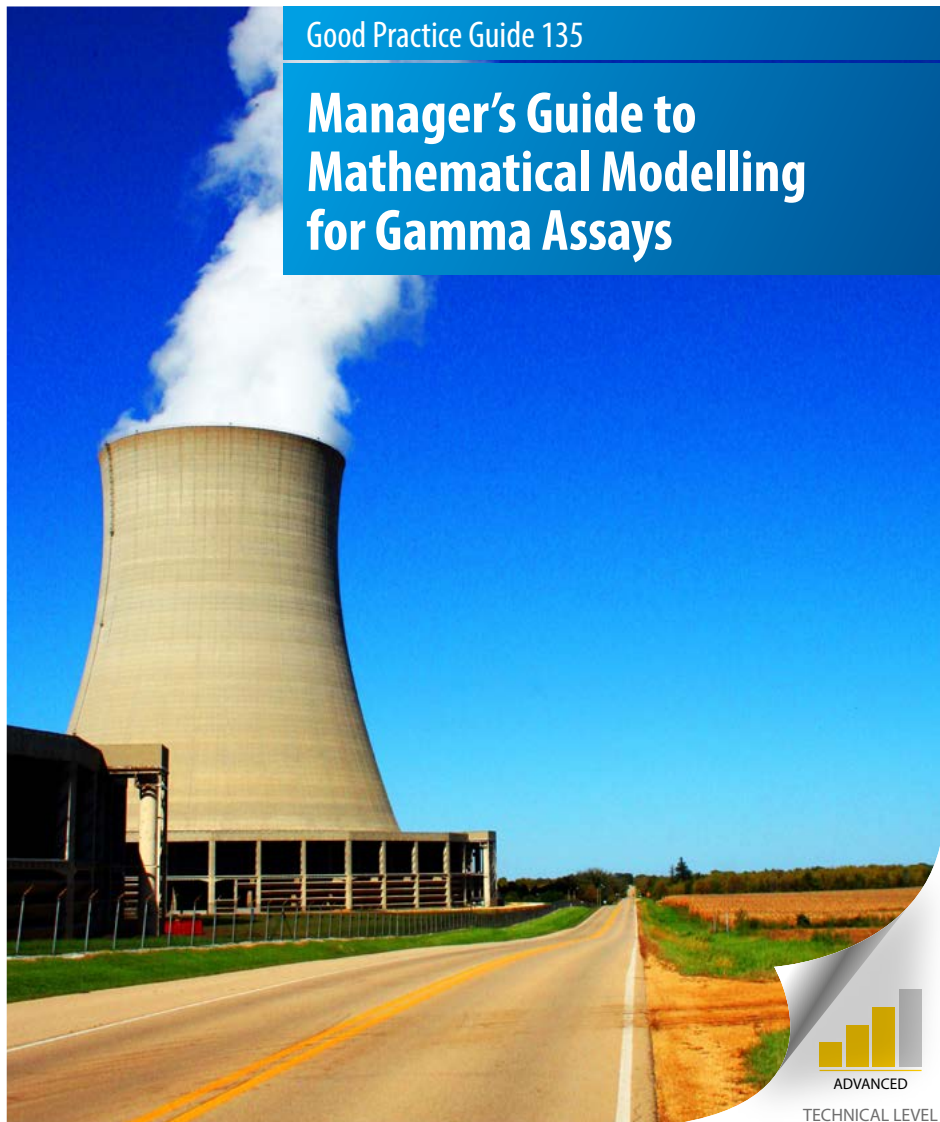




Good Practice Guide 135

Manager's Guide to Mathematical Modelling for Gamma Assays



Good Practice Guide 135

Version 1.0

Printed in the United Kingdom, 2015

© Queen's Printer and Controller of HMSO, 2015

ISSN

1368–6550

NPL Authors

Mark Bailey

Julian Dean

Andy Pearce

Graeme Taylor

Other Contributors

Pete Burgess (*Radiation Metrology Ltd*)

Barrie Greenhalgh (*Sellafield Ltd*)

Trevor Hatt (*AMETEK ORTEC*)

Bob Major (*AMEC FOSTER WHEELER*)

Neil McMillan (*AWE*)

Ian Pearman (*Nuvia Ltd*)

Martin Rushby (*Canberra UK Ltd*)

This Good Practice Guide has been written by a working group of expert users, technical support specialists, instrument suppliers and NPL specialists.

National Physical Laboratory
Hampton Road
Teddington
Middlesex
TW11 0LW

www.npl.co.uk/contact

Good Practice Guide enquiries: **020 8943 6606 / gpg@npl.co.uk**

Find out more about NPL measurement training at **www.npl.co.uk/training**
or our new online study programme at **www.npl.co.uk/e-learning**

Front Cover Image: *Cooling tower at the Byron Generating Station, a nuclear power plant in Illinois, US (Image courtesy: Thomas Anderson).*

Guide Information

What is it about?

This guide describes various aspects of mathematical modelling procedures used for non-destructive (gamma) assays of radioactive materials. It discusses input data required by the models, types of models, the various sources of uncertainty, and also quality and training issues.

Who is it for?

The guide is for project managers and others with responsibility for commissioning the measurement of radioactive materials in the nuclear industry, in particular for the management of potentially radioactive waste from the decommissioning of nuclear sites. Those involved in the actual measurements may also find it useful.

What is its purpose?

The purpose of the guide is to provide an overview of the use of mathematical modelling procedures in the measurement of radioactive materials such as waste arisings. It should be used in conjunction with the advice of technical experts.

What is the pre-required knowledge?

The guide assumes that the reader has little or no working knowledge of modelling, although a basic knowledge of the principles of radioactive decay is assumed.

Key to icons:



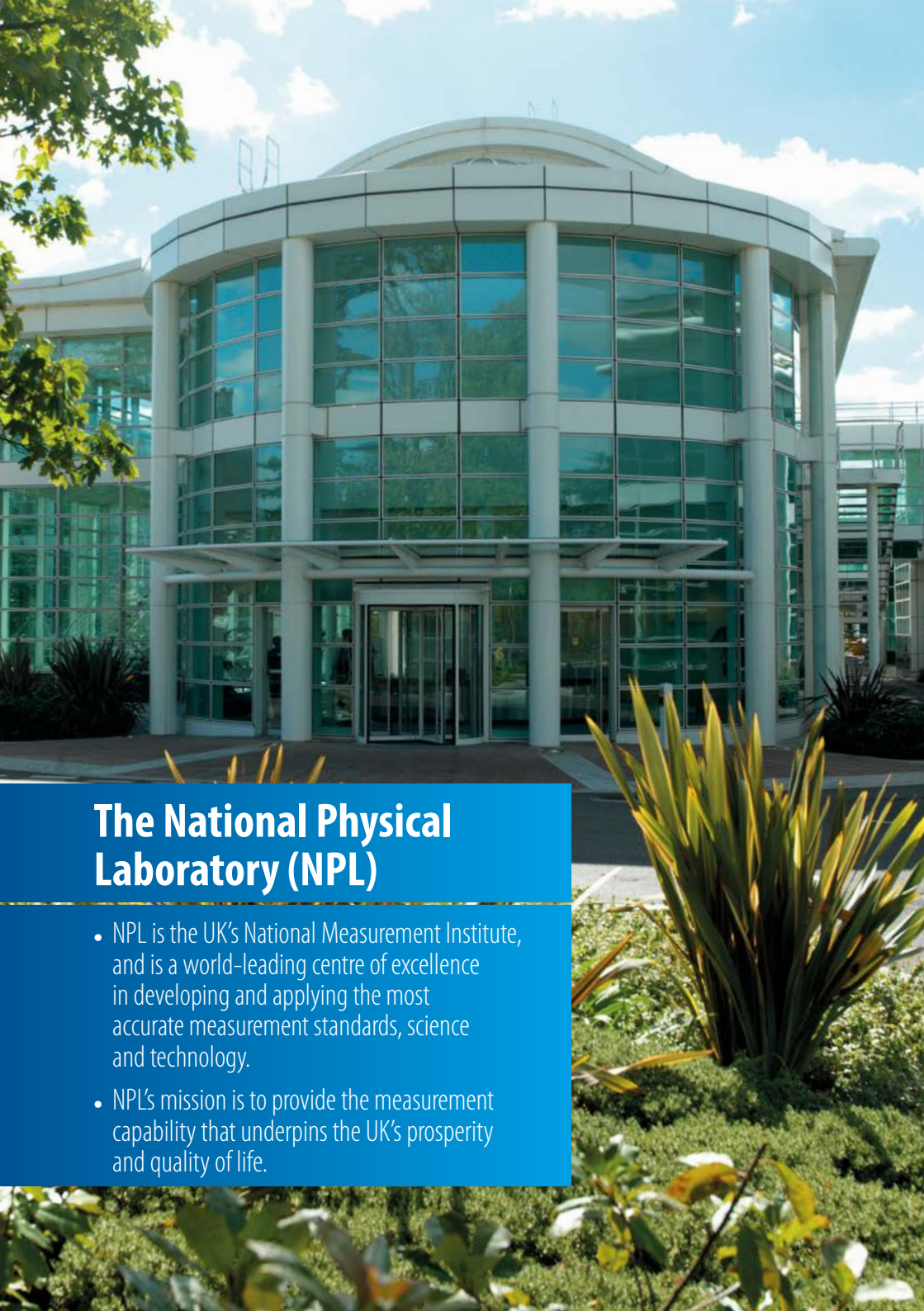
Need to know



Good to know



Checklist



The National Physical Laboratory (NPL)

- NPL is the UK's National Measurement Institute, and is a world-leading centre of excellence in developing and applying the most accurate measurement standards, science and technology.
- NPL's mission is to provide the measurement capability that underpins the UK's prosperity and quality of life.

Contents

Foreword	07	Chapter III	
Defining the need for mathematical modelling for gamma assays	08	Types of models	33
Chapter I		Brief introduction	34
Overview of use of mathematical models for gamma measurements	11	Monte Carlo methods	34
Gamma-ray spectrometers and their calibration	12	Deterministic methods	35
Use of modelling	13	Modelling assay systems	35
Modelling limitations and issues	14	Chapter IV	
Chapter II		Uncertainties	37
Information needed for modelling	17	Brief introduction	38
Brief introduction	18	What is uncertainty?	38
The output required	18	Sources of uncertainty	41
Information about the waste form	19	Practical implications	42
The waste container	19	Uncertainty due to activity distribution	42
The composition of the waste	20	Stable background	43
Degree of fill and waste density	21	Chapter V	
Activity distribution within the waste	22	Quality and training	45
Detectors and environment	24	Brief introduction	46
Detectors	24	Quality Assurance	47
Detector environment	26	Benchmarking	48
Knowledge of radionuclides present	27	Training and competency	49
Interference corrections and fingerprinting	27	Documenting and reporting	50
Other useful information	28	Record keeping	53
Reference data	29	Further Reading and Engagement with NPL	57
Nuclear decay data	30	Glossary	58
Photon and electron interaction data	30	References	59
Other recommendations	30	Contact NPL	61
Summary	31		

Foreword

Mathematical modelling has applications in a wide range of disciplines. One very specific area where modelling is applied is in the measurement of various types of radioactive items in the nuclear industry. Such applications may include the measurement of radioactive waste packages, plant or process design (where there is a requirement to measure process output or performance) and verification measurements.

This guide covers the use of modelling for such applications (the waste measurement application being cited frequently as an example), and is aimed primarily at a Project Manager (PM), a Bid Manager and others who have a responsibility for delivering a project within a fixed budget and timescale. It is intended to provide them with a basic grasp of modelling so that they can better understand the needs of technical staff. It is concerned specifically with modelling for the measurement of γ -emitting radionuclides and deals with modelling from the simple (e.g. spreadsheet-based) to the complex (e.g. Monte Carlo-based). Guidance is given on the type of information the modeller will need from the PM and others, the various types of models they might use, factors contributing to measurement uncertainties and the importance of using good quality nuclear decay data. The quality aspects of the subject are also covered (e.g. the importance of 'benchmarking' of models, staff training and comprehensive documentation).

