SnT 2023
CTBT: SCIENCE AND TECHNOLOGY CONFERENCE
HOFBURG PALACE - Vienna and Online
19 TO 23 JUNE

Comparison of Methods for Analysis of Radioxenon Beta-Gamma Coincidence Spectra Using MIKS Data

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INTRODUCTION

There are several different approaches for estimating radioxenon activity by analyzing beta-gamma coincidence spectra.
It is necessary to compare methods for obtaining more stable and accurate results, especially in the case of poor statistics.

METHODS/DATA

The most important and general approaches to activity estimates based on beta-gamma spectrum data were considered.

START

RESULTS

Some calculations were carried out on the MIKS data using various methods. Comparison between XeMat and NCC showed good agreement.

CONCLUSION

We recommend using methods with ROIs, since they are much more resistant to perturbations. Matrix methods (LS) are very convenient for calculating errors and the MDC. The Bayesian approach allows us to use a priori knowledge about the background activities of isotopes, which potentially provides the best result.

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Introduction

Some detection systems like the Russian MIKS system, create 2D β-γ coincidence energy spectra using a plastic scintillator cell to detect conversion electrons and betas and a NaI(Tl) crystal to detect gamma- and X-rays. These β-γ spectra are used to estimate the activity of the radioxenon isotopes in the air.

Some approaches to estimating the activity using beta-gamma spectra are:

- the Net Count Calculation (NCC) method;
- matrix methods;
- the Standard Spectrum Method.

If the counting rate is high, all methods are consistent and similar. Differences will be visible only for low statistics.

Estimation of the activity at low statistics is a difficult problem. Detector calibration drift distorts spectra. Another important problem is the interference of spectra of relevant xenon and radon isotopes.

It is necessary to compare methods for obtaining more stable and accurate results, especially in the case of poor statistics.

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Standard spectrum method for radioactivity estimation

Linear regression (column-vectors)

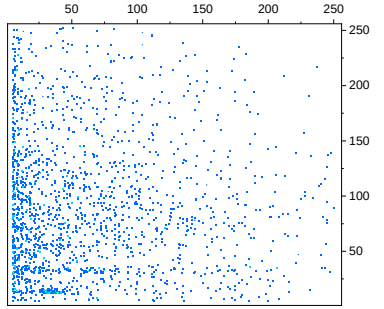
$$\vec{c} - k_s \vec{b} \approx \sum_{i=1}^{5} \beta_i \vec{r}_i = \mathbf{R}\vec{\beta}$$

β_i – amount (unknown) of the *i*th nuclide in the mixture;

