	0.4%	−1.12%
^{75}Se	3.9577	0.1%	3.988	1%[b]	−0.76%
^{88}Y	22.6181	0.1%	22.53	1%	0.39%
^{99}Mo*	2.6757	0.2%	2.689	0.4%	−0.50%
^{99m}Tc	1.2409	0.2%	1.251	0.4%	−0.81%
^{111}In	4.1374	0.1%	4.104	0.4%	0.81%
^{123}I	1.7791	0.2%	1.774	0.4%	0.29%
^{125}I	0.4957	0.2%	0.485	0.6%	2.21%
^{131}I	3.9984	0.2%	4.033	0.3%	−0.86%
^{133}Ba	4.2726	0.2%	4.298	0.6%	−0.59%
^{133}Xe	0.5057	0.3%	0.5055	1%[c]	0.05%
^{134}Cs	15.4777	0.1%	15.59	0.4%	−0.72%
^{137}Cs	5.7156	0.2%	5.741	0.6%	−0.44%
^{152}Eu	10.9677	0.1%	11.00	0.1%	−0.29%
^{153}Sm	0.6853	0.2%	0.6555	1%[b]	4.55%
^{192}Ir	8.5210	0.1%	8.481	0.1%	0.47%
^{201}Tl	0.9068	0.2%	0.8985	0.4%	0.93%
^{207}Bi	14.6426	0.1%	14.94	1%[b]	−1.99%
^{241}Am	0.2499	0.3%	0.2453	0.2%	1.87%