NSUF 2004

Correlated uncertainties in gamma spectrometry:

Assay of Radium-223

Andy Pearce

CAIR

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Correlated uncertainties in gamma spectrometry: assay of Radium-223

- Introduction
- What are we trying to do and why?
- What is the problem?
- How can we solve it?
- How does this work in practice?
- Summary

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Introduction

NPL occasionally use calibrated gamma spectrometers to produce calibrated radioactive solutions for customers

Why?

- Ionisation chambers are only useful if a calibration figure exists
- Ionisation chambers can only be used if the sample produces a significant current in the chamber, which requires high activities
- Coincidence counting also requires a minimum activity, is very expensive (in staff time) and requires high purity solutions
- Gamma spectrometry, however, only requires a calibration curve and knowledge of the decay scheme (gamma emission probabilities etc)

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Introduction

^{223}Ra Assay

- Recently requested by a customer to assay ^{223}Ra and provide a certified activity concentration
- This work highlighted some of the complications in analysing data in gamma spectrometry
- A brief demonstration follows of problems encountered in obtaining a reliable mean value and associated uncertainty
- Am ignoring for the moment some more serious issues with the ^{223}Ra measurement (apparent sticking, limited nuclear data, inconsistent liquid scintillation results etc.)

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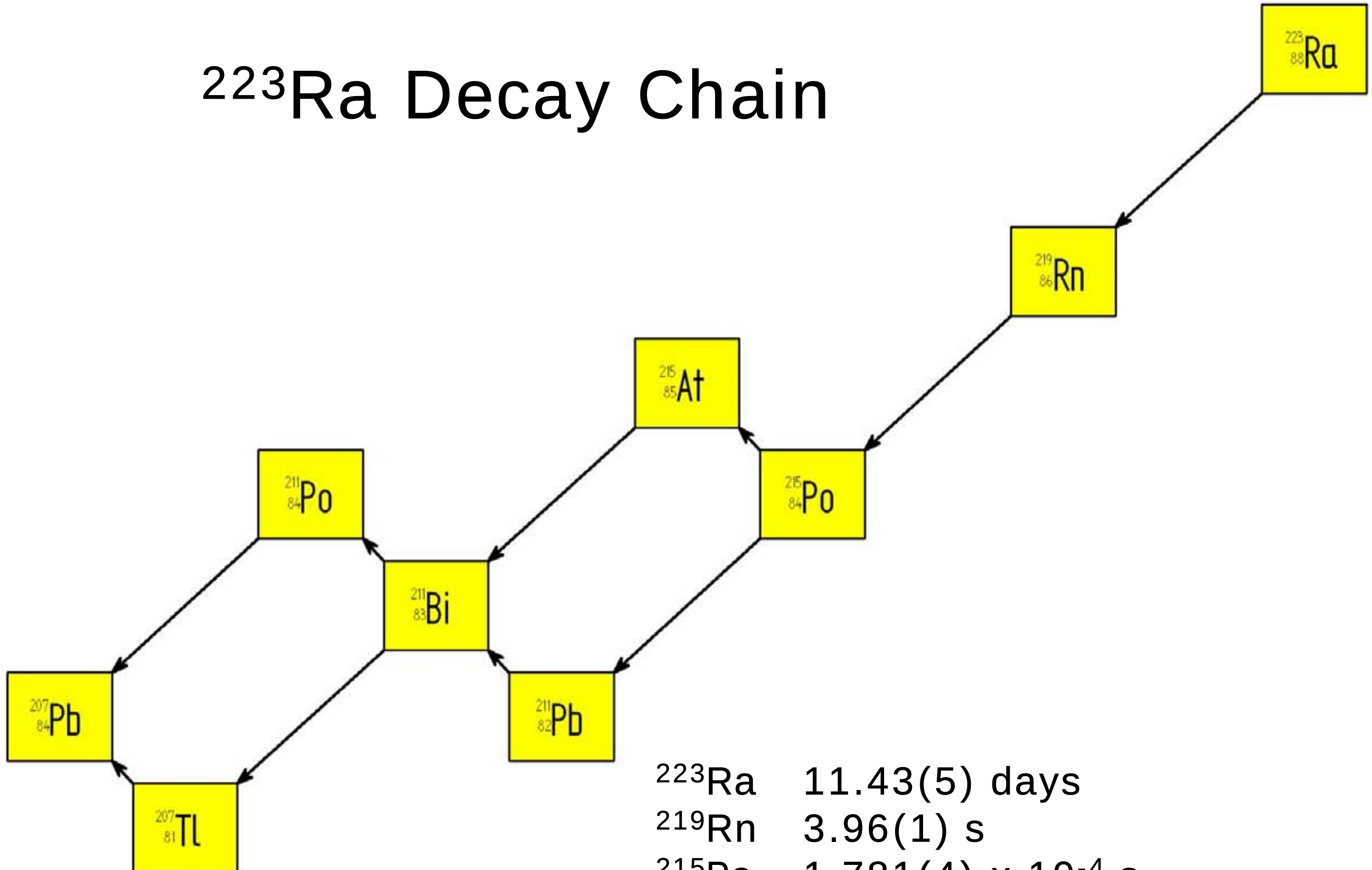
^{223}Ra Decay Chain



