U.S. DEPARTMENT OF ENERGY

PNNL-22308

# Advanced Spectral Analysis for Radioxenon Project

# Final Technical Report on Radioxenon Event Analysis

JH Ely
MW Cooper
JC Hayes
TR Heimbigner
JI McIntyre
BT Schrom

March, 2013

Pacific Northwest NATIONAL LABORATORY

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# Executive Summary

Automated radionuclide samplers to monitor for nuclear explosions are being installed as part of the International Monitoring System. Data from these systems is being generated and tools to analyze large data sets and complex operational parameters are required. In addition, the processes to efficiently screen and categorize the samples need to be developed. This Advance Spectral Analysis for Radioxenon (ASAR) project supported the U.S. National Data Center (NDC) radionuclide sample collection objectives by researching and developing algorithms and analysis approaches and building analysis tools that can be used with the NDC database. These tools were designed initiated on the previous Data Center project, and extended and upgraded during the ASAR project. These tools provide an independent calculation approach and can be used to provide confidence in the data and activity concentrations provide by the International Data Center (IDC).

In addition to the main review toolset, the ASAR project supported the enhancement of the calibration tool, which allows semi-automated calculation of the calibration data collected from beta-gamma systems. And the ASAR project supported the development of a stand-alone analysis tool, using a different analysis approach using standard spectra fitting. This was developed in conjunction with the University of Texas, with the support of Dr. Biegalski during his sabbatical at PNNL. This tool is useful for low activity concentration samples and can provide increased accuracy in the calculated values.

With the increased capability in analyzing the data files and providing high-confidence values, the ASAR project focused on exploring the approaches to effectively screen and categorize samples. There are many samples collected every day, with many activity concentrations below the minimal detectable concentration. However, there are also detections every day which need to be reviewed and resolved. Almost all of these are from emissions from nuclear power plant reactors or medical isotope production facilities, but they require careful inspection and review; making this process as efficient as possible was a main focus of the ASAR project. This is an active research area and no simple approach is satisfactory. Although the systems measure four radioxenon isotopes which can provide much more information when combined in ratios that when analyzed individually, they are not always detected above the minimal detectable concentration (MDC) of the collection and measurement system. Even in cases where several or all isotopes are detected, the radioxenon data can't discriminate nuclear explosions from medical isotope production or reactor start-up in all scenarios. In these cases, additional information is required to support resolution of radioxenon detections, such as seismic detection or other information. The radioxenon data can be screened to narrow down the number of detections that require further analysis and combined with other data for resolution, and methods for this have been explored. It is likely, however, that combining additional data, such as atmospheric modeling results or particulate measurements, *prior to categorization,* may make the process more efficient.

A more holistic approach to the data screening and categorization is the focus of the follow-on Integrated Nuclear Signature Interpretation of Global Happening Toolset (INSIGHT) project, which will research approaches to combining data prior to screening and categorization as a possible method to increase screening efficiency and identify the appropriate high-value events for further review.

# Acronyms and Abbreviations

| | |
|---|---|
| ARIX | Analyzer of Xenon Radioisotopes |
| ASAR | Advanced Spectral Analysis for Radioxenon |
| ATM | Atmospheric Modeling |
| HPGe | high purity germanium |
| IDC | International Data Center |
| IMS | International Monitoring System |
| INGE | International Noble Gas Experiment |
| INSIGHT | Integrated Nuclear Signature Interpretation of Global Happening Toolset |
| JDBC | Java DataBase Connectivity |
| MDC | minimal detectable concentration |
| NDC | US National Data Center |
| NG | Noble gas |
| PNNL | Pacific Northwest National Laboratory |
| QC | quality control |
| SAUNA | Swedish Automatic Unit for Noble Gas Acquisition |
| SDAT | Spectral Deconvolution Analysis Tool |
| SoH | state of health |
| SPALAX | Systéme de Prélèvements et d'Analyse en Ligne d'Air pour quantified le Xenon |
| UI | user interface |
| Xe | xenon |

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# 1. Introduction

Detection of xenon isotopes is a proven and important method for distinguishing nuclear from conventional explosions and earthquakes and is particularly well suited to detecting undeclared underground testing. The radioxenon isotopes $^{131m}Xe$, $^{133}Xe$, $^{133m}Xe$ and $^{135}Xe$ are of particularly high-value in identifying nuclear explosions and thus are the focus of current noble gas detection systems. The U.S. National Data Center (NDC) receives radioxenon measurement data from a variety of sources including the noble gas monitors that are part of the International Noble Gas Experiment (INGE), where stations are being installed and evaluated for certification into the International Monitoring System (IMS), with the data being distributed by the International Data Center (IDC). There are three types of radioxenon collection and measurement systems being installed for the IMS including the Swedish Automatic Unit for Noble gas Acquisition (SAUNA), the Russian Analyzer of Xenon Radioisotopes (ARIX), and the French Systéme de Prélèvements et d'Analyse en Ligne d'Air pour quantified le Xenon (SPALAX) [1-3].

Pacific Northwest National Laboratory (PNNL) has been developing software tools to support the NDC. In particular, software tools were developed to allow the NDC to automatically receive data from the IDC, calculate the concentrations, and provide a viewing functionality for review, while storing all the data and results into a database. These software tools were initially developed under the Data Center project, and continued development under the Advanced Spectral Analysis for Radioxenon (ASAR) project.

There were several areas of research for the ASAR project. The first was to support the development of independent analysis tools for the NDC to provide additional confidence in the activity concentrations provided by the IDC and also to provide calculation capability for other systems such as have been developed by PNNL. Additional tool development was associated with calibration of the PNNL beta-gamma detectors. The other focus of ASAR research was in sample review and screening for interesting events to investigate further. This categorization of radioxenon measurements is challenging and requires research into background sources of radioxenon and methods for distinguishing them from nuclear explosions. This report describes the research performed on the ASAR project to provide high-confidence activity concentration values, and the approaches explored for efficient screening and categorization.

# 2. Activity Concentration

For the categorization of the radioxenon events, the activity concentration is one of the most important quantities, providing information on whether there was detection of an isotope and at what confidence. However, the activity concentration value may not always be precise or accurate, and can cause incorrect categorization of an event. To provide a high confidence in the value calculated, a number of data quality checks and comparisons are required to verify accuracy and optimize precision. These include verification and calculation of the calibration values for the systems collecting the data, performing a data quality check on the data and making adjustments if needed (for example adjustment of the detector gain for beta-gamma systems), and verifying the concentration calculations and values by using several different approaches. The ASAR project has researched and developed software tools to support analysis of the data and provide confidence in the calculated concentration values. This tool and capability development will be described in the following sections.

Once the data is checked to ensure high quality, the sample measurement can be screened and graded and flagged for further review if necessary. This project investigated a number of approaches in categorization and methods to support efficient screening of sample data; these will be discussed in detail as well.

There are a number of data files produced by the radioxenon systems, the most important for the concentration calculations are the raw spectral (pulse height) data files. This data includes the calibration values for the specific nuclear detector system used to collect the data, allowing a user to review the data and calibration constants, and calculate the concentration calculation independently of any other files.

## 2.1. Nuclear Detector Calibration

When the systems are installed in the field, the nuclear detectors need to be calibrated to ensure the data is collected from a calibrated system to allow correct calculation of the activity. The initial calibration is typically followed by periodic calibrations, although many of the IMS systems are relatively new, there have been few re-calibrations. The detectors can change over time, and go out of calibration, but the majority of the changes can be accounted for by using the quality control (QC) data that is collected before every sample. However, periodic calibration is required to account for larger changes, such as aging effects, that can occur over time.

