NKS REMSPEC: An emergency preparedness exercise utilising a complex synthetic gamma spectrum

Dowdall, M.; Nielsen, Sven Poul; del Risco Norrlid, L.; Palsson, S.E.; Renvall, T.; Sidhu, R. Singh

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A Nordic seminar for users of gamma spectrometry

Oslo 15-16 September 2009

Paula Nunez (ed.) 1
Seppo Klemola 2
Sven P. Nielsen 3
Sigurður E. Pálsson 4
Carsten Israelson 5

1 Institute for Energy Technology, Norway
2 Radiation and Nuclear Safety Authority, Finland
3 Risø National Laboratory for Sustainable Energy, Denmark
4 Icelandic Radiation Safety Authority, Iceland
5 National Institute of Radiation Protection, Denmark

March 2010
Abstract

The project GammaSem was proposed to provide a forum for discussions and sharing of information on practical issues concerning gamma spectrometry and to establish a network of users of gamma spectrometry in the Nordic countries, thereby strengthening the collaboration and improving all participants’ competence in practical gamma spectrometry.

The seminars’ focus was practical challenges met by the users themselves, rather than theoretical matters. Scientists and users of gamma spectrometry from all five Nordic countries were invited to the seminar, as well as scientist from the Baltic countries. A total of 75 people participated; representing 34 different universities, commercial companies, research institutes and also all Nordic authorities.

During the seminar several key issues for follow-up were identified and working groups for addressing the identified problems were established. The working groups were:
- Uncertainties and detections of limits
- True summing coincidence
- Monte Carlo simulations and efficiency transfer
- Absorption (density corrections and geometries)
- Mobile gamma spectrometry systems
- Nuclear forensics (on special samples and special parts of the spectra)

The identified topics will form the basis for the agenda of the next seminar in 2010. There, the different working groups will be invited to present their ideas/solutions to the relevant problems.

Key words

gamma spectrometry
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Introduction
The project GammaSem was proposed to provide a forum for discussions and sharing of information on practical issues concerning gamma spectrometry and to establish a network of users of gamma spectrometry in the Nordic countries, thereby strengthening the collaboration and improving all participants’ competence in practical gamma spectrometry. It is acknowledged that during the last two decades there has been a dramatic decrease of new students in nuclear disciplines, and that many organisations, including the IAEA and the NKS, are concerned about this decline.

Two seminars for users of gamma spectrometry were proposed to be arranged in 2009 and 2010. This report is a description of the first seminar, held in September 2009.

The seminar
The seminars’ focus was practical challenges met by the users themselves, rather than theoretical matters. Scientists and users of gamma spectrometry from all five Nordic countries were invited to the seminar, as well as scientist from the Baltic countries. The interest for the seminar was far greater than anticipated by the organisers and a total of 75 people participated; representing 34 different universities, commercial companies, research institutes and also all Nordic authorities. For a complete list of participants see attachment 1.

Issues to be discussed at first the seminar included:
- Lessons learned from the NKS laboratory intercomparisons
- User experience with commercial software, presentations of independent evaluations
- Calibration and corrections, i.e. recent trends (Monte Carlo methods) and practical solutions (e.g. experience with calibrations for true coincidence summing corrections), density corrections etc...
- Ensuring quality in gamma-ray spectrometry
- Accreditation
- Discussion of the form of a contact group for the participants
- Possibility for a common Nordic standard for nuclide libraries
- Common Nordic geometries for better emergency preparedness

The well-known expert in gamma spectrometry, Gordon Gilmore, was invited to give a presentation at the seminar. He gave an overview of some of the most common difficulties and sources to mistakes when using gamma spectrometry. His presentation is found in attachment 3.

The subsequent part of the seminar was divided into four sessions covering:
- general descriptions to the gamma spectrometry activities in some of the big participating organisations
- detail descriptions of project specific challenges
- discussions on uncertainties, corrections and quality assurance
- experiences from intercomparison exercises.
See attachment 2 for the seminar agenda and attachment 4 for abstracts from the presentations. The full presentations are available at the NKS website.

During the seminar several key issues for follow-up were identified and working groups for addressing the identified problems were established. The groups are listed below (subgroup leaders highlighted in cursive letters, the rest of the participating organisations follow).

**Uncertainties and detections of limits**

IFO  
Lund University  
TVO  
ICT  
SKB  
STUK  
RISØ  
Ringhals NPP  
Forsmarks Kraftgrupp AB  
Icelandic Radiation Safety Authority  
Danish Decommissioning

**True summing coincidence**

IFK Kjeller  
NRPA  
RISØ  
SIS  
NRPA Tromsø  
Icelandic Radiation Safety Authority  
Ringhals NPP  
FOI  
OKG AB

**Monte Carlo simulations and efficiency transfer**

NRPA  
Göteborg University  
STUK  
FOI  
Icelandic Radiation Safety Authority

**Absorption (density corrections and geometries)**

IFE Halden  
Lund University  
ICT  
IFE Kjeller
Mobile gamma spectrometry systems

NRPA
IFE Kjeller
Danish Decommissioning
FOI
DEMA

Nuclear forensics (on special samples and special parts of the spectra)

RISO
NRPA
STUK
FOI
IFE Kjeller
Danish Decommissioning

The identified topics will form the basis for the agenda of the next seminar in 2010. There, the different working groups will be invited to present their ideas/solutions to the relevant problems. This will ensure that the identified key issues are not forgotten but will be followed up and hopefully lead to solutions that will increase the performance of the individual laboratories. Each working group will also contribute to a report that will be the final delivery from the project.

A working group for planning the follow-up seminar in 2010 was formed and includes now both the original organisers as well as all subgroup leaders not previously involved.

Concluding remarks

There is a big need for more cooperation and for training within the field of gamma spectrometry. This fact has been proved at this first seminar, both by the many different topics that were discussed, but also by the huge interest for participating by the organisations within the different fields that are end-users of gamma spectrometry. The seminar has thus provided a much welcomed starting point for a broader Nordic collaboration.

There was not enough time at the seminar to discuss all possible topics for collaboration. The seminar was instead a first recognition of common difficulties and challenges, and through the formation of working groups some issues were prioritized as more important, relevant and acute to the participants. This will provide a basis for future collaboration between end-users and hopefully lead to more precise discussions at future seminars, now that basic common issues have been recognized.

Most importantly, the participants agreed that the seminar was a good idea and that it had been successful in providing a forum not otherwise present in the Nordic countries. With this in mind the organizing committee strongly recommends the NKS that seminars of this kind are supported also in the future.
### Attachment 1: List of participants