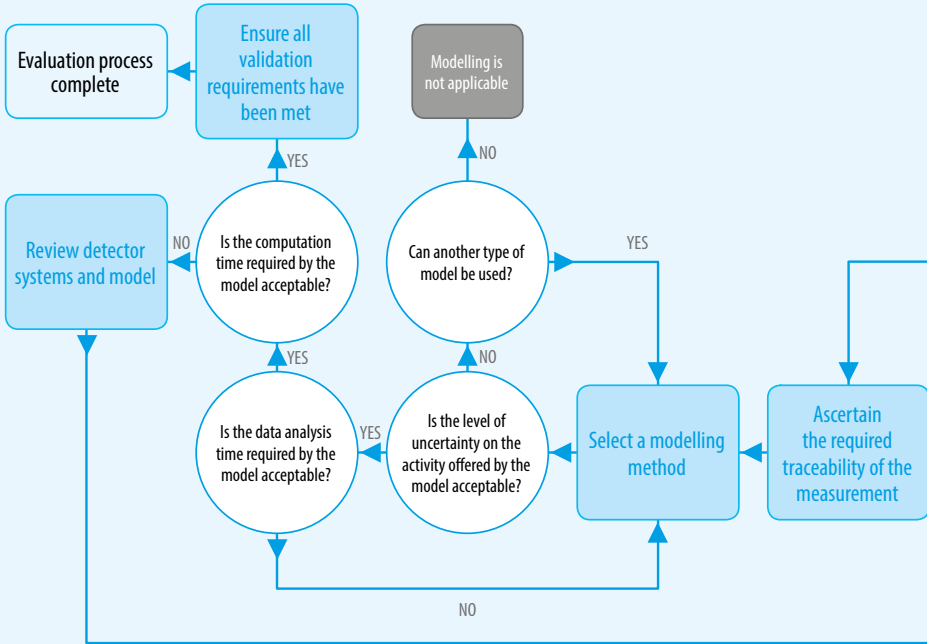
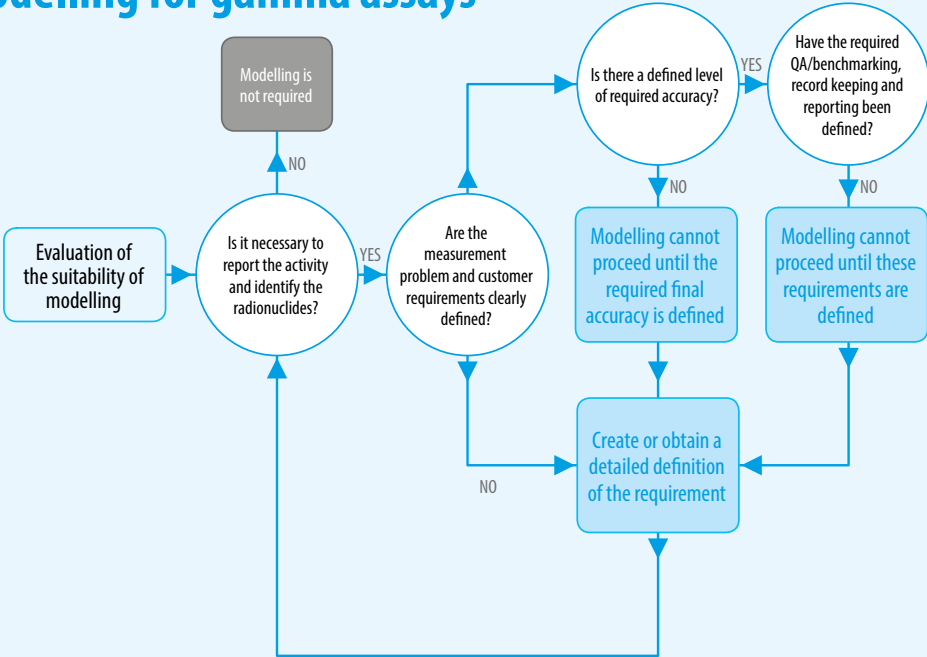
The guide is intended to complement existing publications in this field, such as Measurement Good Practice Guide No.34 ('Radiometric Non-Destructive Assay')¹ and 'A Good Practice Guide for the Use of Modelling Codes in Non Destructive Assay of Nuclear Materials'². It is intended only to provide an overview of the subject and the reader is encouraged to consult with technical experts throughout any project.

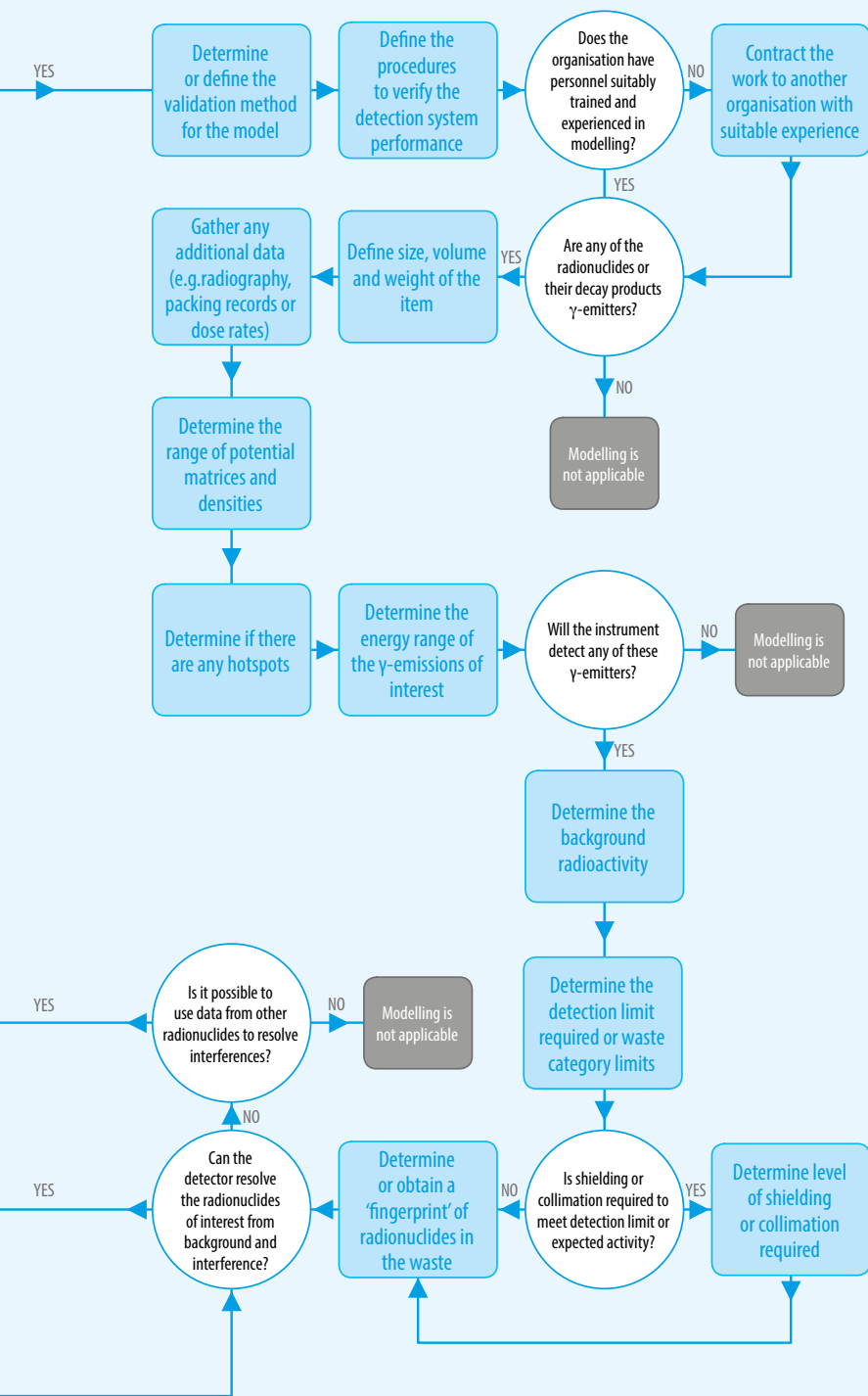
The **flowchart overleaf** gives guidance on whether modelling is applicable to a particular measurement scenario. It should be used in conjunction with later chapters of this guide and should aid discussions with technical staff.

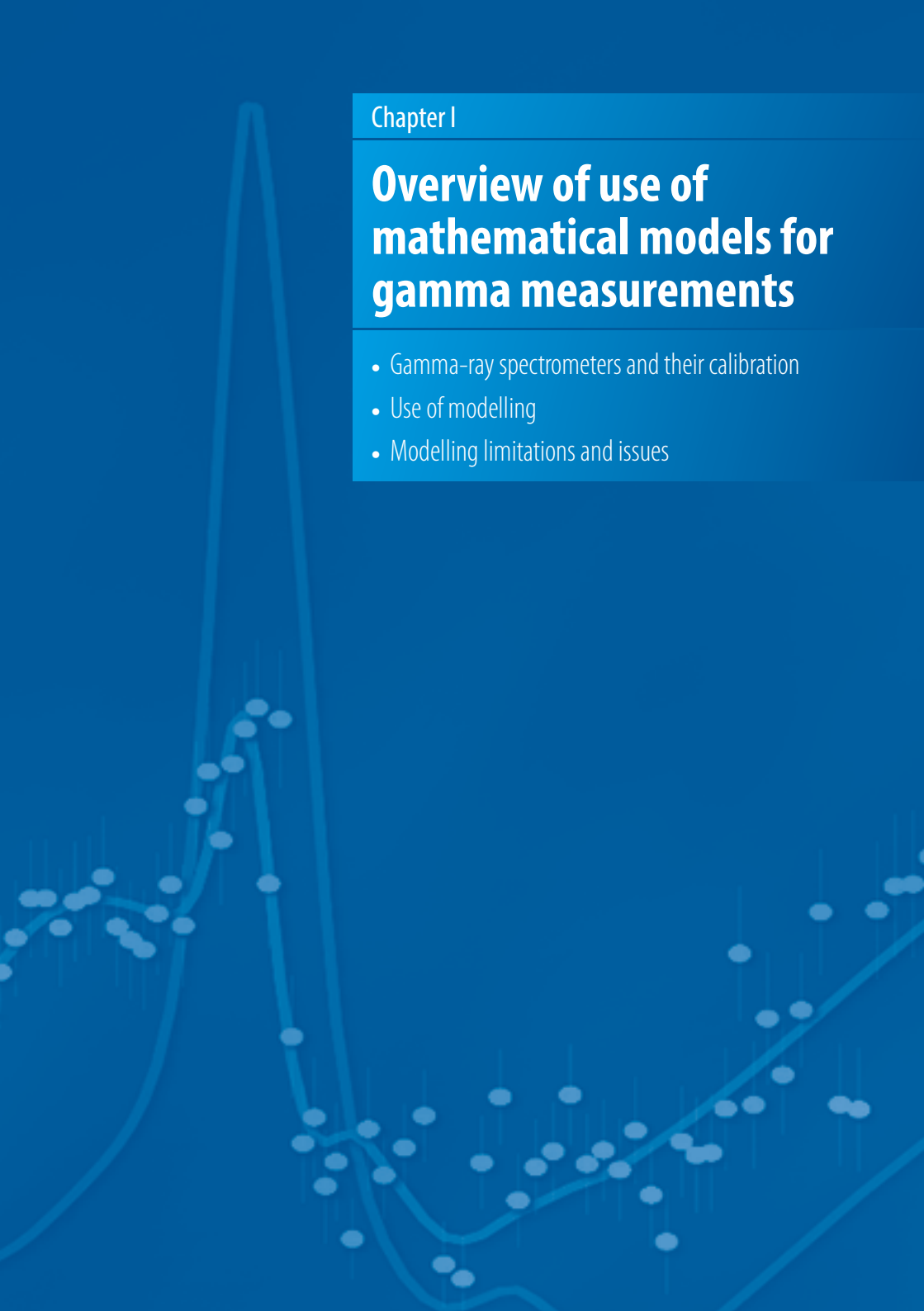
Dr Julian Dean,

*Senior Research Scientist, Radioactivity Group,
Acoustics and Ionising Radiation (AIR) Division, NPL.*

Defining the need for mathematical modelling for gamma assays







Chapter I

Overview of use of mathematical models for gamma measurements

- Gamma-ray spectrometers and their calibration
- Use of modelling
- Modelling limitations and issues

Gamma-ray spectrometers and their calibration

This chapter very briefly summarises the types of gamma-ray spectrometers which are available, their calibration and the advantage and limitations of modelling.

Methods for the measurement of γ -emitters are covered in many texts, such as Gilmore³, but essentially there are two principal techniques:

- High-Resolution Gamma Spectrometry (HRGS)
- Low-Resolution Gamma Spectrometry (LRGS).

HRGS systems are usually built around detector crystals formed of hyperpure germanium (HPGe) whereas LRGS systems are generally based on crystals of thallium-activated sodium iodide (NaI(Tl)). Note that other detector materials are increasingly used, such as cadmium zinc telluride (CZT) semiconductor detectors, and lanthanum bromide (LaBr₃), bismuth germanate (BGO) and plastic scintillation detectors.

