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ISOTOPICS FROM PEAKS

Relative Efficiency Analysis in InterSpec

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20240728

U.S. DEPARTMENT OF ENERGY | NNSA

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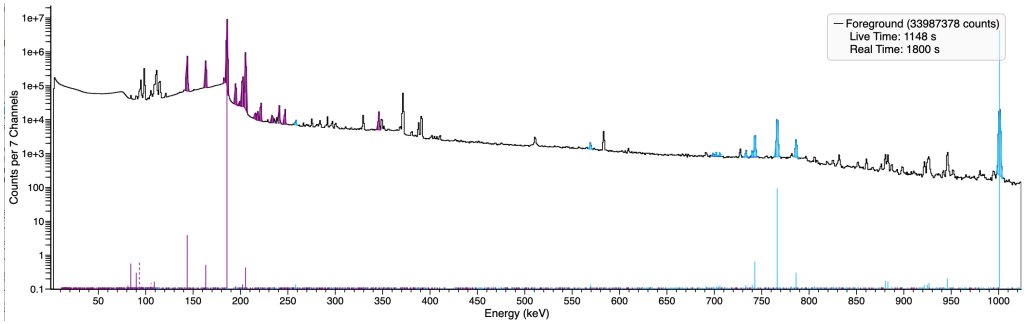
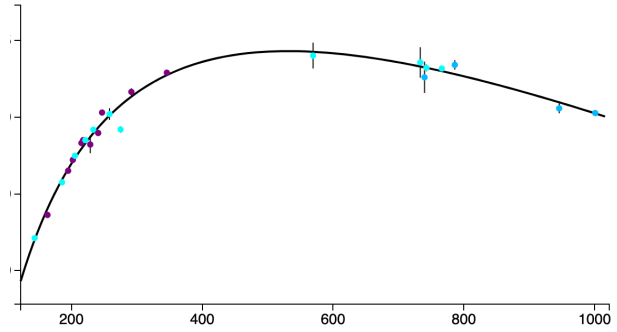
RELATIVE EFFICIENCY ANALYSIS

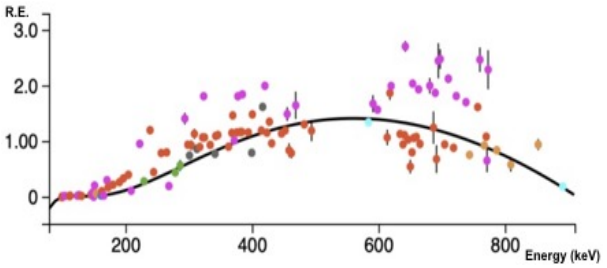


$$\frac{Peak\ Area}{Act. \times B.R.}$$

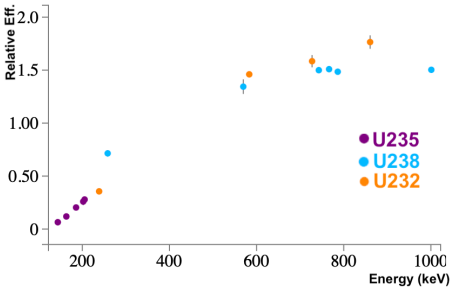


- Relative Efficiency Analysis finds the relative-activities for the nuclides in a spectrum, so that (peak area) / (activity × branching ratio) makes a smooth curve, as a function of energy
- It gives you the nuclide activities/masses, relative to one another – does not give absolute activities/masses
- *Does not* require knowing a detectors efficiency, the distance, shielding, or geometry
- *Does* require that all nuclides and shielding are homogeneous

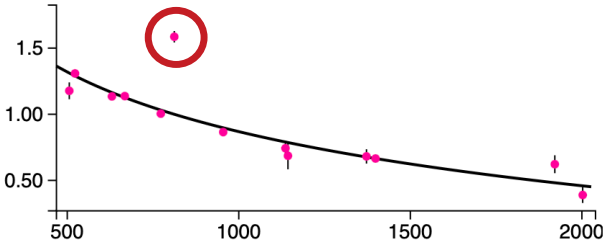
RELATIVE EFFICIENCY ANALYSIS – SIMPLE AND USEFUL!



A Plutonium measurement showing the sample is non-homogeneous due to different isotopes (different color dots) following different rel. eff. curves



A Uranium sample containing enriched, and natural or depleted U
(best fit line removed)



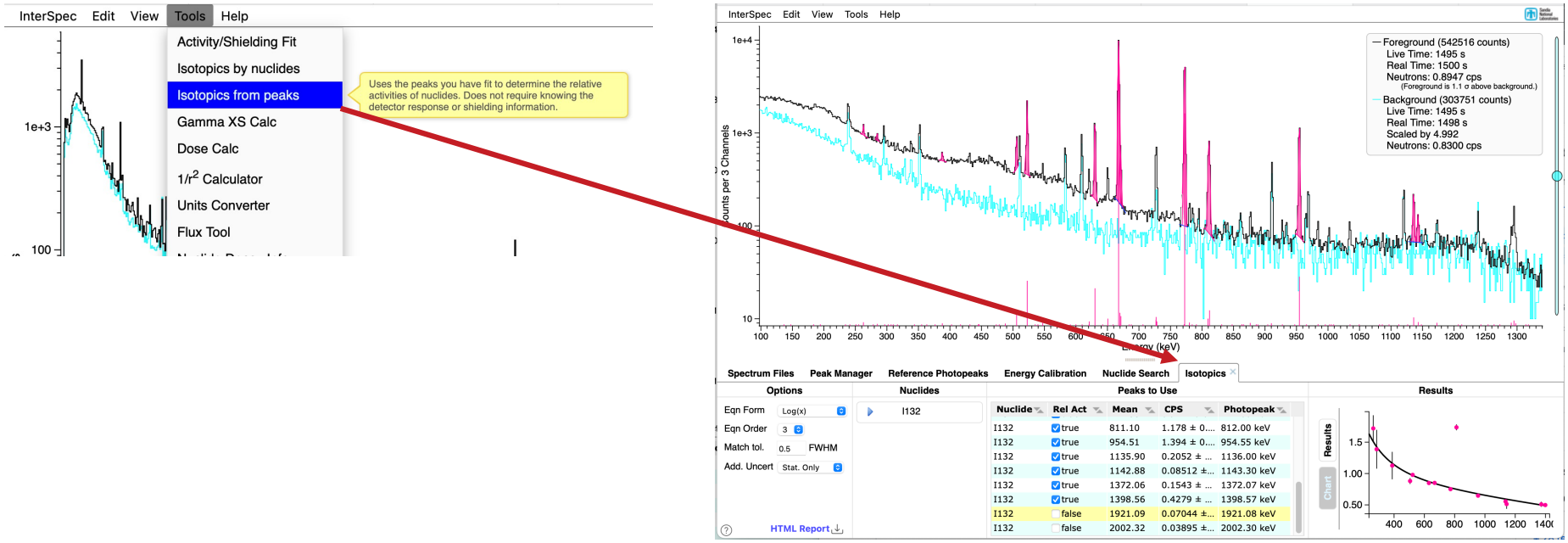
An interfering source – here a I-132 spectrum has an outlier efficiency. The Co-58 source was otherwise not obvious

- Most commonly used to determine enrichments for SNM
- But more generally gives ratio of activities, when you may not know distance, or detector efficiency, or geometry
  - Works with any gamma-producing nuclides, you just need nuclide(s) with enough energy points to fit the relative efficiency curve
- Often times also super useful to make sure sure you have the correct nuclide ID, or that there are no interfering sources present, or your peak fits are good, or that your sample is homogenous, or the shielding doesn't have holes in it, or that you haven't mis-assigned a peaks nuclide, …

OTHER REFERENCES

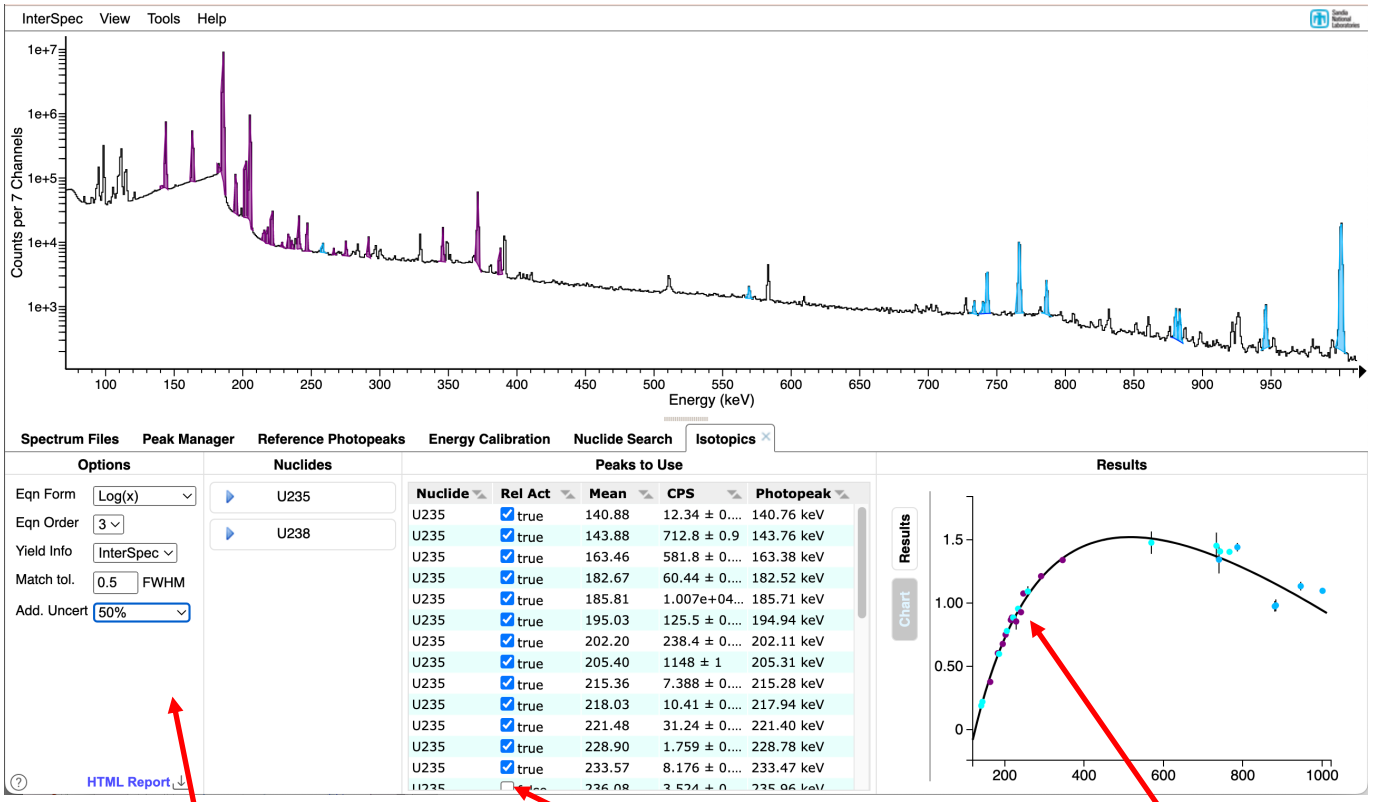
- *Relative Efficiency Curves Demystified*, by Michael Enghauser, SNL, July 2016
  https://www.osti.gov/servlets/purl/1399186
  A a great and concise introduction, especially for Uranium enrichment
- *FRMAC Gamma Spectroscopist Knowledge Guide*, by Michael Enghauser, Aug 2019
  https://www.osti.gov/biblio/1763003
  An all around great gamma spectroscopy manual; see Section 14 for Relative Efficiency
- *Application Guide to Gamma-Ray Isotopic Analysis Using the FRAM Software*, by T. Sampson, T. Kelley, and D. Vo, LANL, Sep 2003
  https://cdn.lanl.gov/files/app-to-isotopic-analysis-using-fram_06e9e.pdf
  FRAM is considered by many to be *the* software for U and Pu enrichment analysis from gamma spectra
- *Peak-Based Relative Efficiency analysis in InterSpec*, W. Johnson, SNL, Aug 2022,
  https://sandialabs.github.io/InterSpec/tutorials/#relative-efficiency-analysis
  Similar to this presentation, but focusing on U-enrichment

