



# A Modelling Approach to Defining Radiochemical Separation Requirements for Gamma Spectrometry Analysis

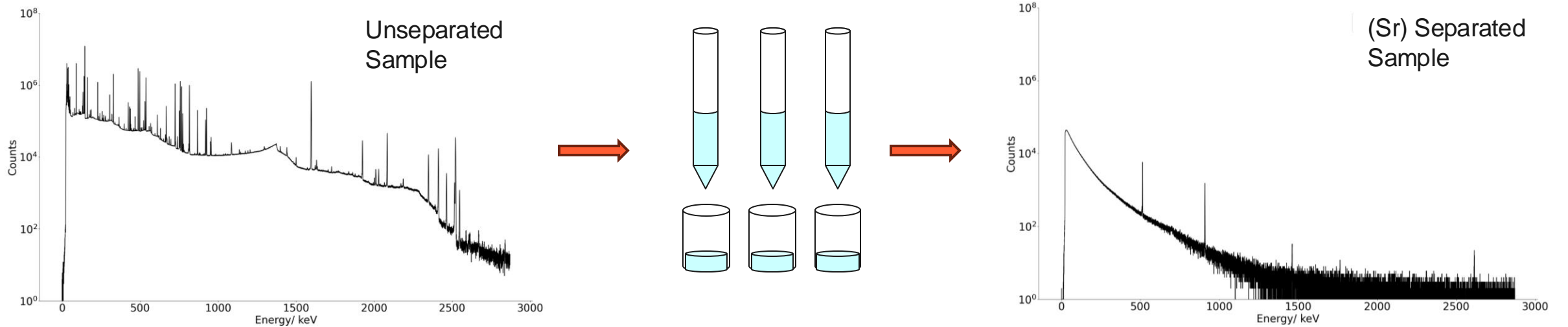
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# Introduction

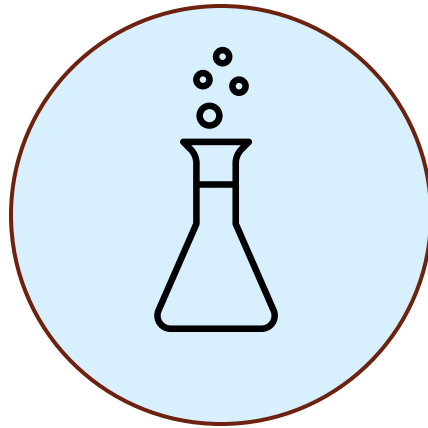


*An example flow of a chemical separation with analysis by gamma spectrometry*

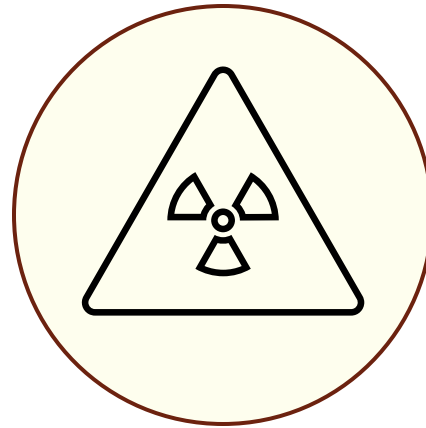
Gamma-emitting impurities can impair the measurement of target nuclides in two ways:

1. Direct interference with peaks of interest
2. Elevating the background continuum from incomplete absorption (via Compton scattering interactions)

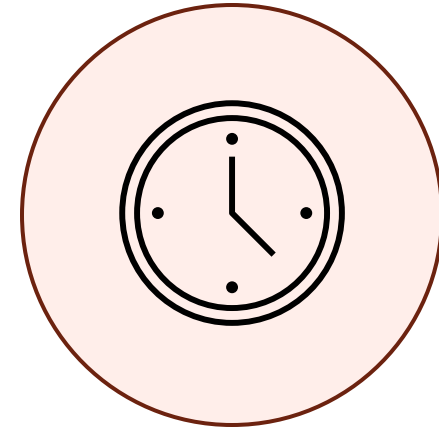
# Background – Radiochemical Separations