Some systems are laboratory-based, containing single or multiple detectors; many are fitted with movable, collimated detectors which 'scan' the sample. In addition to fixed systems, many portable units are now in use.

The usual method of calibrating a γ -detector is to use standard radioactive sources of known activity in containers identical to those used for samples. The standard sources used must produce γ -emissions which span the energy range that will be encountered with actual samples, and the matrix (i.e. the bulk material surrounding or supporting the radioactivity) must match that of the samples. The standard is then measured in the same counting geometry as the samples (i.e. in the same position relative to the detector). The observed count-rates for the various γ -peaks from the standard can then be used in conjunction with the known activities of the radionuclides present to derive a detection efficiency calibration curve for that particular container type, matrix, counting geometry and detector system. If any of these parameters are subsequently changed, another calibration has to be undertaken, possibly involving the manufacture of a new standard source.

Calibration of a detector for the wide range of container types and materials encountered in industrial environments would require an extensive range of calibration standards; even then, a sample dissimilar to any of these standards may be encountered. In such circumstances, the laboratory could process the sample in some way to make it match an existing standard. Alternatively, the sample could be measured as it is and a judgement made as to whether the sample is sufficiently similar to one of the standards for an existing calibration to be valid. All of this would represent a significant investment in time and resources, and is usually impractical. Discussions with the customer at the early project planning stages (e.g. to discuss

measurement capability and sensitivity for the sample types that may be generated) are essential, and may result in some experimental work to generate suitable standards.

Use of modelling

Using modelling, it is possible to determine the response of a detector to a very wide range of:

- **Sample matrices and densities** (e.g. soils, concrete, metals, plastics and varying mixtures of these and other materials)
- **Sample container shapes and volumes** (e.g. from millilitres to cubic metres)
- **Measurement geometries** (e.g. from very close to the detector to many metres away with any of a range of materials in between, or even immersed in water or another material)
- **Activity distributions within the sample** – from a uniform distribution ('homogeneous') to activity as 'point sources' ('heterogeneous').

Both simple and complex models are possible. Once the model has been set up, it is very simple to change the parameters and thereby determine the change in detector response. In this way, it is possible to optimise the measurement configuration when designing a plant-based system. It is also easy to accommodate unexpected changes in sample types and so process 'non-standard' sample types.

It is important to note that modelling can be used not only to establish a 'calibration' for a range of sample types/shapes but also to help establish the most suitable detector for a given sample type.

Other important benefits from modelling are:

- The capacity to build a system to measure radioactivity in items that could not be measured using traditional empirical techniques
- Minimising the use of radioactive materials to establish calibrations, thereby minimising radiation doses to staff and the cost of disposing of the materials
- The capacity to define system responses to a wide range of variables (e.g. to engender confidence in a particular application or to demonstrate professionalism)
- The ability to analyse a large volume of bulk material in situ rather than have to take many sub-samples for laboratory analysis – this reduces sampling and analysis costs and avoids the problem of 'non-representative' sampling
- For the 'waste' application, the ability to segregate wastes between categories

more effectively (and thereby reduce waste volumes) by generating sample-specific calibrations

- The ability (e.g. at the project bid stage) to answer questions regarding detection sensitivity and measurement viability with a high degree of confidence
- Savings of cost and time (e.g. by avoiding the need for sampling and subsequent analysis).

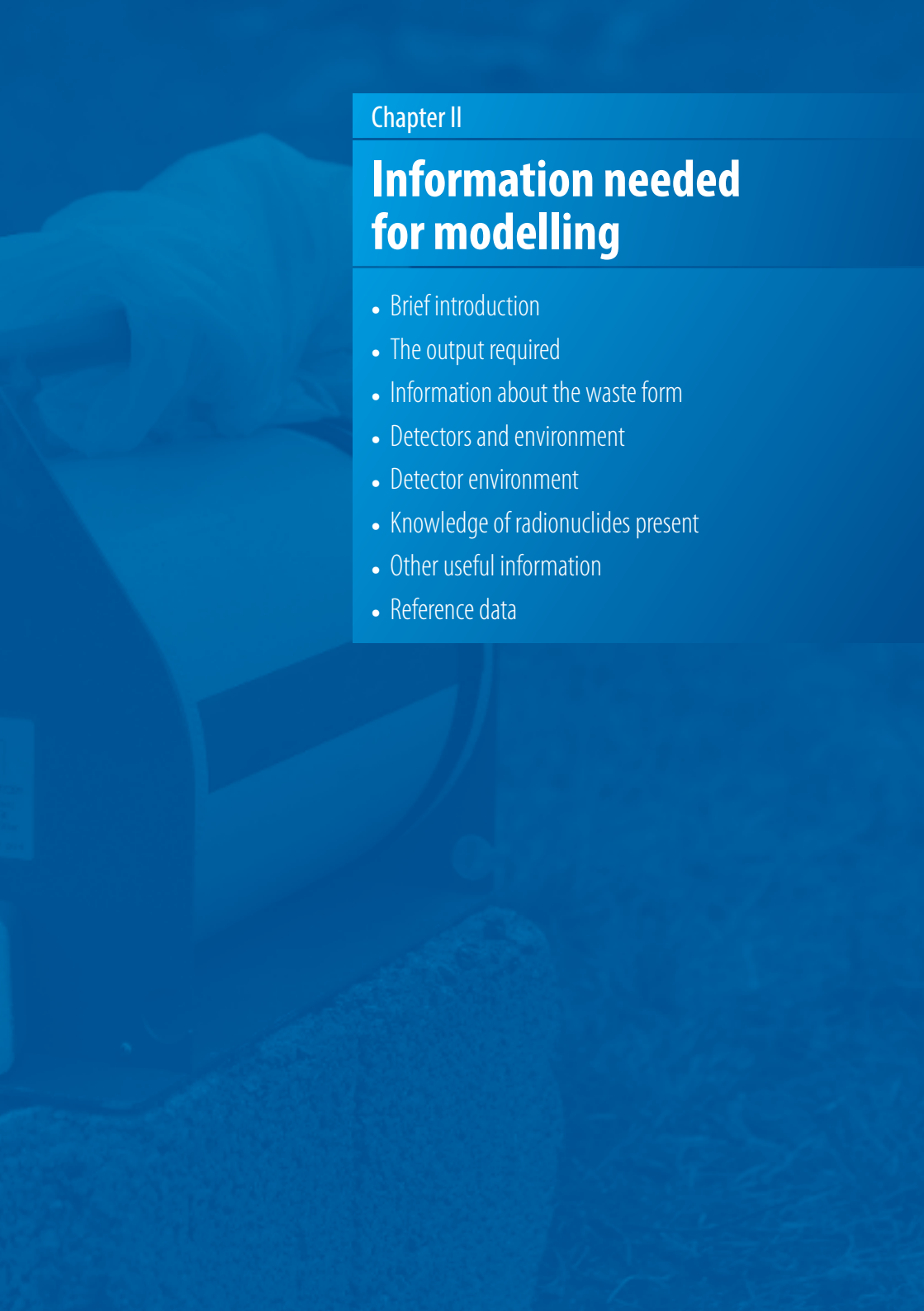
Modelling limitations and issues

There are, however, important issues with modelling that the PM should bear in mind:

- There is a limit as to how well the model can represent the measurement configuration. Some modelling software can allow quite complex configurations to be represented, whilst others may be limited in their capabilities (e.g. it may not be possible to include more than two intervening materials between the source and the detector). Some software allows the user to input materials (and combinations of materials), whilst others may have only a limited library, restricting the user. Before opting for modelling, the PM should first ascertain whether the modelling technique proposed or offered is fit for purpose. Whilst there are no guarantees with any technique, a competent modeller will be able to provide some indication of how well their model will reflect the actual measurement configuration (and perhaps more importantly indicate where it will not).
- The phrase '*rubbish in, rubbish out*' applies to modelling. Assuming the software has no limitations on representing the measurement configuration, the inputs to the representation must be based on accurate and reliable information (where this is unavailable, educated guesses or assumptions will need to be made). The modeller may seek such information via the PM, who should either provide the data or provide a range of possibilities. The PM should see in the output of the modelling process a report of the data used with the model including any assumptions made. One must be wary if this is not present, as it may lead to issues regarding justification of any results based upon it.
- Any detector for which modelling has been undertaken will have had its detection characteristics defined. Demonstrating that these have not changed over time will be a requirement and will need to have been planned into the project.
- Some form of 'benchmarking' will be required to demonstrate that the output is valid. This might consist of a comparison with a calibration carried out using

radioactive sources (for simple configurations) or a comparison with more sophisticated modelling software (for complex configurations). The type of benchmarking required depends on the application and the practicability of doing it.

The above points will be developed further in later chapters.



Chapter II

Information needed for modelling

- Brief introduction
- The output required
- Information about the waste form
- Detectors and environment
- Detector environment
- Knowledge of radionuclides present
- Other useful information
- Reference data

Brief introduction

The information required to define a measurement configuration depends on the modelling technique and software. Not all the information presented below as being required will always apply (i.e. less sophisticated modelling techniques and software will not allow the user to incorporate the information even if it is available). However, the more information that is available, the more representative the model will be. Disadvantages are that the analysis of uncertainties will be more protracted (though this will add to the model's validity), and, given that the model will be more complex, there will be a greater possibility of errors. Conversely, simple models may be inflexible.

The output required

The most likely first questions asked by the modeller will be:

- What do you want to know, and by when?
- How will the results be used?
- What financial resources are available?
- Who are the points of contact for further information?

If the measurement system is required to provide an activity (or activity concentration) value, then the output will be expressed in either Becquerels (Bq), Bq l^{-1} or Bq g^{-1} . The units required will be those dictated by the relevant authorisations or compliance documentation being worked to. Note that the data generated by the measurement system may be added to later (e.g. the measurement system will only be able to detect γ -emitters, so further work may be required to establish α and/or β activity if present, enabling a total activity assessment to be made).

Sometimes, the required output will be a 'Yes' or 'No' (e.g. is the sample radioactive or not). In such cases, the modelling requirements will differ in that the uncertainties will need to be defined to enable an upper limit to be calculated. If this upper limit lies below the 'radioactive' limit, then the answer will be 'No'.

In many cases, the requirement will be to model the response of a detector to containers of radioactive waste. This is discussed below as an example, although similar principles apply to other applications.

Information about the waste form

This includes the following:

- The waste container
- The composition of the waste
- Degree of fill and waste density
- Activity distribution within the waste.

The waste container

The waste container is usually defined at the beginning of a project and is governed by the acceptance criteria of the waste repository or upstream processing plant. Within the nuclear industry, a typical container type is a 200 litre (nominal) waste drum. Drum types vary considerably in specification (e.g. 205, 210 and 220 litres drums are all known as '200 litre drums'). The modeller will need to know exactly which type will be used and whether it contains anything unusual in the way of liners or materials not normally associated with standard waste drums. Other container types are used, and can be modelled (e.g. 1 m³ 'builder's bags' for large-volume wastes such as soil and concrete rubble). However, note that these too can vary in size - if the modeller is expecting 1 m³ bags to be used but 1 Te bags are used instead, then the same model will not apply and the answers will be wrong.

Individual objects can be modelled and, depending upon the sophistication of the modelling software, can either be approximated to a set of templates within the software or modelled more precisely by inputting the dimensions and material composition.

It is possible to model any size of waste container, including very large ones like half-height ISO containers ('HHISOs'). The main problems with very large volume containers are that it becomes more difficult to define the enclosed material, and the distribution of activity within the container may vary considerably. If the waste were to consist of only, say, one material type (e.g. contaminated concrete), then there would be no problem with material definition - only one material type would be present, and it would be a simple parameter to control operationally whilst filling the containers. However, some work would have to be undertaken to show that the activity was distributed more or less uniformly throughout the bulk of the concrete. This aspect is further discussed later in this chapter.

The composition of the waste

Waste can consist of any of a wide variety of materials (e.g. concrete, plastic, metal, wood, paper, etc). Furthermore, there are variations within each material group, for example:

- ‘Plastic’ could be PVC, polyethylene (high density or low density), polypropylene, polystyrene, etc.
- ‘Metal’ could be steel (various types), copper, aluminium, etc.

These various forms can be modelled provided they are known. However, it is highly unusual to know the waste composition to this level of detail and the best information normally available for waste is percentage (by weight) composition data (e.g. 30 % steel, 40 % plastics, 20 % paper and 10 % wood). In most cases, such proportions are estimated rather than measured and the modeller may opt to undertake a sensitivity analysis. This is done by varying the material types (and their proportions) in the model and observing how this changes detection efficiencies in order to obtain an uncertainty (Chapter IV, ‘Uncertainties’, p. 37). In most such cases, the material variations do not generate large uncertainties, but they can be significant if the waste composition includes a high proportion of high-density materials, or materials with high atomic number, and the nuclide(s) of interest are low- to medium-energy γ -emitters.