<table>
<thead>
<tr>
<th>Name</th>
<th>Organization/Institution</th>
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<tr>
<td>Aleksi Mattila</td>
<td>STUK</td>
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<td>Alexander Mauring</td>
<td>NRPA</td>
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<td>Anette Grundin</td>
<td>Forsmarks Kraftgrupp AB</td>
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<td>Ann Helen Haugen</td>
<td>IFE Kjeller</td>
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<tr>
<td>Anna Nalbandyan</td>
<td>NRPA, Environmental Unit in Tromsø</td>
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<td>Ari-Pekka Leppänen</td>
<td>STUK</td>
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<tr>
<td>Bente Lauridsen</td>
<td>Danish Decommissioning</td>
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<td>Bjørn Lind</td>
<td>NRPA</td>
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<td>Carsten Israelson</td>
<td>National Institute of Radiation Protection</td>
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<td>Charlotte Lager</td>
<td>Forsmarks Kraftgrupp AB</td>
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<td>Christopher L. Rääf</td>
<td>Lund University</td>
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<td>Daina Riekstina</td>
<td>Institute of Solid State Physics, University of Latvia</td>
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<tr>
<td>Daniela Markocsan</td>
<td>Department of Radiation Physics, Sahlgrenska Academy at Göteborg University</td>
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<tr>
<td>Dr. Gerhard Fritz</td>
<td>Canberra GmbH Germany</td>
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<td>Elin Enger</td>
<td>Norwegian Defence Research Establishment</td>
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<td>Elisabeth Strålberg</td>
<td>IFE Kjeller</td>
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<tr>
<td>Eva Henell</td>
<td>OKG AB (Oskarshamn)</td>
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<tr>
<td>Gordon Gilmore</td>
<td>Nuclear Training Services Ltd</td>
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<td>Gro Elisabeth Hjellum</td>
<td>Algeta ASA</td>
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<td>Gro Salberg</td>
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<td>Gustaf Ullman</td>
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<td>Hanna Rannemalm</td>
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<td>Hannele Hirvonén</td>
<td>Teollisuuden Voima Oy</td>
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<td>Helle Karina Aage</td>
<td>Danish Emergency Management Agency, Nuclear Division</td>
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<td>Henrik Ramebäck</td>
<td>Swedish Defence Research Agency</td>
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<td>Håkan Pettersson</td>
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<td>Håkon Jensen</td>
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<td>Ingrid Svären</td>
<td>Institute of Marine Research</td>
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<td>Jacek Koziorowski</td>
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<td>Jan-Erik Jönsson</td>
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<td>Janis Berzins</td>
<td>Institute of Solid State Physics, University of Latvia</td>
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<tr>
<td>Jenny Nilsson</td>
<td>SU/Sahlgrenska, Avd för radiofysik</td>
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<td>Jens Søgaard-Hansen</td>
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<td>Kjell Johansson</td>
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<td>Knut K.F. Eitrheim</td>
<td>IFE Halden</td>
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<td>Kristina Veirø</td>
<td>Risø DTU</td>
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<td>Lars Nygård</td>
<td>Nerliens Meszansky AS</td>
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<tr>
<td>Lena Wallberg</td>
<td>Swedish Radiation Safety Authority</td>
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</table>
Lilián del Risco Norrlid  Swedish Radiation Safety Authority
Magnus Kronberg  Swedish Nuclear Fuel and Waste Management Company
Marie Carlsson  Linköping University Hospital
Mark Dowdall  NRPA
Mattias Olsson  Forsmarks Kraftgrupp AB
Mojtaba Ghiasi  Forsmarks Kraftgrupp AB
Morten Sickel  NRPA
Odd Fredrik Laastad  IFE Halden
Óskar Halldórsson  Icelandic Radiation Protection Authority
Patrik Konnéus  Ringhals NPP
Paula Nunez  IFE Kjeller
Per Roos  Risø DTU
Pertti Niskala  STUK, Regional Laboratory in Northern Finland
Peter Holm  OKG AB (Oskarshamn)
Peter Möller  Danish National Institute of Radiation Hygiene
Pia Hedberg  Barsebäck Kraft AB
Qin Svantesson  Westinghouse Electric Sweden
Rajdeep Sidhu  IFE Kjeller
Ronny Nelvik  IFE Kjeller
Seppo Klemola  STUK
Sigurður Emil Pálsson  Icelandic Radiation Safety Authority
Stefan Isaksson  Gammadata Instrument AB
Stefan Mårtensson  Gammadata Instrument AB
Sven Poul Nielsen  Risø DTU
Thommy Ingemann Larsen  Danish Decommissioning
Tore Ramsøy  IFE Kjeller
Trygve Bjerk  IFE Kjeller
Tung Si Ho  IFE Kjeller
Attachment 2: Agenda GammaSem 2009

Tuesday 15th of September

09.00 – 09.30  Registration and coffee
09.30 – 09.40  Opening of GammaSem
09.40 – 10.40  Gordon Gilmore, Nuclear Training Services Ltd.
               Does our software tell us the truth (or should we burn the Black Box)?
10.40 – 11.00  Open discussion
11.00 – 11.20  Sven Poul Nielsen, National Laboratory for Sustainable Energy at the
               Technical University of Denmark
               Gamma spectrometry at Risø DTU
11.20 – 11.40  Rajdeep Sidhu, Institute for Energy Technology
               Gamma spectrometry at IFE
11.40 – 12.40  Lunch
12.40 - 13.00  Hanne Breivik, Norwegian Defence Research Establishment
               General overview of gamma spectrometry activities at FFI
13.00 – 13.20  Seppo Klemola, Radiation and Nuclear Safety Authority, Finland.
               Gamma spectrometry at the accredited laboratory of STUK
               Gamma scanning of nuclear fuel rods
13.40 – 14.00  Patrik Konnéus, Ringhals NPP
               Nuclide specific surface activity measurements at Ringhals NPP
14.00 – 14.20  Per Roos, National Laboratory for Sustainable Energy at the Technical
               University of Denmark
               Gamma- and X-ray analysis of Pu and U in single Thule particles
14.20 – 14.40  Kjell Johansson, Institute for Energy Technology
               Gamma spectrometry as a tool for global disarmament
14.40 – 15.00  Coffee and fruit
15.00 – 15.20  Ari-Pekka Leppänen and Pertti Niskala, Radiation and Nuclear
               Safety Authority, Finland.
               Background problems when measuring NORM's
15.20 – 15.40 **Johan Byegård**, Geosigma  
*Chemical separation methods applied on complex water samples, implications on detection limits for gamma*

15.40 – 16.00 **Open discussion**

18.00 **Joint GammaSem Dinner**

**Wednesday 16\textsuperscript{th} of September**

09.00 – 09.20 **Gerhard Fritz**, Canberra GmbH Germany  
*Sources of uncertainty in gamma spectrometry*

09.20 – 09.40 **Henrik Ramebäck**, Swedish Defence Research Agency  
*Implementing combined uncertainty according to GUM into a commercial gamma spectrometric software*

09.40 – 10-00 **Jonas Boson**, Swedish Defence Research Agency  
*Uncertainty assessment by latin hypercube sampling for in situ gamma spectrometry*

10.00 – 10.20 **Sigurður Emil Pálsson**, Icelandic Radiation Safety Authority  
*Calibration (including TCS) and use of commercial gamma spectrometric software – lessons learned*

10.20 – 10.40 **Coffee and fruit**

10.40 – 11.00 **Gustaf Ullman**, Linköping University  
*A general Monte Carlo model for calibration of the HPGe detector “Detective EX 100”*

11.00 – 11.20 **Liliah del Risco Norrlid**, Swedish Radiation Safety Authority  
*Improving the results of the analysis of a complex gamma spectrum by applying the cascade summing correction*

11.20 – 11.40 **Open discussion**

11.40 – 12.40 **Lunch**

12.40 – 13.00 **Daina Riekstina**, University of Latvia  
*Quality assurance*

13.00 – 13.20 **Mark Dowdall**, Norwegian Radiation Protection Authority  
*NKS REMSPEC: an emergency preparedness exercise utilising a complex synthetic gamma spectrum*

13.20 – 13.40 **Christopher L. Rääf**, Lund University  
*Evaluation and validation of national standard geometries for gamma spectrometry*
13.40 – 14.00 Open discussion

14.00 – 14.20 Coffee and fruit

14.20 – 16.00 Identification of key issues for follow-up and establishment of working groups
INVITED SPEAKER

Gordon Gilmore
Nuclear Training Services Ltd.

Does our software tell us the truth
(or should we burn the Black Box)?
Does our software tell us the truth?
(Should we burn the black box?)

Dr Gordon Gilmore
Nuclear Training Services Ltd., Warrington, England.

Disclaimer:

The results quoted in this presentation were generated using particular versions of the programs used. It is possible that later versions of the same program perform differently – for better or worse. That applies to their manuals as well.

I make no absolute value judgements on these programs; I merely quote experience. It is possible that other people using different parameters might achieve better results.

GammaVision was version 5.30
FitzPeaks was version 3.63
The Genie manual was for Genie-PC

© Dr Gordon Gilmore, September 2009. The author asserts all intellectual property rights over this presentation and notes.
When I was asked to give this talk, it was suggested that I share my experience of different gamma spectrometry software systems. I can only do that to a limited extent. Like most people, my experience, and the systems I have used, was dictated by the commercial situation I found myself in.

In my early gamma spectrometry days, from 1966 to 1993, I ran a small Activation Analysis Service. It was up to me to provide the spectrum analysis software – by which I mean to create it. At the time, the commercial programs were only just appearing, and certainly were not aimed at Activation Analysis. That program, or rather the successor to it, is called CompAct, standing for Comparative Activity measurement.

The experience of writing spectrum acquisition and analysis software means that I can judge other programs with some understanding of the underlying difficulties that those other programs have had to overcome. It also means that I feel justified in being somewhat critical of programs that do not function as well as they might.

Much later, in 2000, I was asked to help set up the gamma spectrometry lab for NIRAS (now part of AMEC) in the UK. That lab was committed to using GammaVision, and so I gained considerable experience of the inner workings of that program.