Chemical Yield



Impurity Activity

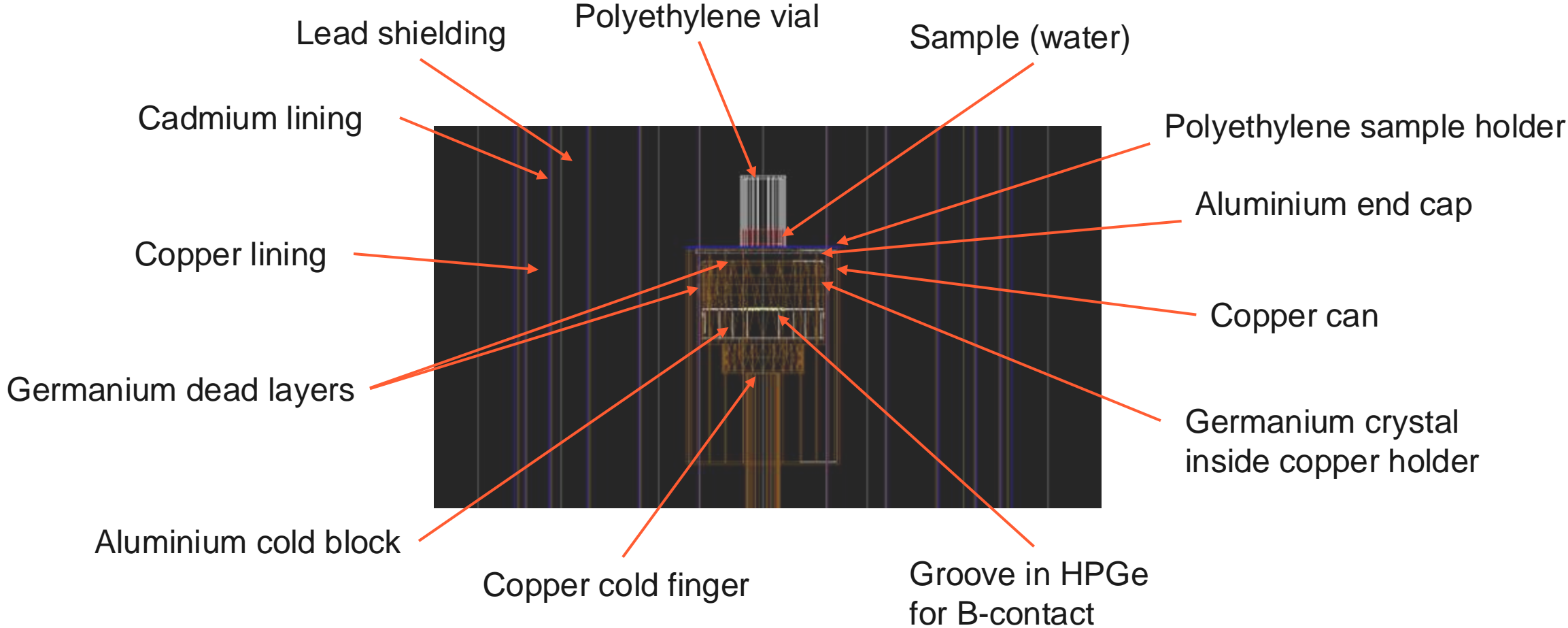


Elapsed Time

**What are the success criteria for a radiochemical separation such that a nuclide of interest is measurable at a given activity?**

*This work aims to use modelling techniques to assess potential radiochemical outcomes to deduce requirements.*