Radioxenon system calibration involves measuring radioactive isotopes and collecting information to determine the nuclear detector resolution, efficiency, and channel-to-energy conversion factors. Typically all four of the radioxenons of interest are measured, and in some instances, radon as well. The measurements provide the dataset to calculate the calibration values for each isotope's region-of-interest. In addition to the above calibration values, the data is also used to quantify possible interferences of one isotope into the region-of-interest of another. For example, quantifying the amount of the $^{135}$Xe that may show up in the (30 keV) energy region of interest for $^{133}$Xe due to the small (5%) branching of $^{135}$Xe via a conversion electron and x-ray. All of these values are calculated from the calibration measurements and are stored with the system and written to every sample data file. This allows for the activity concentration to be calculated from the raw data using only the information provided in the data file.

The calculation of the calibration values from calibration data is a challenging task, and if not performed accurately, could result in errors in the activity concentration calculation. The calibration of the detectors is therefore very important and the ASAR project has investigated various approaches, especially in the area of determining the detector efficiency. The project also developed a semi-automated software tool for calculating the calibration values given the appropriate calibration data.

The calculation of the channel-to-energy conversion factors and the energy resolution is fairly straightforward. For the gamma detector, there are either gamma or x-ray emissions from the various radioxenons that provide peaks in the pulse height spectra that can be associated with a known energy. The mean of the peak can be used to provide the conversion factor at that particular energy, and a number of these factors fit with either a linear or possibly low-order polynomial expression to provide conversion for any channel, which is then used to define the regions of interest for the concentration analysis. For the beta detectors used in the standard beta-gamma detectors for radioxenon [2-5], the process is more challenging, since only the conversion electrons are mono-energetic and provide a peak in the beta detector energy spectra. However, they can be combined with the beta energy point energies of the various beta distributions to provide the channel-to-energy conversion values.

The detector energy resolution values are used along with the channel-to-energy conversion values to determine the appropriate region-of-interest in the gamma detector response, and also for the conversion electron regions in the beta detector response. The various peaks are fitted with a Gaussian function and the full width at half maximum used along with the mean to determine the resolution of the specific detector at that particular energy.

The detection efficiency of both the beta and gamma detectors is a challenging aspect of the calibration process, primarily due to the short half-life of the radioxenon isotopes. Typically, efficiency calibrations are performed by using a source of known activity, preferably the same isotope in gaseous form to be measured subsequently. However, due to the short half-life, it is challenging to measure the isotope, quantify the activity, and then use it for the calibration source for the nuclear detector, especially if the measurement is performed at a different facility requiring shipment of the standard. For the IMS systems using high purity germanium detectors, the typical approach is to measure longer lived sealed sources at a number of relevant energies, which are subsequently used to determine an efficiency curve as a function of energy.

For the beta-gamma coincidence systems, this approach could be used, but is challenging since the beta detector is also the gas cell and therefore the calibration source needs to be external to the beta detector making a precise efficiency calibration challenging. However, since the beta-gamma system provides three estimates of the same activity with the beta detector the gamma detector, and the beta-gamma coincidences, it is possible to extract the absolute activity of a gas sample. This allows the calibration to be performed with radioxenon gas of unknown activity, and removes the requirement of a calibration standard. The process is challenging in practice, as there are different decay branches to account for, the detectors are not entirely independent, and there is a need to have single isotope samples. The ASAR project has supported increased development of the efficiency calibration process for beta-gamma systems, with expansion of the calculations to include second order effects and the use of the "extrapolation method". The extrapolation method is a more precise method of obtaining the absolute activity value using a beta-gamma detector system, and has been the approach used for absolute measurement in metrology. The project explored using this approach for the radioxenon measurement to calculate the activity, and from there, the efficiency of the detector [6, 7]. This approach is being further explored and is being adopted by the IDC, who has implemented the extrapolation method to validate efficiency calculations for the installed beta-gamma detectors.

Finally, interference terms need to be calculated as part of the calibration process. These are fairly straightforward when using the current region of interest approach, where the ratio of counts in one region to another is calculated. The interference ratios are currently calculated for radon, $^{135}$Xe, and $^{133}$Xe interference into the other regions of interest, but may be expanded in the future. The addition of interference terms for $^{135}$Xe was developed and implemented with support from this project providing for more precise calibration of the PNNL developed beta-gamma systems [8].

Along with the development of the calibration methodologies and algorithms, the project supported the development of a stand-alone calibration tool. A calibration tool that had limited capability was developed under a previous project. A new tool has been enhanced from previous work to have additional capability and provide an easy-to-use interface that allows calibration values to be calculated and written to a configuration file automatically for PNNL developed systems. The input to the calibration tool are the data files from the measurements of the four radioxenon isotopes (and radon if available) and once the appropriate files have been selected by the user and loaded into the system, the calibration values are automatically calculated. The user also loads in the previous calibration file (or similar if the detector is new) and the new values are compared to the old ones, and if significantly different, are flagged for review. The user can visually review the automatic peak fitting in the gamma and beta distributions as shown in Figure 1, and can change the boundaries if the fit is poor and recalculate the values. The tool is currently in the verification and validation process.

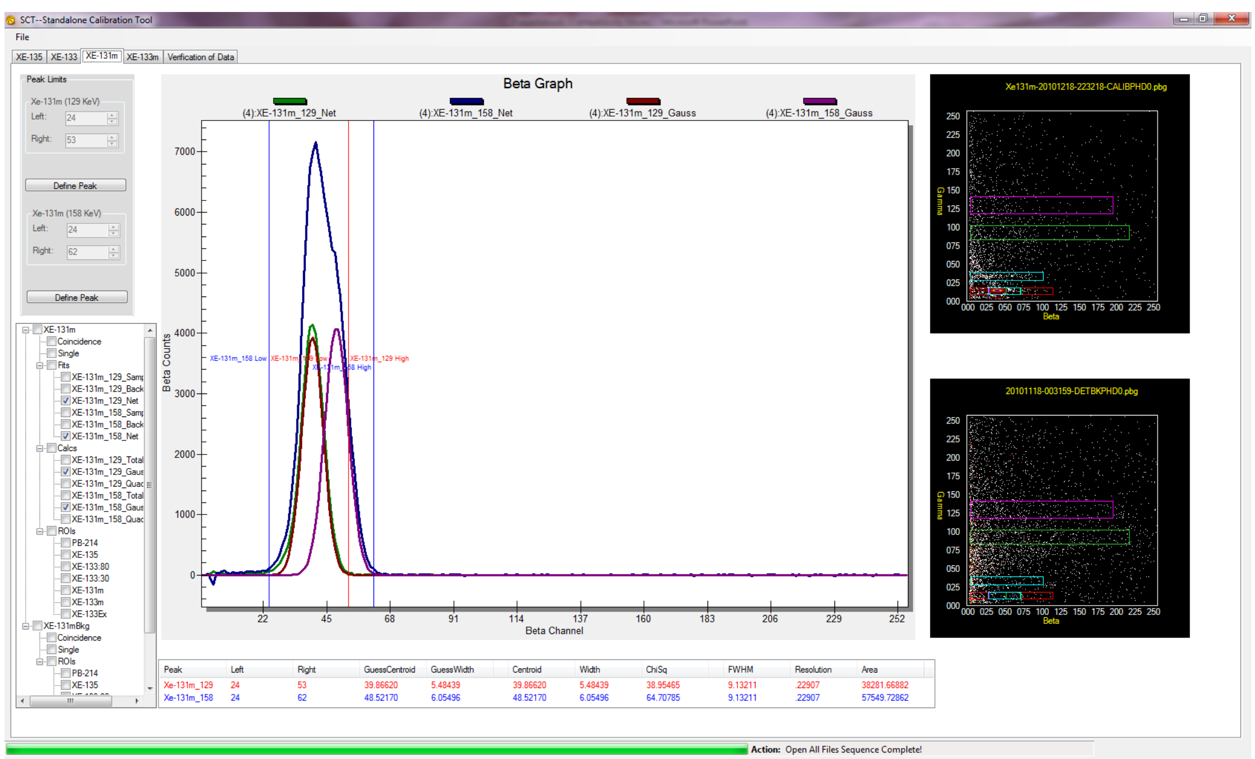


**Figure 1. Screen shot of one of the review pages for the calibration tool showing the fit to the conversion electron distribution of $^{131m}$Xe.**

The research into the calibration methodology and the development of techniques and tools to perform the calibration provide increased confidence of the data being used to calculate the concentration calculations. Currently the calibration tool only processes files from PNNL developed systems, but could be extended to include other beta-gamma systems such as the SAUNA or ARIX.