If the waste type and composition are poorly understood or unknown, significant time and effort will need to be expended by the modeller and others. For poorly-understood waste, the modeller will need to spend much time understanding potential compositional variations and their effects; when the material content is unknown or the confidence in the available information is poor, additional work will be required to understand the material. This might be achieved by opening a sample of drums and examining the contents, or by carrying out radiography on the drums. If this is not possible, pessimistic assumptions would have to be made, frequently leading to large overestimates of activity.

If the PM is in a position to control how waste is generated, the accuracy of the results can be improved. The simplest measure is to segregate similar materials into the same drum, although there will be other considerations to bear in mind, such as weight; for example, a drum containing only metals will be very dense and is likely to be incompletely filled (‘Degree of fill and waste density’, p. 21) so as not to exceed weight limits. This will result in a waste sample for which the detector may not be sensitive enough to achieve the required detection levels; moreover, it would not make good use of the available volume. Consequently, some mixing of waste types is inevitable for an efficient waste packing operation. This is acceptable provided an accurate record of the contents is available.

Degree of fill and waste density

In an efficient waste packing operation, the aim will always be to fill the waste container to capacity. It maximises the efficient processing of waste and reduces waste disposal costs. It is something that the client, the regulators, and the waste repository will expect to see carried out. The measurement configuration will be chosen to maximise the detector's response, which for a single static detector will mean the detector is 'aimed' at the mid-point of the waste container. This will coincide with the mid-point of the waste fill within the container. In the absence of other information, the modeller will assume this to be the case.

If the waste container is only partially full, not only will the modelled calibration be incorrect, but the detector measurement position will not be optimised. This aspect is less critical for systems involving multiple detectors or detectors that vertically 'scan' the waste, though their detector's responses still need to be adjusted according to the degree of fill. In spite of best efforts to ensure the above, there will be instances where the waste container is not filled to capacity (e.g. the final waste container from one area may not be completely filled and it may not be permissible to mix it with waste from a different area). Provided the modeller is made aware of these particular containers, the calibration (and possibly the measurement position) can be adjusted accordingly.

The modeller will be interested in how the materials are distributed within a container. The simplest case is when it is assumed that all the materials are mixed uniformly throughout the container volume. This is rarely the case in practice, but if measurements are carried out appropriately (e.g. by rotating a drum during measurement and measuring at a distance), uncertainties due to making this assumption can be minimised. However, extreme cases should be avoided; if they cannot, the modeller will need to know how the materials are distributed in order to generate an appropriate detector calibration. An example is when materials are added to a container as layers (e.g. a drum is partially filled with plaster and then plastics are added in order to fill the drum without exceeding the drum weight limit). The γ transport characteristics will be very different for each layer, and a model in which the two materials are uniformly mixed will not be an accurate representation of the sample. Another complication is that the *activity* distribution within the drum may not be uniform – e.g. the activity in the plaster may be different from that in the plastics ('Activity distribution within the waste', p. 22). It is important that the PM either ensures the various waste materials are evenly mixed during the drum fill process, or ensures that appropriate records are maintained so that the modeller can account for such non-uniformities.



Bulk density

A key parameter calculated by the model (or entered by the modeller) is the 'bulk density'. This is defined as the mass of the waste divided by the volume it occupies. Note that, if the container is only partially filled, this parameter may be incorrectly calculated. Determining the mass of the contents is a routine measurement for waste processing, and one can expect this to have been done. Note that bulk density (e.g. typically 300 kg m^{-3} for 'soft waste' and 1200 kg m^{-3} for steel) is different from the material density listed in reference books; the former takes account of voids in the container contents (e.g. gaps between or within individual items). This is very important because γ -emissions from the contents will not be as strongly attenuated as they would be if the container was completely full of the material concerned. The use of bulk density takes this into account.

In summary, the more variation there is between waste containers (e.g. different filling procedures and weights), the more modelling work will be required. In many cases this is unavoidable, but where there is scope to control these factors and minimise the variation, the overall process of measurement, analysis and reporting can be simplified (e.g. a single calibration may apply to many waste containers and the process can be automated). For widely varying waste compositions, a unique calibration must be undertaken for every measurement.

Activity distribution within the waste

The response of a detector to radioactivity either within the bulk of a material or residing on a surface is heavily dependent upon how that activity is distributed. Models to calibrate a system will usually assume that the activity is uniformly distributed. This is not always true in practice, and sometimes the activity will be located in 'hotspots' (i.e. localised accumulations of activity) that are distributed in a random manner throughout the bulk volume: it is good practice to remove hotspots during a survey of the waste prior to the filling of waste containers, but this may not always be possible.

There can also be cases where all the activity on a surface such as a wall is *localised in one area*; if this wall is knocked down and placed into a 1 m^3 builder's bag, then a single hotspot may exist within the container and its location will be unknown. The regulator will allow some averaging of activity over a bulk volume but only where the activity is reasonably uniform.

If the activity within bulk waste is close to uniformity (e.g. if there are no hotspots but there are nevertheless some areas of enhanced activity as well as areas of no activity within the waste container), then the measurement procedure can attempt to take this into account, for example:

- Take a number of measurements around the container and take a mean value. If a very conservative estimate of the activity is required (e.g. to ensure that

the activity lies below disposal limits), use only the maximum value. Note that, if the waste container is large, the practicalities of doing this are significant; measurement systems are very expensive, are easily damaged and take time to repair.

- Place the waste container on a turntable (Figure 1) and rotate it during the measurement. This will 'even out' non-uniform horizontal activity distributions. To even out the vertical distributions, place the detector as far from the container as practicable. Note however that this will decrease the detector's sensitivity to activity within the container and make it more susceptible to changes in background levels. Modelling will assist in either obtaining an optimal measurement position or showing that this will not achieve the objective.
- Use a turntable as above, but mount the detector on a platform that can be moved up and down as the container rotates. This will maximise the detector's response but will rely on the waste container being uniformly cylindrical and centrally positioned on the turntable for close-geometry measurements. Note also that the mechanisms required to achieve the above will be expensive and will require regular maintenance and relatively clean conditions in order to operate continuously without adverse programme implications.

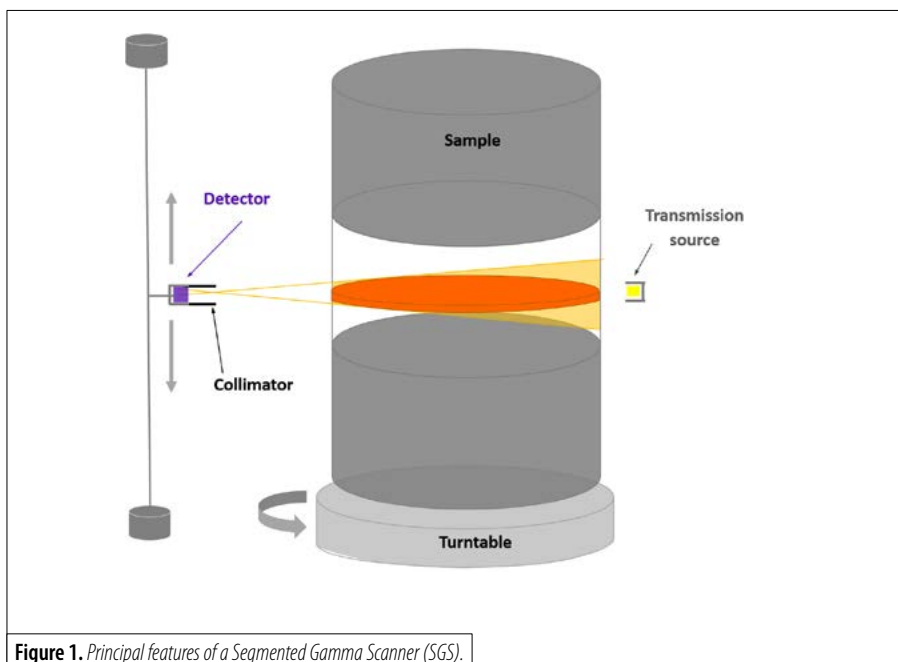


Figure 1. *Principal features of a Segmented Gamma Scanner (SGS).*

Where there is the potential for a hotspot to be present within a bulk volume and if the location and size of the hotspot can be determined, the modeller can take this into account and calibrate the system accordingly. However, it is highly unlikely that this will be known. It may be possible to estimate the location of the hotspot by ‘scanning’ the bag with sensitive probes and then develop a number of modelling scenarios and select the one that appears most representative. To do this on a large number of containers will be very time consuming and is unlikely to be practicable. If hotspots cannot be ‘filtered out’ during the waste generation and container filling process, emptying out the waste and sorting through it may need to be considered.

To summarise, it is possible to use modelling to take account of non-uniform activity distributions but the actual distribution is usually unknown. If it cannot be demonstrated (either by reason or by measurement) that the activity in the bulk waste is uniformly distributed, the PM should try to ensure that the way in which waste containers are filled eliminates hotspots, and that the activity is homogenised within the sample volume (without undue mixing of contaminated materials with non-contaminated materials).

Detectors and environment

So far, the form of the activity and the waste parameters the modeller will consider have been discussed (i.e. how the γ -emissions ‘escape’ the waste container). This section considers the information the modeller will need to determine which and how many of these emissions will be detected.

Both HRGS and LRGS detectors (Chapter I, ‘Overview of use of mathematical models for gamma measurements’, p. 11) are capable of discriminating γ -energies, allowing the activity from different γ -emitting radionuclides to be measured with one detector. However, if the γ -emissions from two different radionuclides have similar energies, HRGS will be required to resolve the two as separate ‘ γ -peaks’; they would appear as one broad peak if LRGS were used. Consequently, LRGS tends to be used only for simple γ fields, or where all the γ -emissions originate from one radionuclide.

Detectors

LRGS detectors have certain advantages over HRGS detectors. For example:

- The detectors operate at ambient temperatures, whereas HRGS detectors require cooling to around 115 K (–158 °C) or lower using either liquid nitrogen or an electrical cooling system
- There may well be little difference in γ -detection efficiencies between detectors of identical sizes from the same batch, so a model created for one detector may

be applicable to a second of the same size (although the second detector should be validated with a suitable radioactive source to confirm that its response is identical to the first).

The modeller will be interested in any differences in detector casing from one LRGS detector to another as this will have an impact on overall detector response (e.g. one detector may have a ‘thick’ aluminium detector window whereas another may have a beryllium window). Different windows will attenuate the γ -emissions from the sample by different amounts. Note also that it is difficult to model the shape of the gamma spectrum for an LRGS detector without carrying out experiments or referring to the literature.

If LRGS detectors are appropriate for the measurements, it may be worth procuring more than one detector if the manner of detector use and the environment in which it will function mean that detector failure is reasonably foreseeable. If additional detectors are procured, obtaining ones with identical crystal assemblies will mean that the modelling may not have to be repeated; however, as indicated above, the new detector should be validated before use.

In contrast to LRGS systems, HRGS detectors are unique in that even detectors of the same type with identical crystal assemblies can exhibit significant differences in detector response. Consequently, changing one crystal for another will always require additional modelling work.

Other detector types can be employed, having varying degrees of peak resolution capability. Table 1 gives examples of the information that may be required to determine γ response if sophisticated modelling software is used.

Some detector manufacturers measure these parameters at the manufacturing stage. They may also determine the response to various γ sources at varying positions with respect to the crystal.

	Information required for γ response
Chemical composition of detector medium (i.e. the crystal); this includes a quantitative listing of any potential impurities	
Density of the crystal	
Dimensions of the crystal	
Active volume of the crystal	
Location of the crystal within the detector housing (distance between the casing end window and the crystal surface)	
Casing materials, including their densities and dimensions	

Table 1. Information typically required as input to sophisticated modelling software in order to determine the γ response of the detector.

Detector environment

In addition to a knowledge of the detector's response to γ -emissions from the sample, the modeller will need information on the environment in which it will function. This may include:

- Whether the detector can view the waste container (or item) fully or only partially
- The presence of intervening materials between the detector and waste container
- The activity levels likely to be encountered.

If the sample activity is very high, the detector system may not be capable of processing the detector output and it may be necessary to increase the distance from the detector to the sample. If it is not possible to increase this distance, or there are physical constraints on detector location, then either the detector or the waste item will require some form of partial shielding (e.g. collimation).

The *materials, thickness, and shape of this shielding* will have a significant effect on the detector response.