# Geometry Generation – 5 mL in a vial on a BEGe 5030 (GEANT4)



# Geometry Validation

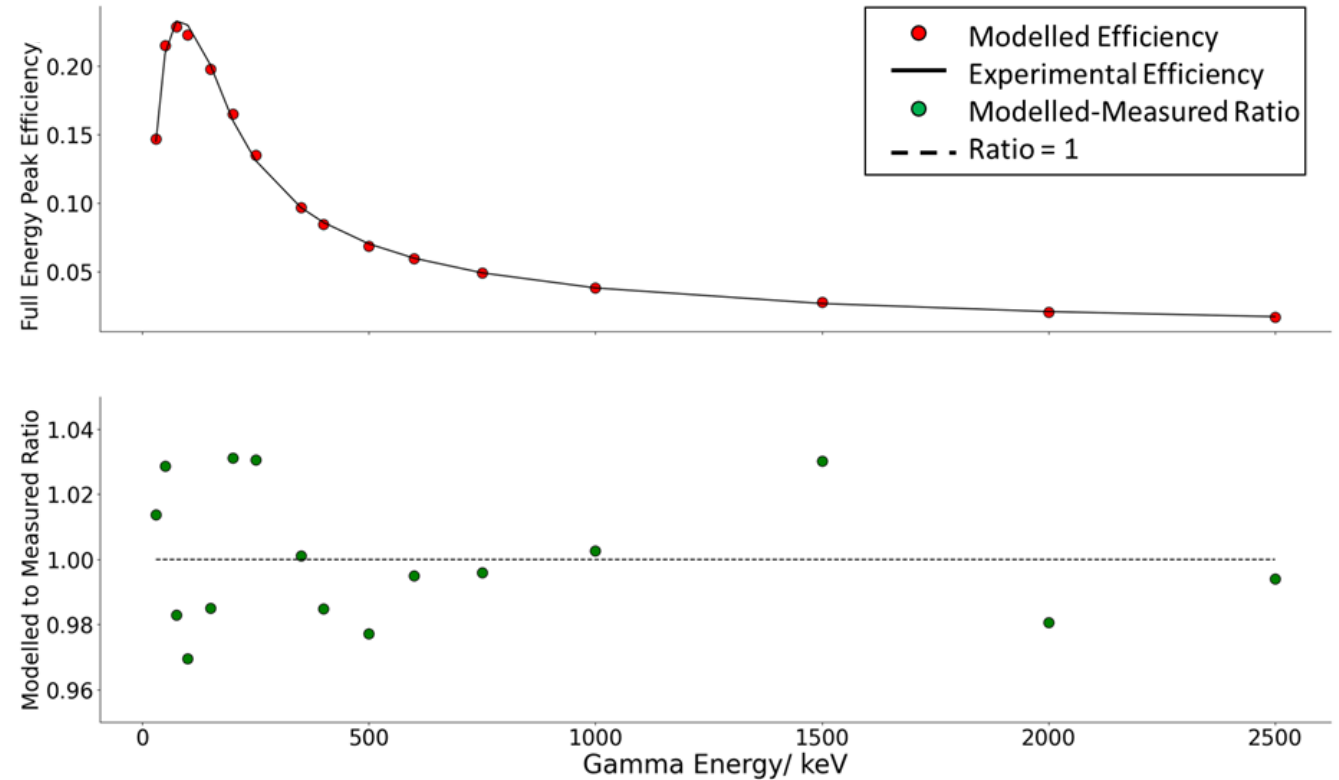
Gamma photons of specific energy emitted isotropically from the sample volume as a G4ParticleGun.

Detection efficiency determined by:

$$\varepsilon_{FE}(\%) = \frac{N_{counts}}{N_{total}} \times 100$$

Key geometry parameters:

- Dead layers (low energies)
- Crystal position (medium energies)
- HPGe crystal size (medium-high energies)



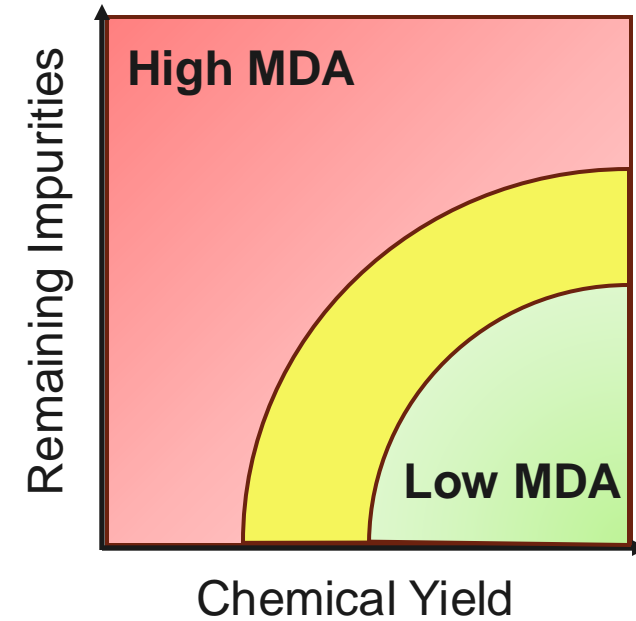
*Measured and modelled full energy peak efficiencies (top) and a residuals plot (bottom)*

# Initial Investigations - Introduction

For a sample containing a radionuclide of interest, a radiotracer/internal standard and two notional contaminants, the effect of different chemical yields and impurity removals were simulated.

