INTERFERENCE TERMS FOR XENON-135

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ABSTRACT

Systems to measure radioxenon concentrations are continually being improved to provide greater sensitivity and precision. As the systems become more capable, additional terms need to be added to the calculations to account for small previously insignificant effects such as minor decay branches. Recently, additional terms present in the ^{135}Xe decay were added to the calculations to account for the small (5.2%) fraction of decays that produce cesium K-shell x-rays with an average energy of 32 keV and the Compton scatter of the dominant 250-keV gamma ray. These events overlap with the regions of interest of ^{133}Xe, ^{131m}Xe, and ^{133m}Xe. In typical field applications, the amount of expected ^{135}Xe is low due to the short half-life (9.14 hours) therefore the small fraction that could be mis-categorized as other isotopes is insignificant in most cases.

However, for much more active samples, such as those recently measured in calibration and testing samples with high activity, the amount of mis-categorization can be significant. The additions to the calculations use calibration values to estimate the amount of a possible interference of ^{135}Xe in the other regions of interest, which are then subtracted out. The calibration values, which are ratios of counts in one region of interest to another, are obtained by measuring with single isotope gases during the calibration process.

OBJECTIVES

Ground-based nuclear explosion detection systems currently available include particulate and radioxenon samplers. There are now several nuclear detector subsystems being used in support of nuclear explosion treaty monitoring. These detectors range from β–γ (Reeder, 1998; Cooper, 2005; Cooper, 2007) to high purity germanium (HPGe) detectors. These detector systems provide count rates that are used to calculate activities, and with the volume measurements of the stable xenon from the quantification subsystem, provide the necessary values for the concentration calculation.

These systems allow for simultaneous measurement of the radioactive xenon isotopes including ^{135}Xe, ^{133}Xe, ^{131m}Xe and ^{133m}Xe. The gamma region of interest for ^{133}Xe is in the 80 and 30 keV regions while for ^{131m}Xe and ^{133m}Xe it is the 30 keV gamma range. The ^{135}Xe isotope has a small branching fraction (5.2%) that provides x-rays in the ~30 keV region, and Compton scattering can contribute in this region of interest as well from the dominate (90%) decay branch that produces a 250 keV gamma ray. Due to the short half-life, the expected amount of ^{135}Xe will be small for most measurements, and therefore, the contamination to the 30-keV region of interest negligible, and has been ignored in the calculations to date.

However, when measuring highly active samples, such as during calibrations or measurement exercises, the small fraction of ^{135}Xe in the 30-keV region can be misidentified as one of the other xenon radioisotopes (in particular ^{133}Xe). In order to reduce error in calibrations and confusion in interpretation of exercise results, additional terms in the calibration and concentration calculations need to be included. For the PNNL developed detector systems and software, these additional terms have recently been added. This improves the precision on the calibration parameters ascertained during the calibration procedure, and will improve comparisons to other types of radioxenon measurement systems during round robin exercises.

For PNNL developed systems, the calculations of the beta-gamma detector response use seven two-dimensional regions of interest (ROIs). The seven ROIs are defined by a range in both γ and β energy. The regions are ^{214}Pb (ROI-1), ^{135}Xe (ROI-2), ^{133}Xe (ROI-3), ^{133}Xe (ROI-4), ^{131m}Xe (ROI-5), ^{133m}Xe (ROI-6), and an exclusion (ROI-7) region (see Figure 1). Each region corresponds to an important β-γ signature or combinations of signatures for each isotope. The ^{214}Pb region (ROI-1) contains just radon daughter products and no radioxenon isotopes. The ^{135}Xe (ROI-2), and ^{133}Xe (ROI-3) regions are free from major interferences of the other radio xenon isotopes, but can contain interferences for example from Compton scattering. Regions 4, 5, 6, and 7 are admixtures of ^{133}Xe, ^{131m}Xe and ^{133m}Xe. Region 7 represents a region with maximal possibility of meta-stable (^{131m}Xe and/or ^{133m}Xe) interference while ROI-4 minus ROI-7 represents a predominantly ^{133}Xe signature.