FitzPeaks I have a little experience of, but I am generally impressed by it. The author, Jim Fitzgerald, used to work for Canberra in the UK providing software support. The nice thing about FitzPeaks is that Jim is resident in the UK and is approachable – unlike the software writers for the large companies.

Genie I have little real practical experience of, but I have, from time to time, sought to compare its performance against other programs.
Does our software tell the truth?

- The short answer is...
- ...probably most of the time,
- but not all of the time

So to the point…
Does our software tell the truth?
The answer is probably most of the time, but not all of the time.
I should explain what I meant, in the sub-heading to my talk, by ‘the Black Box’. It is the system, be it spectrometer, program, whatever, that unaware people use for their measurements. The sample, or spectrum, goes in at one end and results come out of the other and are accepted with complete and utter, uncomprehending and untested, trust. The box is black because no light – or should we say, enlightenment – penetrates it.

It is a great concern to me that the ‘Black Box’ mentality is rife in many gamma spectrometry labs. In my opinion it will not do!

You must always bear in mind that the manufacturers live by selling systems and software – not by generating results. They don’t lose jobs or clients if their systems give the wrong results – but YOU might.

You may think that is a very cynical view of the manufacturers, but I have spent many years observing and trying to communicate with them.
As some people here may remember, on my gamma spectrometry course, I present one of my articles of faith as
‘All software will let you down under the right circumstances’. The trick is to understand what those circumstances are.

That knowledge can only be acquired by reading the manuals (which, as we shall see, may themselves tell lies!) and playing with the software; changing parameters to see what happens to the analysis. It might be necessary to do some manual calculations, both of activity and uncertainty, to check whether the program gives justifiable results.

The most profound learning experiences are to keep one’s eyes open for inconsistencies: situations where the results look odd, or the peaks don’t look right, or something just doesn’t seem right. When that happens, don’t just say ‘Oh, how odd!’ - find out why it’s odd!

Resolving inconsistencies can be particularly satisfying.

By way of example, I want to bring to your attention a few inconsistencies and, while discussing them, point out various pitfalls in trying to understand your software.
This is a calibration – it doesn’t matter what calibration (although any experienced gamma spectrometrist should recognize it). Look at it from a simplistic point of view – what calibration line would *you* fit to the data?
Well, I’m a simple soul! A straight line looks fine to me.
In fact, of course, this is a peak width calibration – in fact it is part of the data within the calibration spectrum provided as an example with the Genie-2000 software.

Do you think it fits? I certainly don’t. As far as I’m concerned the data and the fitted function are inconsistent.

Why do Canberra use that function?
Canberra Peak Width Calibration

2. Peak full-width at half-maximum (FWHM in units of channels) as a function of energy

\[ \text{FWHM} = \frac{F_0 + F_1 \sqrt{E}}{C_1} \]

where \( E \) is the energy in keV, \( C_1 \) is the “gain” term from the energy calibration equation, and \( F_1 \) and \( F_0 \) are the coefficients of the FWHM equation.

This is the appropriate page from the Genie manual…

The calibration is a function involving the square-root of the gamma-ray energy: an Intercept + a gradient times square-root of the energy.
In their book, Debertin and Helmer suggest the square-root function shown in red: the square-root of an Intercept + gradient times the energy. There is theoretical justification for this equation. There is no theoretical justification for the Genie equation.

I suspect that the Canberra programmer thought he/she was using a rearrangement of this Debertin and Helmer equation. That is not the case and there is no theoretical justification at all for it. As far as I am concerned it is wrong and, bearing in mind that our programs make judgements based up peak width, this must have unwanted consequences for our analyses.
Again I invite you to judge whether the Genie function fits the data properly.

UPDATE:
In discussion with Gerhard Fritz, of Canberra, at this Seminar, he admitted that perhaps there had been a mistake in the FWHM calibration but insisted that the Genie algorithms are relatively insensitive to the actual form of the FWHM calibration.
Having looked at this matter in some depth while preparing the second edition of ‘The Book’, taking theoretical considerations into account, I have come to the conclusion that a square root quadratic function best fits FWHM data. There is reasonable theoretical justification for that view.

GammaVision uses an un-square-rooted quadratic and for some time I have been critical of that choice as being an easy, unimaginative choice by a badly advised programmer. There is no theoretical justification for it but, to my surprise (and some embarrassment) in practice it appears to fit actual data almost as well as the square-rooted quadratic function I propose.
So, we had there an example of a fundamental inconsistency within the software itself.

But sometimes even the manuals may frustrate our attempts to understand the software.

This is another page from the Genie manual – the statement made is simply incorrect.
Canberra again…

\[
F_i = H e^{-\frac{(x_i-c_p)^2}{2\sigma^2}}
\]

where

- \(F_i\) is the value of the peak model function at channel \(x_i\),
- \(H\) is the height of the peak,
- \(c_p\) is the peak centroid, and
- \(\sigma\) is the width of the Gaussian (approximately 2.355*FWHM, where FWHM is the width of the peak at half height after the subtraction of the continuum).

No! FWHM = 2.355 \times \sigma

this is what it should be. In this case, I suspect that within the software the correct relationship is used. The problem is the manual compiler. But it is not helpful is it?

UPDATE:

Gerhard Fritz pointed out to me that the current versions of the Genie manual do have this function defined correctly. Excellent!
Another example…

This is from the GammaVision 5.30 manual.

While trying to create an uncertainty budget for the AMEC lab, I found that the uncertainties given by GammaVision did not agree with manual calculations based upon the list here…
– unless, that is, I left out the calibration source uncertainty.

I contacted Ametek and, on my behalf, they contacted ORTEC in the USA who went back to the source code for GammaVision and agreed with me – GammaVision does not take that into account.

They then said ‘We will alter the next version of the manual’! Hmmmph. As it happens they haven’t. And, of course, what they should have said is that they would alter the program!

There is another problem with this uncertainties list, which is not immediately obvious. GammaVision assumes all random uncertainties have a Gaussian distribution, and all systematic uncertainties as having a rectangular distribution.

This is not ORTEC’s fault, they were following common usage at the time, where uncertainties were split into random and systematic. Experience has shown that there is no clear-cut distinction between two categories. Modern usage is to split into Type A, determined by repeated practical measurements (counting uncertainties are a special Type A case) and Type B, determined by any other means – including guesses!

Clearly, when setting up an uncertainty budget, all that should be borne in mind.
Let’s have a look at a spectrum inconsistency…

This is a spectrum with the peaks detected by GammaVision superimposed. This is a spectrum created by SpecMaker, for which I happen to know exactly what the mean background is – that is represented by the red line.

The thing to note is that the lowest point of most of the peaks is below that mean line. Anybody who has worked with GammaVision and bothered to examine their spectra will recognize that this is very common for GammaVision.

But, if the base of the Gaussian is below the mean background level it must mean that the area of the peak is overestimated. Quite so! The problem is proving that…
How do you know that your program gives you accurate peak areas?

It is not good enough to say ‘Well, ORTEC or Canberra or Jim Fitzgerald or anybody else says so’. You really have to convince yourself by doing some sort of testing.

This is more difficult than it sounds. You need spectra with peaks at known positions, with known peak areas and known widths. There are a number of sets of test spectra available, but none of them are without criticism by somebody.

The problem is that if the spectra are created mathematically, some people will cry ‘not real detector peaks’. If the spectra are created by counting, one can never know how many counts in a peak region are due to background and how many actual counts. You can easily estimate those numbers of counts, but never measure them.
Test spectra available

- IAEA G1 1976 – tests peak area measurement
- Sanderson 1992
- IAEA 1995 – tests peak area measurement
- NPL 1997 – tests activity measurement
- IAEA 2002 – tests activity measurement
- Intercomparison Exercises - e.g. NPL – test activity measurement
- SpecMaker – prepare your own test spectra

These are sets of test spectra available. Most of them are available for download on the gammaspectrometry.co.uk website.

The G1 spectra were created mathematically from real detector spectra counted for such a long time that statistical scatter was negligible. They test the basic peak detection and area measurement of the software.

The Sanderson 1992 spectra were mathematically generated spectra and again test peak area measurement.

IAEA 1995 spectra were created by Menno Blauuw of Delft. They test area measurement and Menno claims that the quoted areas are absolute and traceable.

NPL 1997 spectra were created by counting and are designed to test the whole analysis process. Unfortunately, there are problems with these spectra. In a report dealing with an assessment of various programs using the spectra, the originators criticise the lack of true coincidence summing corrections, but their calibration spectra supplied did not include the spectra needed to make those corrections. Some of the spectra are also subject to random summing, although that has gone unrecognized in the assessment report by NPL, the software manufacturers and the NPL’s expert analyst. The means to make those corrections are present, but not obvious.