Sometimes, the *level of background radiation* will be high, and some form of collimation will be used to enable the sample to be measured. If the background cannot be reduced by other means (e.g. by removing nearby sources or by constructing a shielded enclosure), then the detector itself may be shielded to minimise its response to γ -emissions not arising from the sample. In practice, this procedure may affect the detector's response to the sample itself.

The *sample-to-detector distance* is a critical factor affecting the detector's response. The modeller will either define this distance so as to optimise system response (taking all the other factors mentioned in this chapter into account), or it will be imposed by the size of the measurement area available and therefore simply incorporated into the model. If the sample-to-detector distance changes for any reason it will render the modelled calibration invalid.

Finally, note that *extreme variations in environmental conditions* (e.g. ambient air temperature, pressure and humidity) may affect measurements of low-energy γ -emissions. One situation where changes in the sample environment must obviously be accounted for is measurements of items under water (e.g. in fuel storage ponds).

Note that modelling can be used to design a measurement configuration to achieve a particular sensitivity.

Knowledge of radionuclides present

Any model must be valid over the range of γ -energies which you wish to measure. Depending on the modelling software or technique used, there is usually a need to ensure that the γ -energy range falls within the range for which the software is valid (as stated by the software or system supplier). In practice, this means that the modeller will list γ -emissions that span the range of those anticipated from the sample and which are also within the 'range of validity' for the software being used. The model will use this information to calculate the detector efficiencies for the γ -energies listed. Another approach is that the modeller inputs the radionuclides of interest and the software generates the detector efficiencies for the γ -emissions of those particular radionuclides. Some software is capable of predicting the γ spectrum that would be observed by the detector. To achieve this, the modeller will need to input the relative proportions (by activity) of each radionuclide expected to be present in the waste. This must be done with care; for example, if the radionuclide mix includes relatively short-lived species then the relative proportions may change significantly over the timescale of the project. If 'unexpected' radionuclides are detected, then the suitability of the model should be re-examined.

Note that it is essential to use reliable nuclear decay data, such as that recommended by NPL (See Example, p. 29).

Interference corrections and fingerprinting

One problem which can arise is that of *overlapping peaks*: the spectrum provided by the system may include γ -emissions from different radionuclides which are so close in energy that not even HRGS detectors can distinguish between them. An example of this is where ^{235}U and ^{226}Ra are present (See Example, p. 29).

This is quite a complex measurement scenario, which is not uncommon. There are others. The message to take from this is that if a system is based upon the detection of a single nuclide with a single γ -emission, or a combination of γ -emissions for a low-resolution system, then such a system should not be used if there are interfering γ -emissions from other nuclides present. The only exception to this is if it can be shown by alternative measurement, modelling, or reasoning that the ratio of the emissions from the radionuclide of interest to those from the interfering radionuclide remains constant.

The above leads on to a common technique used in waste assay, namely radionuclide 'fingerprinting'. The radionuclide fingerprint is just a radionuclide activity composition (e.g. 70 % ^{137}Cs , 20 % ^{60}Co , 5 % ^3H and 5 % ^{59}Fe), such that being able to measure one of the nuclides means that all the others, as well as the total activity, can be inferred. This is especially useful when, as in most cases, there are radionuclides present that

cannot be measured using field γ techniques. Deriving a nuclide fingerprint has to be done with care and to a large extent, is outside the scope of modelling γ -detection systems; its major application is in subsequent analyses of data provided by those measurement systems. The key feature of applying a fingerprint is maintaining confidence that the fingerprint has not changed with time. It will certainly change if a chemical or wash process is applied (in which case the fingerprint should be derived after the process). In practice, the uncertainties on a fingerprint can dwarf those associated with measurement and modelling so all that needs to be undertaken is to establish that key radionuclides (i.e. those being measured and those inferred that have a 'limiting factor' such as low disposal authorisation limits) are still within the range expected.

Other useful information

Level of confidence and activity limits: depending on the application, it is essential for the modeller to be aware of the required level of confidence and any activity limits (e.g. those arising from waste categorisation limits). This links into the required output as it will enable the modeller to design the system in such a way that activities at these limits can be measured taking into account the uncertainties.

Waste volumes: a subsequent output (depending on the sophistication of the modelling software employed) will be the estimation of waste volumes that could fall into more than one waste category, depending upon the model parameters used. If this value is too large, the uncertainties may be reduced by providing more information (if available) or by a change in waste preparation methodology. Undertaking modelling at an early stage of project development (ideally at the bidding stage, if there is one) will provide early warning of any problems with the proposed techniques.

Package content: Techniques such as radiography can assist in defining the content of packaged waste. If this is provided at an early stage (along with other waste records), an early assessment of the validity of modelling (and associated uncertainties) can be generated.

Radiation dose-rate information: this is useful as an alternative assessment methodology. However, its usefulness is highly dependent upon the reasons for the measurements being made. If the measurements are for compliance with transport regulations or for general Health Physics purposes, the levels recorded will probably be maximum readings obtained by monitoring very close to the container wall. The use of these data for waste assessment will be very pessimistic (i.e. they will tend to overestimate the activity present) and are therefore not usually appropriate. Their only use would be as a crude check (e.g. of the dose-rate one would predict for a given activity and waste composition).



Example: ^{235}U and ^{226}Ra

The energy of the γ -emission emitted by ^{235}U is approximately 185.7 keV whereas that emitted by ^{226}Ra is approximately 186.0 keV. In this case if there is sufficient ^{235}U present then this interference can be resolved as ^{235}U exhibits γ -emissions at other energies and one in particular at approximately 143.7 keV. Therefore the ^{235}U activity can be determined from this latter energy γ -emission. Knowing the ^{235}U activity and the detector 'efficiency' for this energy then it is possible to calculate how many counts from the ^{235}U 185.7 keV emission will be detected in a given count time. Subtracting this value from what is observed will give the counts due to ^{226}Ra . So it is possible, under specific circumstances, to measure the activity of differing nuclides even if their γ -emissions 'interfere' with each other.

Taking this particular example further, if activity levels of ^{235}U were so low that the other emissions could not be detected within a reasonable timescale, then the solution just presented is unavailable. This can happen in land remediation projects where uranium and/or ^{226}Ra may be contaminants of concern. An additional complication arises when naturally occurring uranium is present as well. Where the levels are too low for ^{235}U / ^{226}Ra corrections to be applied as indicated above then providing the composition of uranium (effectively, the ratio between ^{235}U and ^{238}U) is known, then it is still possible to carry out a correction. If not, then an alternative analytical method will be required (e.g. sampling and radiochemical analysis).

As ^{226}Ra has γ -emitting daughters, it is tempting to infer ^{226}Ra by measuring the daughter(s). This needs careful consideration as the first daughter of ^{226}Ra is a gas (^{222}Rn) which can escape the sample. Therefore the γ -emitting daughters that the system is trying to measure are actually being created outside the sample (i.e. there will be an under representation of ^{226}Ra using this method). There are techniques to correct for this as well. For example, a ^{222}Rn 'escape fraction' could be agreed with the client which could be based on laboratory sample measurements, or the sample could be stored so that ^{222}Rn does not escape. This allows the measurable daughters of interest to build up and enter 'secular equilibrium' with ^{226}Ra . However, this requires a storage period of 30 days and obviously incurs a significant time penalty in analysis turnaround time.

Reference data

Modelling inevitably requires input reference data. In the context of this guide, reference data will generally consist of:

- Nuclear decay data
- Photon and electron interaction data (where appropriate)

Material data (e.g. densities, atomic compositions and molecular formulae) will also generally be required.

It is important that there is a defensible rationale for the selection of any data used, and that there are adequate records of the input data used and the sources of the data ('Documenting and reporting', p. 50). It should be possible for the data to be

retrieved at a later date (e.g. during a technical audit), and it should be possible to demonstrate that the data has been assembled correctly from the reference data source(s).

The use of good quality nuclear data is essential in modelling. There are many sources of such data available, but it is essential that data recommended by National Measurement Institutes like NPL are used.

Nuclear decay data

In the context of modelling covered by this guide, the nuclear decay data required will consist primarily of γ - and X-ray energies and the photon emission probabilities per decay, although in some cases additional data may be required. A detailed review of the current state of knowledge in the field has been written by Nichols⁴. Nuclear decay data are generally well characterised and it is possible to select data such that the confidence in the values is high. A list of recommended data sources for measurement-related applications may be found in NPL Report⁵ IR 6.

Photon and electron interaction data

Photon interaction data used in modelling generally consists of either *mass attenuation coefficients* (in units of $\text{cm}^2 \text{g}^{-1}$) or cross sections (in barns atom^{-1}).

Interaction data is often less well characterised and inevitably carries higher uncertainties than decay data. These uncertainties may not always be apparent to the user; for example, a given database may not contain uncertainty data, or a modelling code may not process uncertainties, thus leading to an incomplete overall uncertainty on the model. It is recommended, therefore, that in the case of interaction data, the user should consider the impact of reference data uncertainties on the numerical results and provide the PM with details of how this assessment was made.

Other recommendations

In general, later publications should be favoured on the assumption that, as the body of available knowledge grows, the published data becomes more reliable. This is not invariably true, however, and it is advisable to compare the results obtained using data from different sources.

Reference data are often provided by equipment manufacturers and software suppliers. The quality of these data is variable and may not be up-to-date or subjected to an appropriate level of quality control. The onus is very much on the user to provide evidence of the validity of such data; where practicable, it is advised that this data should be replaced with data compiled by the user from recommended data sources.

Summary

The selection of reference data is an integral part of any modelling calculation and will have a significant impact on the validity of the results. Nuclear decay data is well-characterised and its selection is straightforward. For interaction data, uncertainties are higher, and care should be taken to correctly evaluate any related uncertainties.

NPL maintains an enquiry service for nuclear decay data and can provide free advice on nuclear data (subject to reasonable time constraints and continued funding), such as whether a particular data source is fit for purpose.

Chapter III

Types of models

- Brief introduction
- Monte Carlo methods
- Deterministic methods
- Modelling assay systems

Brief introduction

Computer-based simulations have been used for over fifty years to aid the understanding of the interactions of radiation with matter. During this time, two very distinct approaches have evolved: Monte Carlo methods and deterministic methods:

- Monte Carlo methods simulate the physical processes of radiation transport through matter. The probabilistic nature of an individual particle's passage (commonly called its history) through the system of interest is simulated interaction-by-interaction, and the results are collected (or tallied) over many particle histories. Interaction probabilities are dependent on: (a) the material traversed, (b) the type of particles, (c) the energy of the particles.
- Deterministic methods are based on the classical Boltzmann equation (which describes the radiation transport process in general terms). Deterministic codes look to solve the Boltzmann equation by breaking it down and approximating it with a set of linear equations and then solving them at a set of discrete points using an iterative process.

Each approach has its advantages and drawbacks. Both methods are extremely flexible in their scope and are capable of modelling anything from the amount of attenuation produced by the passage of radiation through a metal foil, to criticality calculations for entire nuclear reactors. However, given the opportunity for user error in programming the models, it is important that such models are benchmarked in some way ('Benchmarking', p. 48), either by carrying out supporting experimental work or finding reports of such experimental work in published literature.

Monte Carlo methods

Monte Carlo codes can achieve high precision answers by accumulating events and driving down the statistical uncertainty (Chapter IV, 'Uncertainties', p. 37); however, the accuracy of any results depend on various factors, such as:

- How well has the system under consideration been modelled? Are all the dimensions correct? Are the material compositions known exactly? Is the radiation source well understood?



Interaction-by-interaction method

The interaction-by-interaction simulation approach in Monte Carlo is only strictly true for neutral particles. For charged particles, so-called 'condensed histories' are used, as the simulation of individual excitations and ionisations, slowing the charged particle down event by event, would require computing power beyond that accessible to most users.

- Are all the physics processes well-simulated? Are the cross-section sets accurate?

Sensitivity analysis: it is all too easy to overlook these factors when the statistical precision of Monte Carlo tallies are reducing nicely. The sensitivity of the results to uncertainties in many of these parameters can be investigated: it is possible to perturb dimensions, densities, compositions, source parameters and even cross-sections in order to perform some sensitivity analysis. However this all takes time, over and above the already relatively time-consuming ‘base’ Monte Carlo run.

Biasing the process: time can be saved when running Monte Carlo models by biasing the process, influencing the generation and transport of particles to accelerate the tallying process. However, this presupposes that the outcome is more or less known beforehand, and runs the risk of distorting the results if applied inappropriately.

Deterministic methods

Deterministic codes are less accurate than Monte Carlo methods, due to their use of discrete points to solve the transport equations. However, as discussed above, unless the system under investigation is extremely well understood, this is not necessarily as limiting as it may at first appear. The main issue with deterministic codes is the introduction of errors when selecting the number and position of discrete points to use in the model (usually referred to as the grid or mesh): fine or intricate structures may need a finer grid of points to represent them if significant modelling errors are to be avoided. Although generally much quicker than Monte Carlo, certain problems can still take considerable time to iterate to a solution, particularly when finer grids (i.e. with large numbers of points) are being used.