## 2.2. Activity Concentration Calculations

Once the calibration values provided in a calibration file are verified, activity concentration calculations can proceed. Sample and background data files are required, as well as the gas background and QC data files for beta-gamma systems. In addition to the raw data and the calibration values, the volume of stable xenon (provided in the sample file) that was quantified is needed to be within the normal collection range. If the volume is too low, the quantification could be suspect and will also have a large uncertainty associated with it. If the volume is significantly higher than normal, there are likely issues with the gas separation and the sample is contaminated with other species, such as carbon dioxide.

The raw IMS sample data is provided by the IDC, and the calculated concentration activities produced by the IDC can also be obtained. In order to provide an independent calculation method, and to provide a user interface that is consistent with the normal screening process at the NDC, a database, calculation, and viewer toolset has been developed. On the previous project, a first version of the tool was developed that provided a database and tool to load IMS data into the database, and to calculate and view data from the SAUNA type systems. Under the ASAR project, the basic capability was expanded to include loading of SPALAX and ARIX into the database with capability for viewing, and including concentration calculations for the SPALAX system.

### 2.2.1. SPALAX Activity Concentration

The SPALAX system employs a high purity germanium (HPGe) system, which has a thin entrance window to allow capture of the low energy (~30 keV) x-rays from the four radioxenon isotopes. The SPALAX doesn't have a beta detector on and so only gamma-ray and x-ray data are collected, however, the HPGe detector is the standard in gamma-ray spectroscopy with excellent energy resolution. The calculations at the IDC use both the high energy gamma-ray response as well as a fit to the 30-keV energy region in order to determine the activity concentrations.

For the ASAR tool, the SPALAX concentration calculations were researched and methods developed based on the gamma-ray energies of the radioxenons. The analysis of the 30-keV energy region was also explored, but appeared to provide little additional information to the gamma-ray results. This is due to the fact that the 30-keV region is a convolution of a number of x-ray emissions from the isotopes and the uncertainty on the fit tends to outweigh the additional information obtained. Additional research needs to be conducted to map out the full activity space; however, initial indications performed for a few select cases indicate incorporating the 30-keV region is of limited value [9].

The gamma-ray analysis for the SPALAX data proceeded with assuming the calibration information in the data file was appropriate. The channel-to-energy conversion process used the values provided in the file and assumed a linear conversion curve. The detector resolutions provided in the file were used to help determine the expected widths of peaks for the fitting routine. The efficiency at different energies is provided and used to define a lognormal polynomial distribution which is appropriate for HPGe detectors. Once the spectrum has been converted to energy space, the net counts could be determined by fitting the regions where the radioxenon peaks are expected, with the energies shown in Table 1. For the initial calculation, a peak search is executed within a small energy region around the expected energy for each isotope. If a peak is located, the counts in the peak are integrated, and the background subtracted. The background is estimated by a linear background assumption under the peak anchored by the data in three channels on either side of the peak. If a peak is not located within the energy region, the net count is set to zero.

**Table 1. Gamma-ray emission energies for the four radioxenons.**

| Xenon Isotope | Emission energy (keV) | Intensity (%) |
|---|---|---|
| $^{133}$Xe | 80.997 | 37.0 |
| $^{131m}$Xe | 163.93 | 1.96 |
| $^{133m}$Xe | 233.221 | 10.3 |
| $^{135}$Xe | 249.794 | 90.0 |

Once the net counts *n* are calculated for the gamma-ray peaks in the HPGe spectrum, the activity concentration *C* can be determined at the beginning of the collection period (assuming a constant activity of the collected air) by the usual formulation:

$$C = \frac{n}{\varepsilon BR} \frac{\lambda^2}{\left(1-e^{-\lambda t_C}\right) e^{-\lambda t_P} \left(1-e^{-\lambda t_A}\right)} \frac{t_C}{V}$$

where λ is the decay constant for the isotope, and *BR* the branching fraction into the gamma-ray energy, ε is the efficiency of the HPGe detector at the appropriate energy and the times for collection ($t_C$), processing the sample ($t_P$), and detector acquisition time ($t_A$), and V is the volume of air sampled. This equation is appropriate for $^{131m}$Xe, $^{133m}$Xe, and $^{135}$Xe, but needs an additional treatment for $^{133}$Xe if $^{133m}$Xe is present, since the $^{133m}$Xe decays to $^{133}$Xe. If the $^{133}$Xe value is to be decay corrected properly to a previous time, the contribution that came from the $^{133m}$Xe needs to be accounted for. This correction needs to be applied for the activity concentration for the SPALAX analysis as well as the other system types.

Research was also done on the 30-keV region with the HPGe spectra. This region is populated by x-rays generated from an internal conversion decay process. A conversion electron is also generated, but not detected with the HPGe detector. These generated x-rays are produced in a higher abundance for the $^{131m}$Xe and $^{133m}$Xe than are gamma rays, and are helpful in determining low activity concentrations. The 30-keV region x-rays are used by the IDC in activity concentration and minimal detectable concentration (MDC) estimation [10].

The energies of the x-rays are characteristic of the element instead of the particular isotope, and are therefore associated with the decay element, which for the four radioxenons of interest, are either xenon or cesium. The $^{133m}$Xe and $^{131m}$Xe generate ~30 keV xenon $K_\alpha$ and $K_\beta$ x-rays and the $^{135}$Xe and $^{133}$Xe produce ~30 keV cesium $K_\alpha$ and $K_\beta$ x-ray which are at slightly different energies. The dominant x-rays are shown in Table 2 below, and there are five in each set. The intensities depend on the radioxenon, with the metastables having an x-ray emission in the 30-keV for approximately 55% of the decays, while it is about 50% for $^{133}$Xe and 5% for $^{135}$Xe.

The 30-keV energy region in the SPALAX data is only useful for metastable detection and only when the activity of the sample is low. The $^{133}$Xe and $^{135}$Xe have more sensitivity with the gamma-ray analysis only, and the 30-keV region is not used to estimate activities for these isotopes, but used to help estimate activities for the metastables. When the metastable activity is high, the 30-keV energy region is not as valuable for the metastable activity estimate, as the gamma-ray emissions (even with the low branching fractions) are adequate.

For samples with a significant amount of $^{133}$Xe, the 30-keV region is also not very useful for the metastable activity estimates due to the masking of the metastable xenon isotopic signature by the $^{133}$Xe. Although there does appear to be value in adding in the 30-keV energy region for the metastable analysis for cases where only $^{131m}$Xe is present at low levels, but $^{133}$Xe is not [11], it is not clear how often this scenario would arise for events of interest, though old “medical” xenon has a high ratio of $^{131m}$Xe to $^{133}$Xe. If both metastables are present, then it can become challenging to determine the relative contributions since the emissions are the same energy. For this case, the preliminary data files (the data is saved every two hours in these systems) can be used to estimate the different contributions based on a fit of the activity versus time using the two different half-lives of the metastables.