IAEA 2002 spectra were again created by Menno Blauuw and this time test activity estimation. This set of spectra does include all the spectra necessary to perform TCS corrections on both Genie and GammaVision.

Of course, all laboratories should be participating in intercomparison exercises. If you get the right answer, you can be confident that everything is working fine. If not, then one may have to go back through the individual analysis stages to seek the problem.
SpecMaker is a spreadsheet based program that will create spectra up to 16k in size with peaks of known shape, position and area with realistic statistical uncertainty imposed upon the channels.

The idea is that you should create a suitable spectrum for your purposes, by specifying where you want the peaks to be and how big they should be. The peak widths are determined by the calibration data supplied by the user.

SpecMaker provides a page to compare the actual versus the measured peak information. The program also exports the actual data into a .CSV file, which can then later be loaded into the SpecMaker Assessment spreadsheet to do the comparison.
I have created a number of simple spectra designed to test the peak search function of spectrum analysis programs.

All these spectra contain 50 peaks on a flat continuum. The first peak is centred at 50 keV and the others are then inserted at 40 keV intervals. In all cases, when calculating limits, the background region widths are assumed to be 5 channels.

1) Three spectra containing peaks containing a number of counts equivalent to the Currie Critical Limit at the particular peak energy and peak width on different continuum levels. We would expect these peaks to be significant in 50% of cases, so our spectrum analysis programs should be able to detect 25 peaks.

2) Three spectra containing peaks containing a number of counts equivalent to the Currie Limit of Detection at the particular peak energy and peak width on different continuum levels. We would expect these peaks to be significant in 95% of cases, so our spectrum analysis programs should be able to detect at least 47 peaks.

3) Three spectra with 1000 count peaks on continuums of 100, 1000 and 10,000 counts/channel. The peaks in the third of these spectra are below the Detection Limit.

4) One spectrum with 10,000 count peaks on a 1000 counts/channel continuum. All these peaks should be easily measured.
At the top is part of a spectrum containing 50 peaks on a 10k background that contain exactly the number of counts equivalent to the 95% confidence Limit of Detection. At the bottom are the peaks added. Statistically, we would expect any program to be able to detect 95% of those 50 peaks – 47 or 48. Would you agree with me that all the peaks at the Limit of Detection are visible by eye?

The red spectrum contains peaks that are at the Currie Critical Limit on an 8k background. We would expect to be able to detect 50% of those peaks, because in half of cases the peak area would be below the critical limit and half above. At least some of those peaks are visible by eye.
This is an overall view of the SpecMaker Assessment spreadsheet.
The left hand four columns specify where the user asked SpecMaker to put the peaks and how big they should be.

The next two columns list the actual peak positions and the actual number of counts in the peaks.

The next four columns are the results gleaned from the user’s spectrum analysis program. The right hand four columns give the significance of the difference between actual and measured areas, the difference in peak position and width.
At the top of the screen are a number of assessment summaries:

Peak Analysis box – how many detected, how many spurious, how many missed
Position and Width box – the mean differences – measured and actual
Area Estimation – the centroid of the distribution of peak area difference (measured – actual) and whether it is significantly different from that expected
A plot of the distribution of area differences – (Measured – Actual) – in units of standard uncertainty.
This shows part of the upper spectrum I showed you before – each peak has the number of counts corresponding to the limit of detection on a 1000 ct/channel background. I would suggest that you can easily pick them out by eye.

Do you agree?
Think about this…

- If you are 95% certain of detecting the peak, do you not think you should be able to see it yourself (in most cases)?
- Based up long experience, I am convinced that that is so.
- If you don’t believe me, look at the example spectra and make up your own mind.

A question…

If I’m right, if you can’t see a peak, the actual number of counts must be less than the 95% Limit of Detection.
So why do our programs quote that limit when they can’t find a peak?

UPDATE
I was informed at the Seminar that both GammaVision and Genie now include upper limit option to be quoted when peaks are not found. Hallelujah!
This is simply another opportunity to see if you can pick out the Ld peaks by eye.
This slide compares the performance of FitzPeaks with my own program CompAct. I have never used FitzPeaks myself in practice, but I have to say that analyzing these test spectra suggests to me that it is well worth looking at.

The general conclusions are:

Critical Limit spectra: both programs find less that expected peaks, but CompAct appears to be significantly overestimating these small peaks areas. CompAct finds rather more spurious peaks than FitzPeaks.

Limit of Detection spectra: both programs find the expected 95% of peaks, at the same time generating few spurious peaks. Unfortunately, for me, CompAct is again overestimating peak areas.

1k Continuum spectra: both programs find all the peaks with few spurious peaks and little or no area bias.

(I am not discussing the 10k continuum spectrum because the peaks are below the Critical Limit by a variable amount, which makes assessment difficult.)
Comparing GammaVision to FitzPeaks is, at first sight, rather worrying.

The general conclusions are:

Critical Limit spectra: GammaVision performs disastrously with these spectra, finding few peaks and measuring them with a possible positive bias.

Limit of Detection spectra: again GammaVision does not find the expected number of peaks. In fact, visual inspection of the GammaVision results reveals many visually perceptible peaks that haven’t been detected.

1k Continuum spectra: GammaVision has very variable performance on these easy spectra and again the result suggest a positive bias on the areas.

GammaVision is generally prone to finding spurious peaks. My experience with the program is that it is not possible to remove these without compromising further the ability to detect peaks.

However, maybe we are being too hard on GammaVision. These analyses were done as a blind peak search. ORTEC themselves suggest that it is better to use a peak library and tell the program where to look for peaks.
So let us compare GammaVision’s performance with and without a library.

Critical Limit spectra: now GammaVision tends to find more peaks than expected. Unfortunately, there is still a large number of spurious peaks found and the positive area bias is definitely significant. Careful examination of the results reveals that some of the peaks GammaVision has found are, in fact, below the Critical Limit area and should be rejected.

Limit of Detection spectra: GammaVision now finds all the peaks, but with the usual positive area bias.

1k Continuum spectra: As above.
GammaVision…

- Blind search is poor
- Library directed better - but gives results for non-significant peaks
- Peak areas are consistently overestimated
- …because of that, many spurious peaks
- Unless peak/continuum ratio is high, then poor estimation of peak position
- Unless peak/continuum ratio is high, then awful estimation of FWHM

To summarize:

One of the hidden problems of GammaVision is that it can easily miss very visible peaks if they are not in the library. It means that manual examination of the spectrum and the analysis results are necessary if the spectrum is in any way out-of-the-ordinary.
This is the distribution of area differences (measured-actual) for a FitzPeaks analysis of the Limit of Detection peaks on a 1k counts/channel background.

Fitzpeaks often seems to measure peak areas more accurately than would be expected statistically. I’m not sure how that can be so, but there it is.
This distribution of area differences generated by my own program CompAct shows graphically the positive bias.
But when peaks are better defined, CompAct shows an area distribution exactly matching the statistical expectation.
GammaVision: $L_D$ on 100 c/ch

This is a typical distribution from GammaVision – peaks areas significantly biased high, regardless of the peak/continuum ratio.
These are very well-defined peaks on a low background. As with all these assessments of GammaVision, small peaks or well-defined peaks, the pattern here common to all. GammaVision consistently overestimates peak areas.

This is an inconsistency – and I haven’t yet explained it.
Let’s return to the Limit of Detection peaks on the 1k background

These are the peaks GammaVision fitted. Note the missing, visually perceptible peaks and the fitted peak shapes dipping below the mean of the background scatter. This was the inconsistency we started off discussing.

The reason for it is revealed in the GammaVision manual. GammaVision does not work out the peak limits using the peak width calibration, which would seem to be the sensible thing to do. It finds the limits by searching for a group of 5 points together forming a minimum point on each side of the centroid. Simple logic tells us that the resulting peak area MUST be at a maximum – hence the positive area bias.
The message is…

- Look at your spectra!
- When you find inconsistencies – follow them up – resolve them
- Test your software – make sure you understand it’s weaknesses – and it’s strengths
- If necessary, arrange some sort of post-analysis correction of results

My messages are:

Look at your spectra! THE MARK 1 EYEBALL CONNECTED TO A SENSIBLE BRAIN IS A REMARKABLE DEVICE – in many ways better than any of the software

Test your software – make sure you understand it’s weaknesses – and it’s strengths
YOU HAVE TO TAKE THE TIME

When you find inconsistencies – follow them up – resolve them
THAT’S THE WAY YOU LEARN

If necessary, arrange some sort of post-analysis correction of results
In practice, you have your software, if you replace it you may have just as many (probably different) problems.
It may be possible to correct some of the problems after the software has done it’s work.

