

International Gamma Spectrometric Assessment Exercises Using Simulated Spectra

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Abstract. Gamma spectrometry is a key tool in effective emergency response but may pose significant challenges even for experienced analysts. The diversity of potential emergency scenarios makes it impractical to exercise with real sources but recent computational developments have facilitated computer simulation of gamma spectra of sufficient quality for exercising. Three international exercises employing synthetic spectra to provide high quality training materials for enhancing the abilities of participating organisations have been conducted by the Nordic Nuclear Safety Research (NKS) in recent years. These exercises involved the analysis of simulated HPGe spectra from an air sample taken in the early phase after a major nuclear accident, spectra acquired in response to malevolent acts involving different types of radioactive materials and spectra from a mobile gamma spectrometry system for location and identification of orphan sources and mapping fallout. Scenarios reflected realistic situations and spectra were generated using Monte Carlo simulations and other methods. All exercises were conducted under time constraints ranging from hours to days. Results of the exercises indicated some general areas of difficulty for participants in the areas of spectrum complexity, summation corrections, mother-daughter relationships and less commonly encountered isotopes. These difficulties impacted upon both qualitative and quantitative results. Many of the weaknesses observed in the results can to some extent be explained by lack of experience and exercising opportunities. This is an aspect which the NKS exercises were intended to address. This presentation will introduce the exercises, provide details of the scenarios and materials involved and discuss the exercises from the participants' point of view. The benefits and disadvantages of exercising with simulated spectra will be discussed and suggestions as to further activities will be presented.

KEYWORDS: *Gamma spectrometry, emergency preparedness, synthetic spectra.*

INTRODUCTION

Although an omnipresent analytical technique in laboratories conducting radioanalytical measurements and a key technique in emergency preparedness and response, gamma spectrometry can present significant challenges on a number of different fronts. In the years after the Chernobyl accident, gamma spectrometry has, for many laboratories and analysts, come to involve the measurement of a relatively limited suite of isotopes at relatively low levels for a limited range of sample types. This fact, combined with the inevitable decline in numbers of analysts who were active during the period of nuclear weapons testing and before the Chernobyl accident, has led to a situation where many analysts are unfamiliar or inexperienced in the analysis of materials and isotopes that may fall outside the range of those encountered on a day-to-day basis. This problem is exacerbated by two other aspects of the situation relating to how gamma analyses are conducted and the applications in which gamma spectrometry may be employed as a measurement method.

The last decade has seen rapid changes in how analysts interface with their instruments. Although the fundamental algorithms are still used in analysing spectra, the way they are applied has changed in that the software tends to, or at least is capable of, proceeding through the analysis with minimal interaction from the analyst. While the majority of analysts will ensure that their software is set up to produce acceptable results with minimum interaction, this is usually only tested for the isotopes of primary interest. There is no assurance that the software will perform equally as well in situations where

it attempts to analyse complex spectra produced by a mixture of perhaps rarely seen isotopes in a sample matrix less often encountered. Similarly, there is no guarantee that all analysts will understand that an instrument setup regularly producing quality data for ^{137}Cs and ^{40}K may not necessarily produce quality data for isotopes of, for example, tellurium, xenon or ytterbium. This problem will of course be exacerbated in a situation where the analyst is not experienced enough to guess *a priori* what isotopes could feasibly be present (or could absolutely not be present) in any particular sample type for any particular release scenario. The second aspect relates to the areas of deployment of gamma ray spectrometry and the nature of the scenarios in which it may play a role. Due primarily to hardware considerations, HPGe spectrometry was for many years confined to the laboratory to a large extent. Field measurements were conducted mostly by NaI detectors. The last decade however has seen HPGe increasingly applied in field measurements as reliance on liquid nitrogen ceases to be a limiting factor and NaI detectors have been supplemented by newer detector types with spectral resolution intermediate between HPGe and NaI. Making field measurements places another layer of difficulty on the analyst as the response functions of instruments in the field can be significantly different to those in the laboratory or the analyst may be asked to conduct measurements with an instrument with which they are entirely unfamiliar. In addition, gamma spectrometry may feature in measurement platforms which present data in ways often entirely unfamiliar to the analyst such as during mobile measurements.