**Table 2. Dominant x-ray energies and intensities for internal conversion of xenon and cesium. [12]**

| Element | Dominant x-rays | Energy |
|---|---|---|
| | $K_{\alpha2}$ | 29.458 |
| | $K_{\alpha1}$ | 29.778 |
| | $K_{\beta3}$ | 33.563 |
| | $K_{\beta1}$ | 33.624 |
| Xenon | $K_{\beta2}$ | 34.414 |
| | $K_{\alpha2}$ | 30.625 |
| | $K_{\alpha1}$ | 30.973 |
| | $K_{\beta3}$ | 34.919 |
| | $K_{\beta1}$ | 34.987 |
| Cesium | $K_{\beta2}$ | 35.821 |

Typically, the detection efficiency of the HPGe for radioxenon detection is fairly low in the 30-keV region due to counting geometries and the attenuation of low-energy x-rays in the entrance window and dead layer of the HPGe crystal, and rapid changes could lead to large associated uncertainties. The fit of the data is challenging, involving a possible ten peaks and the background and can also result in a large uncertainty (see Figure 2 below). It appears, from the initial analysis that has been done on the ASAR project, that there are few sample scenarios where the 30-keV energy region analysis would provide benefit. At this time, analysis of the 30-keV region is still being investigated, but the analysis has not been incorporated into the NDC software to date.

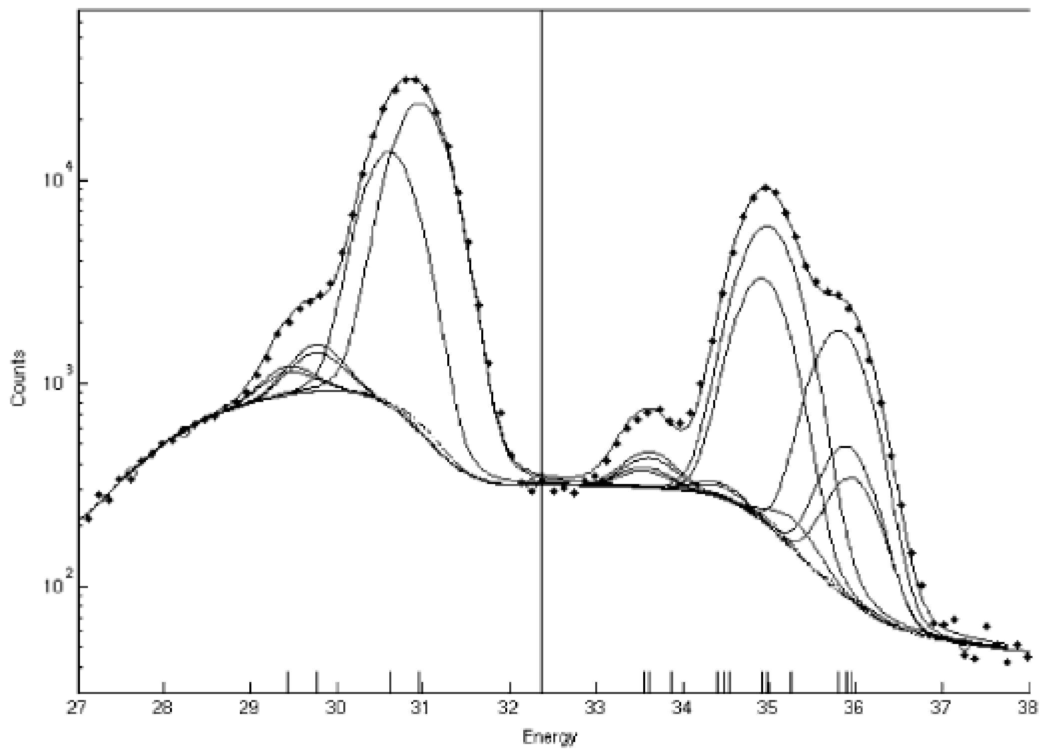


**Figure 2. Illustration of the fitting process in the 30-keV region of an example HPGe spectrum containing $^{131m}Xe$ and $^{133}Xe$ [13].**

### 2.2.1. Additional Software Updates

A loader and viewer program initially developed under the Data Center project was updated under the ASAR project to increase capability. This software toolkit was called the JavaViewer toolkit, as is was developed in the Java programming language, primarily to be as platform independent as possible. During the first two years of the ASAR project, the additional capability for the SPALAX data was added, and beta versions provided to the NDC, with user feedback being incorporated into the software. PNNL installed new releases of the JavaViewer toolkit on the NDC test bed database for their evaluation and feedback. During this evaluation, and due to issues identified at PNNL, it was decided to fundamentally change the JavaViewer toolset to be more functional and easier to use.

The Viewer part of the JavaViewer toolkit was a Java application that was launched on the local computer through a web interface. This allowed the software to be fairly platform independent, and allowed use of the software applet without installing software on the local computer. The mapping of the Java classes to the database was handled using the open source Hibernate tool. The Viewer met the requirements of the NDC, but wasn't very responsive in handling large numbers of data files, especially when reloading a database with all the files. Therefore, a decision was made to have the application operate on the server where the database itself resides, and provide a web-based user interface (web UI) to view the data in a normal web browser. The Viewer application was migrated to the server, and a web UI developed for viewing on remote computers. Hibernate was replaced within the Viewer application with Java DataBase Connectivity (JDBC) calls, with the intent to move to the Lightweight Object Relational

Mapping tool (OrmLite). The loader program was updated, replacing Hibernate with OrmLite, greatly increasing the responsiveness of the program.

In addition to moving the viewer program to the database and changing out the Hibernate tool, the viewer program was upgraded to better reflect the operational usage. Instead of having a view with tabs to all the information of a sample (sample and background files), multiple views were created to allow specific tasks to be carried out more efficiently. For the initial sample review, a simple page is displayed that provides the information required to review the sample and fill out the review form for the majority of the sample cases. New status lights were added to the top of the review page providing color-coded status of a number of parameters or files needed for the review. Information for samples requiring additional investigation can be accessed by an ‘Advanced’ button, which allows the user to view all the data and associated spectra from the sample and backgrounds as applicable. The web UI is more intuitive to use, being similar to contemporary web pages and applications.

Since this change is a significant upgrade to the JavaViewer toolset, the software name was changed from “JavaViewer” to “Watchmen”. A screenshot of the new web UI is provided below in Figure 3 for the initial review of the sample. The color coded status lights are provided at the top, with green indicating the value is within normal range, and red outside the range, or not available, and yellow for the QC file if it has not been reviewed yet. In Figure 3, the detector background file (DetBack) was missing for this SAUNA sample and therefore the status light is red. Below the status lights are spectra, in this case the beta-gated gamma spectra on the left, and the two-dimensional beta-gamma spectrum on the right. The user can change the scale from linear to logarithmic and zoom in on particular interesting features if required. Below the spectra are the parameters and values associated with the sample such as the station, times, and activity concentrations.

The radioxenon activity concentrations are color coded if above the minimal detectable concentration (MDC) in red or, if above the detection limit ($L_C$), in yellow. To the right of the results box is a button to complete the review, which brings up another box where the user can grade the samples and provide comments, and either complete the review or assign it to another reviewer for more in-depth analysis. If the user requires more information to assess the sample, more information can be accessed by pressing the ‘Advanced’ button on the upper right hand corner of the screen. This will bring up another view with additional tabs for viewing all the associated spectra for the specific sample. This structure is consistent with NDC needs and current operating procedures at the IDC.