FOR EXAMPLE…
The link program ‘Fred’ was written for the gamma spectrometry laboratory at NIRAS, now part of AMEC, Birchwood in the UK.

Fred takes the GammaVision .rpt output and performs a number of operations on. Some of them are for convenience, others are necessary.

Of particular note are the following (all are handled by Fred):

GammaVision does not do a correct peaked background correction because it does not take into account the uncertainty on the amount subtracted. This has the effect of leaving false positive peaks.

Although the random summing correction is accurate, the empirical uncertainty is unrealistically high,

If more than one peak is used to measure a nuclide, GammaVision does not use the intermediate results to calculate the final result. It adds all the individual peak areas together to calculate it. This give equal weight to all peaks whether measured with small or large uncertainty. The correct procedure would be to calculate a weighted mean based on the uncertainties of the individual peak measurements.

GammaVision uses the Limit of Detection to calculate MDA instead of upper limit – although I am informed that that may now be an option.
This website was created in tandem with the book ‘Practical Gamma-ray Spectrometry Second Edition’.

It carries the following:

- The appendices from the book
- Fully analysed example spectra: QCYK, long background, NORMS
- Some of the spreadsheets used to generate figures for the book
- SpecMaker and example spectra
- Publicly available test spectra
- Links to other relevant websites
- References and links to Data
- A bulletin board for exchange of information
PRESENTATION ABSTRACTS
A GENERAL MONTE CARLO MODEL FOR CALIBRATION OF THE HPGE DETECTOR “DETECTICE EX 100”

Gustaf Ullman and Håkan Pettersson
Department of Radiation Physics, Faculty of Health Sciences, Linköping University, SE-58185 Linköping, Sweden

The aim of this work was to create a general method for in situ calibration of the HPGe detector Ortec Detective EX 100 based both on measurements and on simulations with the general purpose Monte Carlo model MNCP5.

A device was built that allowed for experimental calibration of the HPGe detector by placing different radionuclides at a distance of 1 m from the detector at different inclination (0-180 degrees) and azimuth (0-360 degrees) angles. In all measurements in the experimental calibration the HPGe detector was placed on a tripod. In this way, also the attenuation in the tripod was taken into account. Four radionuclides were used for the experimental calibration: $^{241}$Am, $^{57}$Co, $^{137}$Cs and $^{60}$Co. Due to the symmetry of the tripod, it was sufficient to make the measurements in a subset of angles. The measurements were therefore performed in a grid of 6 inclination angles (0-90 degrees) and 13 azimuth angles (60-120 degrees) resulting in a total of 78 measurements. Each measurement was performed in approximately 15 minutes in order to obtain a sufficient poisson statistics (approximately 1% error). Monte Carlo simulations were performed in order to account for different source depth distributions as well as attenuation in soil and air. Three different source depth distributions were used, surface distribution, uniform depth distribution and an exponential distribution with mass activity distribution coefficient $0.0625 \text{ kg/m}^2$. A simplified geometry was used for the calculations with allowed them to be speeded up several orders of magnitude. The geometries used for the calculations were either a point source, or a cylindrical source distribution with a small radius of 1 cm, together with a large cylindrical plane detector of radius 15 km placed 1 m above ground level. The soil consisted of a slab with radius 15 km and depth 1.5 m. The results from the simplified geometry could be converted to a conventional in situ geometry by dividing the results with the cosine of the incident photon angle. For this work, calibration coefficients were calculated for three radioisotopes: $^{134}$Cs (605 keV), $^{137}$Cs (662 keV) and $^{131}$I (365 keV) for the three different depth distributions mentioned above.

For a surface source distribution the calculated calibration coefficients are:
1) $^{134}$Cs $k=0.87 \text{ kBq m}^{-2}/\text{s}^{-1}$, 2) $^{137}$Cs $k=1.00 \text{ kBq m}^{-2}/\text{s}^{-1}$ and 3) $^{131}$I $k=0.77 \text{ kBq m}^{-2}/\text{s}^{-1}$.

For a uniform source distribution (1.5 m layer of soil) the calculated calibration coefficients are 1) $^{134}$Cs $k=39.8 \text{ kBq m}^{-2}/\text{s}^{-1}$, 2) $^{137}$Cs $k=45.9 \text{ kBq m}^{-2}/\text{s}^{-1}$ and 3) $^{131}$I $k=19.9 \text{ kBq m}^{-2}/\text{s}^{-1}$.

For the exponential source distribution the calculated calibration coefficients are 1) $^{134}$Cs $k=1.30 \text{ kBq m}^{-2}/\text{s}^{-1}$, 2) $^{137}$Cs $k=1.50 \text{ kBq m}^{-2}/\text{s}^{-1}$ and 3) $^{131}$I $k=1.21 \text{ kBq m}^{-2}/\text{s}^{-1}$.

These results where compared with literature data, and a relatively good agreement was found. However, the results presented here are preliminary, and our model needs further validation.

Our Monte Carlo based model presented here is a fast, general, and feasible method for calibration of the in situ HPGe detectors such as Ortec Detective EX 100. The general nature of the model also allows for calibration of different detectors as well as different types of soil and radionuclides.
BACKGROUND PROBLEMS WHEN MEASURING NORM’S

A.-P. Leppänen and P. Niskala
Radiation and Nuclear Safety Authority - STUK, Regional Laboratory in Northern Finland,
Louhikkotie 28, FIN-96500 Rovaniemi, Finland
email: ari.leppanen@stuk.fi

Traditionally the behaviour of background has been left to little notice in gamma radiation measurements from environmental samples. The impact of variations in the natural background in measuring anthropogenic nuclides has little significance. The background has always been subtracted in the calculations and the importance of the use of recent background measurement has been stressed but is that enough? In recent years the more emphasis has been put to radiation doses from NORMs. Serious problems arise when low level NORM activities have to be determined from environmental samples. In this case, normal radon progeny nuclides, which are normally present in laboratory air, turn from nuisance into contaminants.

Radiation and Nuclear Safety Authority - STUK, Regional Laboratory in Northern Finland started a project where large numbers of samples had to be determined for NORM activities. In the beginning of the project the importance of the behaviour of the laboratory background was realised. An AlphaGuard detector was placed in the laboratory for long term monitoring Rn-222 concentration in the laboratory air. The backgrounds of the detectors were measured for different time intervals and for different times of the year. A detector background measurements was compared with another detector background measurement done at the same time.

The long term monitoring of the laboratory air for Rn-222 revealed changing patterns. First there were random and sharp rises, of approximately 2-5 fold, in the Rn-222 content which were related to ventilation of the laboratory. Secondly, a slow changing annual trend was also observed where Rn-222 content in the laboratory air was higher during summer and lower during fall and early winter. This pattern was related to the fact that the laboratory is located underground. In summer time, the air in the laboratory is colder than the surface level air and in winter time situation is reversed.

The comparison of background properties of different detector setup revealed great differences. The lead shielding were found to contain varying amounts of NORMs. The NORM contamination can increase the MDA’s significantly.

When calculating the final result the changing background during the measurement creates problems. If the background during a measurement is higher than during the actual background measurements too much counts are subtracted from the peaks and the NORM activities cannot be determined. On the other hand if too few background counts are subtracted unrealistic NORM activities are detected. Although, this only applies to NORM activities close to detection limit.
NKS comparison exercises have previously (and repeatedly) shown that improvements are needed in the use of gamma spectrometry. In the Nordic countries gamma spectrometry is used both by expert users, with highly sophisticated software, and by ordinary users using off-the-shelf commercial software. Even though it is easy to use the basic functions of a commercial software package, many find it difficult to use the advanced functions, e.g. correction of true coincidence summing (TCS).

The greatest opportunity for improvements in gamma spectrometry is in the case of non-expert users using commercial software. It was therefore decided to take a commercial off-the-shelf software, GammaVision 6.0, do a calibration including TCS as the manufacturer recommends and then participate in the NPL Environmental Radioactivity Proficiency Test Exercise 2008, organised by the National Physical Laboratory in the UK. The experience gained should be of relevance for other users wanting to use an off-the-shelf product (in this case Ortec’s GammaVision) for analysing gamma spectra.