The obvious solution to the problems outlined above is practice and training. Regular, focussed training can address many of the problems inherent in an analyst being presented with material with which they have little experience as well as familiarising the analyst with what can actually show up in a sample from a given scenario type. Practicable solutions to such practicing however are less amenable than may first be presumed. This can be demonstrated by considering two separate areas: emergency response and security applications. The isotopes encountered in “classical” emergency response are a disparate bunch of often short lived radionuclides. As such, although representative samples could be generated in a reactor and distributed for measurement, very significant problems manifest themselves in relation to sample distribution (customs, cost, security, time factors etc), limiting exposure of analysts to what may be relatively high activities and the willingness of laboratories that have, over time, become de facto low-level laboratories to work with such samples. Spectra could be taken at the point of generation and then distributed but this presents problems in relation to how the “true” quantitative and qualitative values are derived for the sample. The problems increase for security applications where measurements may have to be taken of isotopes that could never be distributed at activities that could never be deemed safe for situations that could never be replicated. For mobile measurement systems, despite useful large scale exercise in the past, constraints imposed by the suite of isotopes available and the environments in which they can be placed as well as cost of participation can limit the practicability of holding or participating in such exercises.

One possible solution to these problems may be found in synthetic spectra generated by any of a number of processes to produce a fit-for-purpose representation of the instrument response to a chosen set of isotopes. Synthetic or semi-synthetic spectra have been employed before (see Karhu et al (2006) and Nikkinen (2001)) for various purposes and offer a potentially useful means of addressing some of the problems outlined above. Spectra may either be produced using deterministic functions describing a detector or using probabilistic methods best known as Monte Carlo. In previous years the primary limitation in applying such techniques was computer processor power but more powerful computers have negated this issue to large extent. It is now possible to derive spectra for any combination of isotope and detector that can include shielding effects and can account for phenomena such as true coincidence summation. Synthetic spectra offer a high degree of control over the scenario being simulated as well as providing opportunities to derive spectra for situations that could never be replicated for training purposes.

As part of the B-programme of the NKS, three international exercises using synthetic materials have been conducted since 2009 to practice in the following areas:

1. Laboratory measurement of a contaminated air filter by HPGe after a significant release from a power plant (REMSPEC - *Analysis of Remotely Accrued Complex Gamma-ray Spectra – Proficiency Test*);
2. Field and laboratory measurements using a variety of detector types for security/emergency response type applications (MALRAD - *Proficiency Test in the Analysis of Gamma Spectra for Malevolent Radiological Situations*);
3. Field measurements using a typical carborne gamma spectrometry system (ORPEX - *Orphan Sources and Fresh Fallout: Virtual Exercise in Mobile Measurement*).

These three exercises covered the main areas where practice opportunities are limited and included spectra generated for situations which would be difficult to replicate using actual sources.