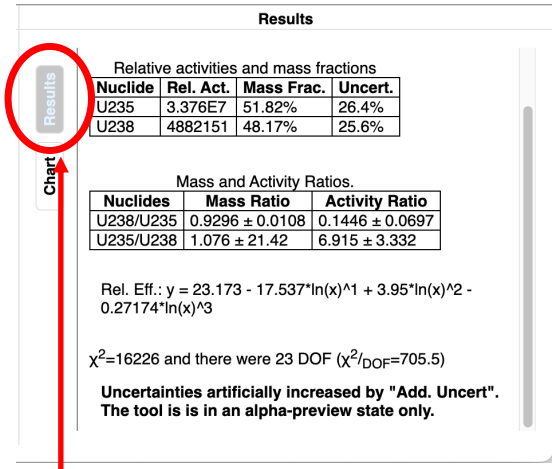
USING THE RELATIVE EFFICIENCY ANALYSIS IN INTERSPEC



- If you are already an InterSpec user, and you associate peaks with nuclides as you go, you are most of the way to doing a Relative Efficiency analysis
- To get to the tool, select the "**Isotopics from peaks**" menu item from the **Tools** menu, and a new tab will be shown at the bottom

ISOTOPICS FROM PEAKS: OVERVIEW OF USING THE TOOL





Take a look at text results

Fiddle with settings until Rel Eff chart looks good

Select which peaks you want, until Rel Eff chart looks okay

You want this chart/fit to look “good”

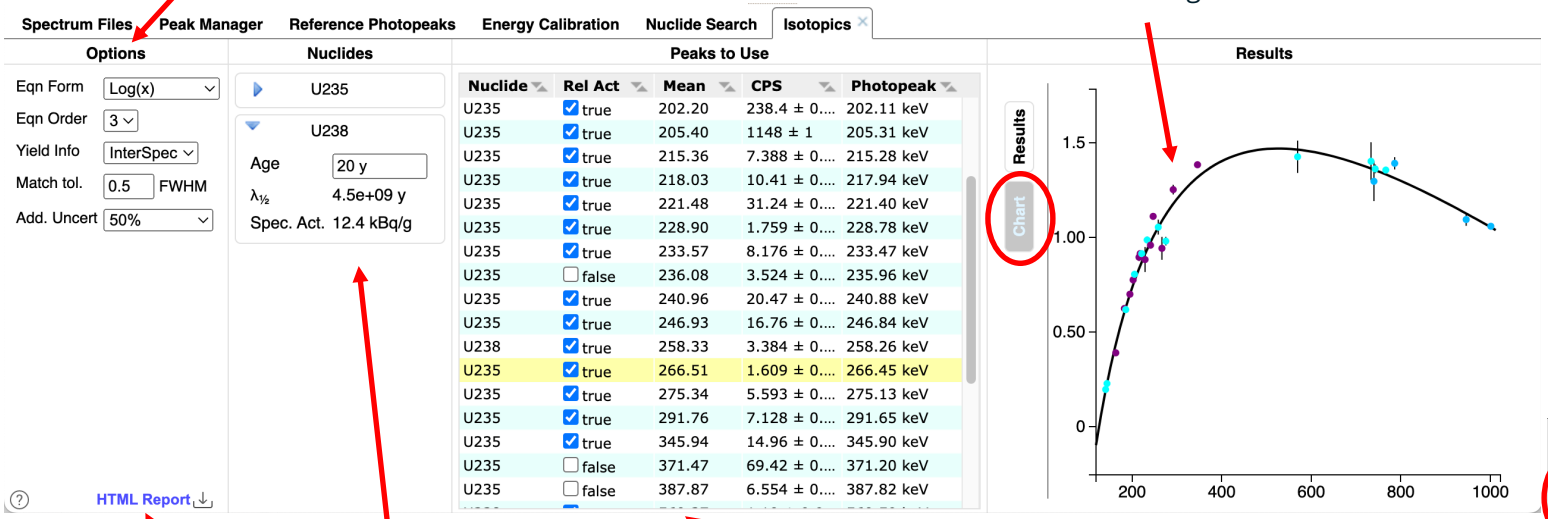
ISOTOPICS FROM PEAKS: PARTS OF THE TOOL

Calculation Options

The current Relative Efficiency curve fit

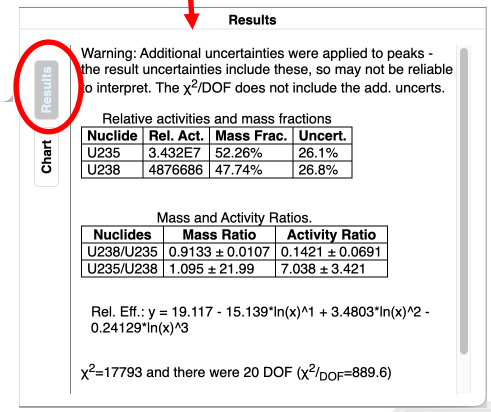
Fit is automatically updated as you change settings, add/remove peaks, etc

Your goal is to make this fit to look good!



The text results of fit

(the "truth" enrichment for this data is 52.49%)



Export a HTML summary

The nuclides for peaks you have selected to use

Basic nuclide info is listed

You can set ages for nuclides that change as they age

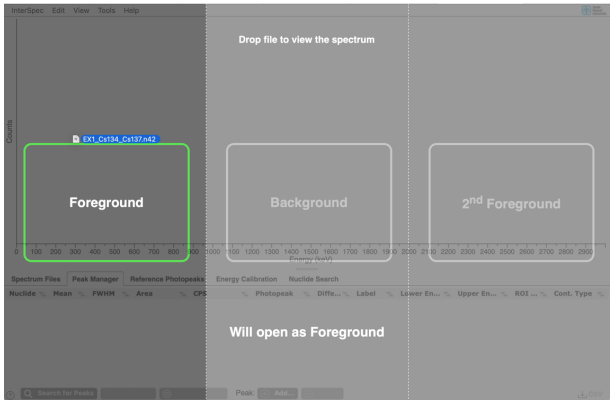
The peaks you have fit in the spectrum

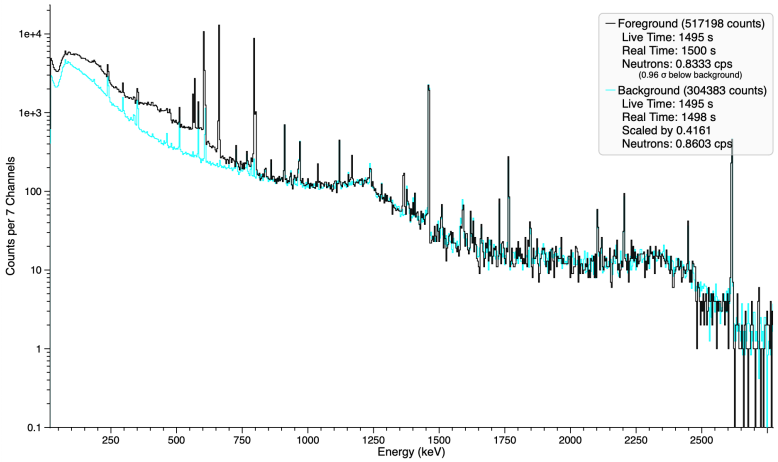
You can choose to use or not-use each peak for the Rel. Eff. analysis

You can also directly edit nuclide info in this table

LETS DO AN EXAMPLE CALCULATION FROM SCRATCH

- Before jumping into details, lets do a simple Relative Efficiency calculation
  - Please load the accompanying "`EX1_Cs134_Cs137.n42`" file into InterSpec, by dragging-and-dropping the file onto the app



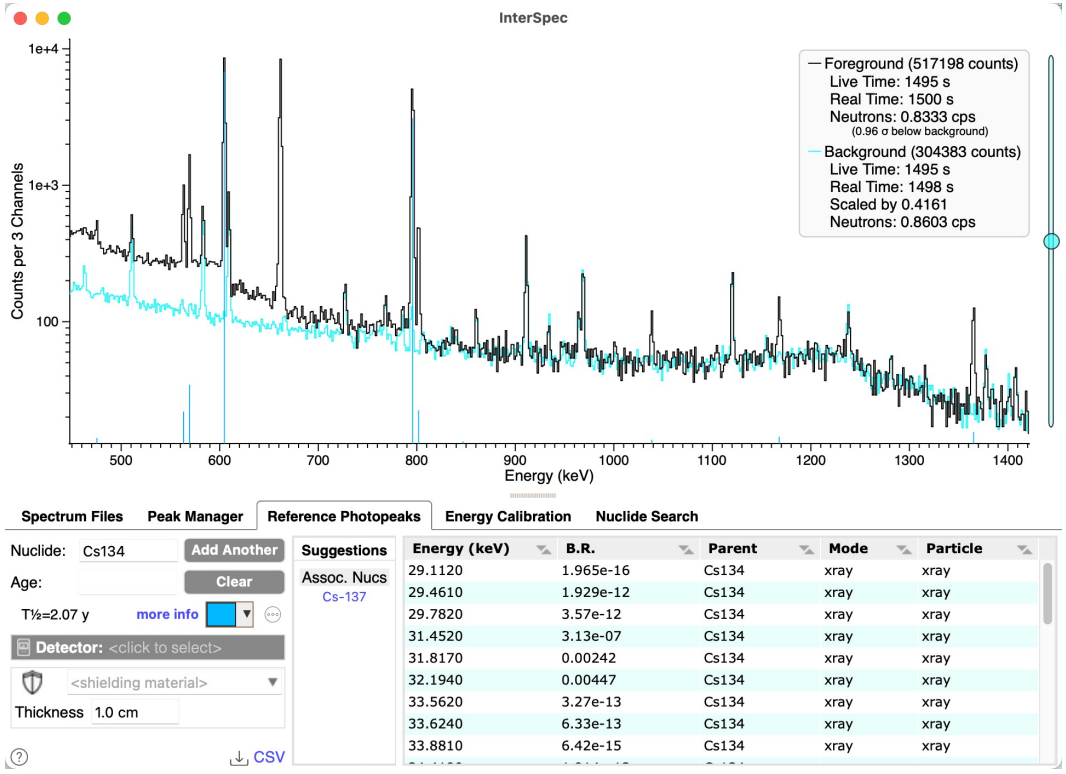


- We have been asked to provide the activity ratio of Cs134 to Cs137
- We are told the sample was an environmental sample to track fallout, but no detector specifications, or source distance/geometry is provided

EXAMPLE CALCULATION: FITTING PEAKS

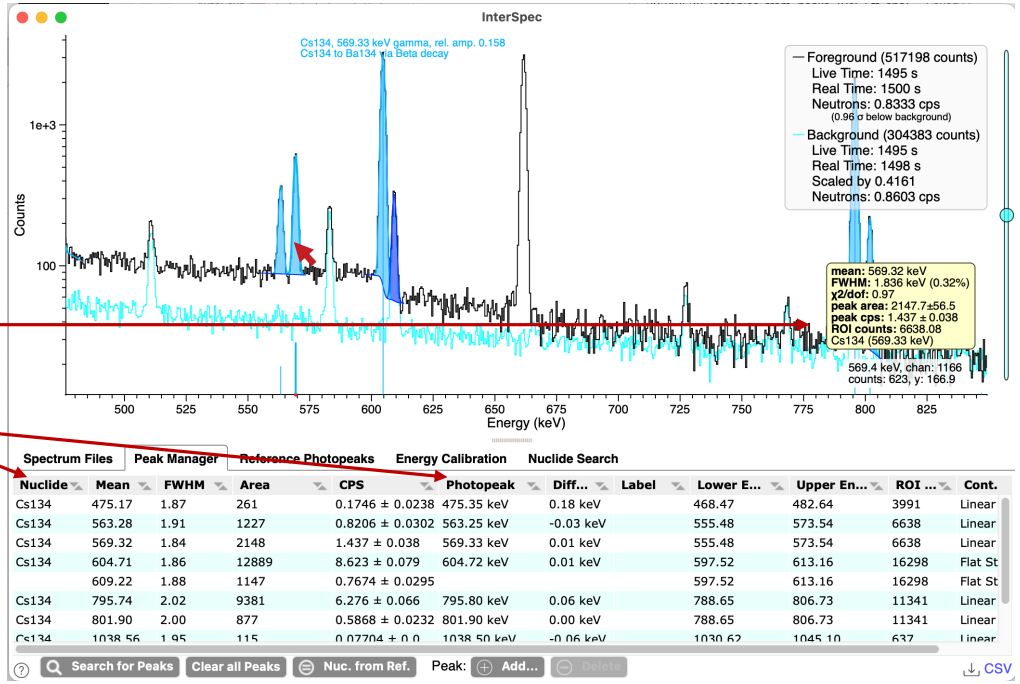
Lets fit peaks for the nuclides of interest – associating nuclides with the peaks as we go