For calibration a special mixture recommended by ORTEC was used, containing both single line and multiple line radionuclides, which produced TCS effects. The TCS library was created using the Ortec Nuclide Navigator software and the procedures recommended by Ortec. Some additional issues needed nevertheless to be taken into consideration: metastable daughter nuclides needed to be removed; the library needed to be checked so the first line of each nuclide was a clearly visible one; after further testing it was decided to remove all low energy (< 50 keV) and low yield (< 1 E-3) gamma rays from the library. Furthermore the country setting of the computer needed to be changed to “USA”. The library thus produced was used for the efficiency/TCS calibration. A simplified version with only the main lines of the parent radionuclides was used for the efficiency/FWHM calibration.

The NPL mixture measured was the “GL”; a ‘low-level’ mixture of $\gamma$-emitting radionuclides and an $^{152}$Eu point source was measured for accurate energy calibration afterwards. The analysis was done using the TCS calibration and a nuclide library including all potential radionuclides in the sample, as listed by the NPL. Low energy and low yield lines were removed from the library as before. Radionuclides not seen in the sample were then removed from the library and the analysis was rerun. Special care was needed when calculating the activity of $^{95}$Nb at the reference time due to the ingrowth from $^{95}$Zr.

Of the 78 laboratories that participated in all the tests, we were amongst the 12 that passed as having all results in agreement. It should be noted, however, that although this exercise tests the ability to deal with TCS and some other corrections, it does not test the ability to deal with attenuation and geometrical corrections.
CHEMICAL SEPARATION METHODS APPLIED ON COMPLEX WATER SAMPLES, IMPLICATIONS ON DETECTION LIMITS FOR GAMMA SPECTROMETRY MEASUREMENTS

J. Byegård, K. Nilsson, H. Widestrand, Geosigma AB, Sweden
M. Kronberg, Swedish Nuclear Fuel and Waste Management Company (SKB), Sweden

An in situ multi-tracer diffusion experiment (Long Term Diffusion Experiment, LTDE) has been performed at the Åspö Hard Rock Laboratory, Oskarshamn Sweden. In the experiment, chemical analogues or homologues to important longlived radioisotopes from spent nuclear fuel were used as tracers. The aim was to study the adsorption and rock matrix diffusion of these elements in a natural environment. For this purpose, an 11 m long borehole was drilled which cross-sected a natural fracture and the tracer cocktail solution was held in contact with the natural fracture surface for half a year. Information about the adsorption was obtained from sampling and measurement of the radioisotopes in the aqueous phase, and (in a later stage) from overcoring and measurement of the penetration depth of the tracers in the rock.

In the tracer cocktail, a total number of 15 γ-emitting tracers were added which were measured using HPGe γ-spectrometry in combination with commercially available evaluation software (GammaVision v 5.31). During the measurements, the following problems were identified:

- Tracers with γ-energy lines in the low energy part of the spectrum suffered from a Compton continuum “pile-up” due to some of the radionuclides with high γ-energies, causing a high background counting rate.
- Tracers that were adsorbed to the rock surfaces to a high extent, e.g., $^{153}\text{Gd(III)}$ and $^{175}\text{Hf(IV)}$, thereby became present in such low concentrations in the groundwater cocktail that they were very difficult to measure in a high background environment caused by less sorbing tracers, e.g., $^{22}\text{Na}^+$, $^{85}\text{Sr}^{2+}$ and $^{137}\text{Cs}^{-}$.

In order to improve the possibilities to measure the low concentration tracers by γ-spectrometry, some chemical separation techniques were outlined and tested. Some examples of tested separation methods are the selective adsorption of $^{137}\text{Cs}^+$ on ammoniummolybdenum phosphate, co-precipitation of $^{85}\text{Sr}^{2+}$, $^{133}\text{Ba}^{2+}$ and $^{226}\text{Ra}^{2+}$ in BaSO$_4$(s) and hydrolysis induced adsorption/precipitation of cations. However, the final technique chosen was a combination of DTPA complexation and eluation through cation exchanger (Dowex 50X8). In this separation process, the cations forming weak complex with DTPA (e.g., $^{22}\text{Na}^+$, $^{85}\text{Sr}^{2+}$, $^{133}\text{Ba}^{2+}$ and $^{226}\text{Ra}^{2+}$) are adsorbed in the cation exchanger while the strong complexation between the DTPA and e.g., $^{153}\text{Gd}^{3+}$ and $^{175}\text{Hf}^{4+}$ prevents the cations from interacting with the cation exchanger. A single step separation process was thereby obtained which could separate the vast majority of the low concentration tracer to a low background activity environment.

In the presentation, separation factors for the different processes will be given as well as the calculated detection limits; both for measurement before and after the separation process. A critical evaluation and general discussion of the benefits of the separation methods will be provided. There will also be a discussion on the applicability of the detection limits provided by evaluation program, in this case the GammaVision.
Within the framework of a programme for enhancing the national emergency preparedness, conducted by the Swedish Radiation Protection Authority, a project was launched in 2005 (project gGEO), with aim of denouncing a set of standard geometries to be used for emergency measurements of gamma spectrometry. The intention has been to have the standard geometries available and to used in emergency situations at all eight Swedish laboratories connected to the national emergency preparedness. Four different geometries, 60ml, 250 ml and 1500ml, with density 1, and one 1500ml-geometry with density 0.2, were proposed, requiring three different types of beakers; 60ml, 250ml and 1500ml with liquid tight covers. Beakers were subsequently purchased by the SSM and then distributed to participating laboratories.

A calibration standard manufacturer made reference sources of each of the four geometries using the standard beakers. The sources contained a cocktail of radionuclides, where each radionuclide (137Cs, 134Cs, 109Cd, 203Hg, 139Ce, 85Sr, 88Y, 241Am, 113Sn and 60Cs) had a specified uncertainty of 1.5% (1 sigma) in their activity contents. These sources were used by each participating laboratory to calibrate their HPGe-detector systems for various prone or remote source detector geometries.

Reference material from IAEA (dried wheat and soil) and NIST (coastal and river sediments) were subsequently purchased in 2007 to prepare validation samples for the standard geometries of the gGEO-project. The reference materials were repackaged into the three types of standard beakers. In all 14 validation samples were then made with various densities (0.7-1.45 g cm\(^{-1}\)) and degree of active volume. The samples were distributed to the participants together with a spreadsheet with all data on the sources. The participants were to report the deviation between their gamma spectrometry measurement and the certificate value, normalized to the total estimated uncertainty of the measurement.

The results showed that 48% of all reported data of the 14 validation samples fell within the 95% confidence interval around a zero-deviation. That is, in those cases the laboratories satisfactory determined the activity concentration and the associated uncertainty in their analysis. Three out of eight laboratories exhibited a significant (p>0.05) normalized deviation, averaged over all samples and radionuclides. A certain tendency to overestimate the \(^{137}\text{Cs}\) concentration was also observed. A systematic underestimation, as averaged over all laboratories, was found for a 1500ml geometry filled soil, with density at about 1.45. All laboratories except the two who performed some form of density correction obtained significant underestimations, thus indicating the benefits of such corrections.

Project gGEO provided Sweden with optimal conditions in terms inter-laboratory agreement. The validation has demonstrated that this approach will achieve an acceptable quality in about 50% of all measurement cases, taking into account differences in performances between laboratories or particular biases for certain radionuclides and matrix densities. However, no particular action towards a structured process for uncertainty estimation was taken. A general approach to also standardize the algorithm for how each laboratory carries out the uncertainty
estimates would probably be beneficiary for the quality of gamma spectrometry assessments, and enhance the overall performance still.
GAMMA- AND X-RAY ANALYSIS OF PU AND U IN SINGLE THULE PARTICLES

Per Roos & Sven Nielsen
Department of radioecology and tracer studies.
Risoe-DTU, Denmark.

Analysis of the plutonium and uranium isotopic composition of individual ‘hot’ particles originating from the Thule accident in 1968 has been conducted. The need to develop methods based on gamma- and X-ray spectrometry is necessary due to the refractory nature of these oxide particles, resisting chemical dissolution, and due to need of elemental/isotopic information without destroying the particles (eg morphological studies). This presentation discusses various advantages and disadvantages associated with gamma and X-ray analysis of Pu and U at these levels, such as choice of detector and energy lines, calibration strategies at different energy regions, spectral complexity and data uncertainty.
HRP offers gamma scanning as part of the post irradiation examination (PIE) program for experimental fuel rods. The advantage of the gamma scanning technique is that no damage is inflicted on the examined object.