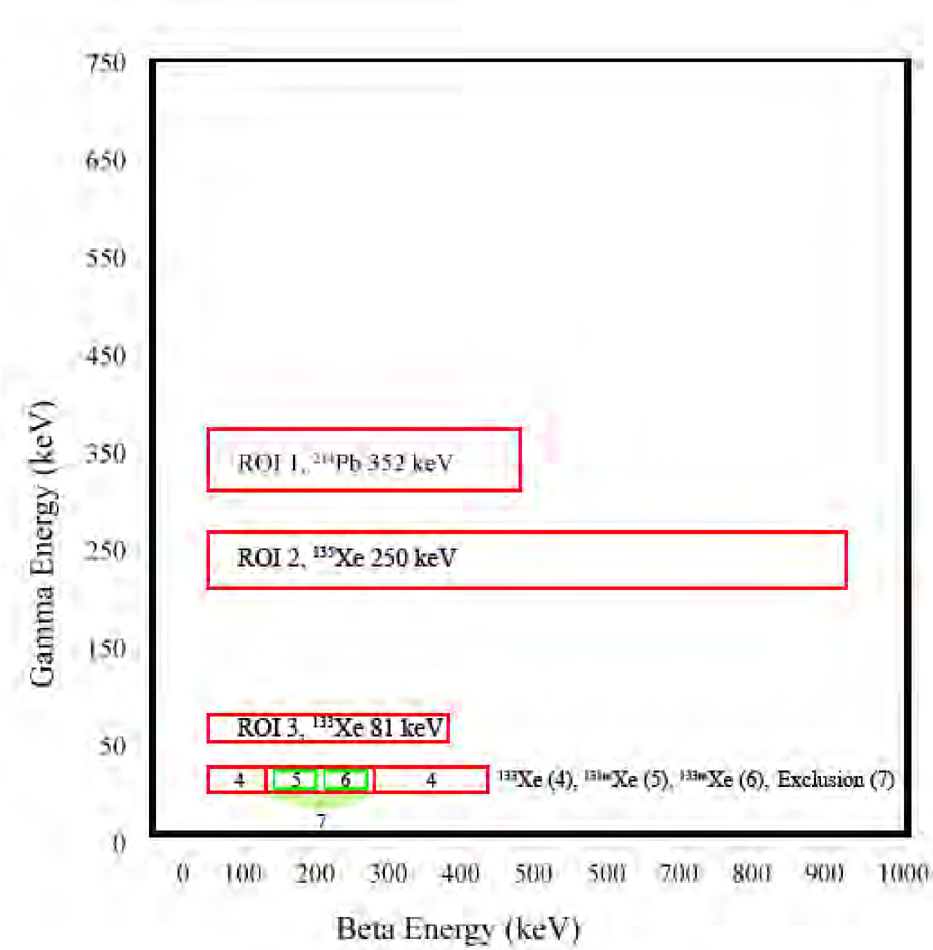

Figure 1. Schematic illustrating the regions of interest (ROI) naming convention that is used for PNNL developed beta-gamma radioxenon detector systems.

Ratios of the ROIs are used to account for interference terms including contributions from radon daughters as well as the two metastable isotopes, and from ^{135}Xe, the focus of this paper. For the radon interferences, the ratios of counts are calculated between ROI-1 and the six other ROI (1:2, 1:3, 1:4, 1:5 1:6, and 1:7) for a pure radon sample (daughter products ^{214}Pb and ^{214}Bi). These ratios are then used in a normal gas measurement to estimate the interference contributions by multiplying the ratios and the measured radon contribution in ROI-1 and then subtracting this estimated interference from the respective ROI. The other important interference ratios are based on ^{133}Xe in the ROI's of the two metastable isotopes. Here the ratios are 3:4, 3:5, 3:6, 3:7, and 3:(4-7). The 3:4 ratio is important for the case where there are no metastable isotopes. The 3:5 and 3:6 ratios are important when there are metastable isotopes present. Finally, the 3:7 and 3:(4-7) are important for determining if there are metastable isotopes present at all.

The ^{135}Xe radioisotope beta decays with a 9.14 hour half-life to one of several excited states of cesium-135 and from this state there are several decay paths to the ground state. The predominant path is via an emission of a 249.8- keV gamma ray and occurs 90% of the time. This path can result in interferences in the lower energy ROIs when the gamma detector does not collect the full energy (Compton scattering). The other interference contribution arises from the decay path where a conversion electron (214 keV) is produced with an accompanying x-ray of around 30 keV (exact energy depends on the shell where the x-ray originates). This decay path occurs in 5.7% of the ^{135}Xe decays, and is a minor contribution to most measurements, but can be significant in calibrations or measurements of high activity ^{135}Xe.