^{223}Ra	11.43(5) days
^{219}Rn	3.96(1) s
^{215}Po	1.781(4) x 10^{-4} s
^{211}Pb	36.1(2) min
^{211}Bi	2.14(2) min
^{207}Tl	4.77(2) min
^{207}Pb	*stable*

Nuclear data

Energy keV	Gamma Emissions per 100 decays	Uncertainty k=1 %
122.3	1.19	2
144.2	3.22	3
154.2	5.62	3
269.5	13.7	3
338.3	2.79	2
445.3	1.27	4
271.2	0.108	6
351.1	13.0	1
832.0	29.3	2

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^{223}Ra Parent

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daughters

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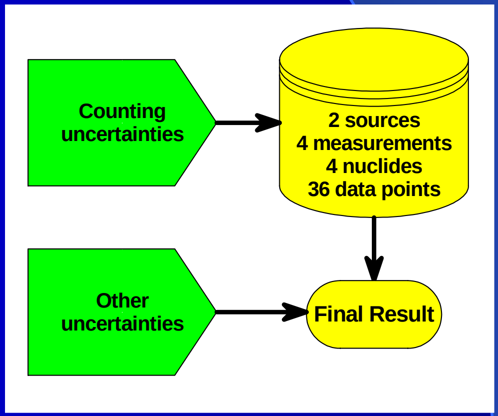
What is the problem?

- Two sources assayed
- Each source assayed twice
- Each measurement contains nine useful data points, corresponding to gamma lines of ^{223}Ra and three of it's daughters
- Total number of data points to combine: 36
- Would typically combine results by weighted mean:



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What is the problem?

Problem:

- Which uncertainties do you use to weight the data points?
- Type A's (counting statistics) do not reflect the *true* accuracy of a given data point
- Including type B uncertainties *before* the weighted mean can severely underestimate the true contribution due to correlations
- In the case of our ^{223}Ra measurements weighted mean suggests an uncertainty <1%
- However we *know* this is unrealistic, because the smallest uncertainty on any of the gamma emissions is around 2 %

