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Is there a potential for further enhancing IDC spectrum analysis methods of CTBT radionuclide measurements after 25 years of progressive development?

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P3.5-507

POSTER

Outlines

OUTLINES

- This presentation gives an overview on radionuclide analysis procedures at the IDC.
- A significant portion of new methods is currently under implementation for the new generation of noble gas systems.
- Based on the achievements of the past 25 years, opportunities for potential enhancements of IDC radionuclide analysis methods are described in view of further improving the quality of IDC RN products.

Outlines

- Introduction
- Analysis methods for particulate and noble gas
- Isotopic ratio analysis
- Reporting analysis results
- Summary

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Which analyses are performed at the International Data Centre (IDC)?

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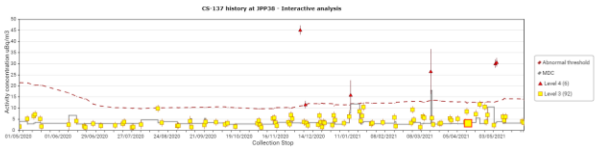
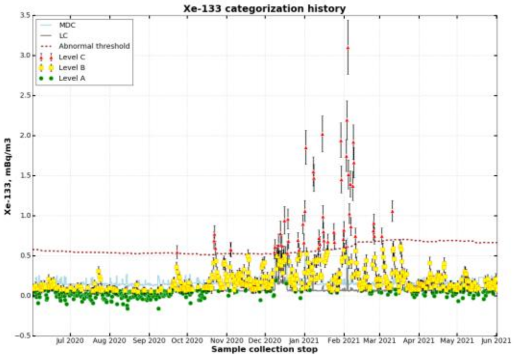
INTRODUCTION

- Special requirements on the IDC analysis
  - It is a single measurement for each sample.
  - Most samples are at low level close to the background.
  - Peak counts might be low to a few counts, especially for metastable xenon.
- Identification of CTBT relevant radionuclides
  - Particulate: 84 fission products and activation isotopes.
  - Noble gas: Xe-131m, Xe-133m, Xe-133 and Xe-135.
- Quantities measured in IMS samples
  - Activities collected in samples;
  - Estimates of activity concentration under an assumption of constant concentration during sampling.
- Analysis results are provided in IDC products
  - Routine analysis, A/RRR
  - Expert technical analysis and Special studies



Cs-137 detections in one year at JPX38



Xe-133 measurements in one year at JPX38

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What are purposes of IMS measurements and IDC analyses?

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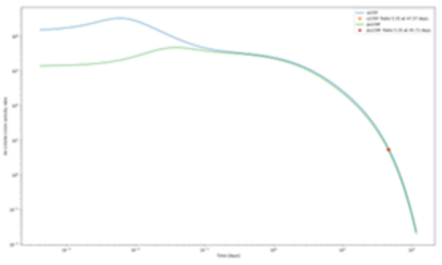
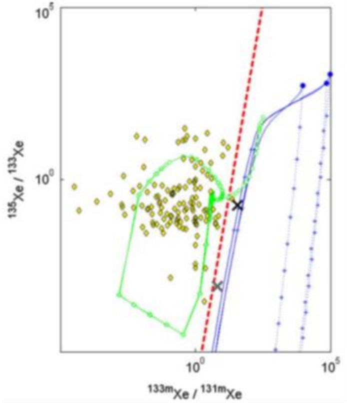
INTRODUCTION

- Event screening of a nuclear explosion from releases of civil facilities are performed based on isotopic ratio analysis.
  - Four xenon relationship of Xe135/133 vs Xe133m/131m
  - Decay curves of paired isotopes, e.g., Cs-134/Cs-137, …
- The explosion time of the nuclear event is estimated by using a function of the isotopic ratio with time based on an assumed scenario.
  - Pairs of parent-daughter decay chains
    - Zr-95/Nb-95, Ba-140/La-140, Xe-133m/Xe-133
  - Pairs of fission products
    - Xe-131m/Xe-133, Ru-103/Ru-106, …



Ratio of Xe133/Xe131m determining explosion time



Plot of four xenon relationship discriminating nuclear explosions (right domain) from releases of civil facilities

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The SCAC method for gamma spectra of particulate 1/3

METHODS

- The method of single channel analyser curve (SCAC), critical level curve (LCC) and baseline (B) was developed for gamma spectrum analysis of particulate samples.
  - SCAC and B are defined based on energy resolutions and "lawn mower" algorithm.
  - Peaks are identified by the detectability (D) when D_i >= 1 at channel i and risk level k.
  - Peak areas (A_i and u_{A_i}) are estimated by $(SCAC_i - B_i)$ and resolution R_i.
  - Energy resolutions are critical for the peak area estimation.

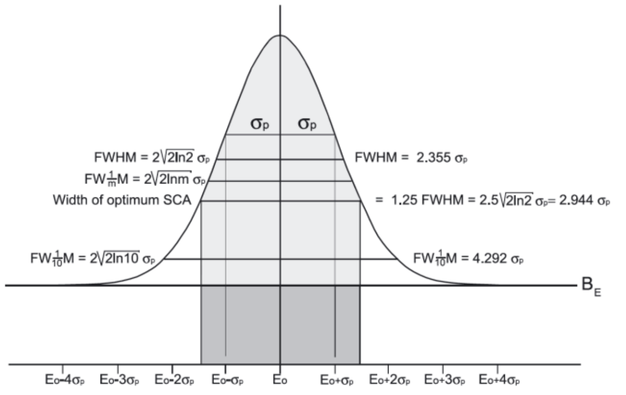
$$LCC_i = B_i + k * \sqrt{\frac{B_i}{1.25 * R_i}};$$

$$D_i = Max\left\{\frac{SCAC_i - B_i}{LCC_i - B_i}\right\}\ ,\ (i-2, i+2);$$

$$A_i = (SCAC_i - B_i) * 1.25 * R_i / 0.85891;$$

$$u_{A_i} = \sqrt{(SCAC_i + B_i)} * 1.25 * R_i / 0.85891.$$

(Right) A Gaussian peak with characteristics indicated. The grey areas show the optimized single channel analyser.



The light grey area within the SCA bounds is erf [1.25√ln2] A = 0.85891 A ~ 85.9 % of A

The SCAC method for gamma spectra of particulate 2/3

METHODS

- Features.
  - It is a unique algorithm fit well to automatic processing of IMS gamma spectra.
  - Based on Monte-Carlo simulations by the code MCNP or GEANT4, efficiencies and interference response functions (IRF) are calculated for the whole energy range and given isotopes, including coincidence summing corrections.
  - Discrimination of peaks around 140 keV: Tc-99m (140.51 keV, CTBT relevant) or Ge-75m (139.68 keV, Germanium activation)
  - Distinguish Tc-99m detection combining Mo-99.