METHODS

Scenario development for each exercise was based as far as possible on real life events. The REMSPEC source term, for a major release from a nuclear power plant, was derived from Larsen et al (1999), source terms for the MALRAD sources being based upon (or employing modified versions of) the situations described in Hibbs (1995), Sevan'kaev et al (1999), Nature (2003), Mullen (1987) and LLNL (2001) amongst others. The ORPEX activity consisted of two situations: the first simulating the search for orphan sources in an urban environment using mobile gamma spectrometry and the second being mapping of local fallout after an accident involving radioactive materials. The sources for the first scenario of ORPEX were typical industrial and medical sources. One spectrum for a ^{90}Sr source in ORPEX was obtained from David Mercer and Benjamin Sapp of the Los Alamos National Laboratory, United States and was not synthetic. The dispersion pattern for the second scenario of ORPEX was based on calculations performed using the using the HOTSPOT code (Homann, 2010). Simulations of spectra for all exercises were conducted using the codes as described by Hensley and Lepel (1998), Plenteda (2002) and using the MCNP code. Spectral conversions for all exercises were conducted using either home made routines, the CAMBIO utility from the US DoE and SPECON 2000 (Hong, 2001). All nuclear data was obtained in the days prior to the exercises from the Table of Isotopes as maintained by Lund University at <http://nucleardata.nuclear.lu.se> and the source of nuclear data communicated to participants. Data was delivered to participants either by email or by download from an NRPA server. The file formats which data were provided in varied between the exercises. For REMSPEC, spectral data was provided in the proprietary formats of the major manufacturers as well as a number of standard formats. For MALRAD spectra were provided in a range of formats similar to that of REMSPEC as well as scenario descriptions and ancillary dose data as simple documents. The situation for ORPEX differed substantially in that a range of data types were required to correctly simulate the information that would be generated by or available from a mobile measurement systems. In excess of 5000 individual spectra were distributed in 4 different formats as well as the data in spreadsheet format. Geographical and spectral data was also distributed as a kml file openable in Google Earth or similar. Custom standalone software was provided as part of the exercise for the analysis of data in a manner typical for carborne systems. The software, described fully by Watson and Smethurst (2011), is based loosely on the Geological Survey of Norway's GAMMALOG emergency measuring system of Smethurst et al (2005) and provides the usual visualisations including spectrum plots, waterfall (or rainbow) plots and charts of data derivatives like estimated air kerma rate as well as some more advanced features.

EXERCISE DESCRIPTIONS

The three exercises were conducted following the same basic principles: synthetic materials were derived for the relevant scenarios, the materials were disseminated electronically to participants with supplementary information as necessary and participants were obliged to respond within a certain time frame, further materials and answers being made available after the exercise closed when appropriate.

The first exercise (2008) was REMSPEC, a simulation of a scenario where a major release had occurred from a nuclear power plant and an air sample had been taken using a charcoal impregnated filter some time after release at a distant point. The spectrum for this filter (taken on a standard HPGe in 7 different data formats) was distributed along with detector efficiency information (peak and total). Participants had to respond with isotopes and activities in three hours. The spectrum included nearly 1000 peaks from 24 isotopes released including traces of noble gases. Mother daughter relationships were included, true coincidence summing was represented although laboratory background was not. Eleven laboratories from 8 different countries returned results in time.

The second exercise, MALRAD (2009), differed from REMSPEC in that 7 different scenarios were devised. Each scenario featured different sources in various configurations and employed different detector types. Scenarios were based where possible upon isotopes, materials and contexts that had featured in real life situations. A summary of each scenario was provided with the information that would have been available to the analyst in real life (such as dose measurements, efficiency data, etc) as well as the spectra (in a range of data formats) and point source energy calibration spectra where appropriate. The seven scenarios were:

1. A 10 year old Am/Be well logging source (300 GBq plus daughters) concealed within a large volume of an explosive mixture under a lead blanket in a sealed truck. Short NaI spectrum taken with dose measurements at two positions. Objective: identify/quantify.
2. A sealed steel cylinder containing 100 g of 25 year old weapons plutonium (93% ^{239}Pu) oxide plus daughters measured by HPGe. Objective: identify, quantify, any other information possible.
3. 11.1 GBq of ^{192}Ir placed under the seat of a car. Spectra taken with CdZnTe detector as well as dose measurements. Objective: identify/quantify.
4. Indoors dispersal of irradiated graphite, measurement by NaI. Objective: identify
5. 3 GBq ^{99}Mo /3 GBq $^{99\text{m}}\text{Tc}$ on a bus. Spectra (NaI) and dose measurements taken. Objective: identify/quantify.
6. A 1480 GBq ^{75}Se source hidden under scrap metal (10 cm aluminium). CDZnTe spectrum plus dose measurements. Objective: identify/quantify.
7. A 10 g sample of enriched (72.7%), reprocessed (^{232}U present), 10 year old uranium plus daughters counted on HPGe. Aqueous efficiency data provided. Objective: identify, quantify, any other information.