RESEARCH ACCOMPLISHED

The radioxenon concentrations are calculated from the beta-gamma detector response using the so-called "Stockholm equations." The Stockholm concentration equations are used for calculation of radioxenon concentrations, and were the consensus of an international experts working in the area of xenon detection. These equations use the net count *C* and correct for decay branches, detector efficiencies, and decay during the collection, processing, and data acquisition times (T_C, T_P, and T_A respectively) to calculate an activity, which is converted to concentration by dividing by the volume of air that was sampled:

$$Concentration\left(\frac{mBq}{m^3 air}\right)=\left(\frac{C}{\varepsilon_\beta \varepsilon_\gamma \beta_{BR}\gamma_{BR}}\right)\left(\frac{\lambda^2 T_C}{(1-\exp(-\lambda T_C))(\exp(-\lambda T_P)(1-\exp(-\lambda T_A))}\right)\left(\frac{1000}{V_{air}}\right)$$

where ε_γ and ε_β are the beta and gamma detector efficiencies, β_{BR} and γ_{BR} are the beta and gamma branching ratios for the particular ROI, and λ is the decay constant for the particular isotope. Each of the four isotopes uses the same equation with the appropriate parameters and values for the constants.

The net counts C, for each ROI are calculated by subtracting the appropriate background, memory effects, and interferences from the sample file:

$$C(net) = Sample - DetectorBackground - MemoryEffect - Interferences$$

For the current beta-gamma systems, there are two background files that are used where the counts in each file for each ROI are summed and corrected for dead-time. The detector background is a long background measured when the memory effect is negligible. This is used as the primary background, and is normalized to the sample acquisition time, before being subtracted. The background is also subtracted from the gas background, after it has been normalized to the sample time. The background subtracted sample file is then corrected for possible interferences, which at present include radon in regions 2-7, ^{133}Xe in regions 5, 6, and 7, and ^{135}Xe in regions 3-7. The (background and interference subtracted) gas background provides an estimate of the memory effect for each region and is decay corrected to the acquisition time to estimate the memory effect during the sample acquisition, and then subtracted from the background subtracted sample counts. Finally, the direct interferences in the sample file are subtracted for each ROI.

The possible interferences of ^{135}Xe, the Compton scattering contributions and the 30-keV x-rays, will only be in ROIs that are of lower gamma energy (below the 250-keV region). Therefore, the interferences only need to be calculated and subtracted in ROIs 3-7. The approach to estimating the interference contributions is to determine the ratio of counts in ROI-2 (250-keV region for ^{135}Xe) to counts in ROIs-3-7 for a high activity pure ^{135}Xe sample. These ratios need to be calculated for each detector system that has been appropriately calibrated with respect to energy (gain) and ROIs. These ratios are then placed in the system configuration file and provided for each data file produced by the system. During the data analysis of a sample, the interference terms for ROIs 3-7 are determined by calculating the net counts in ROI-2 (amount of ^{135}Xe in the sample) and the multiplying by the appropriate ratio to estimate the possible interference contribution in each respective ROI. This interference is then subtracted out from the sample counts of each ROI to determine the net counts, which is then used in the Stockholm equations for calculating the concentration.

The implementation of the ^{135}Xe interference terms occurred in two distinct calculation routines; the calibration calculations, and the concentration calculations. The process to collect data to determine the calibration parameters is the same as prior to the incorporation of the ^{135}Xe interference terms. Data is collected on radioxenon isotopes as before the implementation of the ^{135}Xe interference terms, with data collected for the background, each individual xenon isotope, and radon with enough activity and time to have a statistical uncertainty for the region of interest of less than 1%. Typically, enough time is provided between the measurements to minimize any possible memory effect. Calculation of the calibration parameters is expanded to include interference terms for ^{135}Xe in the lower energy regions.