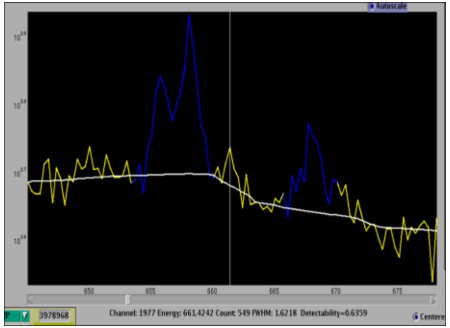
Bayesian statistics behind the SCAC

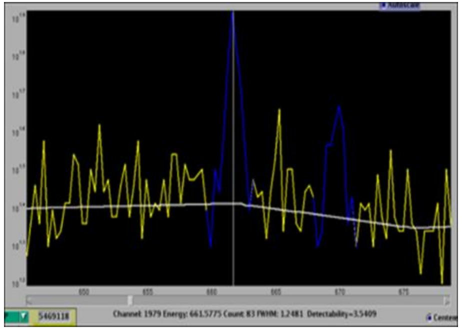
- Numbers of counts ($SCAC_i$, B_i) are somehow the averages of original counts measured.
- Poisson distribution is used for the number of counts in each single channel.
- Variances are equal to the values of $SCAC_i$, B_i although they are not counts measured, which are different from the values derived based on the uncertainty propagation.

The SCAC method for gamma spectra of particulate 3/3

METHODS

- Lesson learned and enhancements needed.
  - An advanced interactive algorithm and software are needed to deal with small peaks and complicated baselines as well as an interactive calibration procedure for analyst review at the IDC.
  - How to apply SCAC to low level baselines and beta/gamma coincidence spectra.





Independent calibration at the IDC

- The energy and efficiency calibration at IMS stations is carried out based on analysis software from equipment providers.
- The calibration analysis performed at the IDC is used for calibration validation only.
- The spectrum analysis at the IDC is performed by using the SCAC method but the calibration data comes from different software at IMS stations.
- There are systematic deviations in between although calibrations are validated.

The peak 661.6 keV (at cursor) was not detected in the station spectrum (left figure, AT=24h) but detected in the laboratory reanalysis spectrum (right figure, AT=72h).

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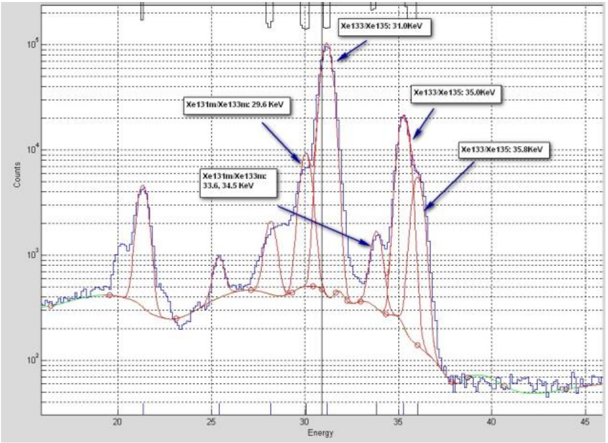
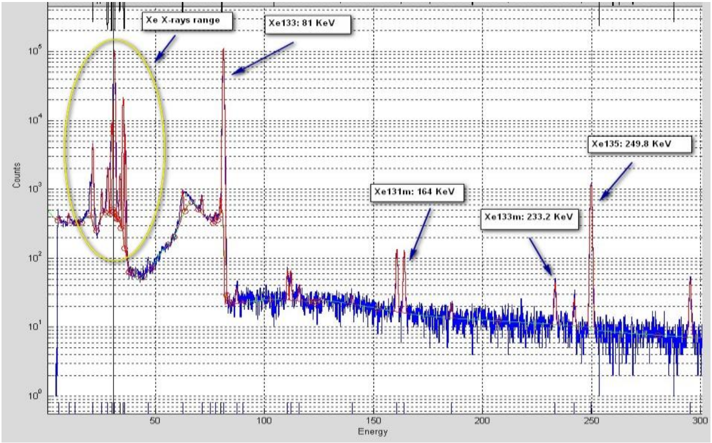
Least squares regression analysis for gamma spectra of noble gas 1/3

METHODS

- The least squares fitting on gamma- and X-rays peaks of xenon isotopes is used for gamma spectra from noble gas systems with high resolution detectors.
  - Peak analysis by SCAC is performed but not used to determine xenon activities.
  - Channel counts vector **B** is composed of four gamma regions and one X-rays region.
  - Response matrix **A** is determined by the efficiency calibration and nuclear data.
  - The peak counts **X** for each xenon are estimated by the least squares fitting $\boldsymbol{A} \cdot \boldsymbol{X} = \boldsymbol{B}$.

gamma- and X-rays peaks of xenon isotopes





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Least squares regression analysis for gamma spectra of noble gas 2/3

METHODS

- Calibration
  - Standard source made of a gas equivalent material is used.
- Challenges
  - The fitted peak counts are referred to gamma peaks of each xenon.
  - Decision thresholds are estimated based on baselines in SCAC but not available for metastable xenon in the regression analysis.
  - The regression analysis was designed in which peak areas of metastable xenon were supposed below their decision thresholds, but X-rays peaks above the related ones due to higher detection efficiencies and branch intensities of X-rays.
  - False positive of Xe-133m were observed occasionally due to a bump around 233 keV, resulting in negative values of Xe-131m, although both were not detected in SCAC.

Least squares regression analysis for gamma spectra of noble gas 3/3

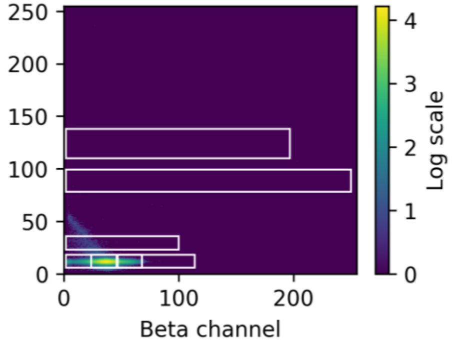
METHODS

- Solutions in the project of Alternative Radionuclide Analysis System (ARAS)
  - **Method 1**: Gamma only analysis. Uses only gamma peaks for activity calculations.
  - **Method 2**: Upper limits for metastable. Uses the 29.7 keV X-ray peak for both Xe-131m and Xe-133m and calculates what would be the highest activity for both of these if all of this X-ray came from that isotope.
  - **Method 3**: Xe-131m from X-rays if Xe-133m gamma is available. The share of Xe-133m in the 29.7 keV gamma peak is calculated based on Xe-133m gamma peak and the rest of this X-ray peak is assumed to be Xe-131m. This method is calculated only if the 233 keV peak of Xe-133m and the 29.7 keV X-ray peak are available.
  - **Method 4**: Xe-133m from X-rays if Xe-131m gamma is available. The share of Xe-131m in 29.7 keV gamma peak is calculated based on Xe-131m gamma peak and the rest of this X-ray peak is assumed to be Xe-133m. This method is calculated only if the 165 keV peak of Xe-131m and the 29.7 keV X-ray peak are available.
  - The ARAS is available at IDC since 2017 for analysts performing alternative analysis for comparison with the regression analysis.