- The easiest way to associate nuclides with peaks, is to show the reference line, ***before*** fitting the peaks
  - On “Reference Photopeak” tab, type in “Cs134”
- Then just double-click on the spectrum near each Cs134 line



EXAMPLE CALCULATION: FITTING PEAKS (CONT)

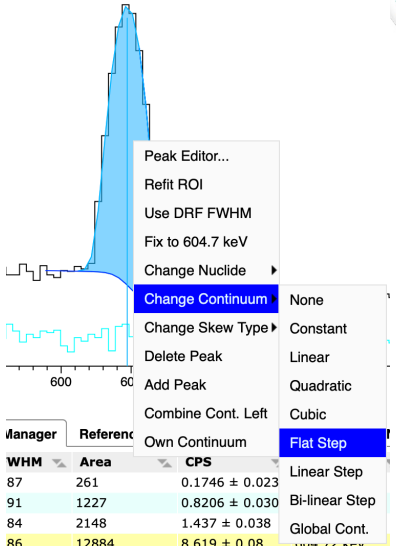
- As you fit peaks, the peaks, you can tell the nuclide association is made by:
  - The peaks will take on the color of the reference photopeak lines
  - You can mouse-over a peak, and a yellow information box will appear
  - The “Peak Manager” tab will list the nuclide and gamma energies associated
- You can also change nuclide association directly in the table, or by right-clicking on the peak and selecting “Peak Editor” or “Change Nuclide”

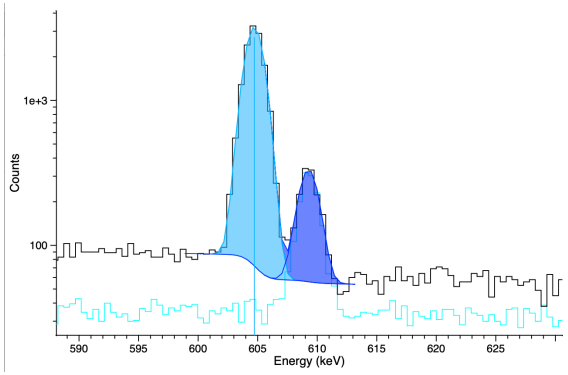


Nuclide	Mean	FWHM	Area	CPS	Photopeak	Diff...	Label	Lower E...	Upper En...	ROI ...	Cont.
Cs134	475.17	1.87	261	0.1746 ± 0.0238	475.35 keV	0.18 keV		468.47	482.64	3991	Linear
Cs134	563.28	1.91	1227	0.8206 ± 0.0302	563.25 keV	-0.03 keV		555.48	573.54	6638	Linear
Cs134	569.32	1.84	2148	1.437 ± 0.038	569.33 keV	0.01 keV		555.48	573.54	6638	Linear
Cs134	604.71	1.86	12889	8.623 ± 0.079	604.72 keV	0.01 keV		597.52	613.16	16298	Flat St
	609.22	1.88	1147	0.7674 ± 0.0295				597.52	613.16	16298	Flat St
Cs134	795.74	2.02	9381	6.276 ± 0.066	795.80 keV	0.06 keV		788.65	806.73	11341	Linear
Cs134	801.90	2.00	877	0.5868 ± 0.0232	801.90 keV	0.00 keV		788.65	806.73	11341	Linear
Cs134	1038.56	1.95	115	0.07704 ± 0.0	1038.50 keV	-0.06 keV		1030.62	1045.10	637	Linear

EXAMPLE CALCULATION: FITTING PEAKS (CONT)

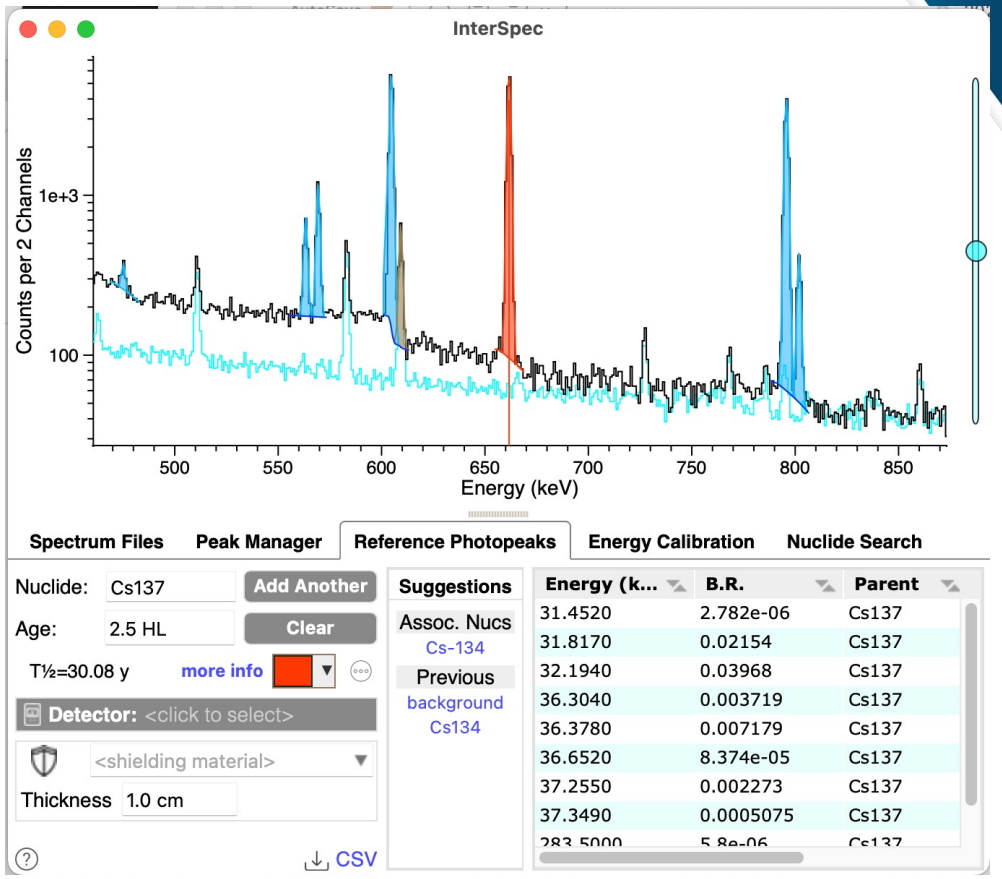
- The 605 keV Cs134 peak has a background peak near it – so we can also fit it (by double clicking on it) to improve fit accuracy.
  - There is no need to associate a nuclide with this 609 keV (Ra226 series) peak
  - If InterSpec doesn't assign both peaks to have the same continuum – right-click on the peak, and select to combine continuum
  - A "step" continuum would also work better, so you could right-click on the peaks, and change the continuum to flat-step (it's a small effect, so you don't actually need to do this)





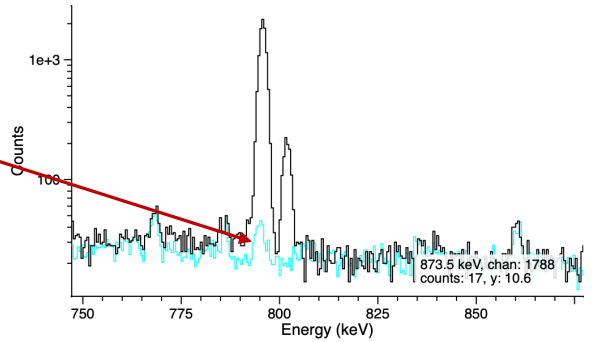
EXAMPLE CALCULATION: FITTING PEAKS (CONT)

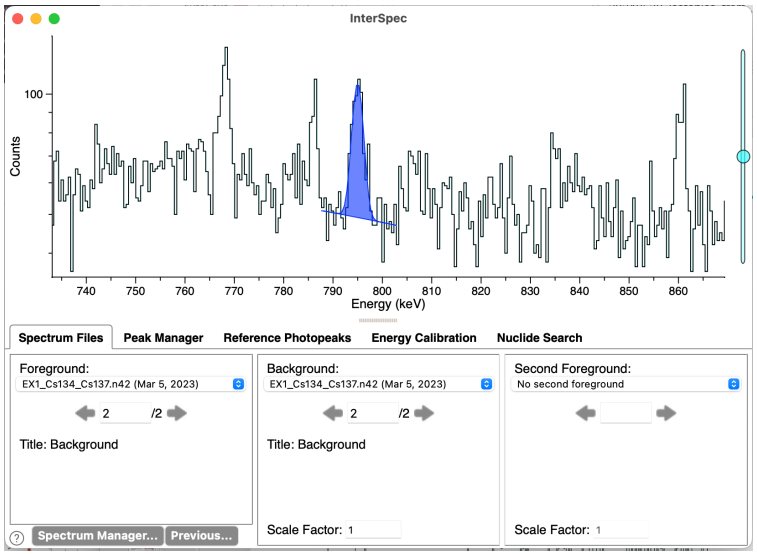
- Now fit for the Cs137 peak by displaying the Cs137 Reference Photopeak line, and then double-clicking on the peak



EXAMPLE CALCULATION: BACKGROUND CORRECTION

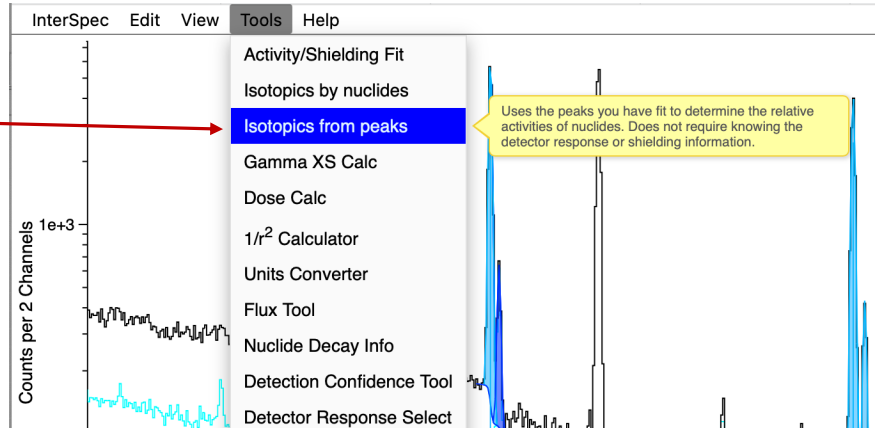
- The 796 keV Cs134 peak has a small background interference.
  Its small enough to ignore – but we can also correct for it
- On the “Spectrum Files” tab, under the “Foreground” section, change to sample 2, the background spectrum, and fit for this peak
  - No need to associate a nuclide
  - You could also do an auto fit for all peaks in the background spectrum using the “Search for Peaks” button on the “Peak Manager” tab, and fit, but you don’t need to
  - You can switch back to foreground on the “Spectrum Files” tab, and all your peaks and everything will still be there

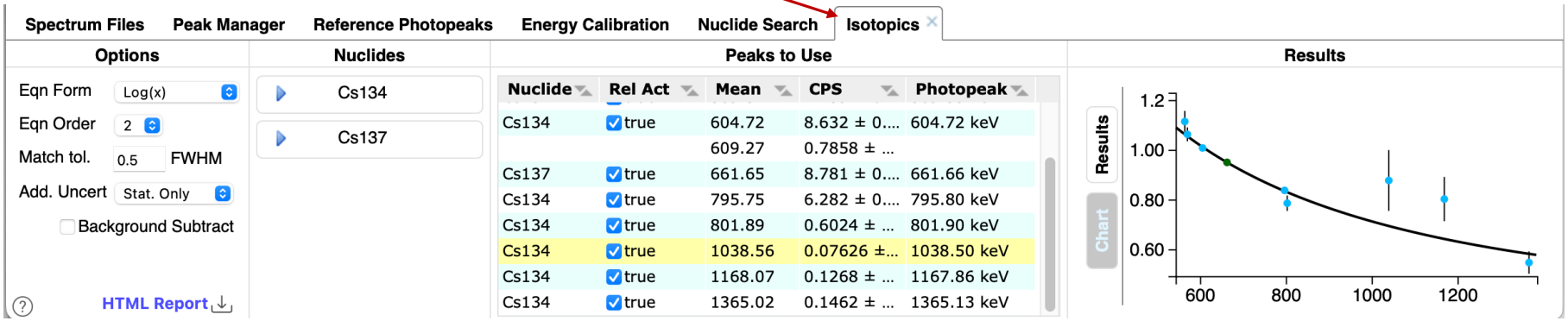




EXAMPLE CALCULATION: SHOWING THE TOOL

- From the **Tools** menu, select the "**Isotopics from peaks**" item
- A new "Isotopics" tab will be added to the tabs at the bottom of the app