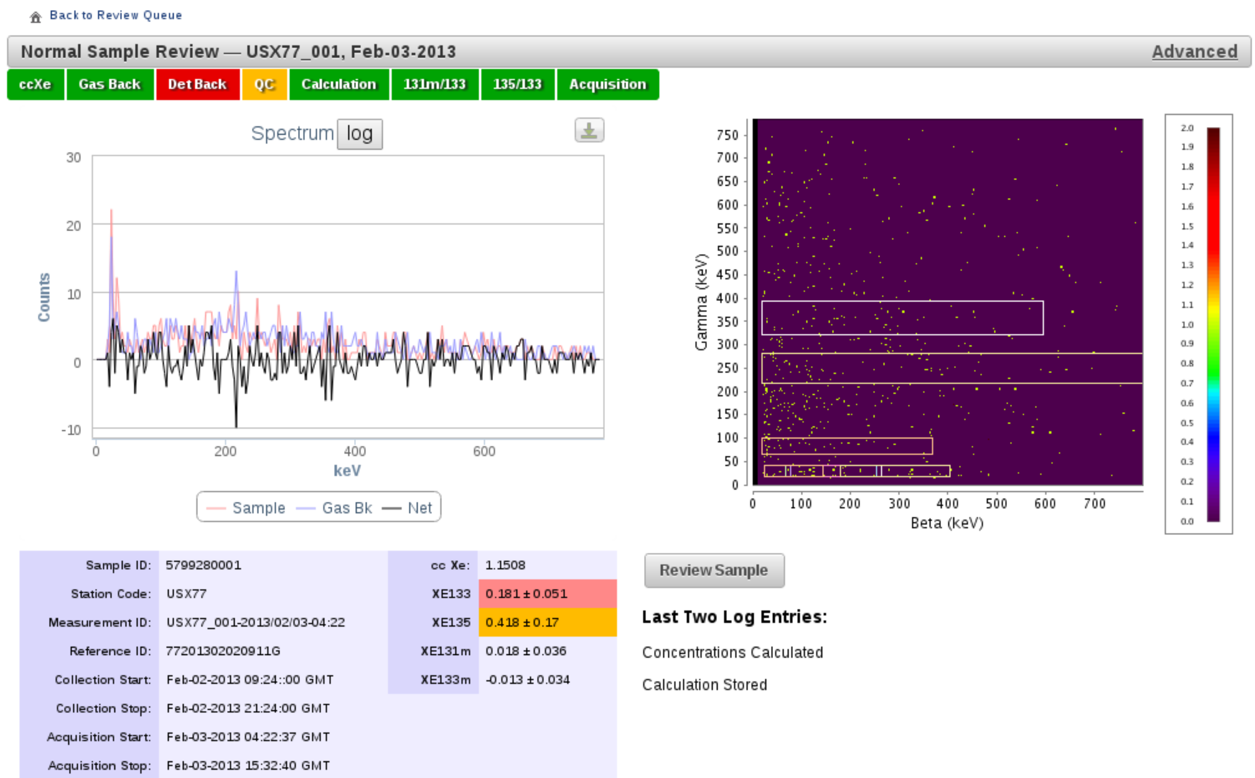


**Figure 3. Screenshot of the Watchmen viewer application showing the review screen for a beta-gamma generated sample.**

Also included in the view below the information on the individual sample are two visual plots that provide the values for the specific sample in terms of previous samples. These two plots are a trending plot, where the values (concentrations or xenon volumes) are plotted for this specific station as a function of time, and a frequency plot, where a histogram of the frequency is plotted on one axis, and the cumulative percentage is plotted on the other axis, with the current sample location displayed with a red line (see Figure 4). These plots help the reviewer to determine if the values of the current sample (volume of xenon and activity concentrations) are typical for this station, or anomalous. This starts to touch on the concept of categorization, which will be discussed in further detail in the section 3 below.

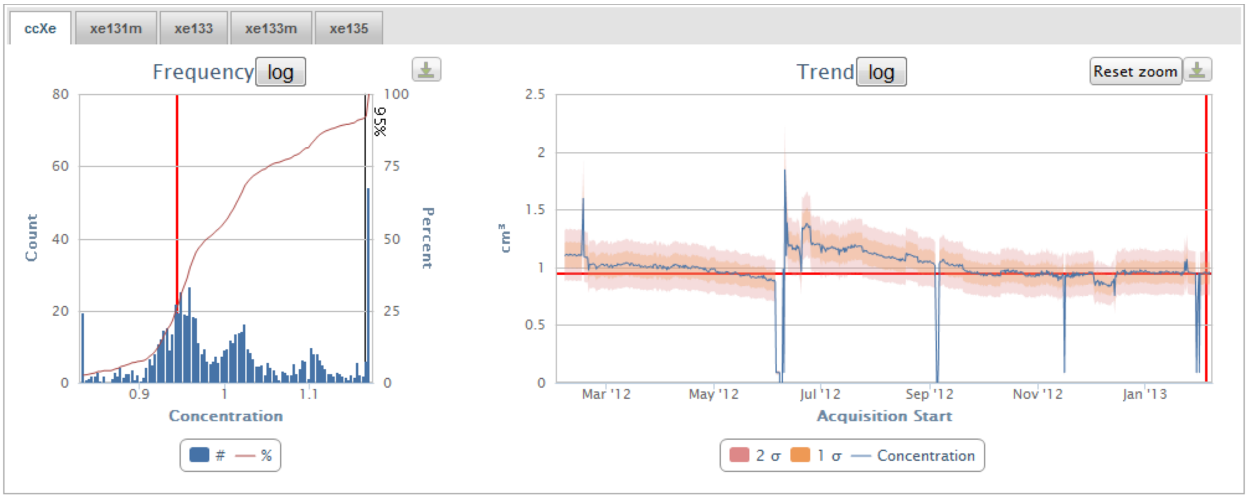


**Figure 4. Screenshot of the frequency and trend plot of the Watchmen viewer tool. The graphs are on the tab showing the stable xenon yields.**

## 2.1. Additional Analysis Approach

The current analysis approach to calculating the radioxenon activity concentrations starts with a determination of the net counts for each isotope. For the SPALAX HPGe system, this takes the form of finding and fitting peaks with energies associated with the particular radioxenon isotope and determining the total number of counts within the peak. For the analysis at the IDC, the backgrounds are estimated by fitting the full spectrum underneath the peaks with a cubic B-spline, and subtracted from the total peak count to obtain the net peak count. Net counts in each peak are summed together for isotopes with multiple peaks.

For the beta-gamma systems, the analysis has more process steps due to the data being coincidence data and the detector technology used. Both the gamma and beta detectors provide energy information, which results in a two-dimensional beta-gamma distribution (as shown in the upper right hand plot of Figure 3). The different radioxenon isotopes provide different distributions in the two-dimensional spectrum and the activity of each can be extracted. The simplest method to do this, and the one currently employed at the IDC and NDC, is to use a simple two-dimensional region-of-interest for each isotope (shown in Figure 3 as rectangular boxes). The counts in each region are simply summed and the appropriate backgrounds subtracted. There are several additional complications; the first complication is that regions of interest overlap for the various isotopes. These are accounted for by using non-overlapping regions to determine the contribution in the overlapping regions, and simply subtracting out the interference. Another complication is due to the beta detector, which absorbs xenon gas during the data acquisition, and is not completely removed when the sample is evacuated. Up to ~5% of the xenon gas can remain in the cell walls (plastic scintillator material), and add to the next sample count rate. To account for this memory effect, a ‘gas’ background data acquisition with an empty cell is taken prior to the sample acquisition. The net counts of this gas background are calculated, decay corrected to the sample acquisition time, and subtracted out of the sample net counts to remove any possible memory effect.