Eighteen laboratories from 9 countries provided results within the one week deadline.

The third exercise, ORPEX, was held in 2011 and was a representation of two situations: trying to find and identify a series of orphan sources in an urban environment and trying to map contamination after a fire involving radioactive materials in the same environment. Both scenarios were conducted using a simulation of a large volume NaI detector in a car as is typical for carborne measurements. The system simulated takes one spectrum over 256 channels every second and includes geographical data, estimates of kerma rates and various other data. Real life data from the chosen route was used as the basis and upon this data were superimposed synthetic signals from 10 discrete point sources for the first scenario and, for the second scenario, data for a simple nuclide mixture in amounts varying by

geographical location. Signals varied with distance in a correct manner and the shielding effect of buildings was approximated. Data was provided in custom format readable using the custom software, provided for the exercise, featuring the typical functions and controls found in such systems. All individual spectra were provided in a range of formats and data was also provided in ASCII format for those wishing to convert/analyse the data to their liking. The data was also provided in Google Earth format with typical information as to kerma etc graphically represented for each point.

The sources for ORPEX were intended to represent the typical difficulties in making such measurements. Sources included ^{137}Cs , ^{60}Co , ^{192}Ir , ^{75}Se , ^{133}Ba , ^{67}Ga , ^{169}Yb , depleted uranium and a ^{90}Sr thermoelectric generator. The sources were placed in various juxtapositions, some were collimated, some were barely visible by the detector and some sources were seen more than once. Participants were asked to locate the sources as best they could and identify them if possible.

RESULTS AND DISCUSSION

The three exercises highlighted a number of interesting aspects with respect to how analysts managed the materials. For REMSPEC a number of issues were noted that have come to the fore in more conventional activities such as the various regular intercomparisons that take place. Although coincidence summation is now a well recognised problem, not everyone corrects for it and for those that do, some only correct for isotopes like ^{134}Cs and ^{60}Co for which the correction is easily achievable empirically or which the analyst may encounter regularly. For some isotopes that are strongly affected by summing but may only be encountered after accidents, the extent of correction appears to be much less. Problems in relation to mother daughter pairs were evident, the presence of one not always being used to infer the presence of the other – a matter which is probably related to the experience of the analyst. In addition, the number and type of false positives being reported was indicative of either over reliance, in some cases, on the suggestions of the software or a lack of awareness of what can actually be present in a sample after a release from a power plant – a matter of no small significance given recent events in Japan. Reported uncertainties varied widely even though all participants had the same data and allowing for some analysts reporting at a 2σ level instead of 1σ . On a practical level, it was apparent that participants were struggling to read data if it was provided in anything other than the proprietary format they use daily (or that their software could read) and in entering efficiency data by hand to construct a calibration as opposed to using the calibration spectra that much proprietary software requires. Again, both of these matters are of some significance in relation to emergency preparedness and assistance. The results indicated that laboratories that could be expected to have a lot of experience in this area or analysts who worked with Chernobyl for example, had a better success rate. More accurate results were correlated with less false positives probably being indicative of the analyst's general level of confidence in that they felt confident in ignoring the more improbable suggestions of their software or were using software that produced less of them. Some participants made the point that the library they were using did not include isotope x or y and therefore they could not identify it but it could be argued that this is less a failing of the software than the analyst who must be in a position to know whether or not the tools they are using are up to the task at hand. After the exercise was finished, the test spectrum was provided to participants with a number of modifications. These included the test spectrum with a super high resolution, the test spectrum without coincidence summation, the test spectrum with just the photopeaks and various combinations of these for training purposes. Provision of these materials was solely for the participants use and was not intended as test materials to be reported upon.