The impact of these changes on the Minimum Detectable Activity (MDA) of the preferred emission of the radionuclide is then calculated.

For these simulations, the count time (68000 seconds) and number of elapsed days (1 day) was kept constant.



*A simplified representation of the expected behaviour. Green to red desirable to undesirable.*

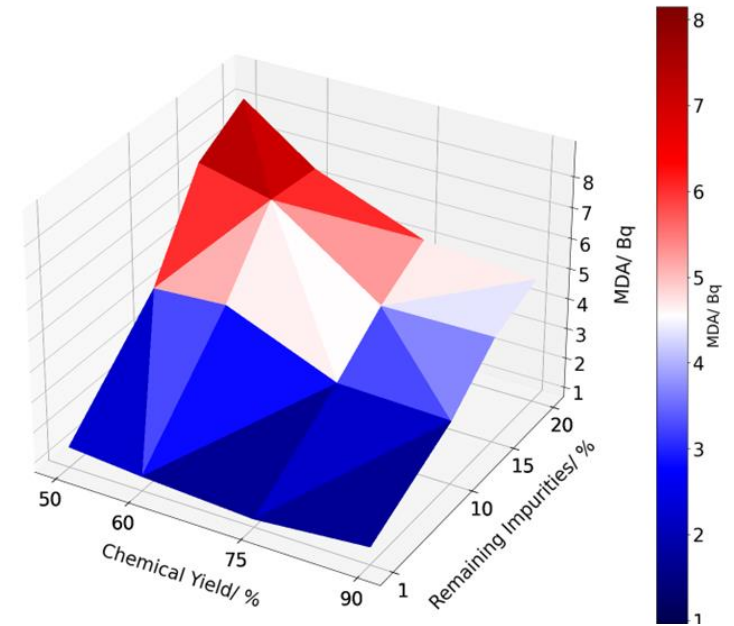
# Initial Investigations – $^{115}\text{Cd}$

Simulation Number	Radionuclide	Activity/ Bq	Signature of interest
01	$^{109}\text{Cd}$	100	$^{115}\text{Cd}$ (336.2 keV)
	$^{115}\text{Cd}$	50	
	$^{140}\text{Ba}$	50	
	$^{140}\text{La}$	50	
02	$^{109}\text{Cd}$	35	$^{115}\text{Cd}$ (336.2 keV)
	$^{115}\text{Cd}$	10	
	$^{140}\text{Ba}$	250	
	$^{140}\text{La}$	250	

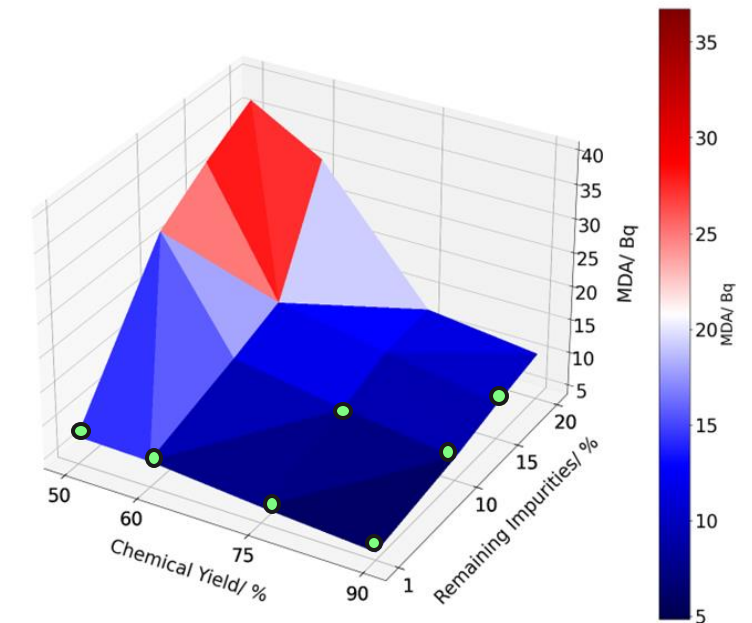
Key points:

1. High MDA at worst chemistry outcomes and vice versa, as expected.
2. Sim 02 –  $^{115}\text{Cd}$  only measurable in 7 of 16 scenarios.
  - Shown by ● markers

Sim 01



Sim 02



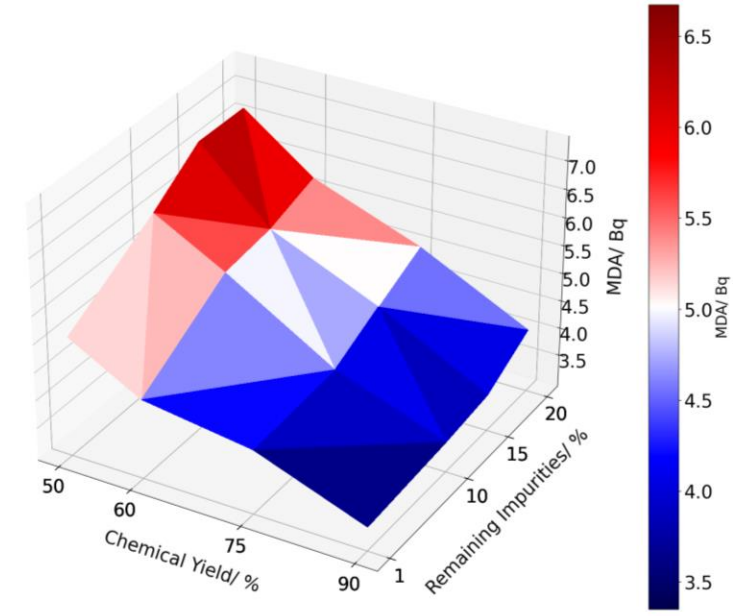
# Initial Investigations – $^{111}\text{Ag}$

Simulation Number	Radionuclide	Activity/ Bq	Signature of interest
03	$^{110\text{m}}\text{Ag}$	25	$^{111}\text{Ag}$ (342.1 keV)
	$^{111}\text{Ag}$	4.5	
	$^{140}\text{Ba}$	50	
	$^{140}\text{La}$	50	
04	$^{110\text{m}}\text{Ag}$	25	$^{111}\text{Ag}$ (342.1 keV)
	$^{111}\text{Ag}$	4.5	
	$^{140}\text{Ba}$	500	
	$^{140}\text{La}$	500	

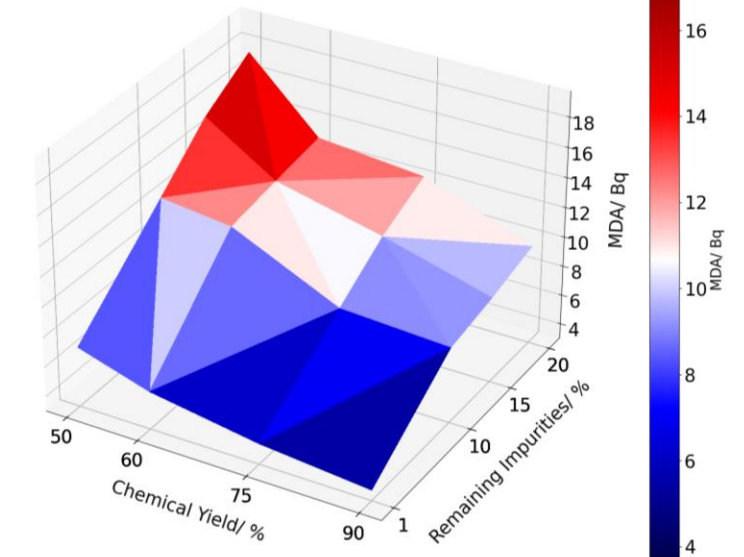
Key points:

1. Sim 03 – Plot indicates a preference to **prioritise yield over purity**
2. Sim 04 – Plot indicates a preference to **prioritise purity over yield**