Although there are more steps in calculating the net count than for the HPGe spectra of the SPALAX systems, the regions-of-interest analysis approach used in the beta-gamma analysis is quite simple and the beta-gamma technique is much more powerful than gamma singles spectroscopy. Sophisticated approaches may provide more accurate activity concentration, especially for mixed radioxenon samples with low activity.  A more sophisticated approach can provide increased confidence in activity concentrations for high interest samples, since it provides an additional independent method of analysis, and may be more accurate.

One approach to a more sophisticated analysis is through fitting a sample with standard spectra.  The standard spectra are the detector response functions for each individual isotope of interest, which includes the four radioxenon isotopes and radon.  The standard spectra or templates are combined together and used to determine the optimal contribution of each to provide the best fit to the sample.  If the overall fit is poor, if might indicate other interfering isotopes are present in the data. The advantage of this approach is that it takes into account the entire two-dimensional spectrum and uses all available information.  This approach was developed for the beta-gamma radioxenon data by S. Biegalski of the University of Texas to provide increased accuracy of activity concentrations for mixed samples [14-17].

The standard spectral analysis approach is of interest to the NDC and the ASAR project sponsored Dr. Biegalski to spend a one-year sabbatical at PNNL.  During his time at PNNL, Dr. Biegalski led the development of a software tool that incorporated the standard spectral analysis approach.  This tool was developed as a standalone tool with a graphical user interface with the anticipation that this could be used at the NDC (see Figure 5 below).  The tool is called the Spectral Deconvolution Analysis Tool (SDAT) and developed in the C# programming language.

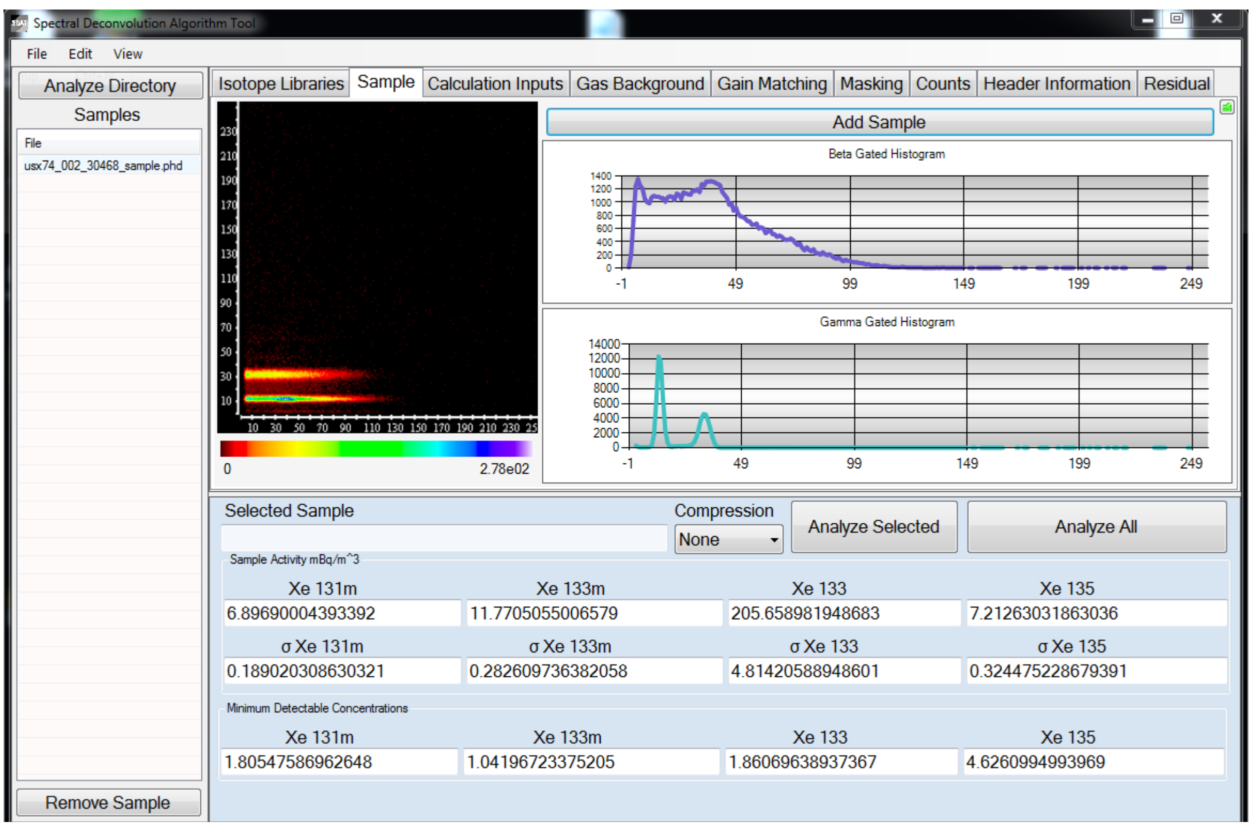

**Figure 5.  Screenshot of the SDAT tool graphical user interface.**

Although the concept is straightforward, there are several processing steps in the SDAT analysis as with the region-of-interest approach, but with added complication of the two-dimensional array instead of a single number. The background, gas background, and decay corrections need to be appropriately accounted. In addition, the standard spectral responses (library) have to be generated and these may need to be generated for individual detectors although the research to date indicates a general library for a single type of detector may be adequate. The standard spectral approach is quite sensitive to gain changes and each sample requires gain adjustment to be consistent with the standard spectra or templates. There is also some sensitivity to sections of the spectrum where there are few counts or only background counts. Since there are few counts in these regions, the fluctuations can be significant, and can affect the final activity concentrations. To mitigate this, the tool has incorporated a method to mask off regions of the two-dimensional spectrum, providing increased weight to the areas with higher counts.

The standard spectrum approach provides an additional method to calculate concentrations and can provide increased confidence in the estimated activity concentrations for samples with activities near the minimal detectable concentration.

# 3. Categorization of Radioxenon Events

The ASAR project has explored the interpretation of detected radioxenon events from the IMS and reported initial findings [18]. There are often positive detections (above the minimal detectable limit) of radioxenon from the stations, with $^{133}Xe$ detection being the most common, arising from benign sources. There are several benign sources of radioxenon in the atmosphere including nuclear reactor power plants and medical isotope production facilities. One of the interesting findings of the International Nobel Gas Experiment has been to determine that medical isotope production contributes far more radioxenon to the atmosphere than is emitted by nuclear power plants. However, it is challenging to discriminate radioxenon produced by medical isotope facilities from nuclear explosions, more so than it is to discriminate nuclear power plant emissions. With the number of xenon detections, it is impossible to fully explore each sample manually and associate it with a benign source. The issue of weeding out the truly benign detections of radioxenon to allow further analysis of suspect (although likely benign) detections is the focus of categorization.

Determination of the source of a detection of a single radioxenon isotope is almost impossible without additional information, which is the reason for requiring the capability to measure four radioxenon isotopes. With the four different radioxenon isotopes, various ratios can be formed and can provide information on the origin of the radioxenon. For example, multiple isotope ratios can discriminate most nuclear reactor power plant emissions from nuclear explosions [19]. An example of this type of analysis is shown graphically in a multiple isotope ratio plot below in Figure 6. Typically only during startup of the reactor with fresh fuel before equilibrium is when a power plant will emit the radioxenon isotopes in ratios that could mimic nuclear explosions. Medical isotope production is more challenging, as the ratios overlap the regions of nuclear explosions under some scenarios.