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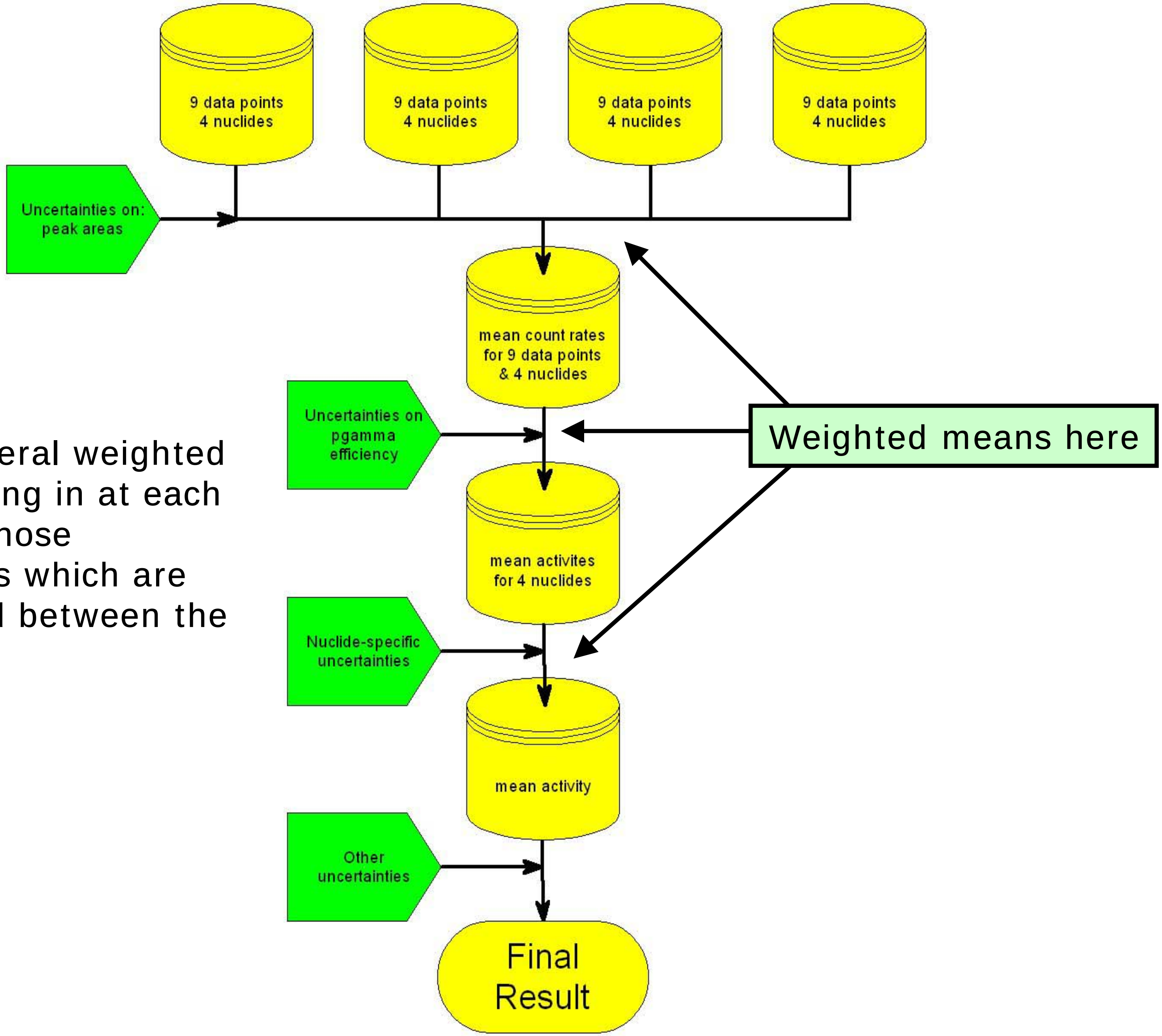
Solution:

Perform several weighted means, folding in at each stage only those uncertainties which are uncorrelated between the data points



However…

Another problem:

- Efficiencies are taken from the same calibration curve and are therefore the uncertainties are partially correlated
- Due to the way gamma emission probabilities are measured, the associated uncertainties are also partially correlated
- Other uncertainties – particularly any source-specific uncertainty such as gravimetric – may also be correlated

Not possible to combine this data using weighted means!

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How to proceed?

Option 1

- Ignore these problems and take a multistage approach similar to the one described above
- Assume the weighting procedure is “good enough”
- Estimate the average uncertainty contribution from the efficiency and gamma emissions
- This has been used successfully in the past; however with relatively large gamma emission uncertainties it is no longer viable

Option 2

- Surely there is a mathematically sound way of doing this?

Alternative procedure

- Use high quality calibration curve software which can both interpolate at the required energies and provide you with the covariances of the interpolated values
- Find some way to estimate the correlations between the gamma emission probabilities
- Combine the data by setting up covariance matrices and applying a Gauss-Markov least squares procedure to obtain a more complete weighted mean
- This approach makes it possible to propagate the uncertainties in a much more rigorous way
- However, it is mathematically complex and requires access to good quality linear algebra software (such as NAG)