Specifically, the ratio of counts in region 2 (the 250-keV gamma region used as the region of interest for ^{135}Xe) to regions 3-7 are determined for the ^{135}Xe data. These ratios are then added to the calibration parameter set, which is installed as a configuration file for each detector system and included in each measurement data file as part of the header information. Interference terms for radon have been part of the calculations, as well as interference terms for ^{133}Xe in the metastable (^{133m}Xe and ^{131m}Xe) regions of interest. Therefore, the new ^{135}Xe interference terms are added to the interference data under the interference header block. The data format allows for additional data to be included into each block, allowing different implementations of the calculations, without change to the file format.

Each data file produced by the detector systems uses the Comprehensive Nuclear-Test-Ban Treaty Organization format that includes the relevant calibration parameters, allowing calculation of the xenon concentrations without any additional information. The ^{135}Xe interference terms are included in each data file and allow the interferences to be calculated without any additional information as well. Since the format has not changed, the calculation software uses the date of the measurement to determine if the ^{135}Xe interferences should be included. Measurements taken prior to the inclusion of these interference terms do not have the interference ratios required to calculate the possible interference of ^{135}Xe, and therefore, the calculation software must determine whether to include the terms or not.

The calculations were not modified significantly as the software was developed using a modular approach, and interferences were already being calculated for radon and ^{135}Xe. Therefore, only minor modifications were required to the calculations module to allow for the inclusion of the ^{135}Xe interferences, using the same routine as the other interferences. Additional modifications were required to allocate the interferences to the appropriate ROIs.

As described above, the software was modified to allow for calculations of ^{135}Xe interferences into the lower energy regions of interest (ROIs 3-7). Interference calculations were already part of the code, since the interferences from radon into other regions (ROIs 2-7) and ^{133}Xe interference into the meta stable regions (ROIs 5-7) have always been calculated. The ^{135}Xe interferences are negligible for most samples, but can be significant if the ^{135}Xe activity is large, as happens for a calibration measurement, or in possible exercises with high activity samples. It was therefore decided to include these contributions into the calculations.

The interference ratios were measured in calibration samples and added into the data file as part of the calibration parameters that are provided as part of each data file. The calculation module is provided the interference ratio values as part of the function call from the main software package, and uses the same routine for the calculation and subtraction of the interference term.

The verification that the software linked with the other main programs and operated without errors or crashing took place during the compiling process of the dynamic linked library and the other software packages. The software was required to be compiled without errors and function within the software package as intended, and this was verified. An additional verification step was required to ensure that the interference ratios were being passed to the calculations module, and that the calculations module was using those values and calculating the interferences correctly. This was verified by providing different values for the interference ratios and observing the concentration results changed in the appropriate manner.

The validation that the calculations provide the correct value for the interference term was performed by executing several test cases. The first test case was a regression test, using four datasets and comparing the results of the previous version of the calculations module with the results of the new version. The results were as expected, with the comparisons shown below in Figure 2. For the first data set, which was a mix of ^{133}Xe and ^{135}Xe, the results were expected to be very similar between the versions, since the contribution of the ^{135}Xe when ^{133}Xe is present is small. The data are consistent, with the new version showing slightly smaller values of ^{133}Xe as expected. The ^{135}Xe value does not change as expected. For the case of mainly ^{135}Xe (indicted pure in Figure 2) the new version correctly subtracts out the ^{135}Xe interference in the lower energy regions where the other isotopes are measured. For the ^{133}Xe dataset, the values do not change, as expected, since there isn't any ^{135}Xe to cause interference. This is also the case for the ^{131m}Xe dataset.