The MALRAD exercise produced an entirely different suite of problems than REMSPEC and these problems also varied depending on which scenario was involved. In general, for isotopes with clear

signatures and no confounding factors, irrespective of the detector employed or whether or not the analyst has used that detector type specifically, accurate results can be obtained. For a situation where

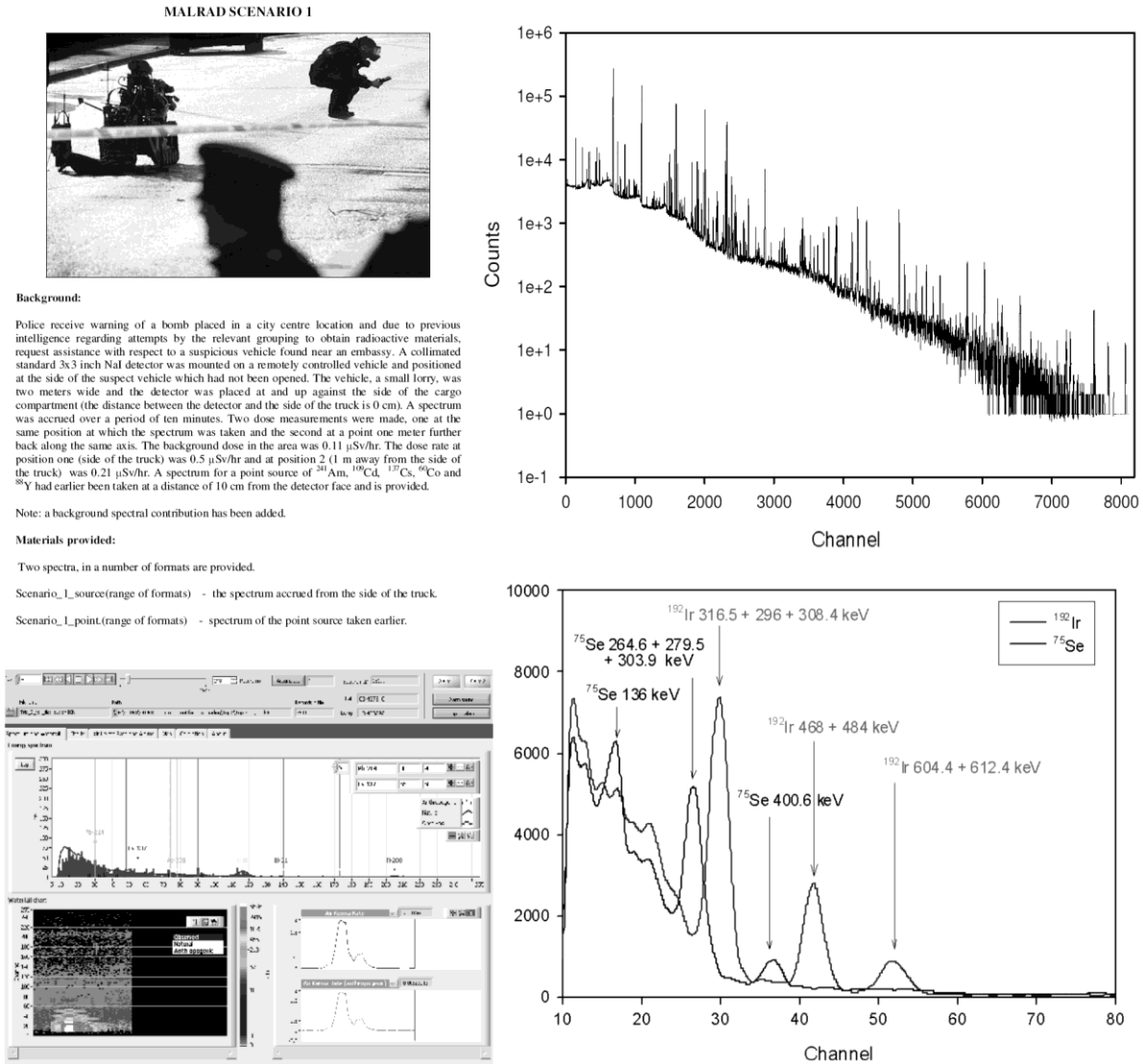


Figure 1. Clockwise from top-left: Scenario description for Scenario 1 of MALRAD; the REMPEC test spectrum; the spectra for Sources 2 and 3 of ORPEX; screenshot of the custom software for the ORPEX exercise.

an easily recognisable isotope is present (such as ^{241}Am) but in the presence of a confounding factor such as shielding, the situation becomes significantly different. In this instance the analyst was presented with a situation where not only was the main peak used for identification diminished but minor, higher

energy peaks came to the fore (being diminished less). This caused obvious problems for some analysts with ^{241}Am being identified as ^{137}Cs or ^{131}I due to the visibility of a usually minor ^{241}Am peak near 660 keV. That a large amount of an alpha emitter was being mistaken for a small amount of a gamma emitter or that an isotope such as ^{241}Am could be thought to be ^{131}I are matters of obvious consequence in further handling of the situation and therefore of some concern from an emergency preparedness perspective. A major factor in this confusion occurring at all was the fact that many nuclide libraries used do not apparently include the lesser known peaks of ^{241}Am (and presumably other nuclides) as they rarely feature at the levels of activity seen by analysts. In this context the value of careful checking of manufacturers default libraries in relation to the possible contexts to which they will be applied is highlighted.

The second major issue identified during MALRAD was in relation to the two special nuclear materials of Scenarios 2 and 7. Spectra for such materials can provide a wealth of information with respect to the age of the material since manufacture or last separation, the nature of the material (reactor grade or weapons grade), the source of the material (reprocessed uranium or not) and the form of the material (Pu oxide or metal). There appeared to be a general lack of awareness amongst participants in relation to what type of information could be derived and how to derive it despite the fact that some of the participants would probably have a role to play in the event of interdiction of such materials or at least in the follow up to an interdiction. Participants who had more experience in this area were able to provide concise, accurate information as to all aspects of the sources