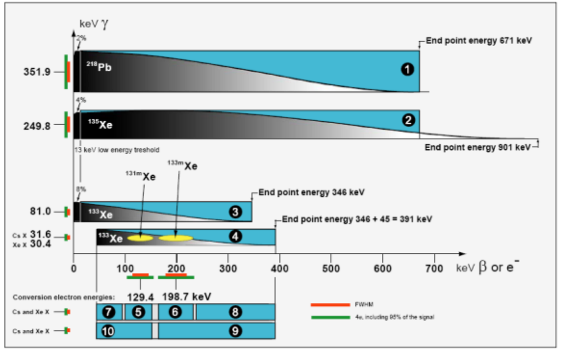
The NCC method for beta/gamma coincidence spectra of noble gas 1/2

METHODS

- The net count calculation (NCC) method is used in analysis of 2D beta/gamma coincidence spectra, quantifying the presence of ^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe in noble gas samples.
  - Net numbers of counts and their variances are estimated by NCC equations or the matrix operation.
  - Uncertainties of interference ratios are not included.
- There are challenges, such as,
  - X-ray's deconvolution,
  - Uncertainty component of interference ratios,
  - Detector background estimation and
  - Estimation of decision threshold for low count level in which the distribution will start to deviate from Gaussian and approach more Poisson.



Xe131m coincidence spectrum



10 ROI definitions

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The NCC method for beta/gamma coincidence spectra of noble gas 2/2

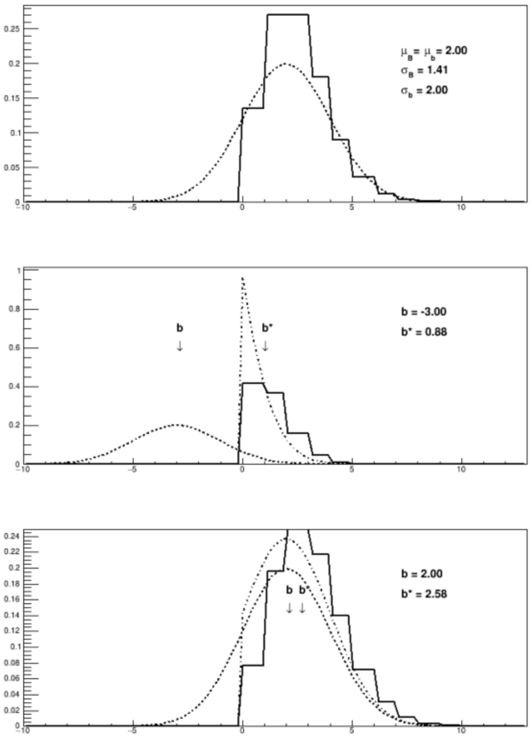
METHODS

- Stockholm equations of the classic NCC method (Used at the IDC until 2018)
  - There are binary decisions on interference corrections from higher to lower ROIs.
  - Memory corrections are performed always.
  - Subtracting negative values of memory corrections might cause decision thresholds under-estimated, resulting in false positive detections.
- Modification of the classic NCC method (Deployed at the IDC in 2018)
  - All binary decisions were discarded.
  - As a result, detection rates of metastable xenon were reduced.
  - Removing binary decisions could cause a mess for low count level, resulting in under- or over-estimation on decision thresholds due to negative net numbers of counts in higher ROIs and the gas background spectrum.
- Along with new generation systems, different approaches of the NCC method were developed. Net numbers of counts are the same in between, but the associated uncertainties might be different with respect to analysis algorithms.

Features of analysis algorithms in new generation noble gas systems: SAUNA III 1/2

METHODS

- BGM algorithm (provided by FOI, IDC implementation 2021)
  - The classic NCC method and associated calibration are applied basically but the matrix operation is used.
  - A dynamic ROI-4 instead of ROI-7 to ROI-10 regarding the X-ray contribution of Xe-133 is introduce based on patterns of metastable xenon detections.
  - Uncertainties related to interference corrections are processed by a special matrix operation or the Monte-Carlo method.
  - Covariances between ROI-4 and other ROIs are derived analytically.
  - The concept of the best estimate is used to handle negative variance in the estimation of decision threshold.



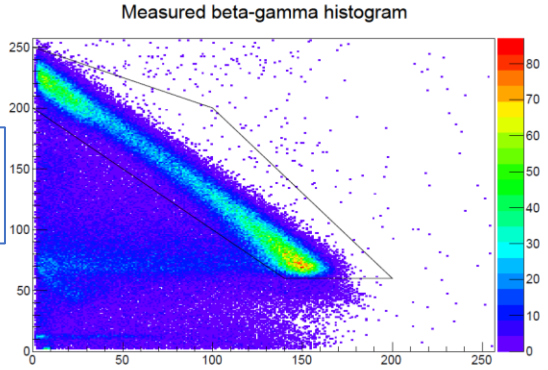
Processing negative variance

Features of analysis algorithms in new generation noble gas systems: SAUNA III 2/2

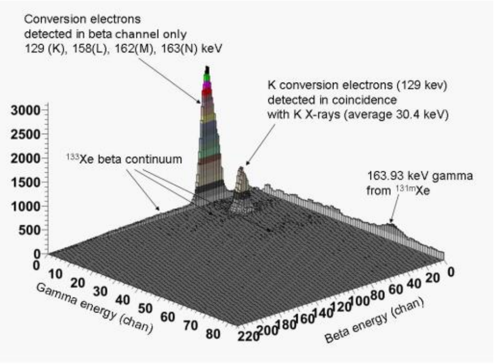
METHODS

- Calibration of SAUNA II and III
  - Two spikes of Xe-131m and Xe-133 and point sources of Cs-137 and Eu-152 are used for gamma energy and resolution calibration.
  - A diagonal band of Compton scatter of the Cs-137 point source is sliced and the projected beta peaks are used for the beta energy and resolution calibration.
  - Efficiencies related to Xe-131m and Xe-133 are determined by their spikes, which are applied to Xe-133m. For Xe-135, it is based on Monte-Carlo simulations.
  - Interference ratios are determined based on a radon spike measurement.

Cs-137 2D diagonal for beta energy and resolution calibration



Simulated Xe-131m 2D coincidence spectrum



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Features of analysis algorithms in new generation noble gas systems: Xenon Int.

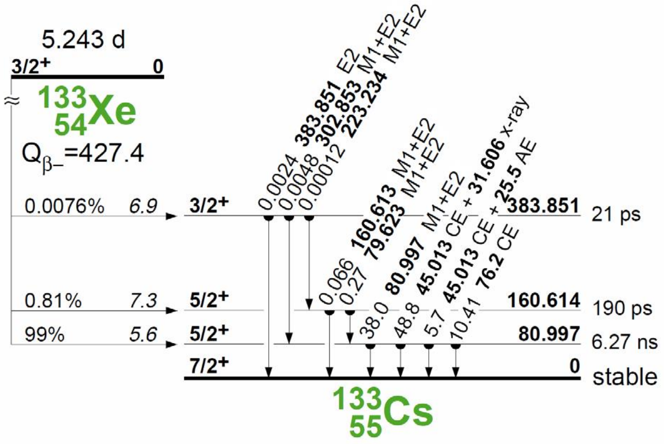
METHODS

Analysis algorithm with 7 ROIs

- A ROI-4(-) of the ROI-4 minus the ROI-5 and -6 is used to process X-rays contributions.
- The uncertainty estimation is processed by using an iterating procedure of interference corrections from higher to lower energy ROIs, but correlations are ignored.
- Interference ratios of ROI-3 to ROI-4 and ROI-6 to ROI-5 need to be optimized further.