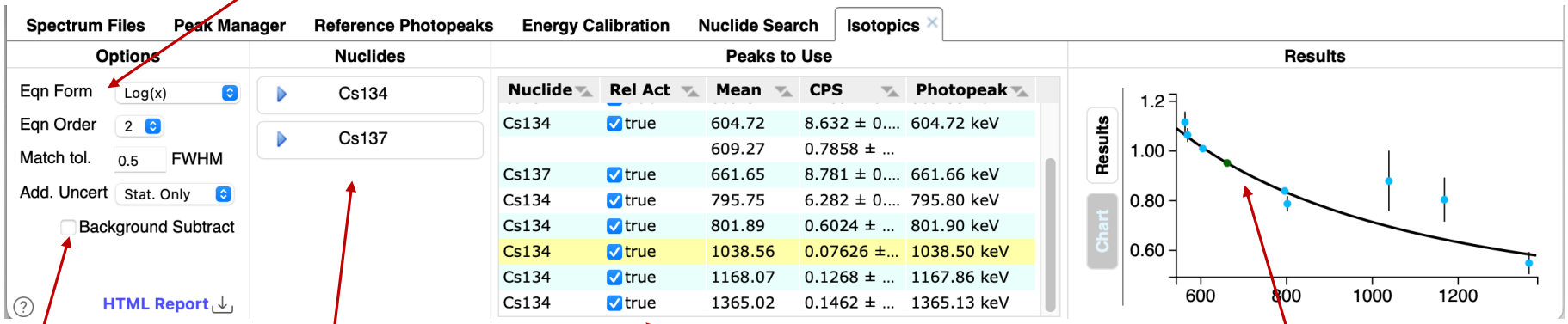


You could have opened this tool up before fitting peaks, or loading a file, it doesn't effect anything else, or get in the way – the primary InterSpec author leaves it open all the time

EXAMPLE CALCULATION: ADJUSTING OPTIONS

- Our goal is to change options to make the Relative Efficiency chart *look* good

Select the functional form and order to fit data to - for this case doesn't matter much



The nuclides for peaks you have selected to use

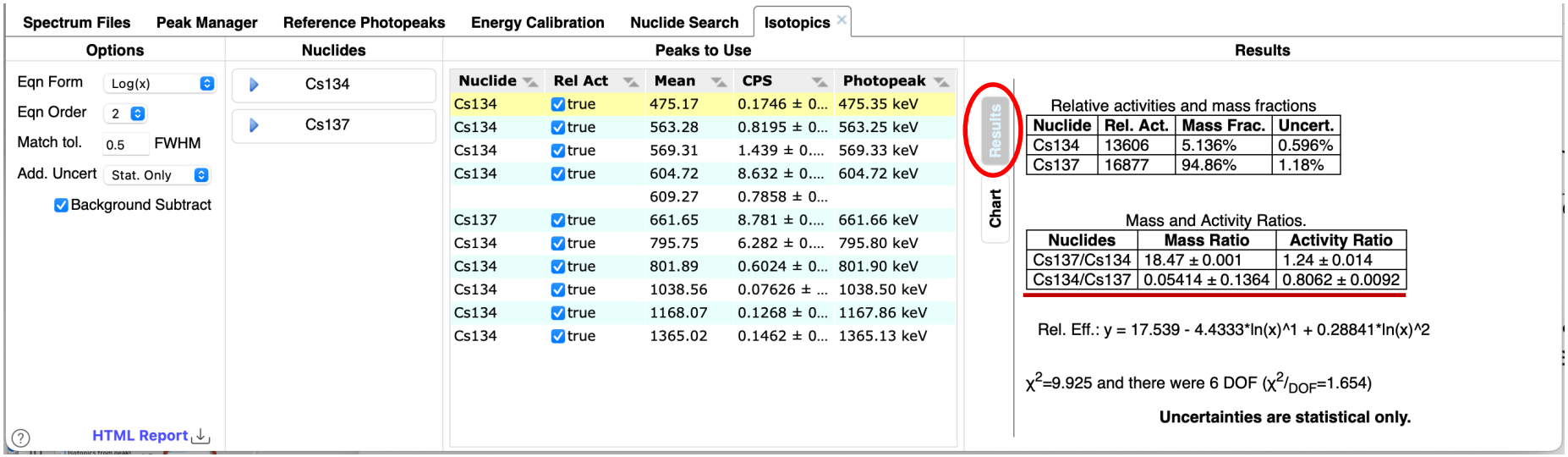
If you fit the background 795 keV peak, select this option

Select which peaks you want to use here

The data, at the fit relative activities, compared with the best-fit line (*mouse over data-points for more info*)

EXAMPLE CALCULATION: RESULTS

Once you are happy with how the Relative Efficiency plot looks, click on the “Results” tab



The ratio of Cs134 to Cs137 activities is 0.806 ± 0.009 – true answer is 0.8

OPTION DETAILS: EQUATION FORM

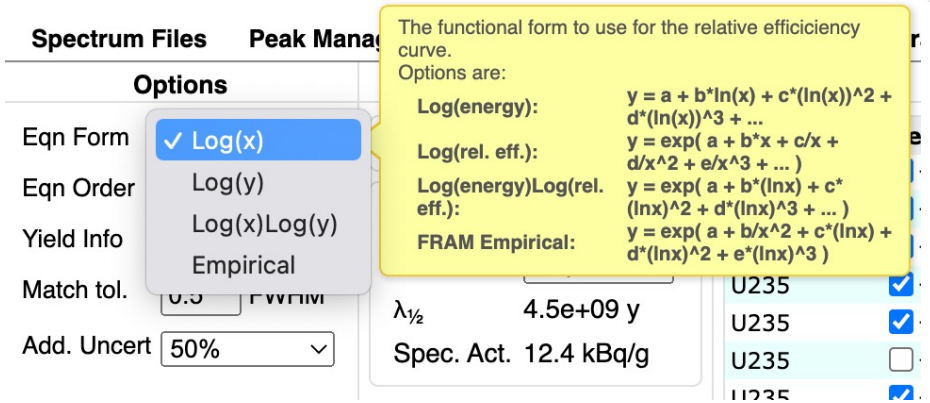
There are a few different functional forms of the Relative Efficiency curve you can choose from:

$$y(x) = c_0 + c_1 \ln^1(x) + c_2 ln^2(x) + c_3 \ln^3(x) + \ \ldots$$

$$y(x) = \exp(c_0 + c_1 x + \frac{c_2}{x} + \frac{c_3}{x^2} + \frac{c_4}{x^3} + \ \ldots)$$

$$y(x) = \exp(c_0 + c_1 \ln^1(x) + c_2 \ln^2(x) + \ \ldots)$$

$$y(x) = \exp(c_0 + \frac{c_1}{x^2} + c_2 \ln^1(x) + c_3 \ln^2(x) + \ \ldots)$$



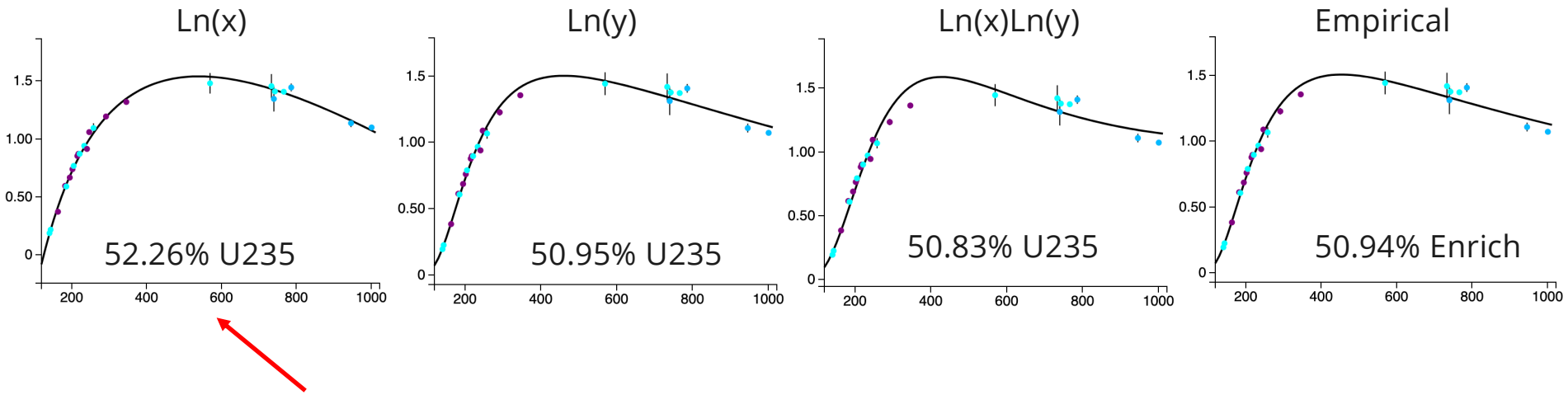
Enable “Help” → “Options” → “Show tooltips” to see descriptions like above yellow box

Discussion: there isn’t a ton of motivation for these functional forms, other than they are kinda like attenuation convoluted with detector response functions – pick the one that *looks* best.

OPTION DETAILS: EQUATION FORM

Generally you want to choose the *Eqn Form* that *looks* the best.

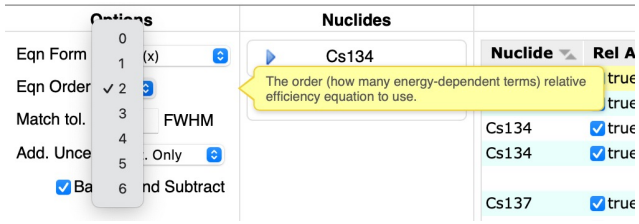
Here is a Uranium example with known enrichment of 52.49%



Subjectively looks best
– data *looks* to follow line best

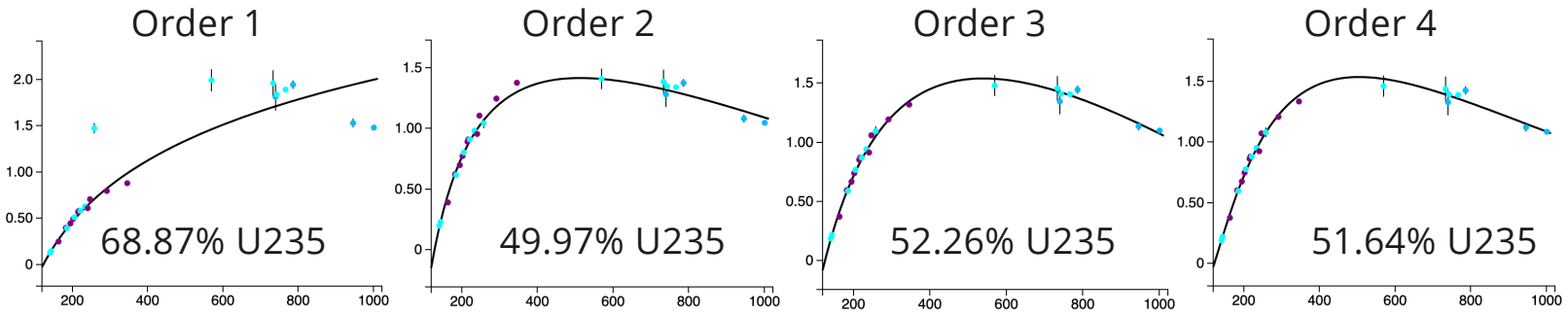
None of these equation forms account for x-ray attenuation edge – so if your source or shielding is Pb, W, U, Pu, etc – do not use peaks both above and below the k-edge (e.g., ~118 keV for U, 90 keV for Bi, etc)

OPTION DETAILS: EQUATION ORDER



- *Eqn Order* is the number of energy dependent terms in the Relative Efficiency equation.
- Generally: use the fewest number of terms that make the Relative Efficiency curve fit *look* good.