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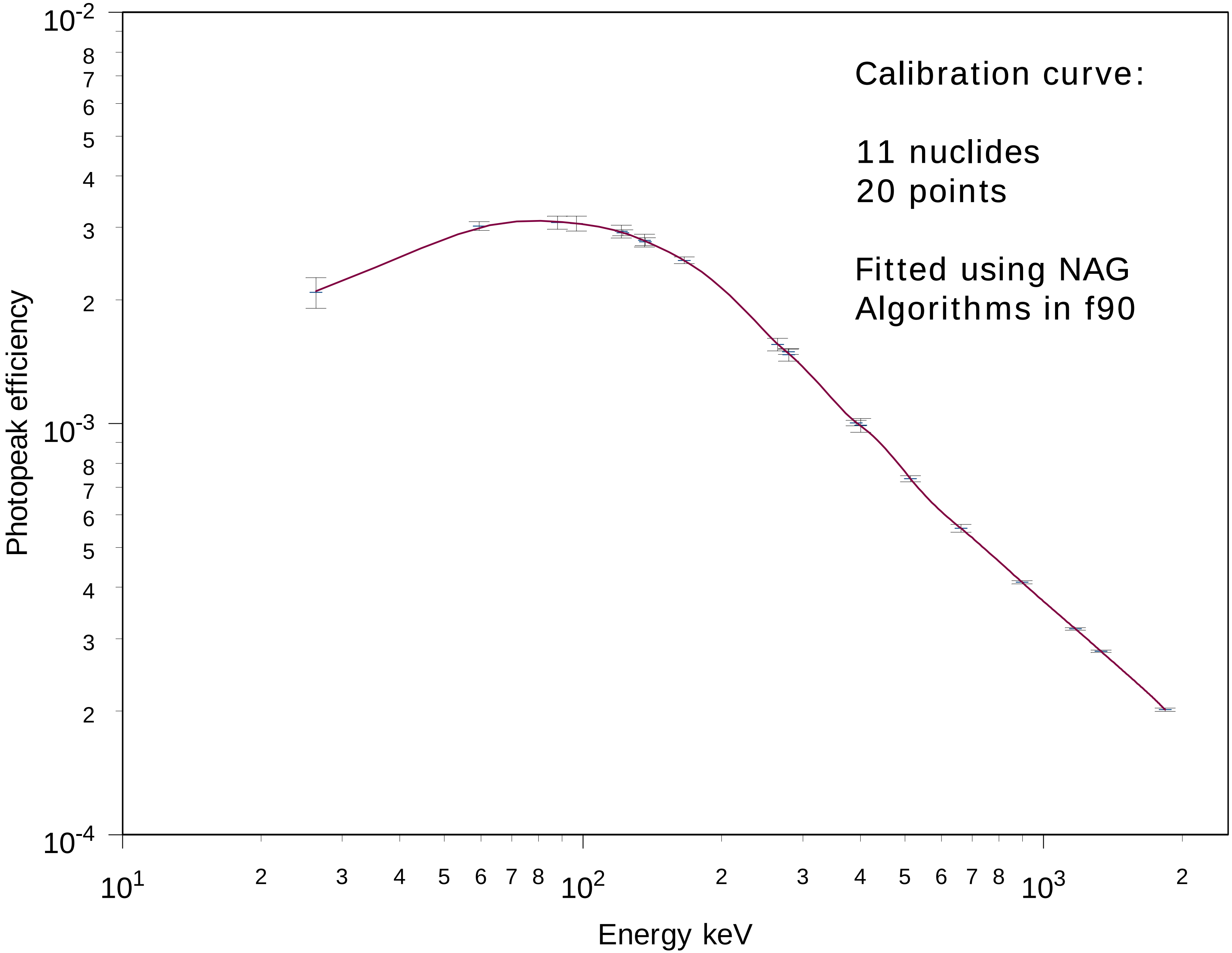
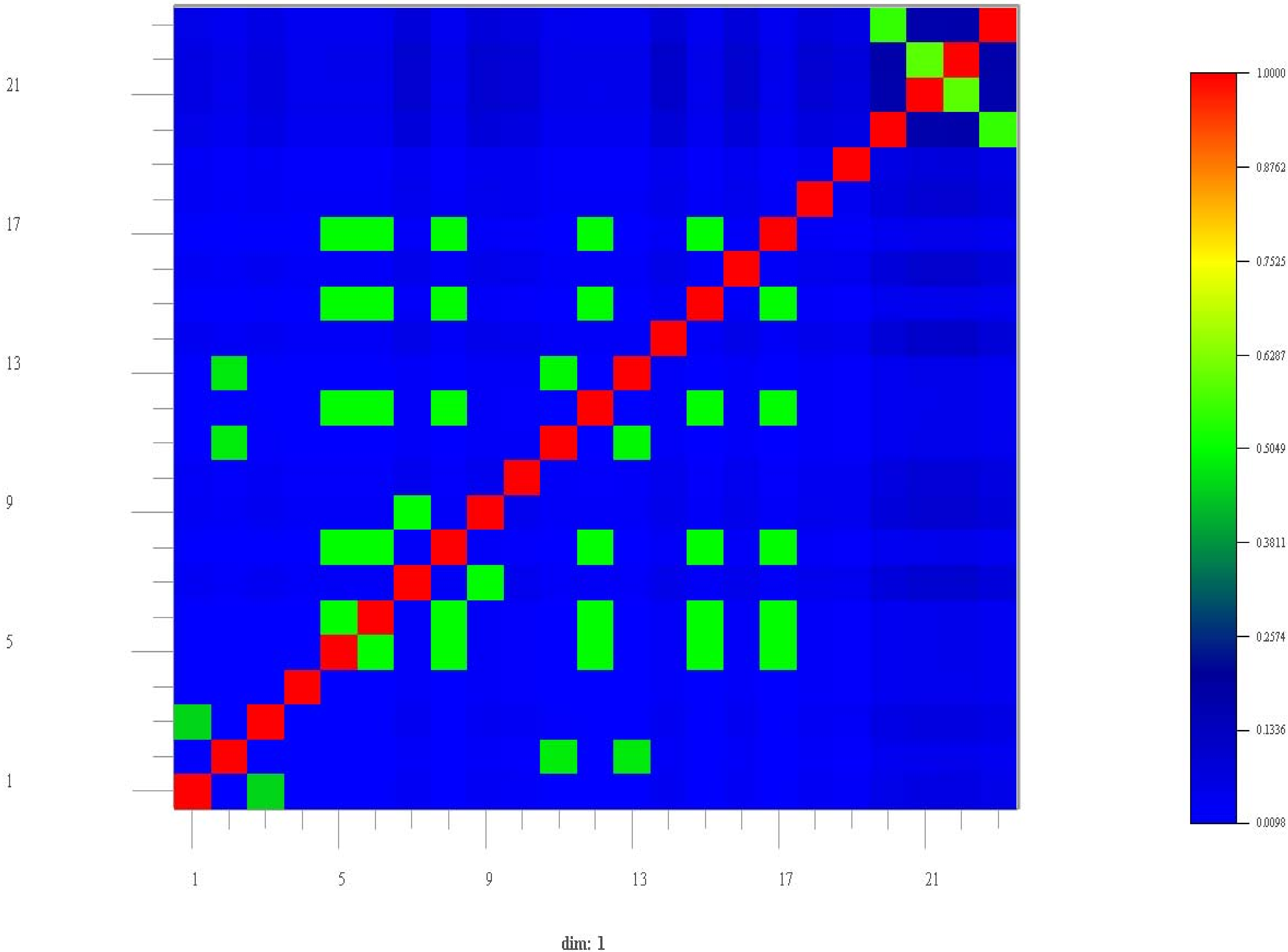