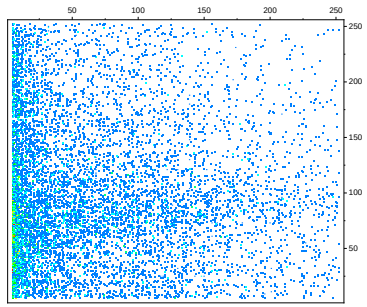
$k_s = t_S / t_D$ – acq. time ratio

Acquired β-γ spectra

$\vec{c}$ = SAMPLE Spectrum

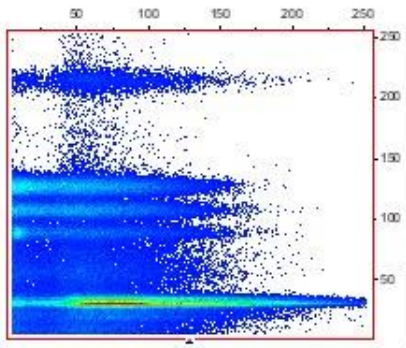


$\vec{b}$ = DETBK Spectrum

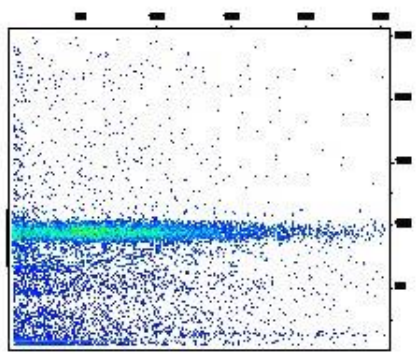


Reference β-γ spectra

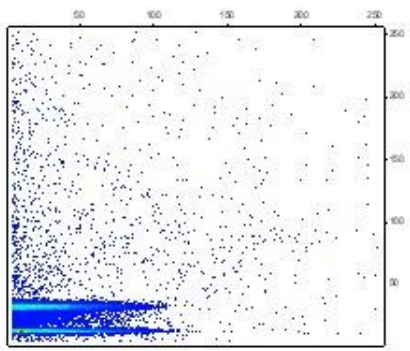
$\vec{r}_1$ = ^{222}Rn daughters spectrum



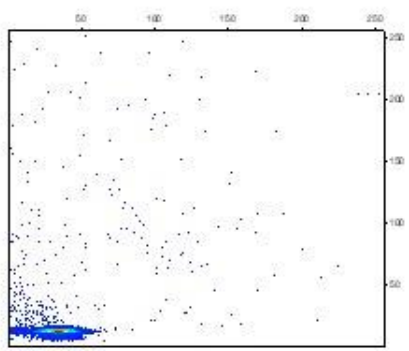
$\vec{r}_2$ = ^{135}Xe spectrum



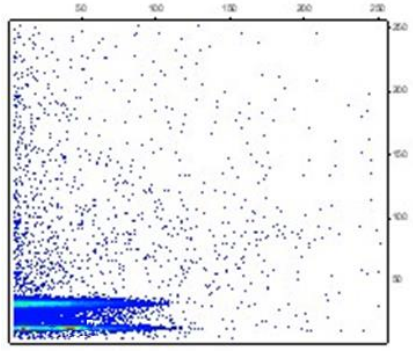
$\vec{r}_3$ = ^{133}Xe spectrum



$\vec{r}_4$ = ^{131m}Xe spectrum



$\vec{r}_5$ = ^{133m}Xe + ^{133}Xe spectrum



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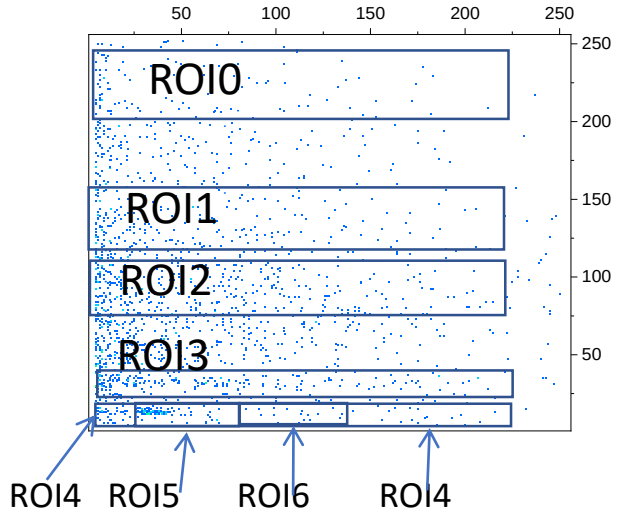
Methods/data

Simultaneous fitting of ROIs is derivative from the standard spectrum method

The region-of-interest approach is more resistant to gain shift

Example: 7 ROIs

One of the possible ROI sets



Improved #Ratios

#Ratios

```
PB214   1  0  0.50000
PB214   1  1  1.000000
PB214   1  2  0.650000
PB214   1  3  1.380000
PB214   1  4  0.080000
PB214   1  5  0.060000
PB214   1  6  0.060000
XE135   2  0  0.103000
XE135   2  1  0.026000
XE135   2  2  1.000000
XE135   2  3  0.042000
XE135   2  4  0.007000
XE135   2  5  0.022000
XE135   2  6  0.103000
XE133   3  0  0.103000
XE133   3  1  0.326000
XE133   3  2  0.136000
XE133   3  3  1.000000
XE133   3  4  0.007000
XE133   3  5  0.022000
XE133   3  6  0.103000
XE131m  4  0  0.103000
XE131m  4  1  0.326000
XE131m  4  2  0.136000
XE131m  4  3  0.042000
XE131m  4  4  0.007000
XE131m  4  5  0.022000
XE131m  4  6  0.103000
```