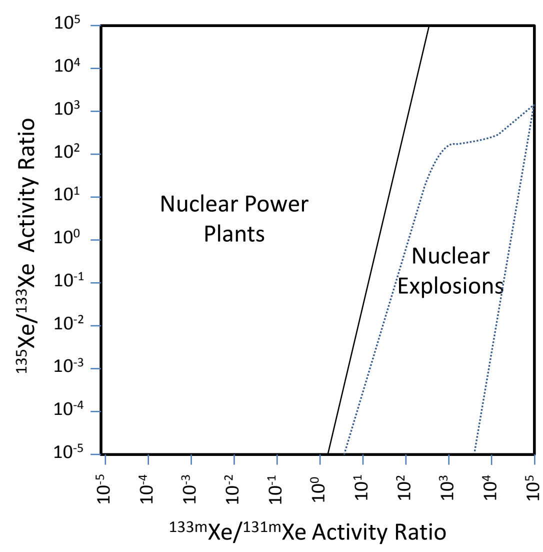


**Figure 6. Multiple ratio isotope plot showing the regions associated with nuclear power plants and explosions.**

One issue of determining the source of radioxenon detection is the many variations that could occur for an underground nuclear explosion scenario (if above ground, detection of particulates will be more useful). The amount of xenon that gets released and how long after the explosion affects the ratios of the isotopes. Xenon can be generated from iodine, if iodine is released as well, it can affect the radioxenon ratios and cause changes over time. The different half-lives come into play depending on how far the source is from the detecting station, and how the atmosphere (weather) transports the xenon to the station. With all these factors, the ratios for a nuclear explosion can vary over a wide range and overlap ranges of medical isotope production (which typically has fewer affecting factors). The overlap is shown schematically in Figure 7 where it is not possible to determine the source without additional information.

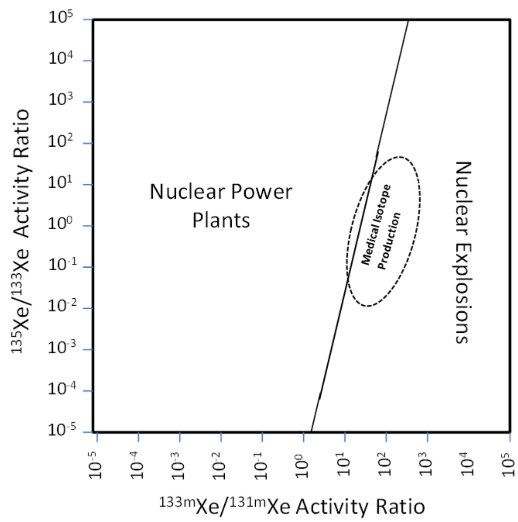


**Figure 7. Multiple ratio isotope plot showing the regions associated with nuclear power plants, medical isotope production and nuclear explosions.**

As a starting point for categorization, the measurements from each station can be compared to historical data. This can help provide an analyst with information whether a measurement above the minimal detectable concentration is abnormal, or typical for the particular station. The ASAR project developed a graphical representation to compare the current sample with historical data as part of the Watchmen tool. An example is shown in Figure 8 for an IMS station where the $^{133}$Xe activity concentration is being compared. On the left hand plot is a frequency distribution of the $^{133}$Xe activity concentration over the last 12 months. The current sample is shown as a vertical red line to provide a relative indicator. The cumulative distribution is also plotted to allow a quick estimate of where the current sample is in terms of the percentile for the station. Notice the several measurements that are in the furthermost right hand bin (overflow); these are the measurements above the maximum plotted concentration value, representing about 5% of the samples. On the right hand side of Figure 8 is the same activity concentrations over the last year now plotted as a function of time. The blue line shows the concentration, while the red line provides the minimal detectable quantity. This station has multiple detections of $^{133}$Xe over the last year; these have been attributed to the Chalk River medical isotope production facility by the station location and atmospheric transport information. However, without additional information such as the atmospheric transport model results, it becomes challenging to resolve detections with high activity concentrations. The particular sample being analyzed, although above the minimal detectable concentration, falls in the middle of the historical distribution and can be associated with the ‘normal’ background for this station.

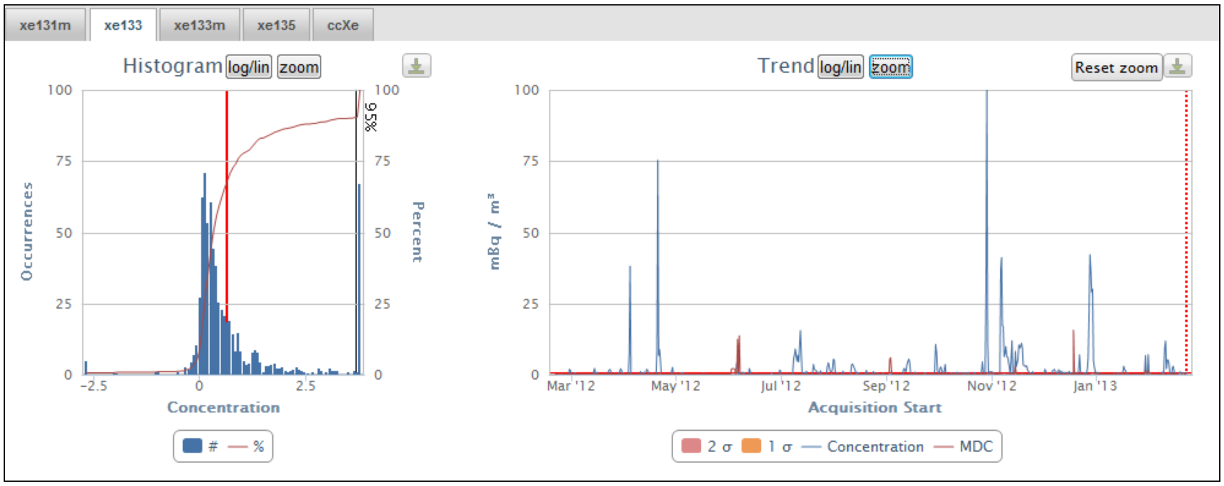


**Figure 8. Example of the frequency histogram and trend series for $^{133}$Xe for an IMS station.**

This type of historical analysis suggests a path forward for categorization. Categorization is important to screen the samples to focus only on the important samples that might be associated with a nuclear explosion. Obviously, the measurements below some value, such as the detectable limit, can be screened out unless other information provides reason to examine further. Measurements that are normal for that particular station could be set aside as well without additional information. The samples that have measured activity higher than the normal for the station should be investigated further. This simple scheme is the method currently employed by the IDC for xenon categorization [20]. There are some challenges with this approach, however. Determination of normal for a station can be challenging since the measured xenon arises from some source such as medical isotope production, which may not be produced continuously, but instead periodically. In this case the transport of the plume to the station is not always the same, but depends on the weather and at times the plume doesn't even reach the station. This results in distributions such as shown above in Figure 8 in which the measurements are not constant but vary with a number of high activity concentrations which may be normal for the station, but are not constant in time and appear as singular events.

Even if the measurements at a particular station were more constant and evenly distributed, it may not make sense to categorize the event as normal and complete the review as there is no way to distinguish the source of the event without further information. However, as a method for screening out the majority of benign samples, the concept of categorizing the event as not detected, normal, or abnormal is a logical first approach. The main issue then is in defining normal for a particular station, and when to declare detection abnormal. One approach may be to set a limit based on the number of events that can be processed by a particular organization. For example, a review team may only be able to examine 5-10 radionuclide events per day in detail. Therefore, a limit could be set on the IMS as a whole to limit the number of abnormal events to the ability of the review process. Another approach would be to define normal as any detected isotope and level that has been observed in the past. This would dramatically reduce the number of abnormal events, but may lead to screening out events that require more in-depth review.