Calibration procedure of Xenon Int.

- Energy and efficiency calibrations are all based on four xenon spikes as well as radon.
- Beta singles are processed for each component, e.g., Auger and conversion electrons and continued beta spectrum (see right figure Xe-133).
- Beta and gamma efficiencies in ROIs are determined for each isotope independently.
- Efficiencies of 30 keV X-rays are different slightly amongst four xenon isotopes.

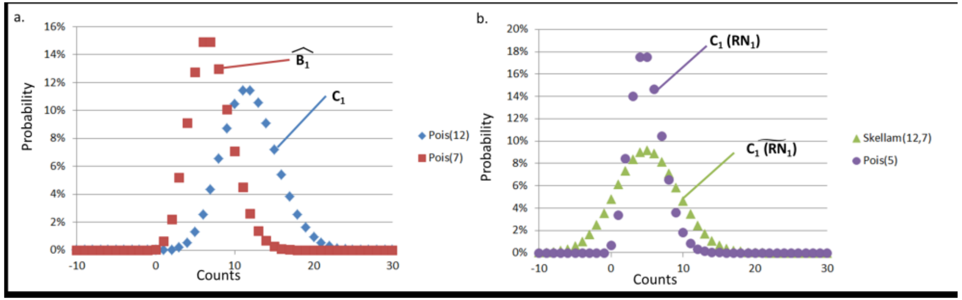


Features of analysis algorithms in new generation noble gas systems: SPALAX NG 1/2

METHODS

- Analysis algorithm
  - X-rays ROIs of Xe-131m and Xe-133m are separated completely due to high resolution beta detectors of Silicon-PIN.
  - There is no gas background measurement due to memory free beta detectors.
  - Correlations of net numbers of counts between ROIs caused by interference corrections are included in the uncertainty estimation.
  - The detector background in sample measurements is estimated based on Bayesian approach, e.g., the variance equal to the mean number of counts.
  - The minimum value of the decision threshold, 3.3. counts, is used for the detector background below 2 counts.

Detector background (B_1) estimated by Poisson distribution

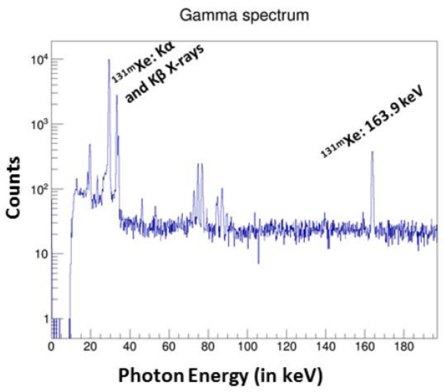


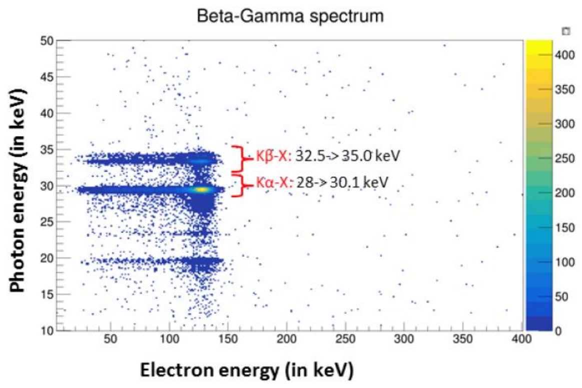
Features of analysis algorithms in new generation noble gas systems: SPALAX NG 2/2

METHODS

- Calibration procedure of SPALAX NG
  - It is not a 4 pie beta detector geometry.
  - A gas equivalent source is used for gamma energy and efficiency calibrations.
  - Beta energy and resolution calibrations are obtained by spikes of Xe1-31m and Xe-133, probably Xe-135 and Xe-133m as well.
  - The activities in spike measurements are estimated by the gamma standard source. Then beta efficiencies are determined accordingly.

The ROI-5 is defined for Ka X-rays only.





Enhancements on the NCC method: Calibration procedure for 4π detector geometry

METHODS

The beta-gamma coincidence measurement in 4π detector geometry is an absolute measurement method and the coincidence efficiency is equal to the product of efficiencies of the beta and gamma singles.

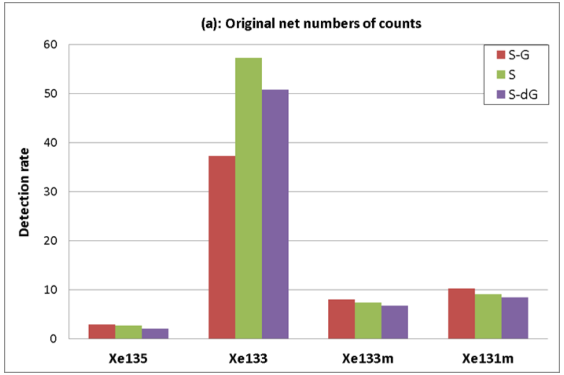
- Numbers of radioxenon decays are the same amongst three measurements of beta-gamma coincidences, beta and gamma singles.
- Gas spikes of Xe-131m, Xe-133, Xe-133m and Xe-135 as well as radon are used.
- Gamma singles are used for gamma energy and resolution calibrations simply.
- Beta energy and resolution calibrations are performed based on single and double peak fitting in beta projections of beta-gamma coincidences and the beta singles.
- Efficiencies of 30 keV X-rays and 129 keV conversion electrons are determined by Xe-131m first. Auger electrons are ignored by adjusting the beta detection threshold. The efficiency of 30 keV X-rays is assumed the same for all xenon isotopes.
- Gamma efficiencies of 81 and 250 keV are derived based on the efficiency of 30 keV.
- Beta efficiencies are determined based on gamma singles and gamma projections.

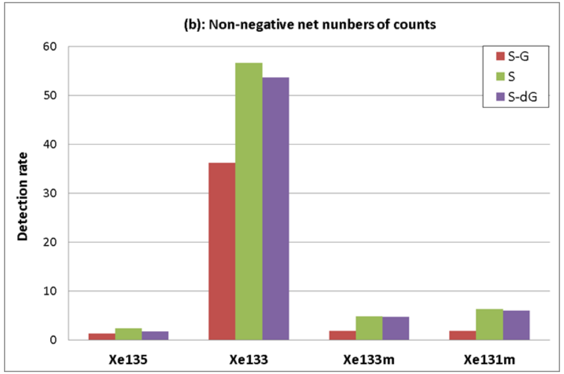
Enhancements on the NCC method: False positive detections

METHODS

False positive detections might be caused by interference corrections and/or memory subtractions.