Calibration matrix (sensitivity matrix)

$$R = \begin{bmatrix} 0.5 & 0 & 0 & 0 & 0 \\ 1 & 0.005 & 0 & 0 & 0 \\ 0.65 & 1 & 0 & 0 & 0 \\ 1.38 & 0.01 & 1 & 0 & 0 \\ 0.08 & 0.02 & 0.41 & 0 & 0 \\ 0.06 & 0.01 & 0.3 & 1 & 0.1 \\ 0.06 & 0.02 & 0.14 & 0.1 & 1 \end{bmatrix} \begin{matrix} ROI0 \\ ROI1 \\ ROI2 \\ ROI3 \\ ROI4 \\ ROI5 \\ ROI6 \end{matrix}$$

$\vec{r}_1$ $\vec{r}_2$ $\vec{r}_3$ $\vec{r}_4$ $\vec{r}_5$

^{222}Rn, ^{135}Xe, ^{133}Xe, ^{131m}Xe, ^{133m}Xe

Matrix R contains most of information about the spectra of nuclides

Creating ROIs and grouping spectral data lead to loss of information and increase the robustness.

- ROIs should not overlap;
- ROIs should contain only the useful signal and give orthogonal spectra vectors;

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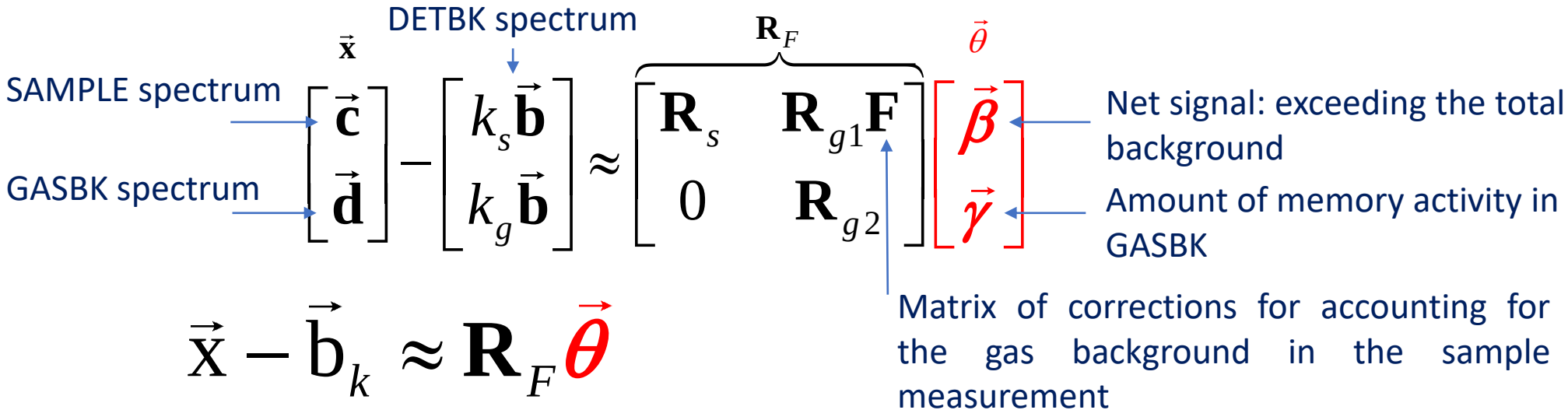
Taking into account the gas background (memory effect) in the regression equation in matrix form



$$\vec{\mathrm{x}} - \vec{\mathrm{b}}_k \approx \mathbf{R}_F\vec{\theta}$$

To account for the gas background, it is necessary to introduce extended spectrum vectors, which consist of the sample spectrum and the gas background. The regression matrix is also transformed. Thus, the problem of taking into account the gas background is also reduced to a linear regression. Usually the sensitivity matrices of the gas background and samples are equal ($\mathbf{R}_s = \mathbf{R}_{g1}$); however, in general it is possible to take into account the difference