Calibration curve:
11 nuclides
20 points
Fitted using NAG
Algorithms in f90
Photopeak efficiency
Energy keV
10^{-2}
10^{-3}
10^{-4}
10^{1}
10^{2}
10^{3}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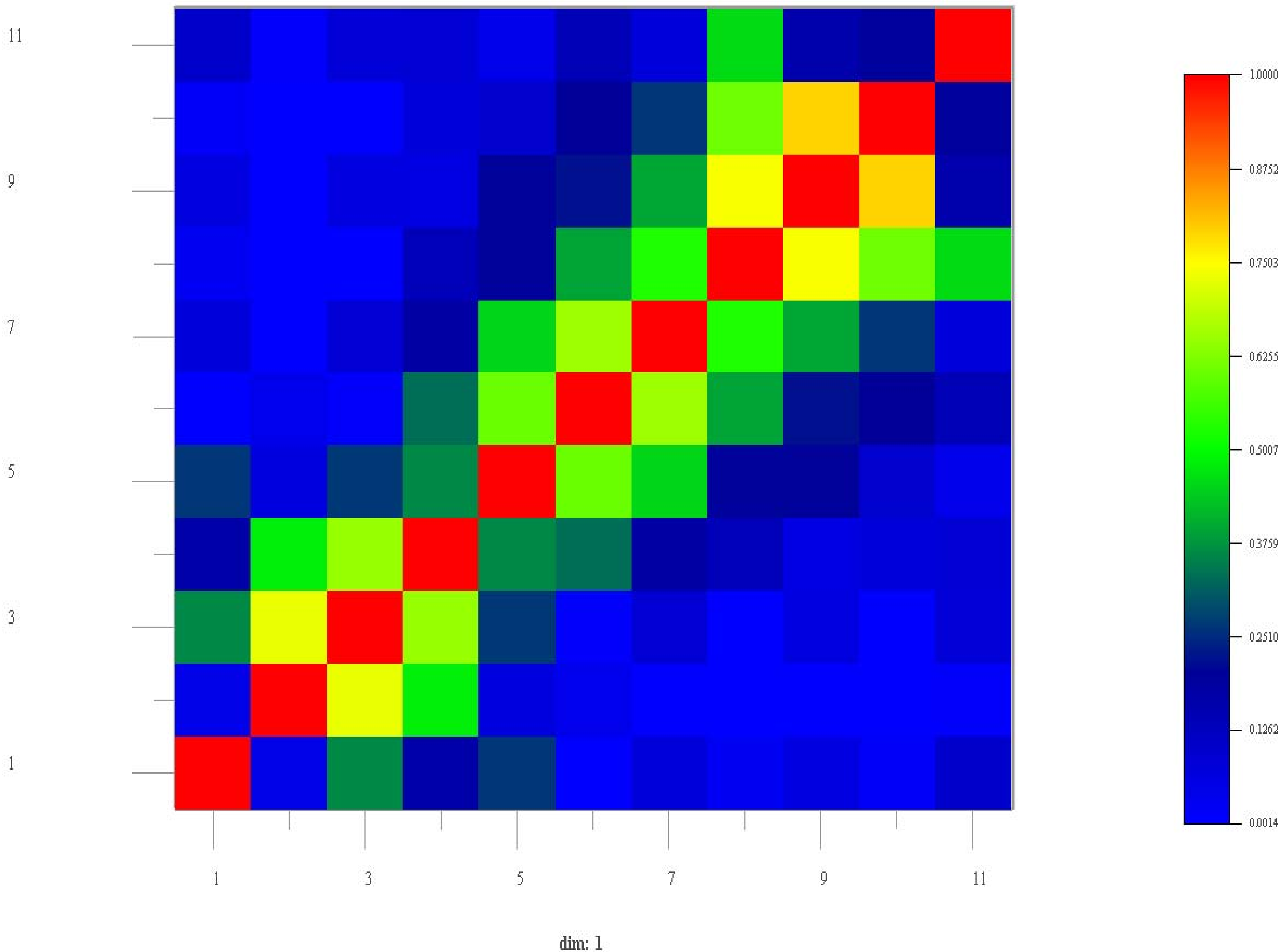