- (Upper) Detection rates of metastable xenon for always subtraction are higher than for samples themselves.
- (Lower) Detection rates were reduced a lot when non-negative net numbers of counts are applied.





967 paired sample and gas background spectra from the detector JPX38_004 during 2014 to 2016 were processed by three modes of gas background subtraction: S: no subtraction; S-G: gas background always subtracted; S-dG: gas background subtracted only if xenon is detected.

Enhancements on the NCC method: Negative variance in decision threshold estimation

METHODS

A negative variance could happen in case there is an under-estimated uncertainty of the detector background in addition of a negative net number of counts.

- By the NCC method without any binary decisions, processing 414 samples from a SAUNA III detector in 2018 resulted in negative variances in 7 samples.
- There was no negative variance in case the detector background in a single sample measurement was estimated by using Poisson distribution directly, variance = mean.

Notice:

- Non-negative approach is consistent with recommendations in ISO 11929:2019.
- Estimation of the detector background in a single sample measurement by Poisson distribution is different from the propagation of uncertainty. This needs to be investigated further.

Enhancements on the NCC method: Potential solutions 1/3

METHODS

Potential solutions in the IDC analysis.

- **Solution 1**: Applying both binary decisions to interference corrections and memory subtractions. This is the simplest way dealing with negative net numbers of counts in decision threshold estimation. It works for SAUNA III too.
- **Solution 2**: Matrix operation with non-negative net numbers of counts
  - Under-estimated decision thresholds can be fixed by using non-negative net numbers of counts, being consistent with ISO 11929-3:2019.
  - Correlations of net numbers of counts between ROIs are processed by the matrix operation, which can be used in isotopic ratio analysis.
  - Addressing uncertainty components related interference corrections by using the numeric derivative approach recommended in ISO 11929-3:2019.
  - Addressing X-rays contributions based on patterns of metastable xenon detections by using the dynamic definition ROI-4.

Enhancements on the NCC method: Potential solutions 2/3

METHODS

Potential solutions in the IDC analysis.

- **Solution 3**: The Monte-Carlo approach of the NCC method
  - The MC method can be used for any complicated spectra as comparison to other methods, including the solution 1 and 2 above and methods from developers.
- **Solution 4**: Optimization regression analysis of ROI counts.
  - Net numbers of counts and associated variances and co-variances are estimated by the least squares fitting or maximum likelihood fitting.
  - The response matrix by the size of 5x6 is composed of interference ratios between 5 isotopes (four xenon and radon) and 6 ROIs.
  - X-rays counts in ROI-4 contain contributions from all isotopes, including all counts in ROI-5 and -6. Related interference ratios, such as ROI-3 to ROI-4, ROI-4 to ROI-5 and -6 are different from the classic NCC method.

Enhancements on the NCC method: Potential solutions 3/3

METHODS

Potential solutions in the IDC analysis.

- **Solution 5**: Standard spectra method (SSM)
  - It is based on full spectrum regression analysis.
  - This method was included in the analysis software '*bg-analyze'* at the IDC but the associated standard spectra are pending, which can be obtained by using Mote-Carlo simulations or the calibration spectra of four xenon spikes.
- **Solution 6**: 1D or 2D peak fitting of beta-gamma coincidence spectrum
  - There is an advantage reducing the false positive detection, especially for complicated spectra with the count level above the background or gain drafting.
  - It is difficult to define proper baselines for low level samples.

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Enhancements on the NCC method: Decision threshold and detection limit

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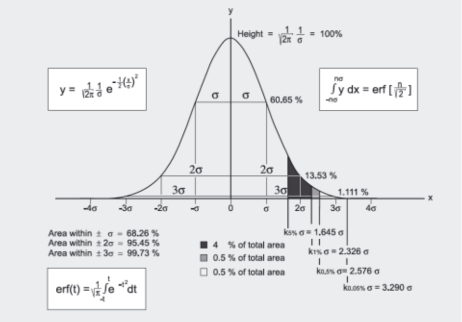
METHODS

- The decision threshold (e.g., LC) is referred to as an a posteriori limit, applied to interpret the measurement after it has actually been carried out.
- The detection limit (LD) (e.g., MDC) is referred as an a priori limit, that characterizes the measurement procedure even before any sample measurement.
- Currie introduced a third limit, the quantification limit (LQ), above which the expected uncertainty is considered low enough for quantitative determination.
- Comparing measurement results with the detection limit is not in accordance with ISO 11929, resulting in that it is decided to conclude too frequently that the physical effect is absent when in fact it is not absent. (ISO 11929)

- LC is defined at a false positive of 5%.
- The false positive at the LD level, LD=2LC, is 0.05%.
- A signal has an one sigma uncertainty of 60% at the LC level, 30% at the LD level, 10% at the 6LC level.
- The best estimate of a signal needs to be reported when the signal has an uncertainty above 25%. (ISO 11929)

Gaussian peak with probabilities indicated.



(De Geer, CTBT/PTS/TP/2005-1.)

Enhancements on the NCC method: Memory correction for non-detections in ATM simulations

METHODS

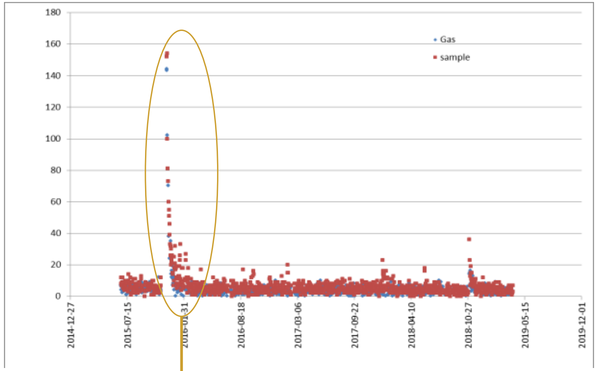
- Detection means that the activity concentration is above the decision threshold instead of the detection limit (e.g., MDC).
- Non-detections can be used to improve source term estimation in ATM simulations, especially uncertainty estimation by using Bayesian approach.
- For SAUNA samples, the gas background spectrum is used to perform the memory subtraction. In the case of both the sample and gas background spectra are at background level, subtracting the memory effect could result in non-physical results of activity concentration. That might make the ATM simulations meaningless.
- A ‘smart’ approach can be used on whether and how to subtract the memory effect with respect to the trend of the gas background in the given period (see next slide).