and the information was provided with the level of confidence that is necessary in effective response. It can of course be argued that the requirement for analytical capabilities in relation to such materials is very limited for most laboratories. However there has never been a real "dirty bomb" incident for which many laboratories actively practice and prepare yet there have been many incidents involving the interdiction and seizure of special nuclear materials, a significant seizure having been made as recently as this spring (2011). Based on such statistics it would appear reasonable that laboratories with presumed possible responsibilities in this area would be in a position to conduct such analyses.

In the weeks after the MALRAD exercise, the materials were used as a practice activity for the US National Nuclear Security Administrations' (NNSA) TRIAGE system, a secure, on-line capability providing remote expert technical support to emergency responders in the event of a nuclear or radiological emergency. Results provided were accurate, precise and prompt indicating the value of this service in providing support for response to situations such as those used in MALRAD.

The third activity, held in 2011, was focussed on the matter of mobile measurements. Although large scale field exercises have been conducted in previous years, obvious limitations for such exercises include the nature of the sources that can be employed and the contexts in which they may be found. ORPEX involved the use of a typical NaI based mobile measurement system in location and identification of orphan sources in an urban environment and for the mapping of contamination after a release event. Participants were asked to locate where they thought sources might be located and attempt identification such that a follow up foot team may localise the source later. The participants were provided with all necessary materials to conduct the task. All participants were in a position to localise and identify some sources although substantially fewer were able to identify isotopes such as ^{90}Sr , ^{169}Yb or ^{67}Ga . Responses indicated that experienced analysts, irrespective of mobile measurement experience, were in a position to identify more isotopes than inexperienced analysts. Analysts with mobile measurement experience were however better positioned to make maximum use of the data and produce more accurate estimates of the sources locations. Obvious problems were present in the use of an NaI detector in detector to identify isotopes with predominantly low energy emissions although some analysts were in a position to utilise the data to maximum effect. All participants were able to describe the main areas of fallout for the second part of ORPEX. Quantification of this fallout, either relative to