Too many terms, especially with smaller number of peaks, can allow the results to be non-physical.

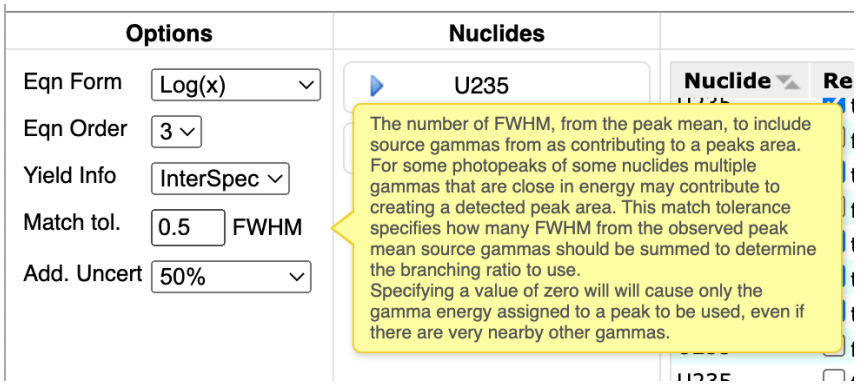


Known Enrichment: 52.49%

(all fits use ln(x) Eqn Form)

OPTION DETAILS: MATCH TOLERANCE

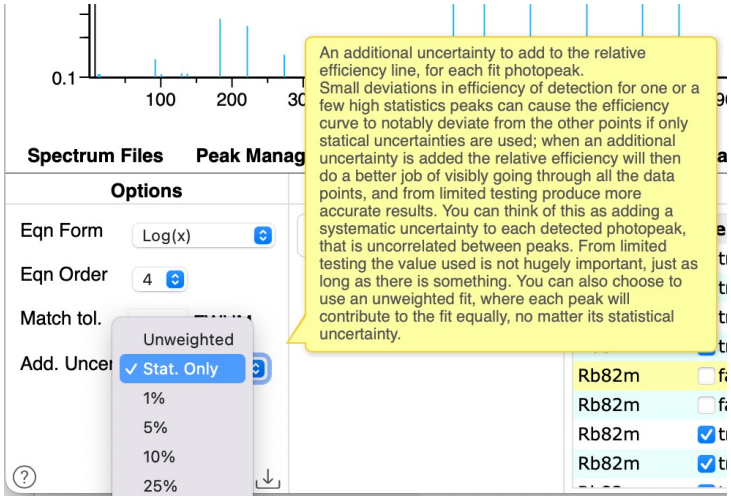
Recommendation: leave this options at the default 0.5



- This option allows InterSpec to correctly account for multiple gammas, maybe from multiple nuclides, contributing to a single observable peak.
- The gamma (i.e., the nuclide and specific energy gamma) you associate with a peak will always be used in the relative efficiency calculation.
- However, the fit peak may have contributions from nearby gammas of either the same nuclide, or any of the other nuclides in the problem.
- The “Match tol.” option defines how many full-width-at-half-maximum a nuclides gamma, from any of the nuclides being used for the Rel. Eff. calculation, can be away from the peak mean, and still considered to contribute to the peak.
- A value of “0” makes it so only the gamma you assigned to the peak will be used.

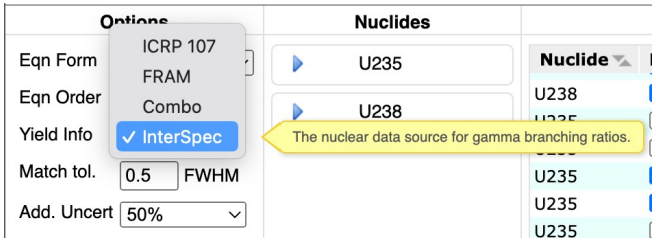
OPTION DETAILS: ADD. UNCERT.

Recommendation: leave this options as “Stat. Only”, except for Uranium problems



- This option is useful for high-statistics samples where systematic errors in fitting peak amplitude (e.g., imperfect continuum or skew modeling) are larger than statistical uncertainties of the fit.
- For high-statistics peaks, fitting an amplitude that is only a very tiny percentage off can be many, many statistical sigma off, so a peak may disproportionately impact the Relative Efficiency curve fit (e.g., cause a “kink”, or discontinuity in it).
- This option allows accounting for this effect – at the cost of making the computed uncertainties no longer easily interpretable
- In limited testing, this option is mostly only **useful for Uranium problems**.
  - For Uranium problems, from a limited test set, adding an additional uncertainty increased the accuracy of the computed answer, in comparison to known values
  - The value used (e.g., 1%, 5%, 10%, etc) doesn’t seem to matter much, just as long as *some* additional uncertainty is used – but using 50% did perform slightly better than other values
  - To quote an uncertainty in enrichment, a reasonable strategy is to take the difference between answers found with “Stat. Only” and with “50%” additional uncertainty, and add it in quadrature to to the statistical error when using “Stat. Only”

OPTION DETAILS: URANIUM YIELD INFO



This option only appears for problems with Uranium in them.

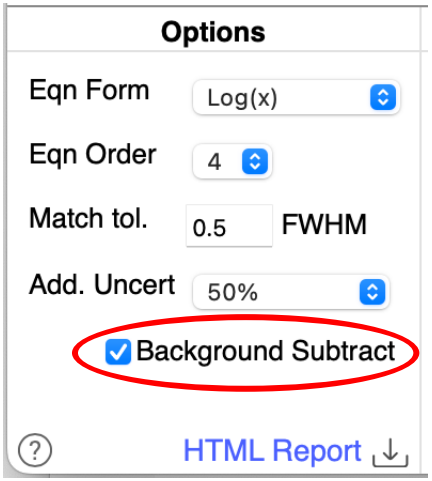
It allows selecting the Branching Ratio source data for Uranium.

ICRP 107, FRAM, and “Combo” are limited to the major ”clean” uranium lines (i.e., not all your peaks may be useable)

“Combo” is a combination of ICRP 107, FRAM, and Sandia branching ratios, that seems to work well

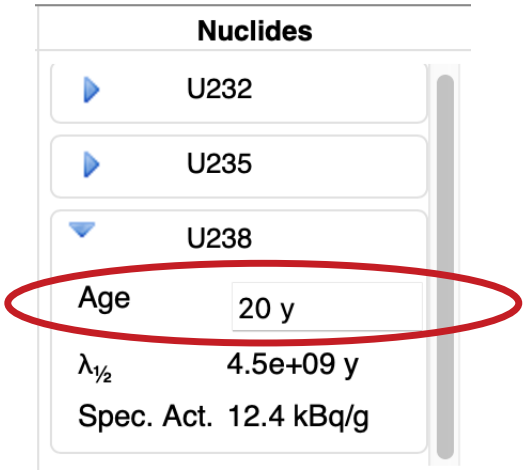
Recommend: use “InterSpec”

OPTION DETAILS: BACKGROUND SUBTRACT



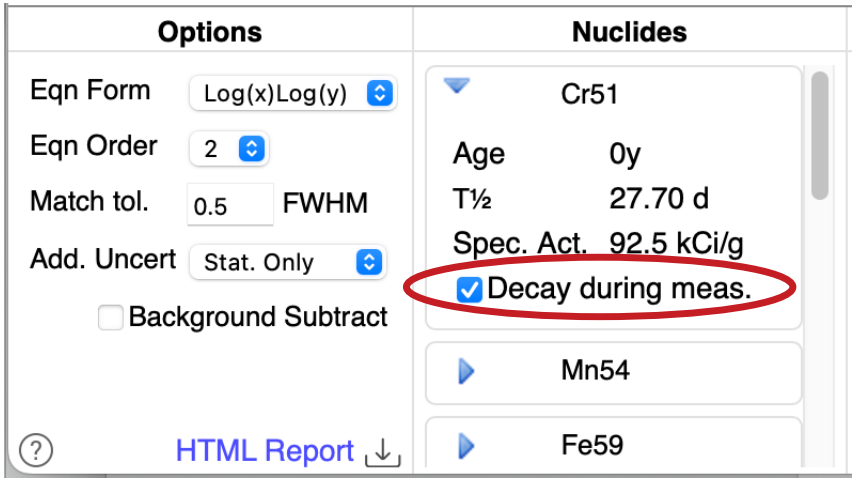
- This option subtracts background peak areas from foreground peak areas.
- You must first load the background spectrum as a foreground and fit the relevant peaks in it (nuclide associations do not need to be made), then use the “Spectrum Files” tab to change the spectrum to the background, then load/select spectrum of interest as the foreground spectrum
- This is not a channel-by-channel subtraction, but instead it matches peaks in the foreground to peaks already fit in the background, and subtracts their area and variances
  - A matching tolerance of about 0.5 FWHM between foreground and background peaks is used
- This option is only shown if there is a background spectrum file loaded, with peaks fit.

OPTION DETAIL: NUCLIDE AGE



You can set a nuclides age by expanding its entry in the “Nuclides” column – if it’s a nuclide whose signature changes as it ages

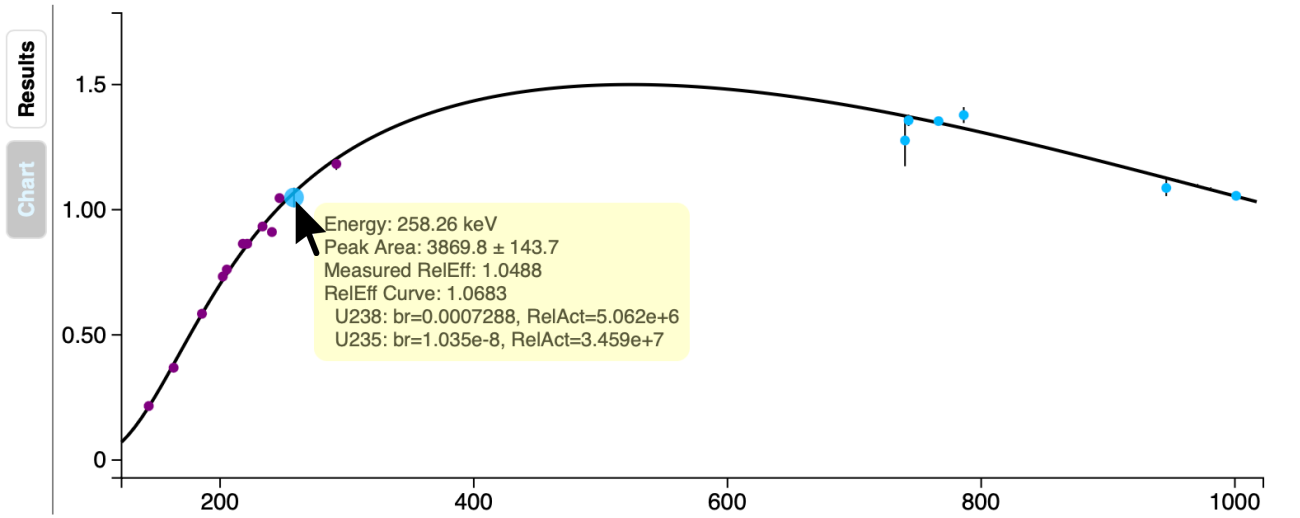
OPTION DETAIL: ACCOUNTING FOR AGING DURING MEASUREMENT



If the measurement time is longer than 0.5% of the nuclide half-life, the nuclides activity will, by default, be corrected for decay during measurement.