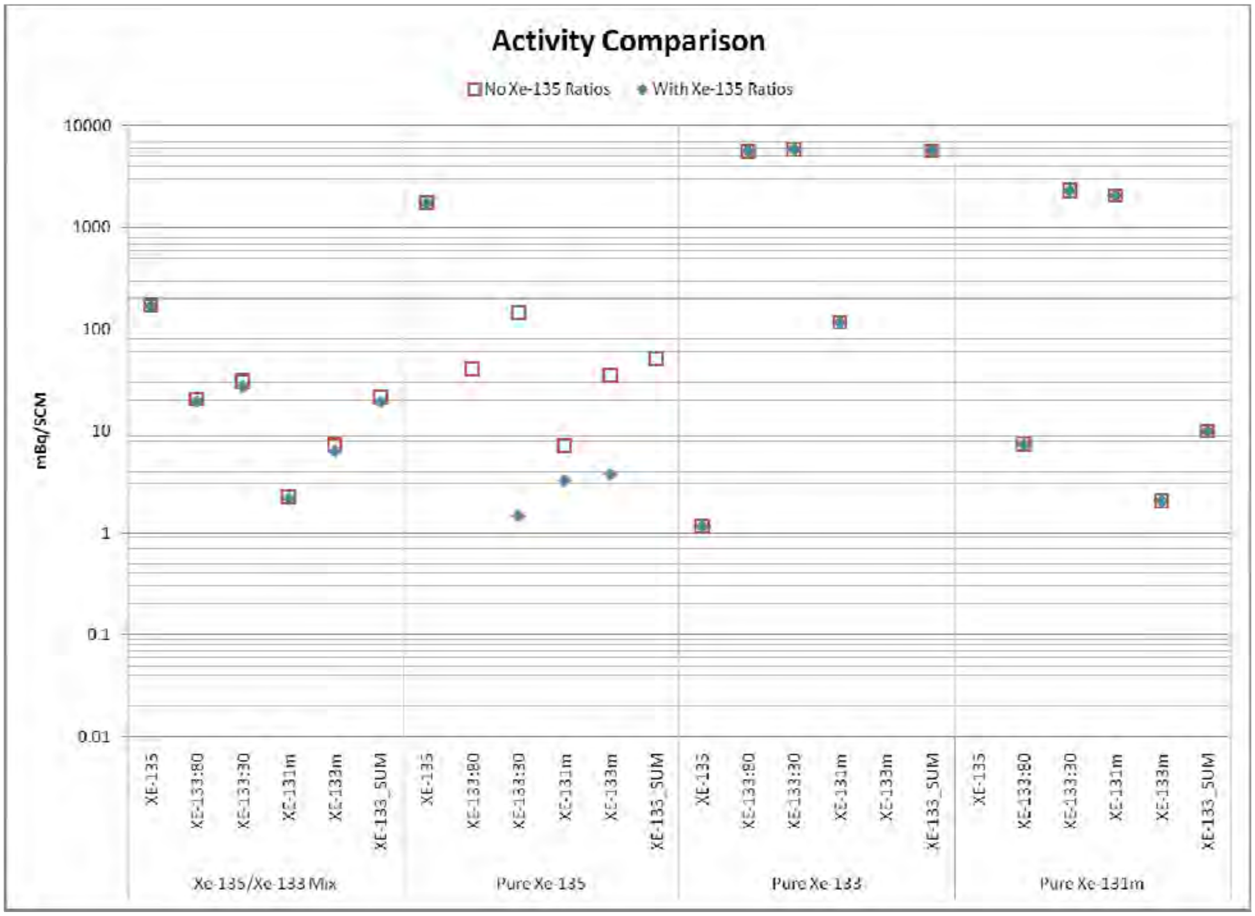

Figure 2. Comparisons of the radioxenon concentration results for the regression testing using four different data sets.

CONCLUSIONS AND RECOMMENDATIONS

Modifications to the calibration and concentration calculations have been implemented to account for the possible interference of ^{135}Xe in other lower energy regions of interest. This allows for a more precise calculation of the ^{133}Xe, ^{133m}Xe, and ^{131m}Xe isotopes, and reduces misidentification when there are high activities of ^{135}Xe present. Although the ^{135}Xe interferences for most samples are negligible, exercises or calibrations with high ^{135}Xe activities can have significant contributions to the lower energy regions due to Compton scattering and the minor decay branch that produces x-rays in the 30 keV range.

The measurements for the calibration data remain the same, only additional calculations are required to determine the ratio of counts in the main ^{135}Xe region compared to the other regions. These interference ratios are then used in the concentration calculation to determine and subtract any ^{135}Xe contributions in the lower energy regions used for ^{133}Xe, ^{133m}Xe, and ^{131m}Xe.

As the radioxenon measurement systems become more precise it will become important to account for the small effects that are currently ignored due to their insignificant contribution in relationship to the current measurement uncertainty. The interference of the ^{135}Xe was one of these types of small effect that has now been properly accounted for in the concentration calculations for PNNL developed systems.

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