The gamma scanning rig with the fuel rod installed is shielded in a hot cell. Remotely controlled, the rig allows for acquisition of both vertical and horizontal data points. The detector is positioned outside the hot cell with the collimator installed through the wall. Several collimator apertures are available and the rod-detector distance is variable in order to suppress the gamma flux and thus limit the detector dead time. Data collection and rig control is operated from a PC using software developed at the HRP. The full energy spectrum up to 2000 keV is collected, making it possible to retrieve data for any nuclide at a later date.

The fuel rod activity profile of $^{137}\text{Cs}$, determined using a slit collimator and axial rod motion, reflects the rod power profile. In one particular HRP experiment series, a fuel rod is installed within a sealed pressure flask and exposed to LOCA conditions. Following extraction from the Halden Reactor, gamma scanning provides a 2D image of the state of the rod with special attention at the $^{103}\text{Ru}$ and $^{137}\text{Cs}$ nuclides.

Generation of a counting efficiency calibration technique enables the determination of activity. A recently irradiated reference rod provides several gamma energies, on which the shape of the counting efficiency curve in the range from 0.1 to 1.6 MeV can be derived. A standard $^{137}\text{Cs}$ reference rod provides the level adjustment factor.
The role of quality assurance in gamma spectrometric measurements of radioactivity is discussed. It is based on the real examples of laboratory practice. The measurements of the gamma spectra of natural radioactive nuclides as well as other radionuclides in different environmental samples (soil, precipitation, different types of water, needles, etc.), various types of samples irradiated in the nuclear reactor and in the radioactive polluted objects are carried out in the laboratory. The laboratory is accredited in the Latvian National Accreditation Service (LATAK). The quality assurance system was implemented in our laboratory in 2000.

The credibility of obtained results is ensured by the quality assurance and control. The main requisitions involved in the quality assurance of the laboratory according to the requirements of ISO/IEC 17025:2005 are:
1) the use of calibrated equipment only;
2) the regular long-time use of reference materials for the control of equipment;
3) the estimation of uncertainty sources and determination of uncertainties within the given interval of credibility;
4) the validation and verification. The very important requirement is a regular participation in the interlaboratory intercomparison exercises, which enables one to estimate and disclose possible sources of non-conformities as well as to carry out the corrective actions.

Since 1999 laboratory is a regular participant in the interlaboratory intercomparison exercises organized by the RISO National Laboratory (Denmark) and IAEA (Vienna). Such nuclides as K-40, Mn-54, Co-57, Co-60, Y-88, Cs-134, Cs-137, Ra-226, and Th-232 were analyzed in following intercomparison samples: soils, sediment, sea weed, hay, meat, dray milk, lake and sea waters. The report presents the laboratory’s system of quality assurance and the process of its implementation. Our internal quality audit program covers all requirements of ISO/IEC 17025:2005 standard, but the main attention is paid to the analysis of results of laboratory’s participation in intercomparison measurements, their evaluation, interpretation and determination of uncertainty sources.

Only credible and justified results can be the basis for further use in any field, thus making it possible to make legitimate decisions.
GAMMA SPECTROMETRY AS A TOOL FOR GLOBAL DISARMAMENT BUILDING AN INFORMATION BARRIER.

Kjell Johansson, Institutt for Energiteknikk
Section head - Electronics – Department of Physics

The presentation is about a UK–Norway Initiative with the objective to develop new technologies, methods and procedures for verification of future multilateral and bilateral disarmament treaties.

In a future verification regime, it is unlikely that inspectors will enjoy unrestricted access to nuclear warheads. Such access would breach mutual non-proliferation obligations of the NPT, as well as reveal national security-sensitive information. An information barrier may be one solution, since it is designed to confirm mutually agreed attributes to the inspectors while preventing the disclosure of sensitive information. During the project two limited prototype devices utilising both a UK and Norwegian design has been built. The instruments were successfully tested in an exercise in June 2008.
GAMMA SPECTROMETRY AT RISØ FOR ENVIRONMENTAL RADIOACTIVITY

Sven Nielsen
Risø DTU, Denmark

Gamma spectrometry at Risø DTU has developed since 1960 when radionuclides in air filters were determined using a 4-inch NaI detector coupled to a 100-channel analyser. Risø started building own germanium detectors but stopped in 1974 when these became commercially available.

At present Risø uses 10 Ge detectors for routine work on environmental radioactivity including 2 Ge(Li) detectors, 2 well-type detectors and 4 low-energy detectors. Sample geometries cover volumes from 1-litre Marinelli beakers to vials holding less than a millilitre. Detector performance is monitored by regular checking of efficiency, energy resolution and background.

Gamma spectra are analysed using software developed at Risø in the early 1970’s. The software includes corrections for sample density and coincidence losses. The reliability of the spectrum analysis was compared in 1998 with other software packages and the results were found to be acceptable. Due to problems associated with maintaining the old software and missing documentation, plans are underway for changing to a commercial spectrum analysis system.
GAMMA SPECTROMETRY AT THE ACCREDITED LABORATORY OF STUK

Seppo Klemola
STUK - Radiation and Nuclear Safety Authority, P.O. Box 14, FIN-00881 Helsinki, Finland
seppo.klemola@stuk.fi

Since 1999 the method of gamma spectrometric sample measurements at STUK has been accredited according to the Standard EN ISO/IEC 17025. Scope of accreditation is defined as analysis of gamma-emitting radionuclides in energy range of 30-2700 keV in environmental samples, biological samples and foodstuffs. After reorganisation of the Department in 2007 all measurements in Helsinki have been performed at Radionuclide Analytics Laboratory and the Regional Laboratory in Northern Finland has continued analyses in Rovaniemi.

The facilities and equipment of the gamma spectrometric laboratory at Helsinki will be described. These include 10 HPGe spectrometers operated in two specially constructed counting rooms. Methods of spectrum analysis and detector calibration are introduced with the emphasis on newly implemented validated software UniSampo/Shaman. Integrated LIMS (Laboratory Information Managements System) databases are also described. Especially LINSSSI database offers new tool to manage large amount of data involved in spectrum analyses.

Applications of gamma spectrometry at Radionuclide Analytics are described. These include surveillance of environmental radioactivity in Finland, radioecological studies of both natural and artificial radionuclides, contracted services for industry, trade, organisations and institutes and emergency preparedness. Radionuclide Analytics is also one of the 10 Radionuclide Laboratories certified by CTBTO (Comprehensive Nuclear Test Ban Treaty Organisation). This status is demanding challenge for quality system, proficiency of personnel and preparedness.

One of the tasks of the Laboratory is to maintain measurement standards which ensure the reliability of radiation measurement. Measurement standards for activity determination of gamma-emitting radionuclides are accurate spectrometers with traceable calibration, validated methods, or radiation sources.
The Health and Safety Department possesses six HPGe detectors for the determination of gamma emitters in various materials. Four of these are low-energy detectors.

The section is responsible for control of radioactive discharges from IFE’s nuclear activities at Kjeller and for monitoring of radioactive contamination in the local environment. In connection with this are other work at IFE we analyse various type of samples: internal control samples, primary water, discharge water, environmental samples (water, sediment, biota, milk, precipitation etc.). These samples are analysed for anthropogenic radionuclides.

We also perform commercial analysis. Most of these samples are analysed for natural radionuclides. Samples include produced water and scale from the oil and gas industry. The detectors are calibrated using certified mixed radionuclide sources. Each detector is calibrated individually for each geometry in use i.e. our calibration is relative to the certified sources.

QA tests on background, efficiency and energy are performed at least every second week. Our radionuclide libraries are based on data from Laboratoire National Henri Becquerel, tuned manually towards our needs. We annually participate in intercomparison and proficiency exercises (IAEA, NIST, NKS, NPL etc.).
GENERAL OVERVIEW OF GAMMA SPECTROMETRY ACTIVITIES AT THE NORWEGIAN DEFENCE RESEARCH ESTABLISHMENT (FFI) GAMMASEM 2009

Hanne Breivik
Norwegian Defence Research Establishment (FFI)

Gamma spectrometry was used as a tool at the FFI up until the mid-1990s, when the activity was put on hold. In 2007 it was decided to revive this capacity, using the existing germanium detector. New electronics and software were procured, and FFI made the system operative in 2008.