- The quoted relative activity is at the beginning of the measurement
- Decay product ingrowth and decay is all properly accounted for

ADDITIONAL FEATURES: REL. EFF. CHART

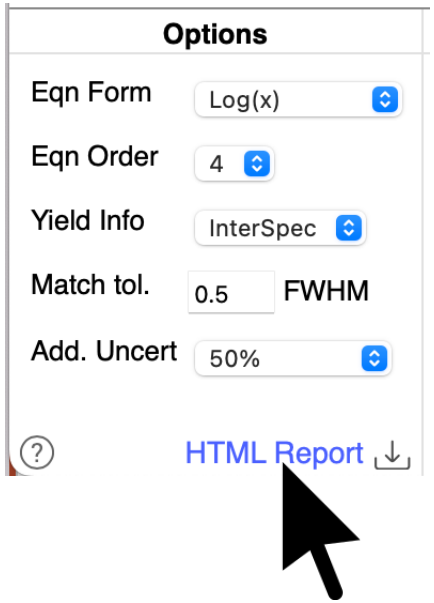


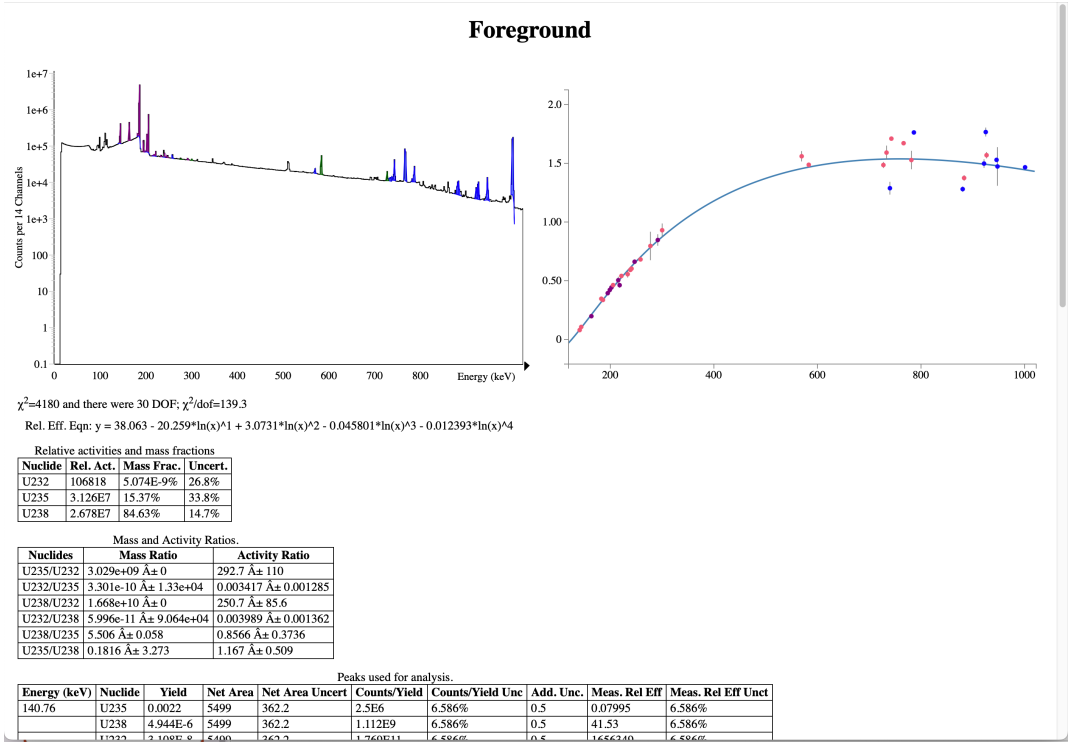
The color of the points on the chart is determined by the majority nuclide contributing to the peak.

If no nuclide contributes over 50% of the peak area, then will be colored the default dark blue peak color.

"Mousing" over data points on Relative Efficiency chart will show a box that gives further information about that data point.

ADDITIONAL FEATURES: HTML REPORT



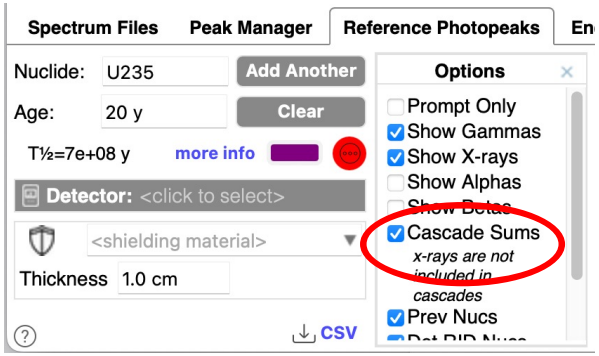


You can export a HTML report that contains an interactive spectrum and relative efficiency charts, as well as additional information not in the InterSpec GUI.

Also if you export to N42-2012 file, all your peaks, and Rel. Eff. settings will be saved in it

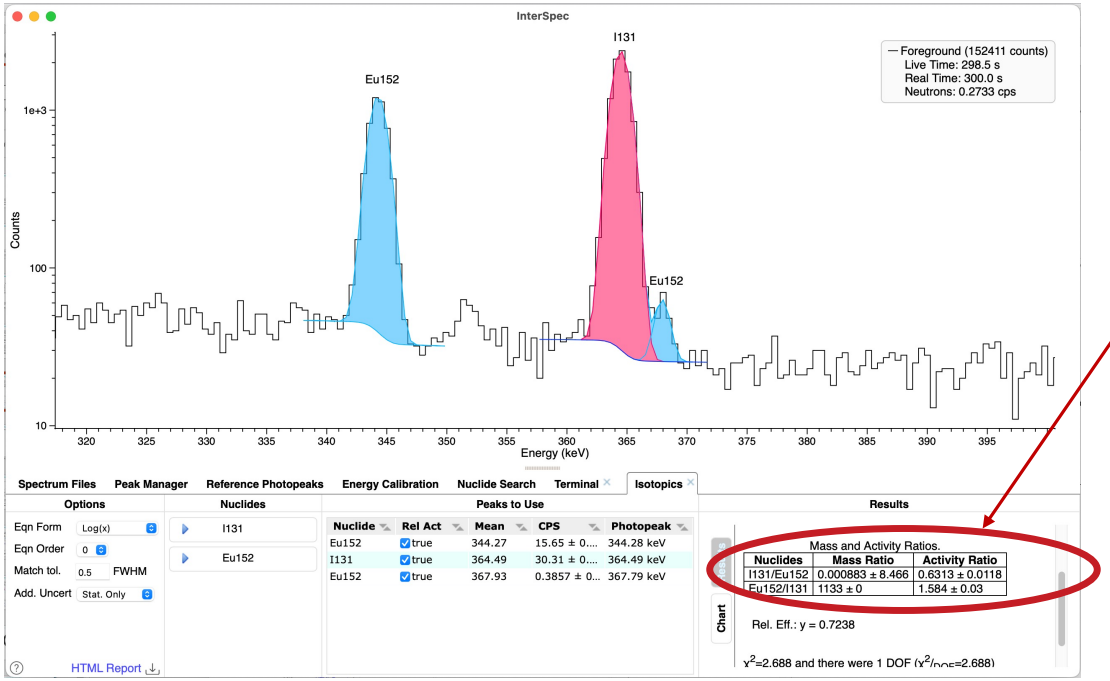
SOME NOTES:

- Interferences, i.e. multiple source nuclides contributing to the same peak, are accounted for in the calculations, as long you are using at least one peak with the interfering nuclide assigned, somewhere in the spectrum
- Coincidence summing is not accounted for – so don't use for cascade decay affected gammas, if the measurement was close to on-contact (≲5 cm)
  - You can check for cascade sums using the **Reference Photopeak** tool
- The source nuclides must be homogenous
  - Only use peaks for nuclides of interest – if you fit some background peaks that don't interfere with nuclides in question, de-select them from being used
- The shielding of the source(s) must be homogeneous
- We are looking for a few weeks of funding to update tool for k-edge, and other improvements…



NEARBY PEAKS EXAMPLE

- People will commonly estimate two nuclides relative activities, using two near-by peaks (where the attenuation and detection efficiencies are about the same).
  - This requires looking up branching ratios, and multiplying and stuff – so instead we can use this tool, with a “Order 0” equation



In “nearby_example_HPGe.n42” we have Eu152 (15 uCi) + I131 (10 uCi).

We get activity ratio (Eu152/I131):
1.58±0.03 (stat. only)

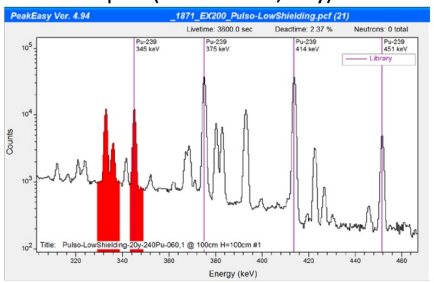
NEARBY PEAKS - PU

From Mike Enghauser's "Plutonium gamma spectroscopy training" (SAND2018-3369: https://www.osti.gov/servlets/purl/1525593):

332/336 keV region of interest

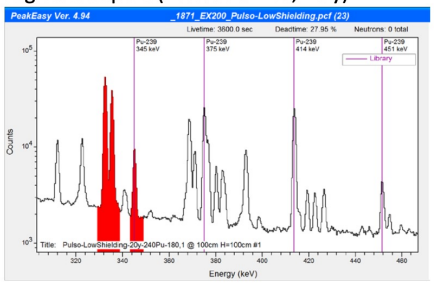
Sandia National Laboratories

Low-burnup Pu (6.0% Pu-240, 20 y)



Energy (keV)	Phot/Decay	Nuclide
332.4	1.49E-06	Am-241
332.4	2.94E-07	U-237
332.8	4.94E-06	Pu-239

High-burnup Pu (18.0% Pu-240, 20 y)

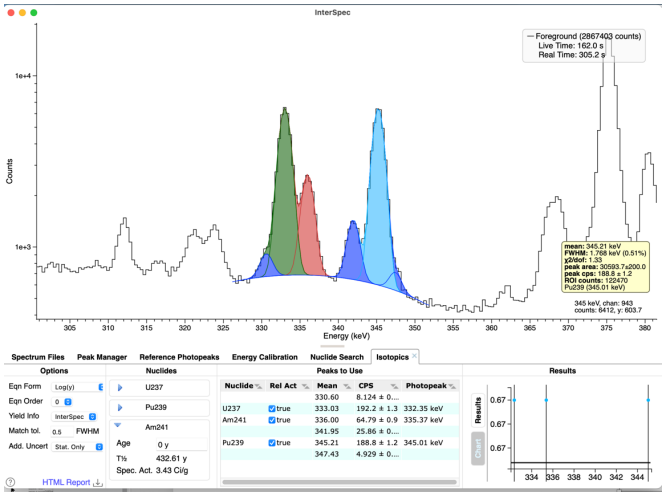


Energy (keV)	Phot/Decay	Nuclide
335.4	4.96E-06	Am-241
335.4	2.33E-08	U-237
336.1	1.12E-06	Pu-239
345.0	**6.06E-06**	**Pu-239**

- The 345.0 keV emission from Pu-239 is interference free.
- If good counting statistics, it is possible to strip the Pu-239 contribution from 332 & 336 keV complexes & solve for the Pu-241/U-237 & Am-241 peak areas to determine the time since chemical separation.

Which is great, but for those of us with a high-barrier to setting this correction up in Excel, and looking all the information up… we can just use InterSpec

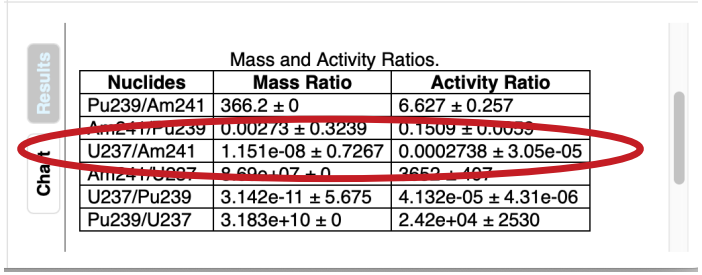
NEARBY PEAKS – PU (CONT – METHOD 1 (HARDER))



Fit peaks – assigning 333 keV to U237, 336 keV to Am241, and 345 to Pu239.

Then select “Eqn Order” 0.