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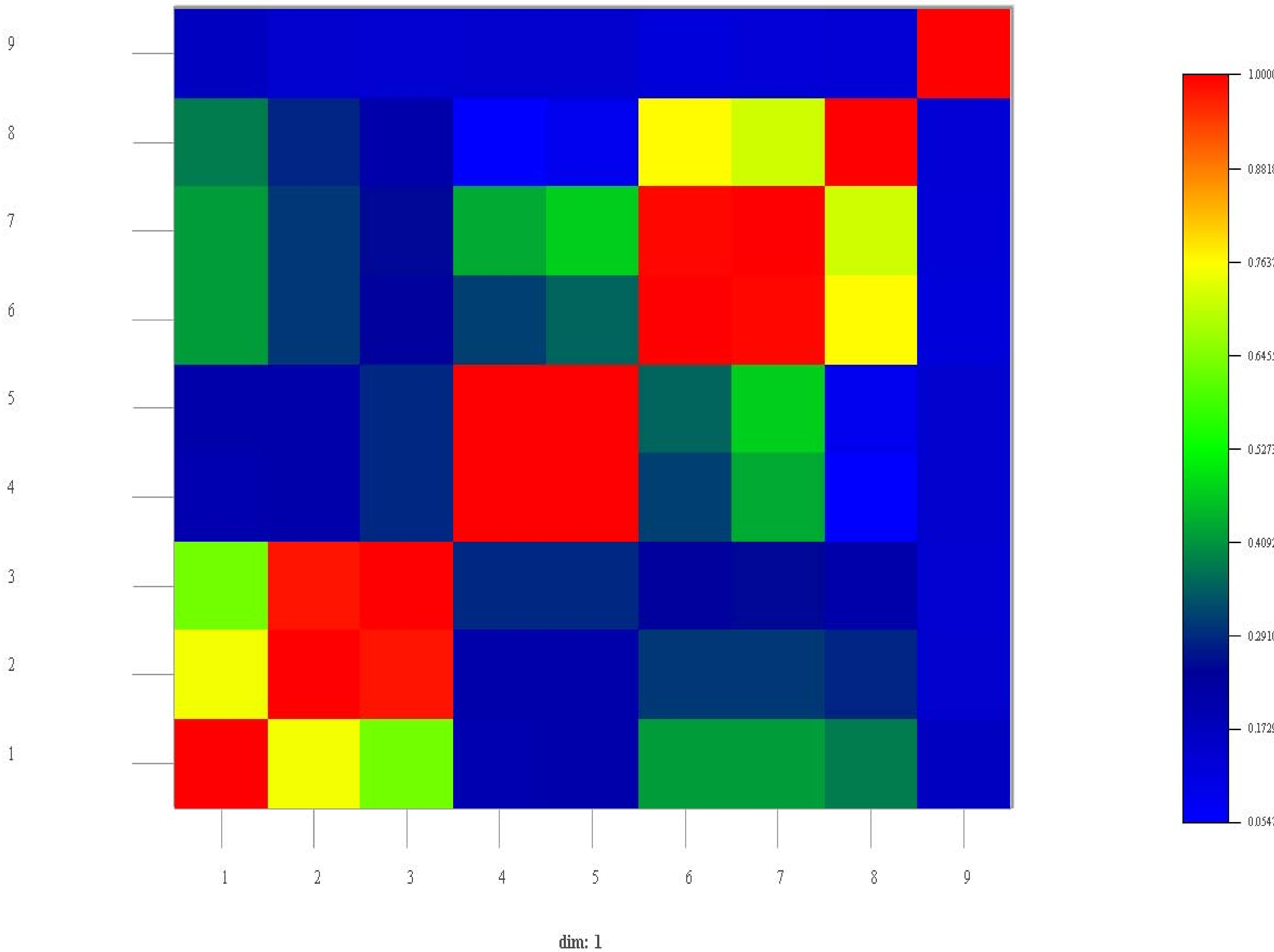