Sim 03



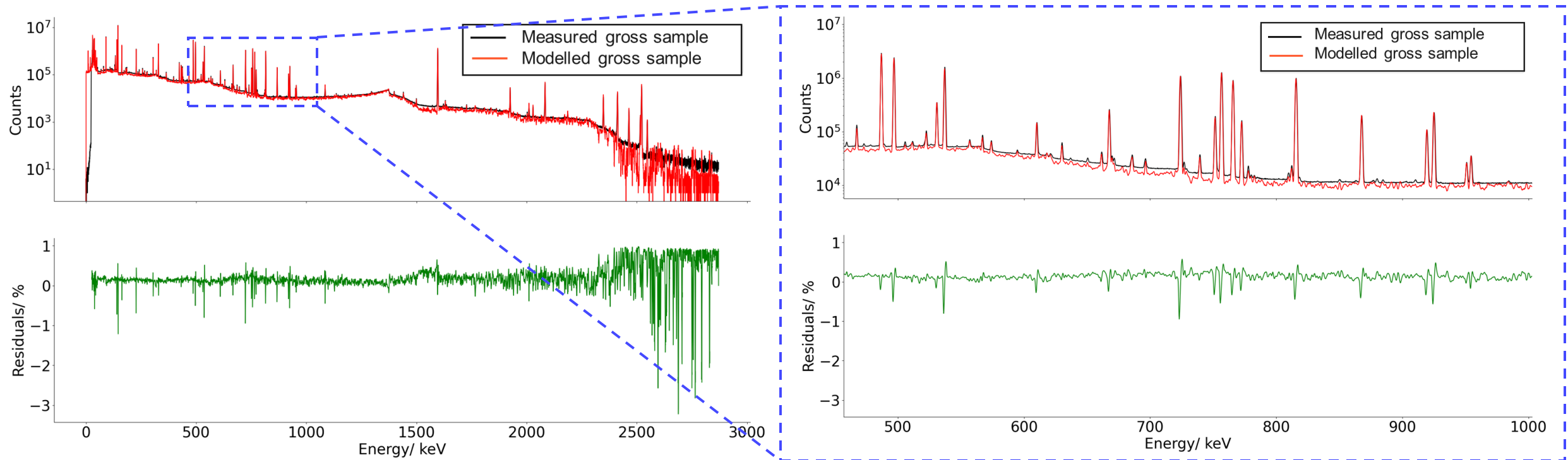
Sim 04



*Manual peak fitting was required due to an interfering Ba-140 X-γ sum peak*

# Real-world Investigation

A sample containing a wide variety of radionuclides was receipted. The initial gross spectrum was replicated in GEANT4 and presented below. Radiochemical separations attempting to isolate neptunium were performed...