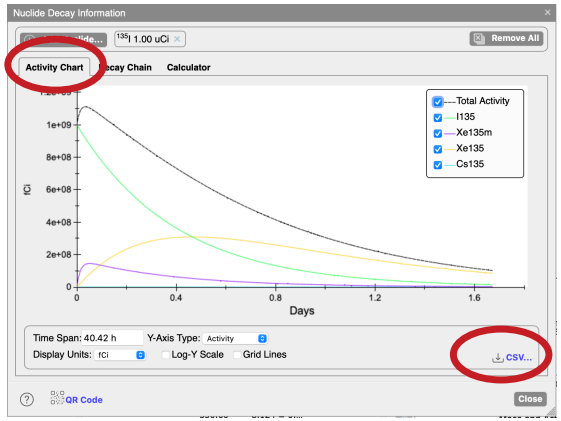
InterSpec will take care of interferences

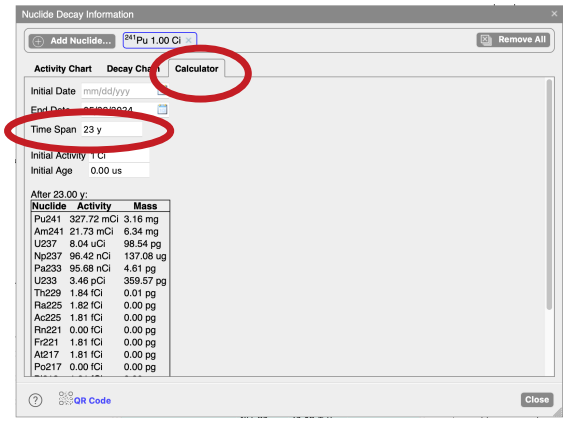


Nuclides	Mass Ratio	Activity Ratio
Pu239/Am241	366.2 ± 0	6.627 ± 0.257
Am241/Pu239	0.00273 ± 0.3239	0.1509 ± 0.0035
U237/Am241	1.151e-08 ± 0.7267	0.0002738 ± 3.05e-05
Am241/U237	8.69e+07 ± 0	3652 ± 407
U237/Pu239	3.142e-11 ± 5.675	4.132e-05 ± 4.31e-06
Pu239/U237	3.183e+10 ± 0	2.42e+04 ± 2530

Then, rather than manually solving decay equations, use the “Nuclide Decay Info” tool.
Either:

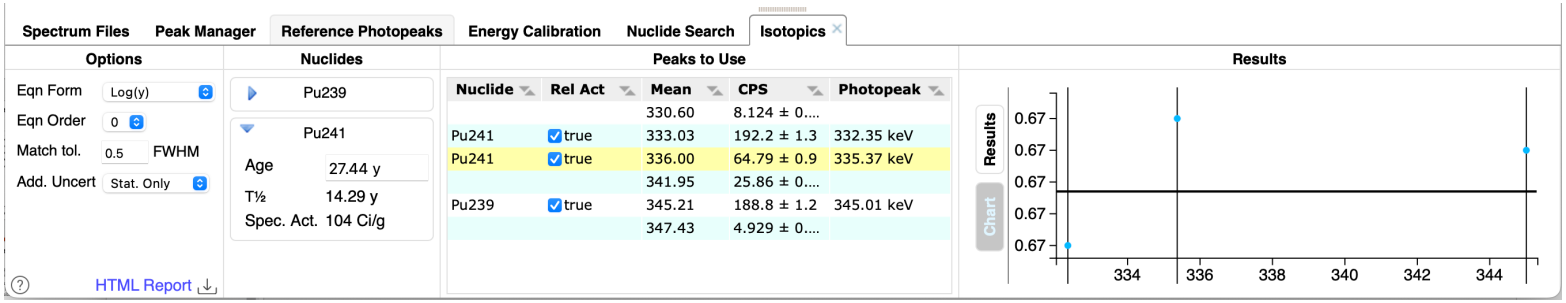
- Export a CSV from the “Activity Chart” tab, for activity over time
- Hunting for the age using the calculator

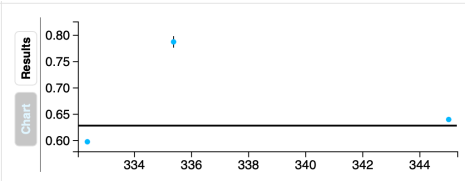




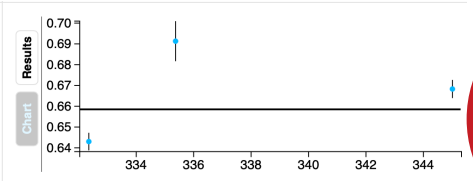
NEARBY PEAKS – PU (CONT – METHOD 2 (EASIER))

Or, even easier, just assign the 333 keV and 336 keV peaks to Pu241, and manually adjust the Pu241 age until the chart looks the best

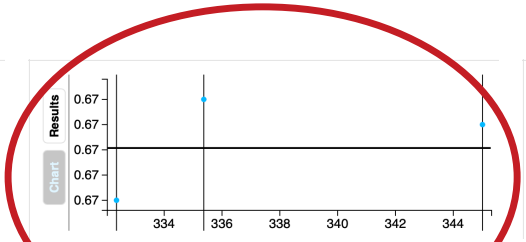




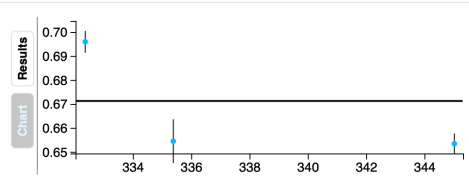
Using Age=10 y



Using Age=20 y



Using Age=27.4 y

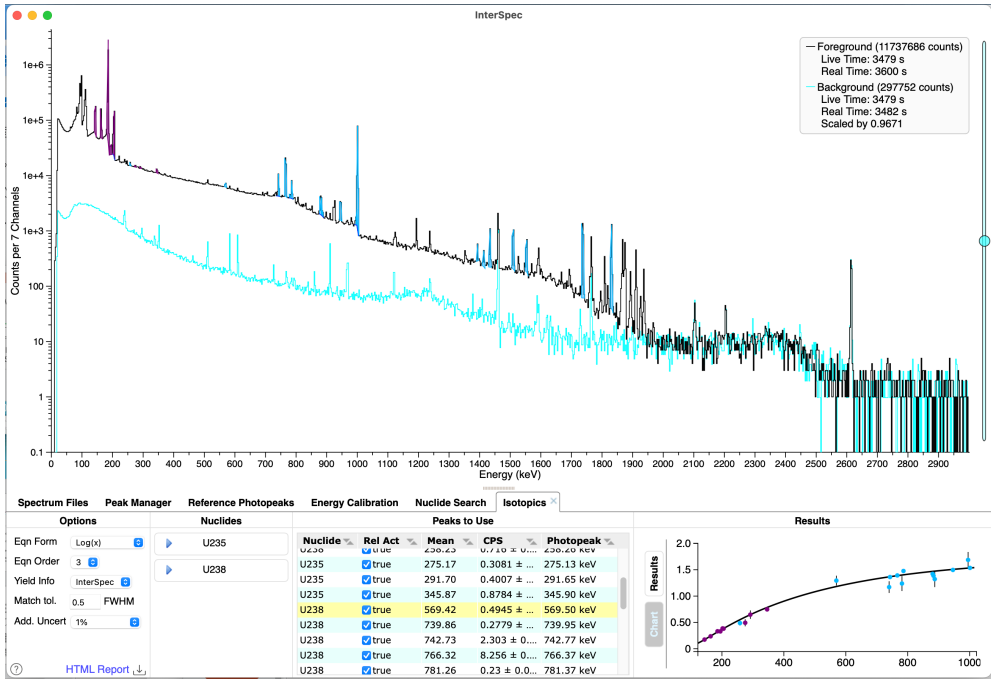


Using Age=50 y

Note: normally in InterSpec you will always assign peaks to the ultimate parent nuclide, unless there is some contamination or disruption of the decay chain – if you find yourself assigning peaks to the progeny nuclides, consider if there is an easier way

URANIUM EXAMPLE

- Load example: EnrichUraniumExample(20%)_withPeaks.n42
  - Should already have U235 and U238 peaks fit.



Results

Relative activities and mass fractions

Nuclide	Rel. Act.	Mass Frac.	Uncert.
U235	1.075E7	19.26%	4.27%
U238	7.01E6	80.74%	1%

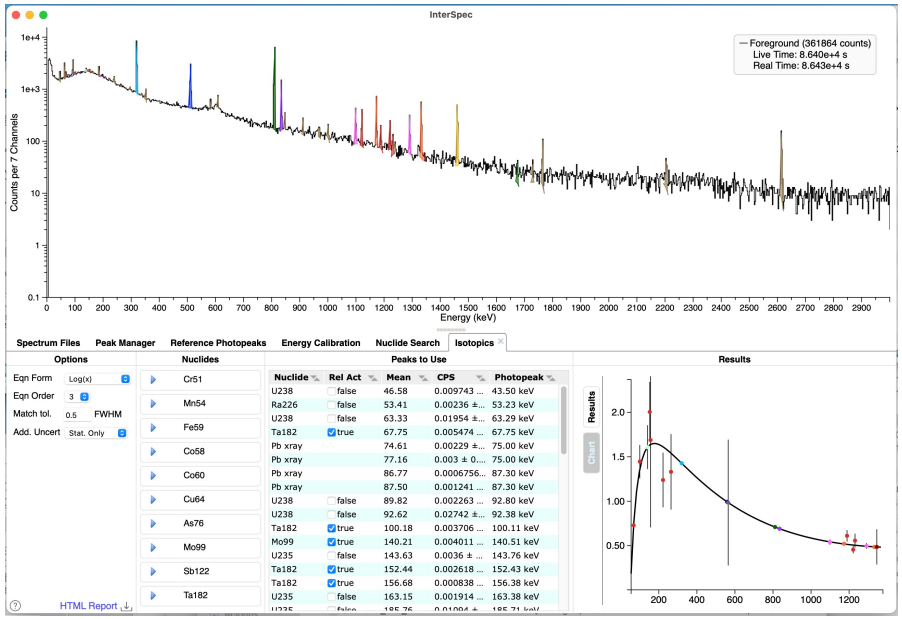
Mass and Activity Ratios.

Nuclides	Mass Ratio	Activity Ratio
U238/U235	4.191 ± 0.0053	0.652 ± 0.0338
U235/U238	0.2386 ± 0.5104	1.534 ± 0.079

Ground Truth is 20% U235 – you should get pretty close

NEUTRON ACTIVATION EXAMPLE

- A small stainless steel sample has been stored next to a Cf252 source for ~5 months, and we want to check if stated material composition, but our low-background HPGe doesn't have an efficiency calibration
  - Example file “Disk C1153a_5 (AE3204).n42” has peaks already fit for you



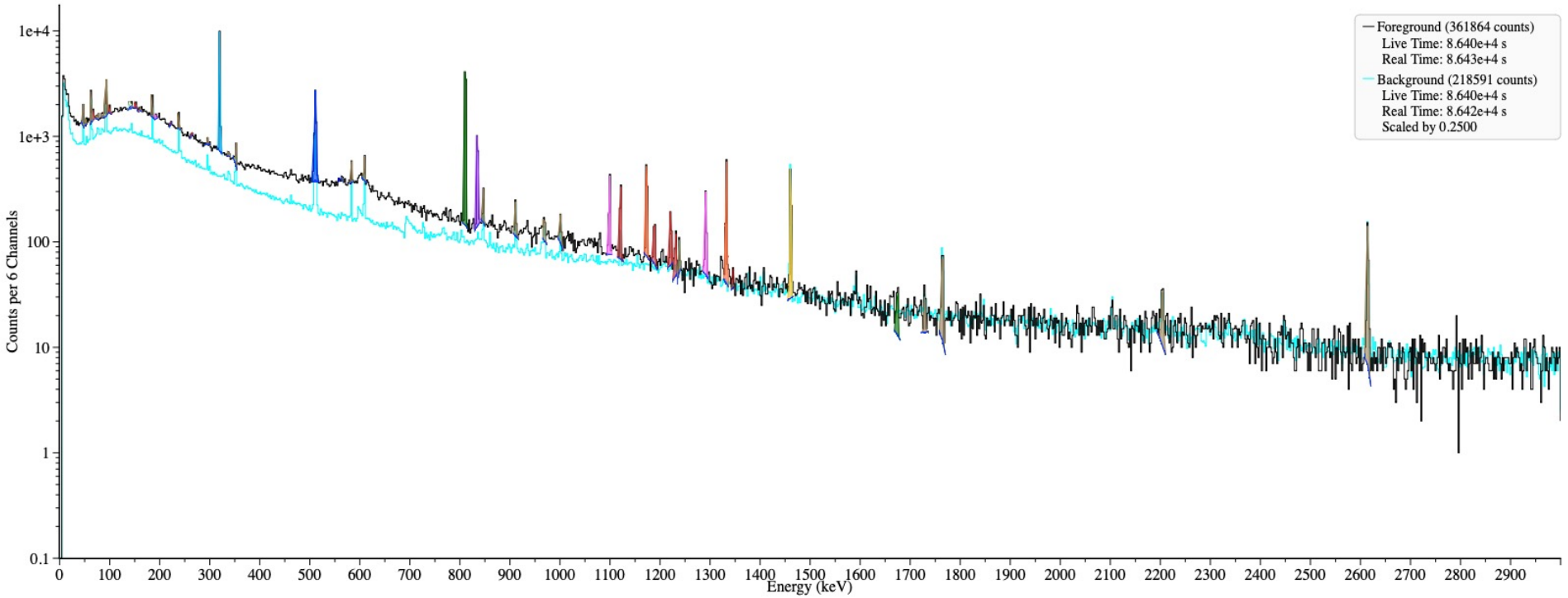
Disk C1153a_5 (AE3204)					
Nuclide	Data Rel. Act.	Stat Uncert.	Predicted Norm Act	n-sigma diff***	
Cr51	1	11%	1	N/A	
Cu64	0.209907144	40.40%	0.00963342	-1.89	
Co58	0.136509933	8.47%	0.145079	0.34	
Mn54	0.027585612	8.54%	0.0167341	-2.12	
Ta182	0.018756243	5.12%	0.0215523	0.99	
Fe59	0.018497281	7.40%	0.0152725	-1.00	
Co60	0.016437917	7.51%	0.00700066	-3.28	
Mo99	0.002532894	20.30%	0.00579295	4.25	
As76	0.00129111	11.60%	0.00481929	12.65	Arsenic content not certified
Sb122	0.001143625	69.70%	0.00431208	3.48	Antimony content not certified