The gamma spectrometry system will be used to analyse environmental samples from the Norwegian Armed Forces and in FFI’s research to support protection of military personnel. Gamma spectrometry is also an integrated part of FFI’s laboratory capacity to screen and analyse unknown and mixed samples, i.e. samples potentially containing one or more of hazardous biological, chemical and radioactive substances.
IMPLEMENTING COMBINED UNCERTAINTY ACCORDING TO GUM INTO A COMMERCIAL GAMMA SPECTROMETRIC SOFTWARE

H. Ramebäck¹, A. Tovedal¹, U. Nygren¹, P. Lagerkvist¹, J. Boson¹, E. Johansson¹, J. Johansson²
¹ Swedish Defence Research Agency, FOI. Division of CBRN Defence and Security. SE-901 82 Umeå. Sweden
² Swedish Radiation Safety Authority, SSM. SE-171 16 Stockholm. Sweden

Measurement uncertainty is an important part of a measurement result that still often is neglected. A complete combined uncertainty budget can be calculated for non-routine measurements. However, for routine measurements, this work becomes time-consuming since every measurement result requires an uncertainty analysis. By analysing the uncertainty on a measurement system level in e.g. high resolution gamma spectrometry, the uncertainty analysis will be universal for a particular measurement geometry. The problem is then reduced to implementing the combined uncertainty into measurement software. This work shows how this analysis can be done and implemented into a commercial software for gamma spectrometric measurements.
The gamma spectrometry lab of the Swedish Safety Authority took part in the NKS intercomparison project called REMSPEC among national laboratories in the Nordic countries. The task for the intercomparison was to analyse a synthetically generated gamma-ray spectrum of a complexity like expected of spectra post nuclear power plant accidents. The results of our analysis made us re-evaluate settings of the analysis software, including the calibration for the complex spectrum analysis and to implement the correction for true coincidence effects.

We relate our experience with analyzing the complex spectra, describe the changes in the analysis and compare the results obtained without and with applying the cascade summing correction. The commercial gamma analysis software we use is Genie (v. 3.1).
This presentation pertains to an exercise conducted as part of the NKS-B programme in which a synthetic gamma ray spectrum was used to simulate the type of data that may be encountered in the early phase of a nuclear accident for the purposes of conducting an analytical exercise. The aim of the activity was to provide participants with an opportunity to exercise in the type of situation and with the type of data that may result after a nuclear accident. The exercise was an attempt to address the fact that analyst demographics tend to be skewed towards those who may never have had the opportunity to analyse complex "fresh fallout" type spectra and the fact that many countries are less able to practice with complex spectra from high activity samples due to their non-nuclear status.

Attempting to conduct such an exercise internationally using actual samples presents practical and logistical difficulties and a synthetic spectrum was employed to negate some of these problems. The spectrum contained a number of typical fallout isotopes and was distributed in a range of formats, along with calibration information, to the participant laboratories. The scenario chosen was a hypothetical large scale release from a nuclear power plant with subsequent air sampling and measurement by HPGe. The participants were required to submit results within three hours of receipt and with the option of submitting further results within one week. The results provided by the laboratories indicate that all laboratories were able to identify and quantify some of the isotopes but only some labs were in a position to identify and quantify virtually all the constituents of the spectrum. Results indicate that there remain some problems with aspects such as true coincidence summation and using file formats or routines with which labs may not be familiar with. The exercise provided a useful opportunity in exploring the possibilities of using synthetic spectra for exercise purposes and offered participants the chance to practice with the sort of scenario that may result after an accident.
NUCLIDE SPECIFIC SURFACE ACTIVITY MEASUREMENTS AT RINGHALS NPP

Patrik Konnéus
Ringhals NPP

The deposition of radioactive corrosion products on surfaces of pipes, vessels and heat exchangers, is one of the most important problems within the reactor environment at an NPP. Increased deposition of corrosion products causes a limited access to the components of the reactor systems due to high radiation levels in spaces close to the components.

Controlling the build up of radiation levels in the reactor systems is one of the main tasks for chemistry and radiation protection departments of NPPs worldwide. To perform this, the chemists must study the whole mechanism of the corrosion process in the reactor systems, the cooling water and material parameters and the deposition process of the radioactive corrosion products on the surfaces. However, the possibility of analyzing samples in the laboratories is limited because the system surfaces are not accessible for sampling.

Therefore, a nondestructive method for analyzing surface activity in the reactor systems using movable HPGe detectors was developed at Ringhals NPP. The method has been used since the early 1990s.

In this presentation, the method including equipment, detectors and calibration procedures will be described. The equipment consists of a movable HPGe detector with a relative efficiency of 4-8 % and a collimator in lead on wheels. The absolute efficiency is calculated on the basis of a surface source calibration, and a program developed by Ringhals called Simba does the geometrical correction of the efficiency. Results from a campaign of measurements are discussed between several departments with interests in health physics, chemistry and core management to follow up changes in the reactor cycle. Examples of results and trends will be shown graphically.
There are a number of factors, which contribute to the total measurement uncertainty (TMU) in spectrometry, especially in Gamma Spectrometry. Among them are the statistical counting uncertainty, the calibration uncertainty, uncertainties in nuclear parameters and any sample preparation uncertainties like random errors or bias.

If the efficiency calibration is performed on the basis of calibration source, this part of uncertainty is in the range of some few %. For mathematical calibrations, the uncertainties are a bit higher at low energies, but have other advantages as explained later. State of the Art Gamma Spectroscopy packages combine the individual uncertainties and generate an overall uncertainty estimate.

Depending on the given measurement conditions like counting time, sample geometry and source activity, the TMU can be any value between 1 and 100% or even more.

One very important source of uncertainty in laboratory Gamma Spectrometry is the Cascade Summing Effect. This effect has a high geometry dependence and is nuclide and even energy line dependant. Genie-2000, the proprietary software of Canberra, has integrated algorithms to correct for these effects. It will be shown in a demonstration example that this effect can be very significant and how it can be corrected in Genie-2000.

Another source of uncertainty arises from differences in the physical properties between the calibration source and the real sample. Incomplete knowledge of sample properties, like density, filling height in a bottle or wall thickness of a container contribute to the TMU, too. Genie-2000 includes a tool (named IUE – ISOCS Uncertainty Estimator) to calculate the additional uncertainties which arise from those not well known physical characteristics. Also, this program can be run in a mode, which gives information about the more or less critical parameters for determining the activity of the sample. In the paper, an example will be given, which demonstrates the steps required to get meaningful information and the magnitude of uncertainty arising from not well know sample parameters.
In situ gamma-ray spectrometry has since the introduction of portable germanium detectors been a widely used method for the assessment of radionuclide ground deposition activity levels. It is, however, a method that is most often associated with fairly large and poorly known uncertainties. In this work an uncertainty analysis of in situ gamma ray spectrometry in accordance with GUM is presented. The analysis is based on a Monte Carlo method known as Latin Hypercube Sampling (LHS). LHS makes use of stratified sampling of parameter distributions to reduce the large number of samples often needed for Monte Carlo calculations. The uncertainty analysis takes into account uncertainty contributions from e.g. the calibration of the detector system; the assumed activity distribution in soil; soil and air density; and detector positioning.
The project GammaSem was proposed to provide a forum for discussions and sharing of information on practical issues concerning gamma spectrometry and to establish a network of users of gamma spectrometry in the Nordic countries, thereby strengthening the collaboration and improving all participants’ competence in practical gamma spectrometry.

The seminars’ focus was practical challenges met by the users themselves, rather than theoretical matters. Scientists and users of gamma spectrometry from all five Nordic countries were invited to the seminar, as well as scientists from the Baltic countries. A total of 75 people participated; representing 34 different universities, commercial companies, research institutes and also all Nordic authorities.

During the seminar several key issues for follow-up were identified and working groups for addressing the identified problems were established. The working groups were:
- Uncertainties and detections of limits
- True summing coincidence
- Monte Carlo simulations and efficiency transfer
- Absorption (density corrections and geometries)
- Mobile gamma spectrometry systems
- Nuclear forensics (on special samples and special parts of the spectra)

The identified topics will form the basis for the agenda of the next seminar in 2010. There, the different working groups will be invited to present their ideas/solutions to the relevant problems.