# Np Investigation

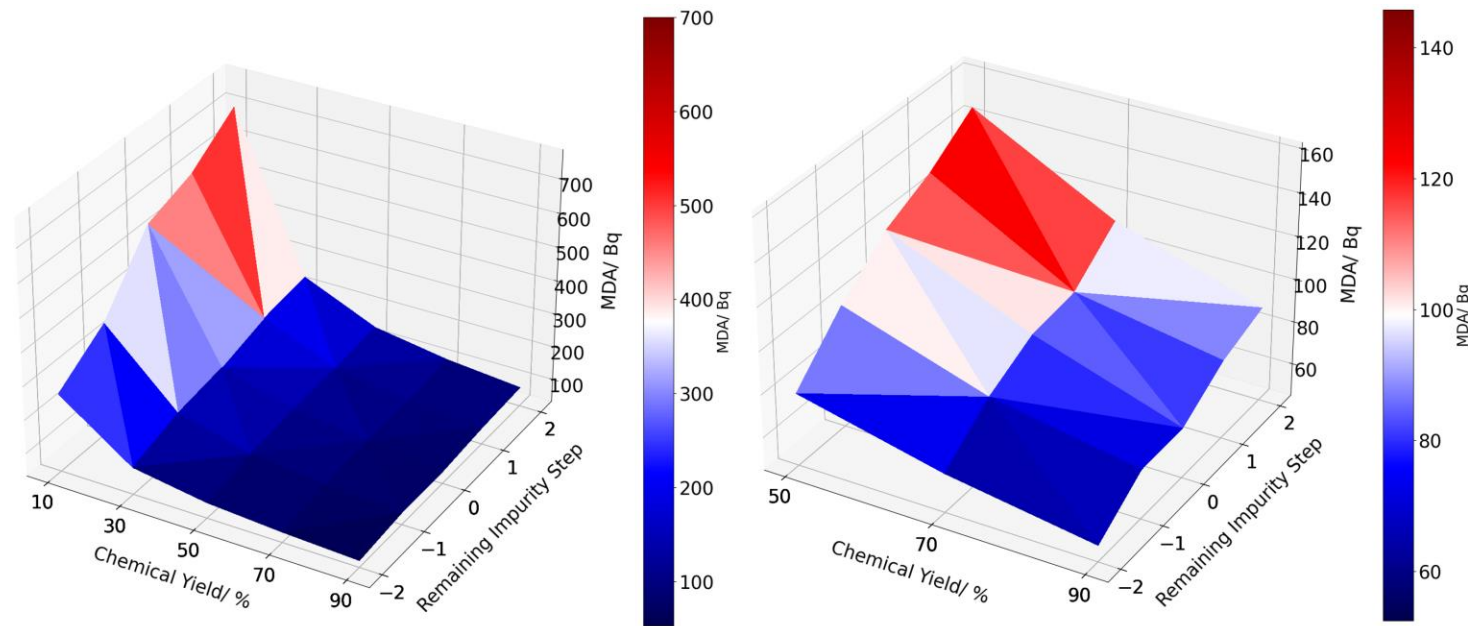
LIVE TIME (S): 26346.2  
 REAL TIME (S): 26358.8  
 ACQUISITION START TIME (DD/MM/YYYY HH/MM/SS/MSMS): 23/08/2024 06/48/48/00  
 REFERENCE TIME (DD/MM/YYYY HH/MM/SS/MSMS): 02/08/2024 18/26/00/00  
 ENERGY CALIBRATION (M,C): [0.17525,-0.4981]  
 SHAPE CALIBRATION (M,C): [0.0358,0.279]  
 SAMPLE MASS (G): 3.73582

DATA TABLE					
RADIONUCLIDE	HALFLIFE(S)	VALUE	UNIT(DECAYS OR Bq)	TARGET_SEP_FACTOR	SEP_FACTOR_STEP
		951.98	Bq/g	0	0
		91049	Bq/g	0.0001152	0.00005
		34322	Bq/g	0.000202	0.0001
		747180	Bq/g	0	0
		3649.6	Bq/g	0.0002282	0.0001
		60.317	Bq/g	0	0
		103.51	Bq/g	0	0
		1237.9	Bq/g	0	0
		235420	Bq/g	0	0
		103460	Bq/g	0.0001188	0.00005
		395550	Bq/g	0	0
		9245.9	Bq/g	0.0002643	0.0001
		37406	Bq/g	0.0001804	0.00005
		2836.54	Bq/g	0	0
		1.8	Bq	1	0
		102210	Bq/g	0	0
		5857.6	Bq/g	0	0
		210.79	Bq/g	0	0
		14324	Bq/g	0	0
		5859.2	Bq/g	0	0
		413160	Bq/g	0	0
		252490	Bq/g	0	0
		160.82	Bq/g	0.0002623	0.001
		17002	Bq/g	0	0
		19066	Bq/g	0.0000366	0.00001

Configuration files were adapted to include a 'target separation factor' and 'separation factor step'.

The step is the amount added or subtracted from the target separation factor when the level of impurities is changed. The number of steps simulated can be defined when running the programme.