Modelling assay systems

Both of the above approaches will yield reasonable results if the limitations outlined above are borne in mind. However, given the large degree of commonality between the systems being used in waste assay, another approach becomes possible.

A class of codes has been developed to cater specifically for the waste assay market. They come in both Monte Carlo and deterministic forms, and essentially offer a limited set of options applicable to waste assay. Table 2 shows the options offered by a typical code of this sort. The code would then calculate what the detector would ‘see’ given the other specified parameters. Given the restricted nature of the available options, the requirement for any external benchmarking is reduced, although some checks on performance are to be strongly encouraged.

Regardless of the type of code used, it is important that the user keeps a record of

which versions of the software were employed, together with details of the model, any benchmarking undertaken, the results obtained and any analysis of the results undertaken. This is necessary in order that a third party can reproduce the results and conclusions if required. See Chapter V, 'Quality and training', p. 45, for further advice on documenting and reporting.

The key point for the PM is to avoid use of complex models if simpler ones will do. Simple modelling approaches are very useful in scoping and providing approximate answers. The use of complex modelling approaches can be applied if required, though having carried out simple modelling to begin with, their use can be more focused and hence more efficient.

	Typical options offered by codes for nuclear waste assays
	Source geometry (box, drum, pipe, etc.)
	Source type (i.e. individual radionuclides or mixtures)
	Source homogeneity (i.e. even distribution of activity or presence of 'hotspots')
	Detector type and size
	Detector collimation
	Source-detector distance

Table 2. Options offered by a typical code (Monte Carlo or deterministic) for nuclear waste assay systems.



Chapter IV

Uncertainties

- Brief introduction
- What is uncertainty?
- Practical implications

Brief introduction

All measurements have an associated uncertainty. The measured value and uncertainty describe an interval which encompasses the range in which the true value of the measured quantity is expected to lie. The magnitude of the uncertainty is dependent on a number of factors; each may or may not be significant depending on the particular process being modelled.

This chapter describes the critical features, sources and implications of uncertainty that may arise during assay work. The modeller should be aware of the numerous uncertainties that may arise and account for them in their modelling work and ultimately in the end result.

This chapter can only serve as an introduction to uncertainty analysis: the reader is referred to the UKAS document M3003 *The Expression of Uncertainty and Confidence in Measurement*⁶ and to the more general ISO/IEC/BIPM *Guide to the Expression of Uncertainty in Measurement*⁷. The lay reader may find the NPL publication *A Beginner's Guide to Uncertainty of Measurement*⁸ an excellent introductory text.

What is uncertainty?

*A Beginner's Guide to Uncertainty of Measurement*⁸ states "The uncertainty of a measurement tells us something about its quality. Uncertainty of measurement is the doubt that exists about the result of any measurement. You might think that well-made rulers, clocks and thermometers should be trustworthy, and give the right answers. But for every measurement - even the most careful - there is always a margin of doubt. In everyday speech, this might be expressed as 'give or take' ... e.g. a stick might be two metres long 'give or take a centimetre'".

*The Guide to the Expression of Uncertainty in Measurement*⁷ defines measurement uncertainty as "a parameter, associated with the result of a measurement that characterises the dispersion of the values that could reasonably be attributed to the measurand".

For example the activity concentration of a sample may be quoted as:

$$(x \pm y) \text{ Bq/g}$$

where x is the estimated value of the measurand, and y the uncertainty. The value of y , the uncertainty, is often quoted at 2σ where σ is the standard deviation of the measurement. This corresponds to a 95 % coverage probability. This means that the activity concentration is expected to lie in the range between $x - y$ and $x + y$ with a

confidence that there is a probability of approximately 95 % that the true value of the measurand will lie within this range.

The *total degree of uncertainty* is dependent on a number of factors which may or may not be significant depending on the particular process being modelled, just one of which is the statistical uncertainty. The total uncertainty for a measurement may be estimated by drawing up an *uncertainty budget*, in which each component of the uncertainty is estimated and accounted for, according to two main categories of uncertainties.



Types of uncertainty

There are two categories of uncertainty to consider⁶⁻⁸.

Type A uncertainties are estimated by statistical means, for example from a statistical analysis of a series of observations. This is described as a *Type A evaluation* of uncertainty. A Type A uncertainty is defined by a distribution of possible values centred about the true value. The distribution function is usually Gaussian in nature (Figure 2). Examples of Type A uncertainties are the uncertainty due to counting statistics or the uncertainty in the results of simulations.

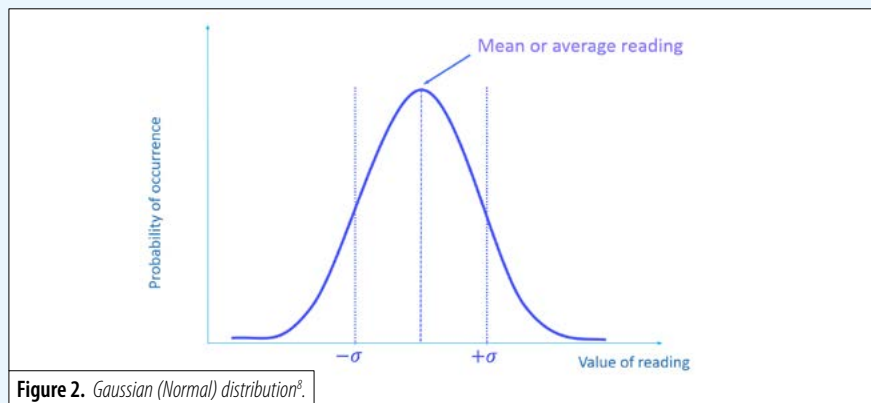


Figure 2. Gaussian (Normal) distribution⁸.

When a set of several repeated readings has been taken (for a Type A estimate of uncertainty), the mean, $\bar{q}$, and estimated standard deviation, $\sigma(q)$, can be calculated for the set. From these, the estimated standard uncertainty, $s(\bar{q})$, of the mean is calculated from:

$$s(\bar{q}) = \sigma(\bar{q}) = \frac{\sigma(q)}{n}$$

where n is the number of measurements in the set. The standard uncertainty of the mean has historically also been called the *standard deviation of the mean*, or the *standard error of the mean*.

Type B evaluation refers to any other method of determining uncertainty. A type B uncertainty may be, for example, a fixed (but unknown) bias in the measurement such that repeated measurements will result in the same offset in the measured and true value (if the bias were known, a correction could be made; the correction would then contribute its own uncertainty to the total budget). Examples of uncertainties evaluated by Type B methods include the uncertainty in the activities of calibration standards and the uncertainty in the composition of a waste matrix. Estimation of an uncertainty by Type B methods is often based on limited available information. Examples are given in UKAS guide⁶ M3003.

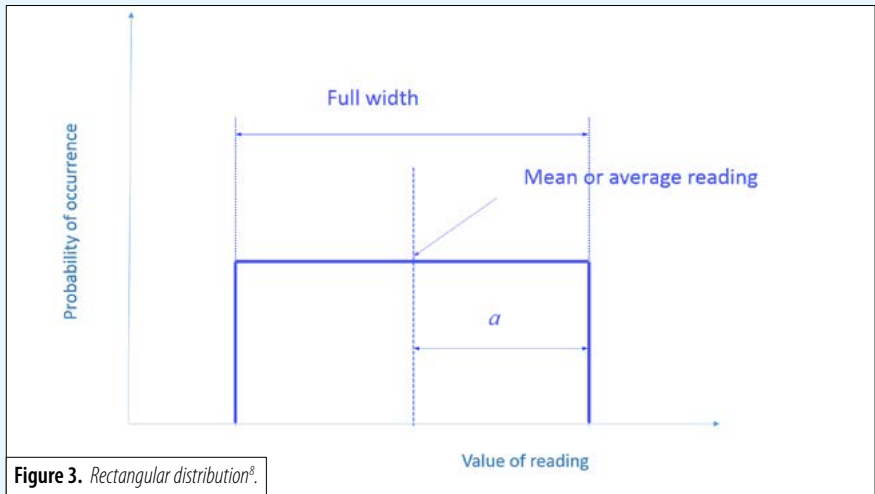
Where the information is more scarce (in some Type B estimates), you might only be able to estimate the upper and lower limits of uncertainty. It may then be assumed, for example, that the value is equally likely to fall anywhere in between these limits.

This is referred to as a rectangular or uniform distribution (Figure 3). The standard uncertainty for a rectangular distribution is found from:

$$\mu = \frac{a}{\sqrt{3}}$$

where *a* is the semi-range (or half-width) between the upper and lower limits.

In some cases a measurement is reported with uncertainties evaluated by Type A methods only, for example reporting only counting statistics or the standard deviation of a set of measurements. This will generally provide an underestimate of the total uncertainty and will provide a false degree of confidence in the measured results.



Sources of uncertainty

In modelled systems there may be many components that need to be considered to derive a precise activity estimate. Each of these components is likely to have their own associated uncertainty.

For illustrative purposes, the main contributors to the overall uncertainty of the measurement of an item using an HRGS detector, measured in an open geometry (i.e. without collimation) are listed in Table 3. This may not be a complete list; users should carry out their own evaluation, taking into account the circumstances of their own measurements and calculations.

Sources of uncertainty for gamma assays by HRGS			
Source of uncertainty	Description of uncertainty componentx	Type A	Type B
Detector calibration	Uncertainty on the characterised detector (for photon energies > 200 keV)	2 %	
	Tabular nuclear data		2 %
	Uncertainty on the reference source activities		2 %
	Uncertainty on the modelling code statistics	2 %	
	Uncertainty on the calibration efficiency function	1 %	
	Subtotals	3 %	2.8 %
Item being measured	Uncertainty in matrix homogeneity (for photon energies between 200 keV and 2 000 keV)		5 %
	Uncertainty in positioning		2 %
	Uncertainty in drum dimensions	1 %	
	Uncertainty in density		2 %
	Subtotals	1 %	5.7 %
Counting	Live time measurement	1 %	
	Random summing correction		1 %
	True coincidence summing correction		1 %
	Background corrections		1 %
	Counting uncertainty	1 %	
	Peaked background correction		2 %
	Subtotals	1.4 %	2.6 %
Grand total (Type A+Type B)		7.7 %	

Table 3. Typical uncertainty budget for measurement of a large item by High Resolution Gamma Spectrometry (HRGS).

The table presents a description of each component of the uncertainty, with typical estimates of the magnitudes of Type A and Type B components. This would be a recommended way to proceed in calculation of an uncertainty budget. Those components which make the largest contribution to the total uncertainty can then be subjected, if necessary, to further work, to reduce the overall uncertainty by reducing their particular contribution. Work to reduce the components making smaller contributions would not significantly reduce the total uncertainty.

Note that the summation of the uncertainty contributions is carried out in quadrature (i.e., the magnitudes of each component are squared, then added, and the square root taken, to find the total uncertainty), as illustrated below:

$$u_{tot} = \sqrt{u_1^2 + u_2^2 + \dots u_n^2}$$

It can be seen that the major contributors to the overall uncertainty are those associated with the item's contents. Note also that some of the uncertainty components are quoted for specific photon energy ranges. This is because there is a strong photon energy dependence, and care must be taken in particular when modelling systems that require the measurement of photon emitters with energies of <100 keV (e.g. for samples containing ²⁴¹Am).

Practical implications

The aim of the modeller is to have sufficient information available to enable the overall uncertainty to be reduced as far as possible to levels commensurate with the measurement uncertainties. However this may be easier said than done. For example, in section 'Sources of uncertainty', p. 41, the most important contributor to the overall uncertainty is shown to be the unknown nature of the distribution of radioactivity in the measured item.

Uncertainty due to activity distribution

Various steps can be taken to more fully account for the distribution of activity within an item (e.g. a drum of waste material requiring assay). These steps may include:

- The use of real time radiography to visualise the distribution of material within the drum
- The use of Segmented Gamma Scanners to enable more appropriate density corrections to be made for each of the measured segments
- The use of tomographic measurement methods to correct more accurately for the self-absorption of emissions from the drum.

Other techniques that may be applied to reduce uncertainties due to non-uniform activity distributions include:

- Drum rotation
- The use of multiple detectors or vertical scanning
- Counting at a distance.

However if the waste stream being packaged is produced on site it may be possible to gain a good understanding of the likely distribution of the activity in the packaged item. This information should be provided to the modeller to enable more reliable models to be produced. The subject of the distribution of activity in a package is also discussed in Chapter II, 'Activity distribution within the waste', p. 22.