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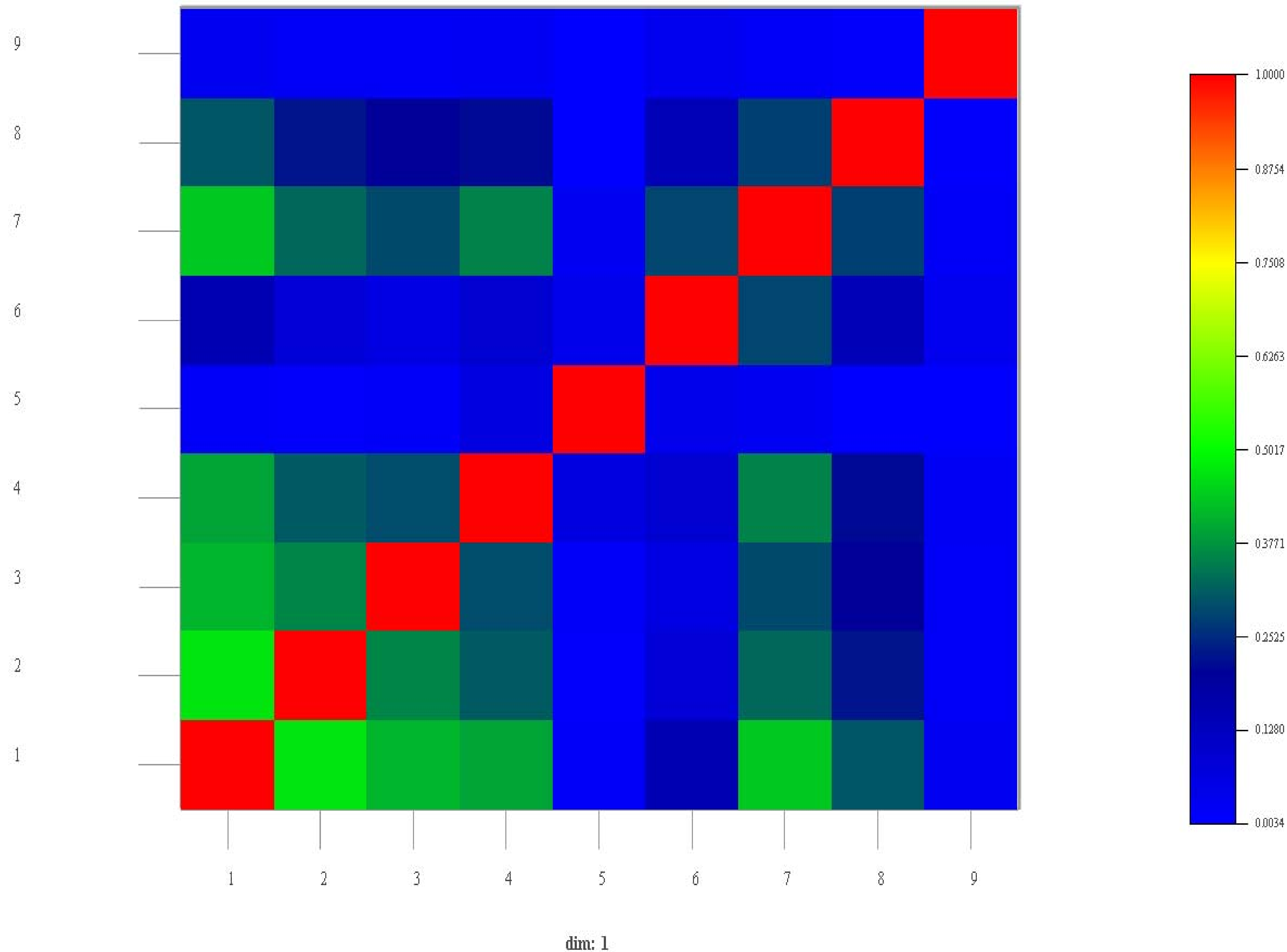