*** A 10% systematic uncert. used – probably an underestimate

SOME THINGS TO WATCH OUT FOR

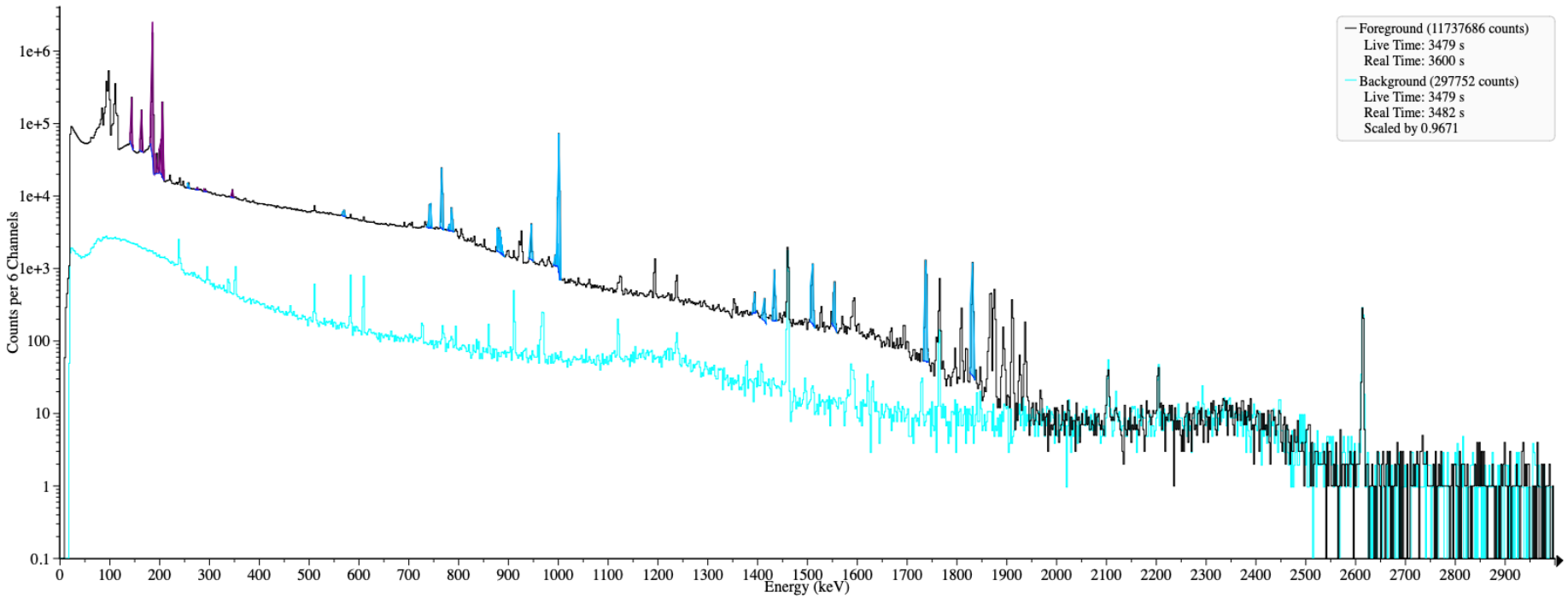
- Shielding k-edge is not currently accounted for – so only use peaks either above this edge
  - By default, peaks, below 120 keV will not be selected to be used
- If a nuclides only peak is at either the left, or right side of the other nuclides peaks – doen't trust that nuclides activity – using the relative eff. curve to extrapolate is not a good idea
  - e.x., If a spectrum only has Ba133 and Cs137, you shouldn't use Rel. Eff. analysis
- All sources should be co-located, and the shielding should be homogeneous (e.g., no holes)

ADDENDUM - SPECTRA PROVIDED – “DISK C1153A_5 (AE3204).N42”



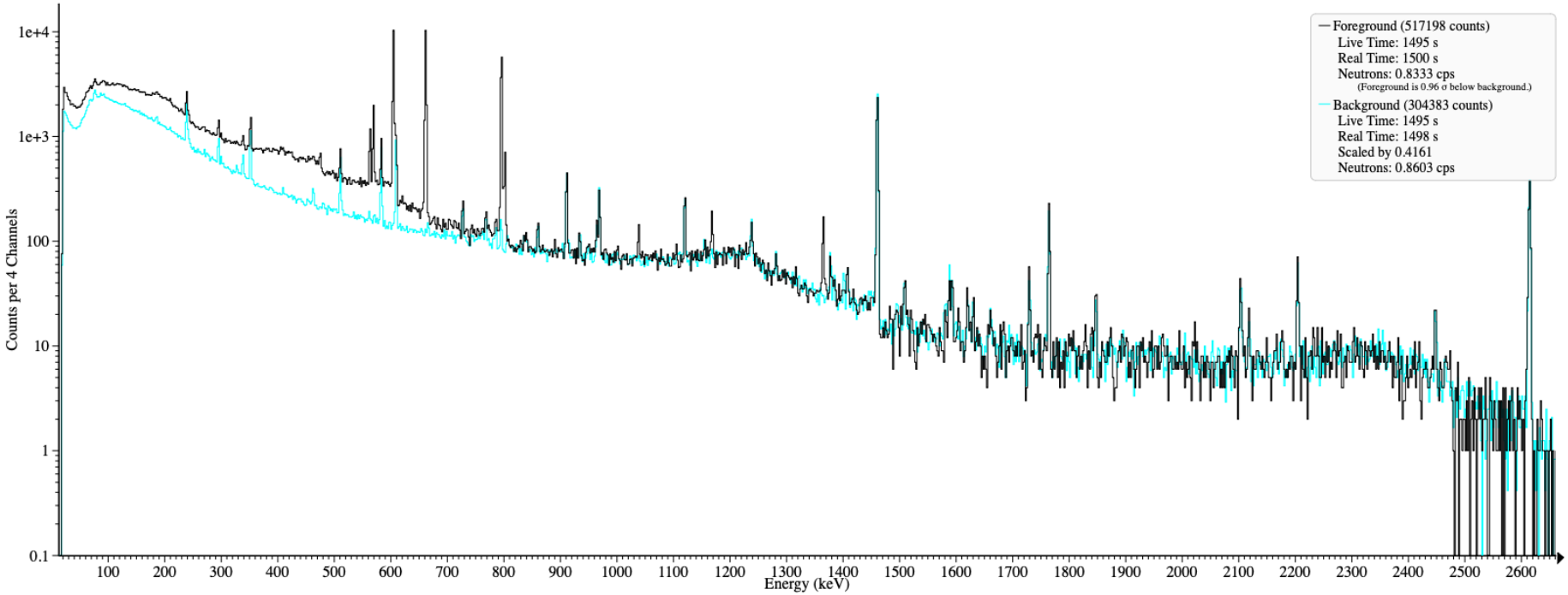
- Measured data of stainless steel disk, irradiated by neutrons for 5 months
- Spectrum file includes peak fits shown above, with nuclides associated with them

ADDENDUM - SPECTRA PROVIDED – “ENRICHURANIUMEXAMPLE(20%)_WITHPEAKS.N42”



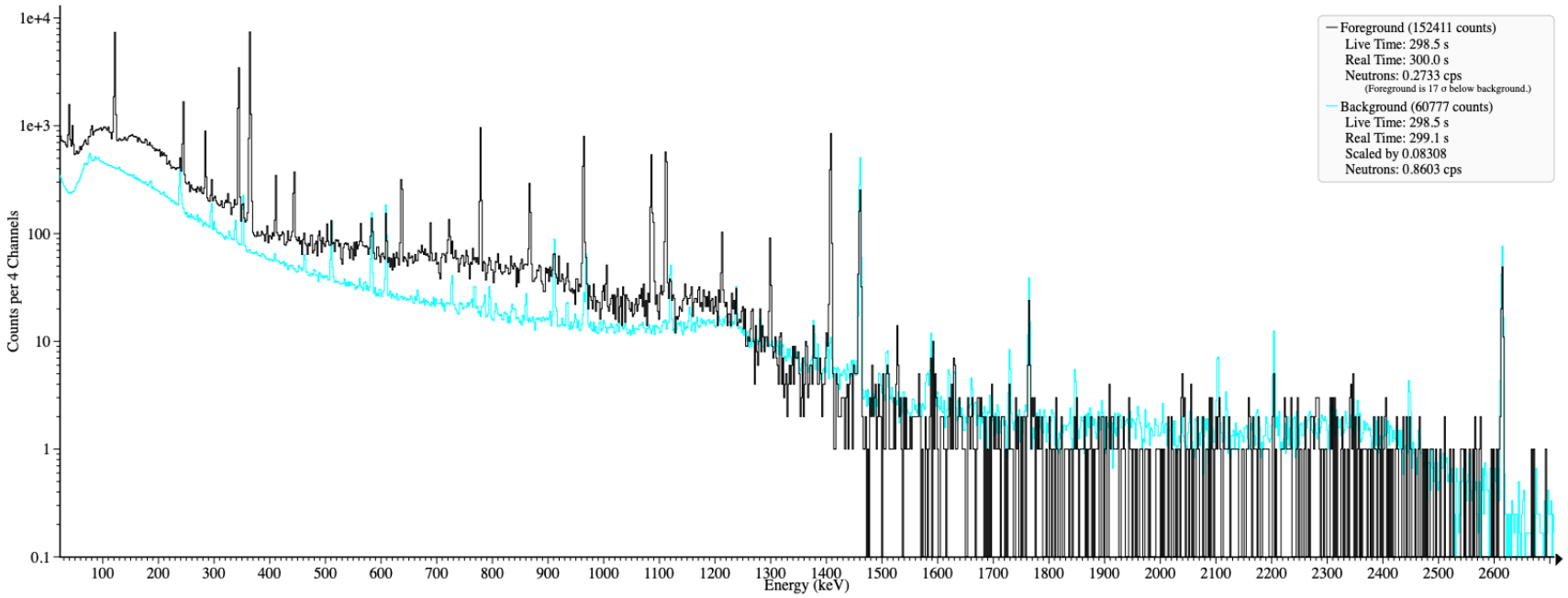
- Spectrum is synthetic data (GADRAS, 20% enriched Uranium + background)
- Spectrum file includes peak fits shown above, with nuclides associated with them

ADDENDUM - SPECTRA PROVIDED – “EX1_CS134_CS137.N42”



- Spectrum is synthetic data (GADRAS, Cs137 + Cs134 + background)

ADDENDUM - SPECTRA PROVIDED – “NEARBY_EXAMPLE_HPGE.N42”



- Spectrum is synthetic data (GADRAS, Eu152,15uCi + I131,10uCi + background)