Trusting the results from modelling

The estimation of uncertainty is an essential aspect of any measurement process. Many factors contribute to the overall uncertainty on a model. Many will be small and not difficult to estimate; however, parameters such as the distribution of the activity in a sample and the composition of the matrix may be poorly defined. Whilst one can assume particular values for those parameters, the modeller must examine the effects of varying the parameters in order to estimate the corresponding uncertainties (e.g. by running the model for two 'extreme' matrix compositions and comparing the results obtained). This may take up significant time but it is essential that the PM ensures that this is done. Such information also enables the PM to respond to 'what if' questions from a client or a regulator. Note that one must be realistic when attempting to estimate uncertainties in this way; needlessly pessimistic variations in composition may produce very large uncertainties. Good communication between the PM, the modeller and others should reduce this effect and provide an end result acceptable to all parties.

Stable background

Another important influence on the reliability of the model is the requirement for a stable background. The background may vary if the measurements are made in or next to waste stores. There is a potential for γ -emissions from the stored items to contribute to the background at the place of measurement and, unless properly accounted for, to be included in the measurement of activity in the item being assayed. This is reflected in Table 1, where the effect on counting uncertainties from the background uncertainty is estimated to be a Type B uncertainty component of 1 %.

In general, claims that a modelled system produces results that have an overall total uncertainty of less than 10 % over the energy range 60 keV to 2000 keV should be critically reviewed. However, for a subset of cases where the material packages

are small and well understood it may well be possible to have lower uncertainties but again this assumption should be examined closely. It should be remembered that a large uncertainty may be acceptable if the measured value is well below any threshold value.

The evaluation and reporting of the overall uncertainty should be agreed with the client at the bid stage.

A background image of a construction worker in a trench, wearing a hard hat and safety vest, with a blue overlay. The worker is positioned on the left side of the frame, looking towards the right. The trench is deep and appears to be under construction. The blue overlay is semi-transparent, allowing the underlying image to be visible. The text is white and positioned in the upper right quadrant of the image.

Chapter V

Quality and training

- Brief introduction
- Quality assurance
- Benchmarking
- Training and competency
- Documenting and reporting
- Record keeping

Brief introduction

Quality Assurance (QA) and benchmarking are critical to the validity of any modelling output. The amount of effort expended needs to be sufficient to engender confidence in the output, but note that too much effort can render the process very time consuming; a balance must be struck.

Essential aspects of quality assurance include:

- *Recording the source of the parameters used* (and the reasoning behind any estimates made) to a degree such that another modeller with the same software can reproduce the results and understand fully how they have been generated.
- *Ensuring that any equipment to which the model relates must have records* to show that its characteristics have not changed significantly since those characteristics were defined and included in the model. If either aspect is not addressed adequately, lack of confidence in the output may occur; moreover, if subsequent changes to the model are required, significant staff time may be wasted in 'reinventing the wheel', especially if personnel have changed since the model was first compiled.

Benchmarking is essentially about relating the model's output to 'reality' (e.g. by using the model to determine the activity of a well-characterised source and then comparing the results) and is therefore very important as, without it, the client or regulator may not accept any of the results. Note, however, that benchmarking must be applied to an appropriate level (e.g. it is unlikely that a PM will want an exact experimental reproduction of the system being modelled without specific funding). This is an aspect that should be raised at the bid stage by either the bidding company or the client. Note that the onus may fall on the bid manager or the PM to determine the level of benchmarking required.

This guide does not provide a prescriptive step-by-step procedure for Quality Assurance and benchmarking of codes. Such codes are too complex to be simplified in this way. Instead, general guidelines are given which, if followed by specialist modelling staff, will ensure a high degree of confidence in the validity of the results produced.

Confidence in the output of a modelling process is dependent on a number of factors, among which is the level of training and experience of the modeller. Previous experience and training of those undertaking modelling work must be borne in mind by the bid manager or PM.

In addition, it is essential to compile reports on any models produced so that the PM can demonstrate to stakeholders that the modelling undertaken is appropriate and

fit for purpose. This section suggests content for any such reports. In most cases, this is the only tangible product of the modelling process.

Record keeping is clearly essential for quality assurance purposes and for revisiting previous models (which may cover a variety of measurement geometries and therefore be of interest to many people) and the results of any modelling.

Quality Assurance

It is important to ensure that careful records are kept of any models produced. This is an essential aspect of traceability⁹, allows models to be reproduced by others, and enables repeat measurements to be carried out. Records can be checked by periodic audits of the modelling specialist's records.

For large modelling campaigns, the modelling specialist may be asked to produce a *Model Specification Document*. This approach is useful if the modelling is to be performed by a team of modelling specialists. The Model Specification Document should contain information such as:

- The geometry models and source descriptions required
- The output data required
- The format of the results
- The precision requirements (relevant for stochastic modelling or Monte Carlo codes).

This approach can greatly assist the peer review and auditing processes, for applications where a large amount of data are created.

Typical contents of a Model Specification Document are:

- Reference to benchmark applications, demonstrating the validity of the code for the parameter range of interest ('Benchmarking', p. 48)
- Reference to an existing benchmarked model from which a new model (for the present application) can be derived with minimal extrapolation or interpolation
- Details of models produced for similar applications in previous work
- Relevant technical reports produced in previous work
- Details of parameter variations required
- Details of output tallies required.

Version control should be used to distinguish between different versions of a model as it is developed. This can be achieved by methods such as:

- Including the version number in model's filename
- Including the version number in a title/description/comment field for each model
- Using the QA system to which the modeller is working.

Any equipment to which a model relates must have records to show that its characteristics have not changed significantly since those characteristics were defined and included in the model.

Evidence should be obtained to demonstrate that models and subsequent measurement results have been checked and validated (i.e. 'peer reviewed') by a specialist qualified in the operation of the modelling code and relevant procedures.

Note that it is not good practice to leave the checking until the work is complete as this puts the checker under pressure to 'sign off' the work, possibly resulting in a cursory level of checking. Model checking record sheets may be used as appropriate to provide evidence that the checking and validation has been carried out correctly.

Benchmarking

One of the most important aspects of the correct use of modelling in γ -spectrometry is to ensure that a model is benchmarked against experiment. In practice, this means that a measurement of a well-characterised sample is performed under conditions typical for a sample of that type, and a measurement of the same item using a model is performed under identical conditions. By comparing the actual activity present with that predicted from modelling, the level of agreement between the two methods can be assessed.

Benchmarking must be applied at an appropriate level. An example is where waste measurements are to be modelled. Given that the client will always have ultimate legal responsibility for the waste generated on their site and given that it is they who will have prime contact with the regulator, it would normally be in the client's best interests to raise this issue. However, for clients who may not be well informed, the onus will fall upon the bid manager or PM to determine the level of benchmarking required. In most cases, documented reports containing details of experimental work on similar waste containers or items (or for measurement scenarios of similar complexity) will suffice. These may have been generated by the company themselves, the manufacturer, or other organisations using the same type of equipment.

It is vital that the validity of the model is checked across the dynamic range of the

parameter space to which the model will be implemented. For example, if modelling is being used to determine the effect of a drum matrix on a γ -spectrometry system response, then one should devise a benchmark experiment using a matrix which leads to a system response representative of the range of real matrices.

If a model has been obtained from a contractor or other external organisation, then an appropriate benchmark should be sought before the model is used. Sometimes, this can take the form of a publication in the open scientific literature, or a referenced communication with the contractor.

It is recognised that it is not always practical to obtain benchmarks that comprehensively cover the full dynamic range for a parameter space of interest. In such circumstances, a technical assessment should be performed to assess the impact of this lack of availability of benchmark experiment data. This may include inspection of nuclear data or an informed assessment of whether the extrapolation to a new value is likely to have any significant impact.

Participation in comparison exercises (e.g. the NPL 'drum comparisons'¹⁰) also act as a suitable benchmarking method.

Training and competency

The quality of the results obtained from modelling a particular physical problem is dependent on a number of factors, many of which have been covered in earlier chapters. In particular, the quality of the results is dependent on:

- The use of a code which can produce accurate mathematical models adequate to describe the physical system coupled with efficient algorithms to solve the mathematical equations
- The use of the best available reference data (e.g. nuclear data) to describe the materials being modelled
- The accurate representation of the physical system using an appropriate geometry model and physical treatments
- An experienced and competent operator.

Whereas the effects of most of these factors can be evaluated by methods such as benchmarking and sensitivity analysis, there is no way of evaluating the effects of operator error or inexperience.

Ideally, the operator should:

- Understand all the implications and consequences of using any parameter that the code allows the operator to pre-set

- Be able to optimise all the possible options and approximations offered by the code and choose the most appropriate option for the given problem
- Describe the physical model to the accuracy for which the problem demands
- Ensure that any assumptions or simplifications affecting the result are made in a conservative manner.

Any QA system requires that any operator performing a task is competent and trained to carry out that task; this applies also to the use of modelling codes and physical models. Training should include elements such as:

- An appropriate educational background
- Participation in specialised training courses
- On-the-job training from experienced operators
- Practical experience of modelling.

Training alone does not guarantee that acquired generic knowledge will be correctly applied to a specific problem; however, the effects of '*human factors*' can be minimised by comprehensive validation of the entire modelling process. For example, several operators could run a limited number of benchmark studies for scenarios that are as close as possible to the modelling problem.

It is important that the PM should be satisfied with the training and competency of modelling specialists. This may be obtained from one or more of the following sources:

- The modeller's CV and training records
- Proof of previous experience
- Published work and references to previous work.

Documenting and reporting

Whether a model is simple or complex, it is essential to document the model in a specific report. The content of the report will depend on the organisation's local QA procedures (or any QA procedures specified by the client); Table 4 shows a number of points that should be normally included in the report.



Typical content of a report on a model

The type of model (i.e. specific instrument) and serial number of the detector modelled:

If the model is not dependent upon a unique detector (e.g. if only 'LRGS' is specified), then details of the detector's dimensions, materials, housing, etc. must be provided (Chapter II, 'Information needed for modelling', p. 17).

Description of any collimation around the detector:

- Dimensions
- Constituent materials and their densities
- Position of the detector within the collimator.

Orientation of the detector with respect to the item being measured:

this should be a 3-D description, including distances from the item.

Description of the item to be measured, for example:

- Dimensions including wall thickness
- Constituent materials and their densities

This should include a description of how these were assessed (e.g. by measurement or by reference to drawings or information supplied by the client).

Description of the modelling software:

this should include the version number.

List of the γ -energies for which the model is valid:

this information should also include any nuclear data used ('Reference data', p. 29)

Description of the distribution of activity within the item:

if any treatment or conditioning is required (e.g. for a waste package) in order to obtain a particular activity distribution (e.g. for which a model is valid), this should be reported.

Input data (and any calculations) related to the density of the material:

some software requires the modeller to input bulk density whereas other software may calculate density 'internally' by combining other details of the material with parameters such as library density values, container packing fractions and internal dimensions. Whichever method is used, the input data and the method of calculation should be defined.

Details of the 'level of fill' in a waste container:

i.e. whether it is completely or partially filled.

continued overleaf

Description of the materials contained:

this must specify the chemical composition of each of the various materials comprising the item, as described in the model. The reference data for each material's composition must be cited. For example, the data may have been drawn from a materials library within the software itself, or may have been drawn from external web sources or manufacturers' data sheets (Also, see 'Proportions of materials present', p. 53).

Details of factors required by the model;

these should include:

- Run-time
- Convergence values
- Any assumed environmental factors.

Default values are always used with some software but a statement confirming they have been used should still be included. Any changes made to default values should be defined and reasons should be given for those changes.

Uncertainty budget:

as a minimum this should be a statement of individual uncertainties and their sources. Ideally, a 'calibration' uncertainty covering all potential credible variations should be determined, which, when combined with uncertainties in measurement will yield an overall measurement uncertainty. This can generate a significant amount of work for the modeller, depending on the software used. Note that the evaluation and reporting of the overall uncertainty should be agreed with the client at the bid stage (Chapter IV, 'Uncertainties', p. 37).

Minimum Detectable Activities (MDAs) achievable with the detection system:

this will involve making some assumptions about the anticipated levels of background radiation, unless 'real' data are available.

Details of benchmarking:

this must be included for applications where benchmarking has been deemed necessary. Alternatively, if similar work has been undertaken elsewhere to the satisfaction of the client or regulator, this should be referenced.

Table 4 . *Points normally included in a report on a model.*

The report is critical for enabling the operator to demonstrate the validity of the model, and facilitates modification of the model at a later stage (if this is required). Revisions can be incorporated either in successive issues of the report or in separate reports. Separate reports have the advantage of being smaller because the initial report can be cited for most of the technical details.