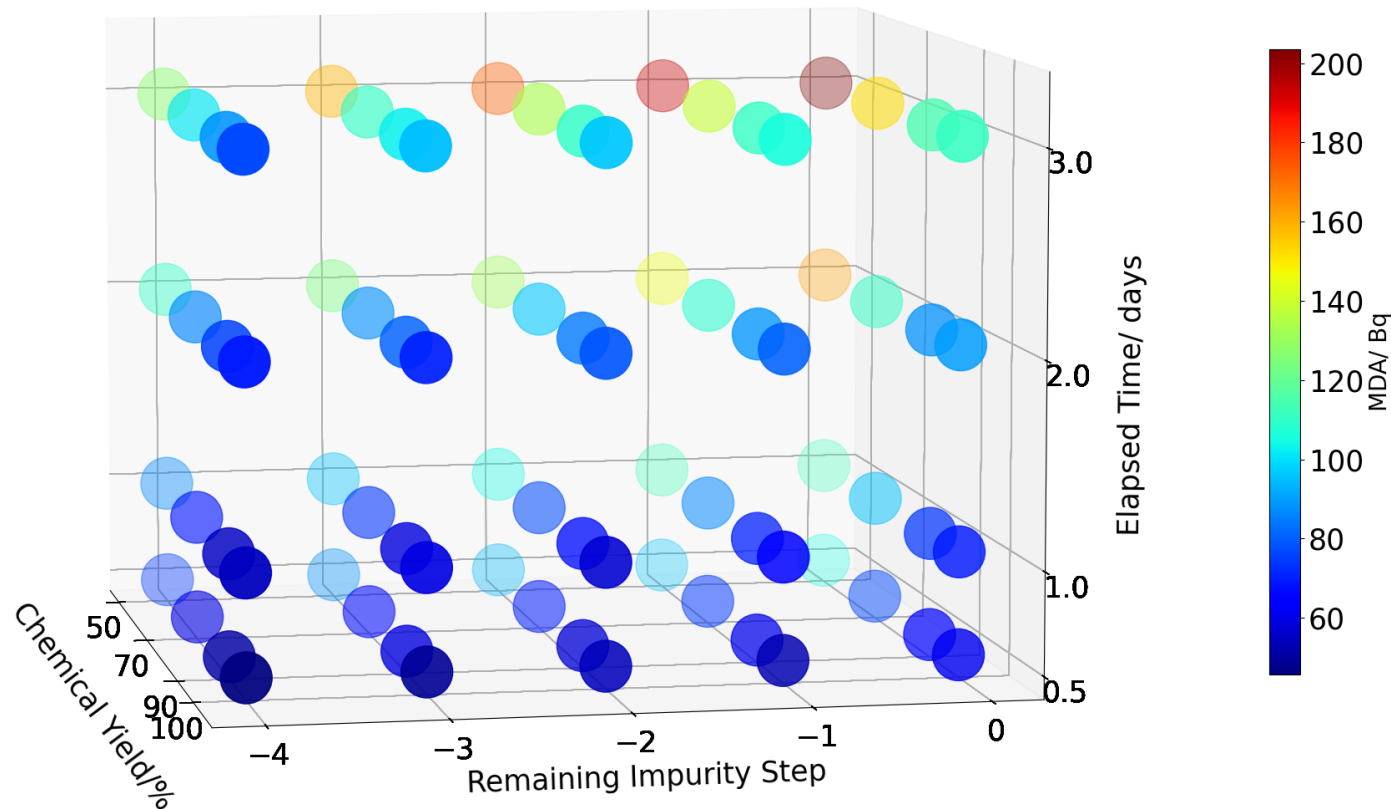
# Np Investigation



If the separation method is currently producing samples of 70% yield and an impurity concentration equivalent to 'step 2', this analysis suggests, broadly speaking that removing the impurities will have a greater impact on improving detection limits than improving yield (across this specific design space).

This analysis provides a semi-quantitative assessment on the interplay between yield and impurity activity for this sample composition.

# Np Investigation



Each impurity step is ~25% of each remaining impurity

For this particular sample, improvements in the MDA of  $^{239}\text{Np}$  is estimated to only be achieved with elapsed time of less than 24 hours and near complete impurity removal. ( $^{239}\text{Np}$  half-life = ~2.4 days)

# Conclusions

1. A model using the GEANT4 toolkit has been developed for a BEGe5030 HPGe detector with accuracy within 4% across a 30-2500 keV energy range.
2. Semi-quantitative detection limits have been estimated with differing outcomes from radiochemical separations by altering chemical yield, impurity activities and elapsed time.
3. Potential uses include:
  1. Predict achievable detection limits without the need for samples
  2. Facilitate radiochemical separation method development
  3. Inform the utility of purification procedures and when longer measurement times are the preferred method to lower detection limits

# Any Questions?

For any follow up comments/questions:  
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