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0.6263
0.5017
0.3771
0.2525
0.1280
0.0034
dim: 1

Results

- Based on the "simplified" single measurement case:

Activity concentration	100 kBq/g	✓ RIGHT
Associated uncertainty	3 %	

Compare with

Activity concentration	99 kBq/g	✗ WRONG
Associated uncertainty	0.8 %	

Obtained by ignoring the covariances

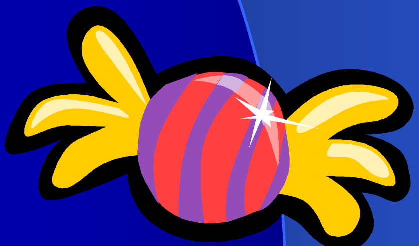
Correlations in P_γ values

Problem:

- Nuclear data sources do not indicate the correlation between the uncertainties on gamma emission probabilities
- This is not the fault of data evaluators as authors don't usually quote correlation coefficients in literature

However:

- Gamma emission uncertainties are often swamped by other uncertainties
- A reasonable estimate can be obtained in many cases by assuming the counting statistics on the main line are negligible and working backwards



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Commercial Software

Does the commercial gamma spectrometry software do the job?

- Have looked at the manuals for three common spectroscopy packages (not necessarily the latest version, but none more than a couple of years old)
- One package appears to handle the calculations quite well – this belief is held up by experience that it generates quite high uncertainties
- One package gives no indication of how the calculations are carried out so it is difficult to assess
- In another package, the manual indicates correlations are taken account of in some circumstances but not others

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Summary

- Neglecting to take account of correlated uncertainties in gamma spectrometry can cause gross errors
- It is possible to deal with these correlations by changing the way the data is analysed and propagating uncertainties correctly
- Dealing with correlation in gamma emission probabilities is still problematic
- Based on the evidence I have, not all manufacturers' software appears to tackle correlated uncertainties very well
- Radium-223 is a monster to measure!

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Finally...

I find these guides useful where difficult uncertainty problems are concerned, produced by the NPL maths section (they are not specific to our area):

SSfM Best Practice Guide 4: Discrete Modelling

SSfM Best Practice Guide 6: Uncertainty and Statistical Modelling

both available from the NPL website:

www.npl.co.uk/ssfm/downloads

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