background or relative to the lowest level of contamination observed was less evident, a distinct minority of the the participants being in a position to categorise the areas of contamination.

The exercises provided a useful chance to test synthetic spectra in terms of their usefulness for this type of exercising. For simple source-detector combinations such as those encountered in the laboratory, irrespective of the actual complexity of the isotopes, synthetic spectra can accurately reproduce the main features of the spectrum extending to features such as coincidence effects. It was interesting to observe that in the reports of the NNSA on the MALRAD material, only one of the spectra were identified as being probably synthetic. Once the complexity begins to increase in relation to the environment in which the simulated measurement is being made, the amount of effort required to accurately reproduce a spectrum increases rapidly. Although modern Monte Carlo codes can simulate any environment, the time to model complex environments and the computational time required to solve the problem can become limiting factors. In addition, certain aspects such as the effects of beta particles can be problematical to simulate as the computational time required for electrons is excessive. For the majority of the spectra featured in the three exercises, “laboratory” background was not simulated. An acceptable solution, as was implemented in ORPEX, was simply to superimpose the synthetic data upon a genuine background although the advantages of being able to accurately synthesize background spectra are obvious. The fine detail of gamma spectra, such as peak broadening and lower energy tails, can be represented although such representations may not be entirely reflective of actual spectra.

In terms of exercising synthetic spectra provided an entirely acceptable means of simulating actual measurements. At no point during the three exercises were problems encountered in relation to the contents of the spectra. It was possible in all cases to derive the correct answer from the spectral data to hand. The dissemination of data electronically proved a convenient method of delivering the data with significant advantages over distribution of a sample material. A problem encountered however was that spectra were required to be delivered in a wide number of formats – participants are often not in a position to convert spectra from one format to another despite the availability of free converters for this purpose. This is not a weakness in the spectra themselves obviously but is a drawback in conducting exercises in this way. It may be argued that it is the laboratories responsibility to be able to convert the data to a suitable format in the same manner that it is the laboratories responsibility to prepare actual samples. However it was noted that participation in the exercises would have been substantially less had the data only been provided in some standard data exchange format.

In terms of future activities, it may be advantageous to focus on the weaknesses evident as a result of the three exercises that may be addressed effectively using synthetic materials. In this regard two aspects are probably of most interest. The first of these relates to corrections – background, decay, mother daughter relationships, sample matrix and summation effects. The second aspect is probably in relation to the materials that proved most problematic and are the hardest to exercise with, namely, special nuclear materials. For this purpose, the derivation of advanced information from gamma spectra (such as time since separation etc) is probably not most critical but rather the identification of samples as being of special nuclear materials . It is this task that is most likely to be the one facing analysts at the national level and yet it is this task that some participants in the exercise proved to be experiencing difficulty with. Although this matter may at first glance appear to be relatively trivial given that such materials are most often uranium or plutonium, the gamma spectra of the various forms of these materials can be radically different depending on the type of material, its age and the source of the material itself.

AVAILABILITY

The materials of ORPEX and MALRAD are available for download at the NKS website.

MALRAD full report and all materials and instructions:

http://www.nks.org/en/seminars/presentations/nks-b_malrad_scenarios_and_spectra.htm

REMSPEC full report and all materials and instructions:

http://www.nks.org/en/seminars/presentations/nks-b_remspec_spectra.htm

As of the time of writing, the ORPEX materials are not available but will be placed on the NKS website before the end of 2011.

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