Another approach is to combine the single isotope information with ratios of isotopes. This has been studied by various groups and provides additional information to focus on events of higher interest [21-23]. One implementation challenge is that many detected events only include single isotope detection, and ratios can't always be calculated. One could substitute the MDC for a non-detect, in order to form the ratio, which may provide some benefit. Another challenge is that the ratios don't necessarily provide the discrimination power since medical isotope production can produce isotopic ratios similar to nuclear explosions. However, implementation of this type of approach could help in sorting out a subset of the detections that should be reviewed further. A schematic of a screening process with the added ratios is provided in Figure 9, where a five-level approach is proposed, with the three levels as stated above are used along with additional ratio information which upgrades the event to either one or two more levels based on the value of the ratios.

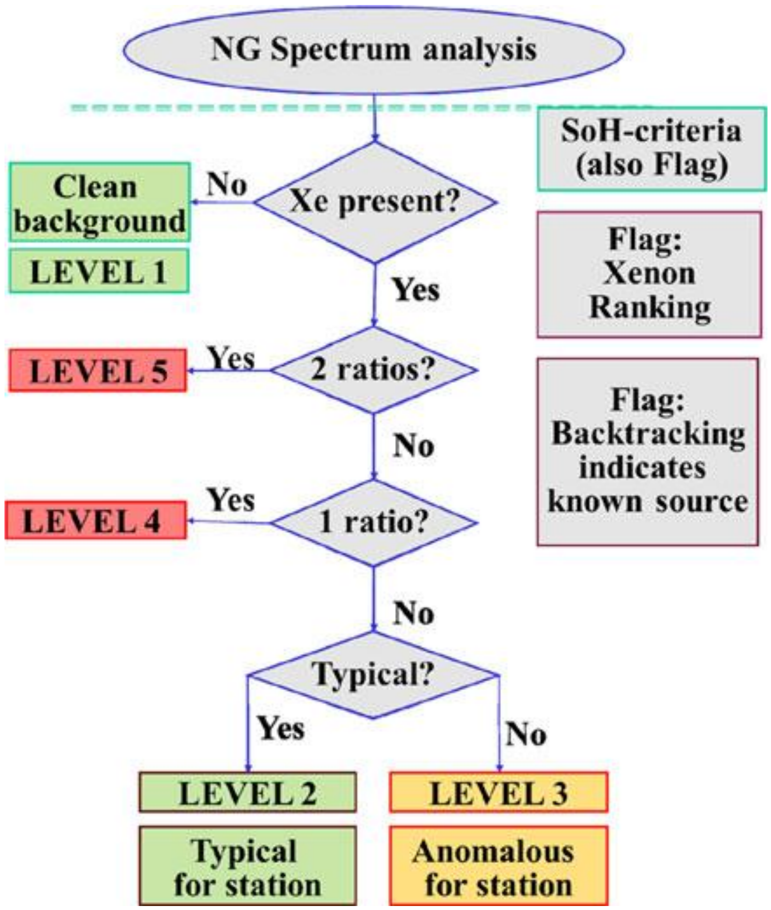


**Figure 9. Schematic of categorization of radioxenon noble gas (NG) events taken from [17]. The decision path can also be influenced by additional flags shown on the right hand side, such as sample State-of-Health (SoH) data.**

Although there are different approaches to categorizing radioxenon data, in some cases there may not be enough information from the xenon measurements alone to be able to provide a high confidence determination of a nuclear explosion. Under conditions in which all four isotopes are not detected because the concentration of one or more of them are below the systems' capabilties, discrimination can be challenging. However, even with all four isotopes measured at levels the MDC, there still can be an ambiguity with medical isotope production and reactor start-up. Therefore, additional data streams must be included in a more holistic fashion in order to fully resolve events. This is already performed for events of interest, where seismic or other information indicates an interesting event, or when the radioxenon measurement incorporates backtracking of atmospheric data using modeling. However, it may be beneficial to develop a framework where the radioxenon is combined with other data streams earlier in the process before categorization takes place. In this way, more information is available to perform a better categorization, which may demonstrate a different, but more interesting set of events selected for further exploration.

The first obvious addition to the radioxenon data in terms of improving categorization would be to include the atmospheric transport modeling (ATM) results. ATM data provides a field-of-regard (possible locations of the release) for each measurement and could be combined with the radioxenon measurements to help determine the normal distribution for a station. For example, if a station has periodic detections arising from an isotope production facility, ATM results could provide further support that a specific detection likely came from the facility. If the ATM indicated the isotope production facility was not a likely source, this may provide additional support to categorize the event as abnormal.

Another related data stream is the particulate data. Since the xenon stations are co-located with the particulate stations, it makes sense to combine the data together, since the ATM and field of regards would typically be the same for radioxenon and particulates. The particulate data is currently categorized by itself, and only if further review is required, other data, such as the radioxenon, is reviewed in combination. However, it would be interesting to research the possible benefit to combine the data, such as radioxenon, particulate, and ATM, prior to categorization. Due to the fact that under most expected nuclear testing scenarios the release of particulates is unlikely, this work hasn't been addressed in the past. There may be a set of events not currently selected for further review, which are illuminated by the combination process. Finally, one could envision a review process where all the data from the IMS including the seismic, acoustic, infrasound, and radionuclide is combined prior to categorization. Obviously, this would require a sophisticated automated approach, but may ultimately provide the most efficient method to process and screen the data from a network such as the IMS.

The work of exploring the optimal framework to combine the various data streams into a more holistic approach is the focus of the follow-on project, the Integrated Nuclear Signature Interpretation of Global Happening Toolset (INSIGHT) project. The primary focus of INSIGHT will be to research the most effective method of combining the particulate and radioxenon data together, perhaps along with the ATM results, in order to increase the efficiency of screening samples and rapidly identify the events of interest for further review.

# 4. Summary and Way Forward

The ASAR project has made significant progress in supporting the NDC in analysis of radioxenon samples. These samples are primarily from the IMS network which is comprised of several different station types and associated nuclear data. The ASAR project has investigated methods and developed tools to help provide confidence in the xenon measurements. This includes confidence in the nuclear detector calibration, and independent calculation approaches that provide confidence in the concentrations provided by the IDC. As part of this, the ASAR project worked on development of an alternative calculation approach for beta-gamma systems based on a standard spectrum fitting or template matching algorithm. This approach has advantages for low activity samples, and provides further confidence in the concentration values.

With the increased confidence in the activity concentration values, approaches to categorizing radioxenon events were explored. This is an active research area and no simple approach is satisfactory. Although the systems measure four radioxenon isotopes, which can provide much more information when combined in ratios that when analyzed individually, they are not always detected above the MDC of the system. Even in cases where several or all isotopes are detected, the radioxenon data can't discriminate nuclear explosions from medical isotope production or reactor start-up for all scenarios. In these cases, additional information is required to support unambiguous resolution of radioxenon detections. However, the radioxenon data can be screened to narrow down the number of detections that require further analysis and combination with other data for resolution, and an approach has been implemented at the IDC. This method has three categories: no detection (below MDC), normal detection for the station, and abnormal detection, requiring further review. The challenge in this simple categorization approach is defining normal for a station, since the activity concentrations can vary significantly and yet still be benign. It is possible that combining additional data, such as ATM or particulate, prior to categorization may make the process more efficient.

A more holistic approach to the data screening and categorization is the focus of the follow-on INSIGHT project, which will research data fusion prior to screening and categorization as a possible method to increase screening efficiency and identify the appropriate high-value events for further review.

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