Enhancements on the NCC method: 'Smart' memory subtraction based on the trend in gas background spectra

METHODS

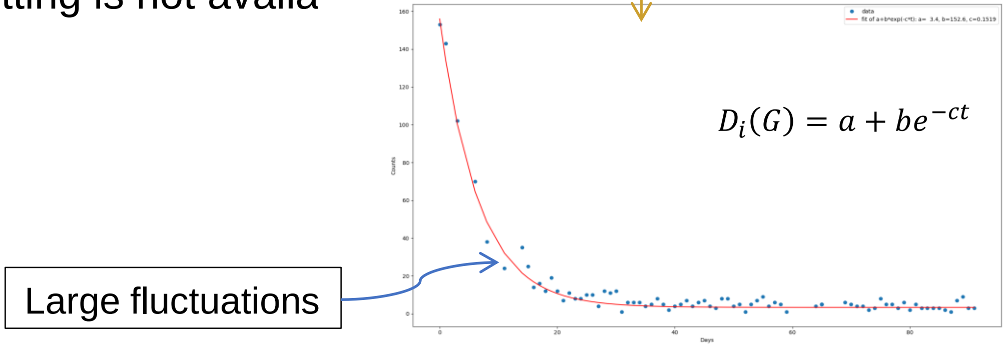
Memory effect in GBX66_004

- Background counts in the ROI-5 (Right upper)
  - Mean value of gross counts of 3.9
  - Decision threshold of gross counts of 8
  - Memory factor of 0.02% in the spike measurement
- Decay curve fitted in a period of 90 days (Right lower)
  - Half-life of 4.3 days fitted < 11.84 days of Xe-131m.
  - The fitted curve can be used for subtraction.
  - No subtraction is needed if the fitting is not availa



$$D_i(G) = a + be^{-ct}$$

	Value	Uncertainty
a	3.3737	0.3011
b	152.6213	9.5229
c	0.1519	0.0082



Bayesian statistics for IMS measurements: Basic concepts

METHODS

It is a single measurement for each sample at IMS stations.

- There is no repeated measurements for a single sample.
- Estimation is based on Bayesian statistics and the a priori distribution derived in conventional frequentist statistics.
- Associated uncertainties are systematic uncertainties.
- The number of peak counts in a single measurement is estimated by using the likelihood function, which has the same formula with the PDF of the a priori distribution.
- The detector background is estimated based on the a priori distribution and related measurements.

Conventional statistics

- The mean and standard deviation are estimated statistically based on observations in repeated measurements.
- The probability density function (PDF) of the distribution can be derived accordingly.

Detector background estimation (see an example in next slide).

- The a priori distribution can be derived based on repeated background measurements.
- The mean number of counts can also be estimated by a longer detector background measurement.
- The variance is estimated based on the a priori distribution derived, i.e., equal to the mean.

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Bayesian statistics for IMS measurements: Example of detector background estimation

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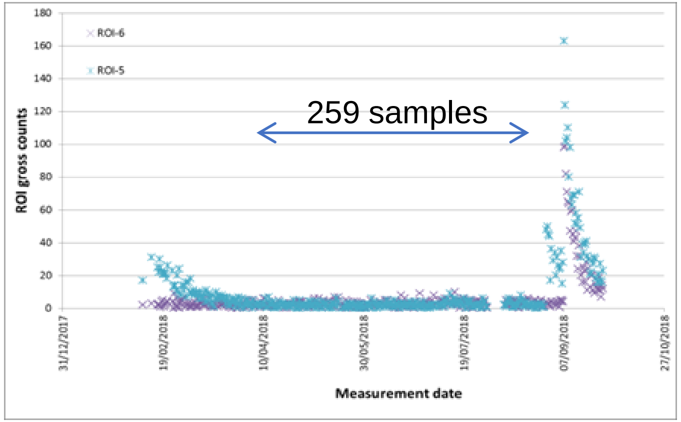
METHODS

Statistical results of a SAUNA III detector

- There are 259 gas background spectra from 8 April to 28 August 2018, 7 half-life after early spike measurements and before next spikes. There is no memory effect observed.
- The mean numbers of counts in the gas background spectra are used to estimate the variances, e.g., equal to the mean numbers by Poisson distribution (P).
- The numbers of counts derived by the detector background measurement are consistent with statistical results, but the associated uncertainties (red) are under-estimated.

Method	Quantity	ROI-1	ROI-2	ROI-3	ROI-5	ROI-6
Gas background spectra	Mean value	73.6	82.8	26.5	2.2	2.8
	Uncertainty	8.2	9.1	5.4	1.6	1.8
	Uncertainty (P)	8.6	9.1	5.2	1.5	1.7
Detector background measurement	Gross counts	1657	1806	596	27	27
	Normalized value	70.9	77.3	25.5	1.2	1.2
	Uncertainty	1.7	1.8	1.0	0.2	0.2
	Uncertainty (P)	8.4	8.8	5.1	1.1	1.1

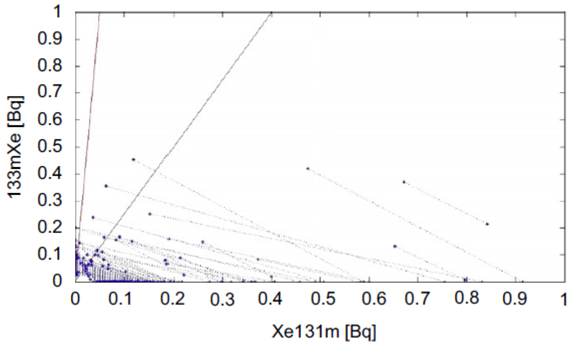


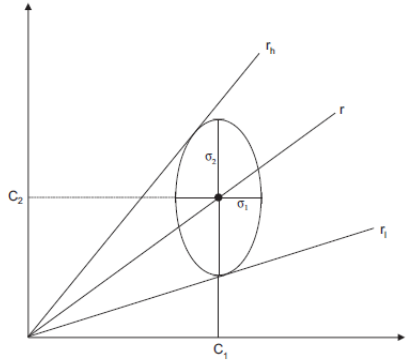
Isotopic ratio analysis: Event screening

METHODS

There are different ways estimating isotopic ratios for event screening.

- Bayesian limits : (Right upper)
  - Current method at the IDC analysis
  - Symmetrical coverage intervals of concentrations
  - Minus and plus values of the ratio
- Fieller's Theorem (Right lower)
  - Suggested and used at FOI
  - The nominal value of the ratio
  - Interprets the coverage region geometrically
- Limits of the coverage interval (LCI)
  - Recommended in ISO 11929
  - The distribution of the ratios
  - Symmetric or shortest definitions
  - Can be estimated by the Monte-Carlo method.





Isotopic ratio analysis: Activity concentrations in the plume 1/3

METHODS

Isotopic ratios ($R(t_2)$) are estimated using activity concentrations in the plume due to the linear SRS.