The level of detail in the final report should be such that the output can be reproduced by anyone with the same software resources and equipment and with a full understanding of the input data and how the output was derived. It is normally necessary to issue the report to the client for comment. The client may not be able to

understand the content (or indeed may not be interested in it) but its production will enable the PM to justify to a third party (e.g. a regulator, or a consultant hired by the client) the project methodology and the results obtained.



Proportions of materials present

The section of the report related to the materials contained should also give the proportion of each material present and detail of how the proportions were arrived at. Examples might be: 'a mixture of PVC (60 %) and cotton (40 %), (...) as defined by the client', or the same mixture '(...) estimated from historical data'. This requires an element of judgement and the reasoning must be made very clear.

However, it should be recognised that where there is a lack of information and where it is not practicable to undertake detailed chemical analysis (e.g. owing to hazards, time or cost constraints), then the modeller may have to simply state that the proportions in the mixture are derived from judgement. In such circumstances, it is very important to undertake a sensitivity analysis ('The composition of the waste', p. 20) by making credible variations to the proportions inputted to the model and noting the effect on the model's output. This must be used in the determination of the overall measurement uncertainty.

Another reason for accuracy in defining the material form is that, should better reference data later become available, it will be easier to incorporate.

Record keeping

Record keeping plays an essential role in any project where modelling has been used. PMs and modellers may later rely on records for reproducing or re-analysing data for a particular project.

The following aspects of record keeping must be addressed:

- Whether there are any legislative requirements
- Whether there are any QA requirements
- Who needs to keep the records
- For how long the records need to be kept (the minimum period will be the duration of the project)
- Whether there should be any protective marking of the records
- Whether printed or electronic copies are required, or both
- How records should be transferred.

Any modelling input and output files on which assessments are made should be retained as per the above. This will enable a re-assessment of the results to be made, if required. Similarly, any measured raw data that are used to validate the model (or which are used during any subsequent measurement campaigns) should be retained.



Case Study: Crate monitoring at Sellafield

This case study is included with the permission of Cavendish Nuclear, who are the owners of the underlying intellectual property.



Figure 5. Sellafield nuclear site (Source: Cat Knott).

The Case Study details the determination of the mass of fissile material in 11 wooden crates, each with approximate dimensions $1\text{ m} \times 1\text{ m} \times 2\text{ m}$, containing legacy wastes associated with the Magnox fuel cycle stored in an outdoor waste compound on the Sellafield site (Figure 6).

The crates were known to be around 25–30 years old and contained waste from historic fuel processing operations. There was a requirement to move the crates to a more suitable location for long-term storage on site, but before this could be undertaken, estimates of the fissile mass had to be made to allow safe transportation across site. To achieve this, an assay program was carried out in order to determine the total fissile mass (i.e. due to plutonium isotopes and ^{235}U). The use of neutron-based assay technology was ruled out as the crates were stored in an outside location with limited access space and an uneven floor, unsuitable for the use of heavy neutron detector slab modules. Therefore, a γ -based technique was chosen as the best means of quantifying the fissile mass. The most suitable instrument available to perform the measurements was an ISOCS™*.

As γ techniques are more sensitive to variations in waste matrix density and homogeneity, the ISOCS™ technique was not deemed accurate enough in its own right to quantify the amount of fissile material.

As the items were sealed and no sampling of the waste matrix was permitted, a series of preliminary measurements were carried out to support the requirement to model each crate for each ISOCS™ measurement.

*ISOCS™ is an HRGS system that uses a characterised detector to facilitate in-field calibrations without the need for radioactive sources.



Figure 6. Crate bearing waste on storage compound on the Sellafield site (Source: Cavendish Nuclear).

The aim of these supporting measurements was to better quantify the distribution of activity inside the crates (in terms of 'hotspot' location) and to better understand the variation in density across the full fill height of the waste matrix.

The first set of preliminary measurements was carried out using a Radscan™** detector. By assaying each crate from three different sides, and examining regions of interest in the γ spectrum collected (specifically, around the 414 keV peak arising from ^{239}Pu decay and the 60 keV peak from ^{241}Am decay), a three-dimensional image containing information of the positions of any hotspots was built up for each crate. It was assumed that no separated ^{241}Am was present in the crates, so that when a ^{241}Am hotspot was detected, it was assumed that Pu would be in the same place.

In addition, each crate was subjected to approximately 12 transmission measurements using a ^{152}Eu source (this radionuclide emits γ photons over a wide energy range, so enables the γ attenuating properties of the matrix to be determined as a function of γ energy). Six measurements in different positions horizontally and vertically were performed along one horizontal axis of the crate and the other six were performed along the other horizontal axis. Where information regarding the specific location of hotspots was available, the transmission measurements were targeted through these regions. Historical records indicated that the physical form of the matrix was mostly PVC and steel. The most appropriate modelled matrix within the ISOCS™ software, in physical composition and density, could then be selected.

** Radscan™ is an LRGS gamma imaging camera which overlays an image of gamma ray hotspots over a photographic image.

After these two preliminary measurement campaigns had been completed, a series of ISOCS™ models was drawn up to specifically reflect the location(s) of the activity and also the most likely bulk matrix compositions and densities as determined from the Radscan™ and ¹⁵²Eu transmission measurements respectively. The variation between a homogenous distribution and a heterogeneous distribution (with known void space and a defined thickness of attenuator) was also considered. Depending on the possible variations in each individual crate, between 6 and 15 different scenarios were modelled and used to determine the efficiency calibration for each scenario. One scenario was chosen to independently benchmark the ISOCS™ modelling process against the Monte Carlo code MCNP™ ***.

ISOCS™ data were acquired for each of the crates. For each crate the spectrometer was positioned approximately 2 m from one side of the crate and data was acquired for 1000 seconds.

The 414 keV peak from ²³⁹Pu was analysed and then used in conjunction with the models to calculate a total activity of ²³⁹Pu. By analysing the acquired spectra through a suitable Pu isotopics code, a total Pu activity (and therefore mass) could be determined for each modelled scenario. Additionally if ²³⁵U was detected (from the 186 keV peak), a final ²³⁵U mass value could be calculated.

To derive a final Pu mass for each crate, the 'best estimate' result was chosen. This result was the one determined from the model which was considered to best represent the activity distribution measured during the RadScan measurements. In addition to using the 414 keV peak from ²³⁹Pu, several other γ photons arising from ²³⁹Pu were used to verify the validity of the 'best estimate' ISOCS™ model chosen for each crate. If the model chosen was a good representation of the crate matrix, consistent results for the ²³⁹Pu activity would be determined for all the additional peaks used for this analysis. Conversely, poor consistency would be observed if the model selected was an inaccurate representation of the true crate matrix configuration.

The possibility of self-attenuation (i.e. attenuation of the γ flux by a 'lump' of material within which the activity is trapped) was also considered. The possibility of this arising would not be seen in the transmission measurements or the Radscan™ measurements. However, the effect of self-attenuation could be seen in the consistency check across the range of γ energies arising from ²³⁹Pu decay. If none of the possible ISOCS™ models yielded consistent results, then the possibility of self-attenuation was considered and taken into account during the Pu activity and uncertainty assessment.

A 1 σ Total Measurement Uncertainty (TMU) value was calculated (as detailed in NPL Good Practice Guide¹ 34) using the variations in activity determined from the full range of modelled scenarios. The variations in activity determined from the modelled scenarios represented the largest contribution to the TMU, although other sources of uncertainty were also included, for example the uncertainty on the acquired number of counts in the γ spectrum.

A final, 'pessimistic' Pu + ²³⁵U mass value for each crate was determined by adding a 3 σ uncertainty to the 'best estimate' mass result. The crates could then be moved to a more suitable long-term storage facility on site.

*** MCNP™ is a Monte Carlo code from Los Alamos: <http://mcnp.lanl.gov>

Further Reading and Engagement with NPL

- Glossary
- References

Glossary

ALARP:	As Low As Reasonably Practicable.
Benchmarking:	Measurements or tests carried out under reference conditions to produce results against which the results of measurements or tests carried out under other conditions may be compared.
Fingerprint:	A list of the radionuclides present in an operational area or sample of interest along with the relative amounts of each present. This may have been derived by analysis of samples or obtained from historical records.
HRGS:	High Resolution Gamma Spectrometry: a technique based on a measurement of the γ spectrum of a sample using a high-resolution detector.
LRGS:	Low Resolution Gamma Spectrometry: a technique based on a measurement of the γ spectrum of a sample using a low-resolution detector.
Matrix:	The material surrounding or holding the substance being assayed. This material may significantly affect the response of the system by attenuation, absorption and other effects on radiation emerging from or entering the substance.
MDA:	Minimum Detectable Activity.
Monte Carlo code:	A computer code that uses random numbers to determine the outcome of a sequence of physical processes in order to simulate a real-world situation.
Nal(Tl):	Thallium-activated sodium iodide: a scintillator material used as a detector in Low Resolution Gamma Spectrometry.
PM:	Project Manager.
Segmented Gamma Scanner:	An instrument which measures γ -emissions from successive segments of a sample.
UKAS:	United Kingdom Accreditation Service.
Uncertainty:	A parameter associated with the result of a measurement, which characterises the dispersion of the values that could reasonably be attributed to the quantity being measured.

References

1. Tattam, D and Keightley, L (2012), *Radiometric non-destructive assay*. Teddington: National Physical Laboratory. Accessible from: <http://www.npl.co.uk/publications/radiometric-non-destructive-assay>
2. Chard P.M.J. (Ed.) (2009), A Good Practice Guide for the Use of Modelling Codes in Non Destructive Assay of Nuclear Materials, *ESARDA Bulletin* 42
3. Gilmore, G.R. (2008), *Practical Gamma-ray Spectroscopy*, 2nd edition, London: John Wiley & Sons
4. Nichols, A.L. (2001), *Applied Radiation and Isotopes*, 55, (pp. 23–70)
5. Pearce, A.K. (2008), *Recommended Nuclear Decay*, NPL Report IR 6, Teddington: National Physical Laboratory
6. United Kingdom Accreditation Service (2012), *The Expression of Uncertainty and Confidence in Measurement*, M3003, Third edition
7. ISO/IEC 98-3:2008, *Uncertainty of measurement Part 3: Guide to the expression of uncertainty in measurement*
8. Bell, S. (1999) *A beginner's guide to uncertainty in measurement*, Teddington: National Physical Laboratory. Accessible from: <http://www.npl.co.uk/publications/a-beginners-guide-to-uncertainty-of-measurement>
9. NPL e-Learning Programme. *Dimensional Measurement, Module 4: Measurement Strategy and Principles of Measurement. Calibration and Traceability*. Retrieved from: www.npl.co.uk/commercial-services/products-and-services/training/e-learning/dimensional-measurement-measurement-user/#stand-alone
10. Dean, J.C.J. (2015), *NPL Nuclear Industry Proficiency Test Exercise 2014*, NPL Report IR 35, Teddington: National Physical Laboratory

Contact NPL

National Physical Laboratory

The National Physical Laboratory (NPL) is the UK's National Measurement Institute. The heart of our mission is to deliver science with impact by disseminating research and measurement best practice for economic and social benefit.

National Physical Laboratory
Hampton Road
Teddington
Middlesex
TW11 0LW

Switchboard **020 8977 3222**
www.npl.co.uk/contact
gpg@npl.co.uk

Keep in touch with us

Sign up to receive regular updates:
www.npl.co.uk/keep-in-touch

Follow NPL on Twitter:
www.twitter.com/npl

Become a Facebook fan:
www.facebook.com/npldigital

Subscribe on YouTube:
www.youtube.com/npldigital

Follow NPL on LinkedIn:
www.linkedin.com/company/national-physical-laboratory

Manager's Guide to Mathematical Modelling for Gamma Assays

Good Practice Guide 135

What is it about?

This guide describes various aspects of mathematical modelling procedures used for non-destructive (gamma) assays of radioactive materials. It discusses input data required by the models, types of models, the various sources of uncertainty, and also quality and training issues.

Who is it for?

The guide is for project managers and others with responsibility for commissioning the measurement of radioactive materials in the nuclear industry, in particular for the management of potentially radioactive waste from the decommissioning of nuclear sites. Those involved in the actual measurements may also find it useful.

What is its purpose?

The purpose of the guide is to provide an overview of the use of mathematical modelling procedures in the measurement of radioactive materials such as waste arisings. It should be used in conjunction with the advice of technical experts.

What is the pre-required knowledge?

The guide assumes that the reader has little or no working knowledge of modelling, although a basic knowledge of the principles of radioactive decay is assumed.