- Source-receptor sensitivity (SRS) fields $M(t_t)$ are simulated using adjoint atmospheric transport Modeling (ATM) and transport time (t_t) from a release to an IMS station.
- Released activities are estimated based on activities measured in samples at IMS stations.
- Activity concentrations in the plume are linear to released activities in ATM simulations.

$$A(t_2) = \frac{C(t_2)}{M(t_t)}$$

↓

$$R(t_2) = \frac{A_2(t_2)}{A_1(t_2)}$$

Linear SRS ↓ $M(t_t)$

$$R(t_2) = \frac{C_2(t_2)}{C_1(t_2)}$$

Simplest case: independent decay →

$$R(t_2) = R(t_1)e^{-(\lambda_2-\lambda_1)t_t}$$

Isotopic ratio analysis: Activity concentrations in the plume 2/3

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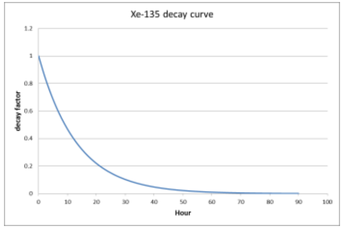
METHODS

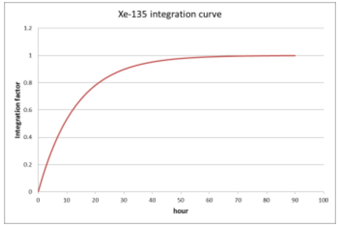
Ratios of concentrations in the plume ($R(t_2)$) is only dependent on decay constants.

- Decay chains are interrupted due to activity accumulation during sampling.
- Ratios of activities collected in the sample ($R_s(t_2)$) is dependent on not only decay constants (λ_1, λ_2) but also collection time (t_c).

During Sampling

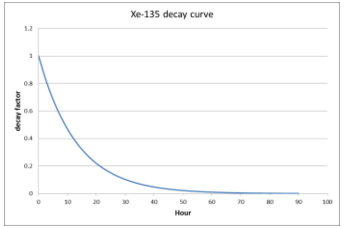


+



$$R_s(t_2) = R(t_2)\frac{\lambda_1}{\lambda_2}\frac{1-e^{-\lambda_2 t_c}}{1-e^{-\lambda_1 t_c}}$$

Before and after sampling



$$\begin{cases} R(t) = R(0)e^{-(\lambda_2-\lambda_1)t}; & t < t_2 \\ R_s(t) = R_s(t_2)e^{-(\lambda_2-\lambda_1)(t-t_2)}; & t > t_2 \end{cases}$$

Isotopic ratio analysis: Activity concentrations in the plume 3/3

METHODS

The explosion time is estimated by a function of the isotopic ratio with time from the explosion time up to the stop of collection, which can be derived based on Bateman equations of decay chains, assumed scenarios and IMS measurements.

- The scenarios, e.g., full in-growth, are assumed not interrupted.
- Bateman equations can be solved numerically or analytically.

Fission yield: $R(0) = \frac{A_2(0)}{A_1(0)}$

Sample measurement: $R(t_2) = \frac{C_2(t_2)}{C_1(t_2)}$

$$N_{m,n}(t) = \sum_{k=1}^{n} [N_{m,k}^{0} \cdot \prod_{l=k}^{n-1} b_{m,l} \cdot \sum_{j=1}^{n} (\frac{T_{m,n}}{T_{m,j}} \cdot \prod_{\substack{i=1 \\ l><j}}^{n-1} \frac{1}{1 - T_{m,i}/T_{m,j}} e^{-ln2 \cdot t/T_{m,j}})]$$

Numerically → t_2

The simplest case: Independent decay

$$R(t_2) = R(0)e^{-(\lambda_2-\lambda_1)t_2}$$

$$t_2 = \frac{1}{\lambda_1 - \lambda_2} \ln(\frac{R(t_2)}{R(0)})$$

Analytically → t_2

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Isotopic ratio analysis: Estimation of isotopic ratios by IMS measurements 1/3

METHODS

Isotopic ratios and associated uncertainties are dependent on not only concentrations but also their uncertainties and covariances.

- For non-linear model, ratios of concentrations and associated uncertainties are estimated by high-order Taylor terms and dependent on uncertainties of denominators.
- For low level samples, it is better to use the Monte-Carlo method, estimating isotopic ratios and their uncertainties based on activities measured in the sample or associated peak counts directly.

Non-linear model of the ratio of random variables:

- Nominal value (linear model): $r_0 = \frac{c_2}{c_1}, u(r_0) = r_0(\frac{u^2(c_1)}{c_1^2} + \frac{u^2(c_2)}{c_2^2} - 2\frac{COV(c_1,c_2)}{c_1 c_2})$
- Nominal plus bias: $\mathrm{r} = r_0(1 + \frac{u^2(c_1)}{c_1^2} - \frac{COV(c_1,c_2)}{c_1 c_2})$
- Variance: $u^2(r) = r_0^2\ (\frac{u^2(c_2)}{c_2^2}\left(1 + 3\frac{u^2(c_1)}{c_1^2}\right) + \frac{u^2(c_1)}{c_1^2}\left(1 + 8\frac{u^2(c_1)}{c_1^2}\right) - 2\frac{COV(c_1,c_2)}{c_1 c_2})$

Isotopic ratio analysis: Estimation of isotopic ratios by IMS measurements 2/3

METHODS

Correlations in SCAC of gamma spectrum analysis

- Basically peak areas estimated by SCAC are independent.
- The overlapped peaks can be identified but peak areas might need to be processed further, such as by multiple peak fitting, resulting in correlations.

Parent-daughter decay chains

- Ba-140/La-140, Zr-95/Nb-95, peaks are independent.
- Correlations are due to decay chains.

Independent decay chains

- Cs-134/Cs-137,
- There is no correlation caused by either decay chains or peak areas.

Correlation due to background subtraction

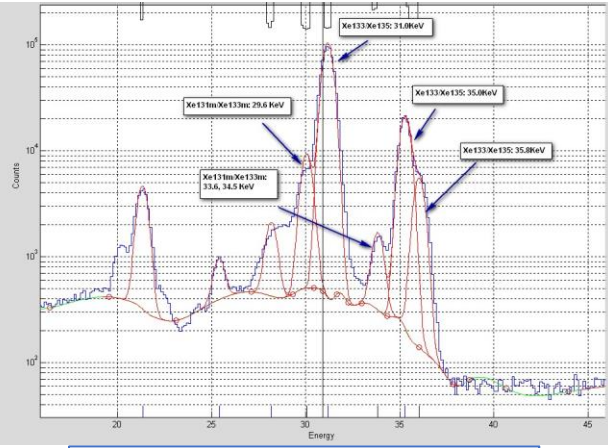
- Baselines might be correlated for close peaks due to "lawn mower“ algorithm.
- The background is subtracted by using single channel, following Bayesian approach. There is no correlation due to the background subtraction.

Isotopic ratio analysis: Estimation of isotopic ratios by IMS measurements 3/3

METHODS

Correlations in the least squares fitting

- There are no correlations between four xenon isotopes by gamma peaks in SCAC.
- For X-rays regions, metastable can be isolated from ground states due to a good resolution of HPGe detectors. X-rays peaks are overlapped in two states.
- Activities of four xenon isotopes are correlated due to the regression analysis.
- For the pair of Xe-133m/Xe133, there is another correlation due to the decay chain.



Overlapped peaks of X-rays of noble gas gamma spectrum

Correlations in the NCC method are due to three ways,

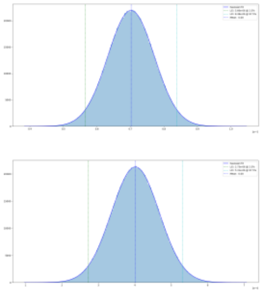
- Interference corrections from higher to lower energy ROIs,
- Subtraction of the same detector background in memory corrections, and
- For Xe-133, the decay correction of Xe-133m to Xe-133 during acquisition.

Isotopic ratio analysis: The Monte-Carlo method

METHODS

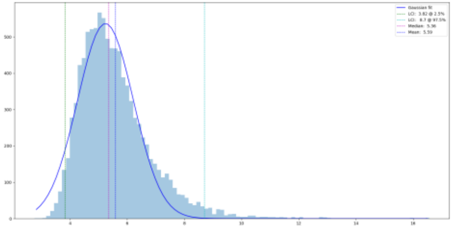
- Probability distributions of measurands, e.g. activity, concentration and isotopic ratio, can be obtained by the Monte-Carlo method, directly based on distributions of peak counts measured, calibration data and related parameters, resulting in realistic estimates, their uncertainties and associated limits of the coverage interval.
- Below is a preliminary example of determining the explosion time by applying Gaussian distribution for concentrations, resulting in distributions of ratios and explosion times.

Gaussian distributions of $C_{133}(t_2)$ and $C_{131m}(t_2)$ are used simply.

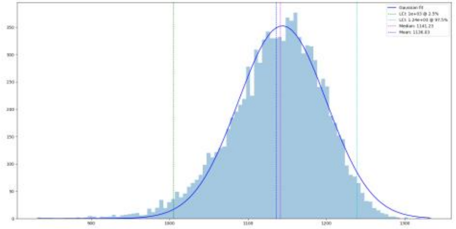


Distribution of the ratios

$$R(t_2) = \frac{C_{133}(t_2)}{C_{131\mathrm{m}}(t_2)}$$



Distribution of explosion times



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Reporting analysis results with respect to the requirements in ISO 11929:2019

METHODS

- IDC radionuclide analysis reports could be enhanced by reporting not only results and their uncertainties but also associated characterization limits.
- Analysis reports should include following results in the NCC method.
  - For current reports such as A/RRR, below results have been included.
    - Net counts and uncertainties
    - Decision thresholds (LC) and detection limits (MDC)
  - For isotopic ratio analysis
    - Covariances of net numbers of counts
  - For expert analysis and special studies, more results need to be estimated.
    - Best estimates and associated uncertainties for low counts
    - The lower and upper limits of the coverage interval

Summary (1/2)
Achievements of the past 25 years

SUMMARY

- In the first half of the past 25 years, only particulate systems were in IDC operations. The basic analysis methods were implemented in the early years and address the challenge of low counts of CTBT relevant radionuclides close to the natural background levels found in most IMS samples.
- In the second half of the past 25 years, noble gas systems came into operation after more than a decade of developing the specific analysis methods.
- There are three sets of approaches currently, the single channel analyser curve for particulate, the least squares regression on gamma- and X-rays peaks of xenon isotopes for high resolution spectra and the net count calculation method for beta-gamma coincidence spectra for noble gas, which are based on conventional frequentist statistics.
- Currently, we are in the implementation phase of analysis methods optimized for the different new generation noble gas systems.

Summary (2/2)
Possible Future Enhancements

SUMMARY

- Most daily IMS spectra have low counts close to background level. Decision thresholds by Currie's definition have been found to tend being underestimated, resulting in false positive detections.
- Enhancements and developments should be consistent with estimation of measurement uncertainty and characterization limits based on Bayesian statistics.
- There are new requirements for noble gas analysis from lessons learned in past years.
  - Under-estimation of decision thresholds due to negative net numbers of counts
  - Uncertainty components related to interference corrections
  - Deconvolution of X-rays contributions of xenon isotopes
  - Covariances of net numbers of counts between ROIs for isotopic ratio analysis
  - Reporting results of sample, gas background and memory corrections separately
  - Reporting analysis results as recommend in ISO 11929:2019

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Main references

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REFERENCES

- Axelsson, A., et al., 2012. Improvement of the SAUNA Noble Gas System Calibration Procedures. FOI-R-3451-SE, July 2012.
- Cagniant, A., et al., 2018. Methods for multimodal analysis applied to environmental radioxenon measurements. CEA/DIF/DASE/SRCE/134/2018/DO.
- Cooper, M.W., et al., 2016. Minimum Detectable Concentration and Concentration Calculations. Pacific Northwest National Laboratory. PNNL-25418.
- Cooper, M.W., et al., 2019. β-γ Absolute Calibration. PNNL-27572 Rev. 1.
- Cooper, M.W., et al., 2019. Radioxenon net count calculations revisited. J Radioanal Nucl Chem. https://doi.org/10.1007/s10967-019-06565-y.
- Currie, L. A., 1968. Limits for qualitative detection and quantitative determination. Appl. Radiochem. Anal. Chem. 40 (3), 586–593.
- De Geer, L.E., 2005. A Decent Currie at the PTS – Detection limit concepts in the PTS radionuclide software. CTBT/PTS/TP/2005-1.
- De Geer, L.E., 2007. The Xenon NCC Method Revisited. FOI-R-2350-SE. https://www.foi.se/report-summary?reportNo=FOI-R--2350--SE
- Deshmukh, N., et al., 2017. Comparison of new and existing algorithms for the analysis of 2D radioxenon beta gamma spectra. Journal of Radioanalytical and Nuclear Chemistry. 311 (3), 1849-1857. doi:10.1007/s10967-017-623 5174-5.
- ISO, 2019. ISO 11929 Standard. Determination of the Characteristic Limits (Decision Threshold, Detection Limit and Limits of the Confidence Interval) for Measurements of Ionizing Radiation – Fundamentals and Application.
- ISO/IEC, 2008. ISO/IEC Guide 98-3. Uncertainty of Measurement - Part 3: Guide to the Expression of Uncertainty in Measurement, Supplement 1: Propagation of distributions using a Monte Carlo method.
- Kalinowski, M.B., et al. 2010. Discrimination of nuclear explosions against civilian sources based on atmospheric xenon isotopic activity ratios. Pure and Applied Geophysics Topical, vol. 167/4-5, S.517–539. doi:10.1007/s00024-009-0032-1.
- Kalinowski, M., Liu, B., 2020. Calculation of Isotopic Ratios of Fission Products Detected at IMS Radionuclide Stations. Proceedings of the INMM 61st Annual Meeting July 12-16, 2020, Baltimore, MD, USA.
- Liu, B., et al, 2020. Enhancements on the decision threshold algorithm of the net count calculation method. Applied Radiation and Isotopes 159 (2020) 109084.
- Ringbom, A., Axelsson, A., 2020. A new method for analysis of beta-gamma radioxenon spectra. Applied Radiation and Isotopes 156 (2020) 108950.
- Wotawa, G., et al, 2003, Atmospheric transport modelling in support of CTBT verification-Overview and basic concepts, Atmos. Environ., 37, 2529– 2537.
- Zahringer, M., Kirchner, G., 2008. Nuclide ratios and source identification from high-resolution gamma-ray spectra with Bayesian decision methods. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment. 594(3), 400-406.