Methods/data

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Approaches to solving linear regression $\vec{\mathbf{x}}_b = \mathbf{R}_F\vec{\theta} + \vec{\varepsilon}$; $\vec{\theta} = ?$

Frequentist

Likelihood function $L(\vec{\theta})$

1. Normal $L_G(\vec{\theta}) = \frac{1}{(2\pi)^{n/2}|\mathbf{\Omega}|^{1/2}}\exp\left[-\frac{1}{2}\left(\vec{\mathrm{x}}_b - \mathbf{R}_F\vec{\theta}\right)^{\mathrm{T}}\mathbf{\Omega}^{-1}\left(\vec{\mathrm{x}}_b - \mathbf{R}_F\vec{\theta}\right)\right]$,
2. Poisson $L_P(\boldsymbol{\theta}) = \prod_{i=1}^{2k}\frac{\left([\mathbf{R}_F\boldsymbol{\theta}]_i + b_{ki}\right)^{x_i}}{x_i!}\exp(-[\mathbf{R}_F\boldsymbol{\theta}]_i - b_{ki})$,

Data β-γ spectra

→ Least squares method/ maximum likelihood

Point estimation

1. $\vec{\theta}^*_{LS} = \left(\mathbf{R}_F^{\mathrm{T}}\mathbf{W}\mathbf{R}_F\right)^{-1}\mathbf{R}_F^{\mathrm{T}}\mathbf{W}\vec{\mathrm{x}}_b$, $\mathbf{K}_\theta = \left(\mathbf{R}_F^{\mathrm{T}}\mathbf{W}\mathbf{R}_F\right)^{-1}$
2. $\vec{\theta}^*_{ML} = \arg\max_\theta L_P(\vec{\theta})$

→ **Confidence interval (region)**

1. $\left(\vec{\beta} - \vec{\beta}^*\right)^{\mathrm{T}}\mathbf{K}_\beta^{-1}\left(\vec{\beta} - \vec{\beta}^*\right) = (\chi^2_k)^{-1}(p)$,

Bayesian

Likelihood $L(\vec{\theta}) = p(\vec{\mathbf{x}}|\ \vec{\theta})$

1. Normal
2. Poisson

Data β-γ spectra

Prior $p(\vec{\theta})$

1. Flat prior $\vec{\theta} > 0$
2. Other priors

→ **Bayes' Theorem** $p(\boldsymbol{\theta}|\ \mathbf{x}) \propto p(\mathbf{x}|\ \boldsymbol{\theta})p(\boldsymbol{\theta})$

→ **Posterior** $p(\boldsymbol{\theta}|\ \mathbf{x})$

Loss function

→ **Point estimation**

The posterior expected loss

1. MSE: $\vec{\theta}^*_{MSE} = E[\vec{\theta}\,|\,\vec{\mathrm{x}}]$
2. MAP: $\vec{\theta}^*_{MAP} = \arg\max\, p(\vec{\theta}\,|\,\vec{\mathrm{x}})$

Credible interval (region)

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Methods/data

Transformation from regression solutions to activities in the matrix form (the XeMat algorithm is used for MIKS)

Activity at acquisition start

$$\vec{A} = \mathbf{H}\vec{\beta}^{*}$$

Activity concentration (average)

$$\vec{A}_c = \frac{1}{V_0}\mathbf{G}_s\mathbf{G}_p\mathbf{\Gamma}\mathbf{H}\vec{\beta}^{*}$$

Standard correction factors in matrix form

$\mathbf{H} = \mathrm{diag}(G_{\lambda_i t_{sr}} / (t_{sl} \cdot \varepsilon_i Br_i))$, – activity factors;

$G_{\lambda t} = \lambda t / (1 - e^{-\lambda t})$ – decay - correction factor of acquisition;

$\mathbf{G}_p = \mathrm{diag}(e^{\lambda_i t_p})$ – decay - corr. of processing;

$\mathbf{G}_s = \mathrm{diag}(\lambda_i t_a / (1 - e^{-\lambda_i t_a}))$ – decay - corr. of sampling;

$\mathbf{\Gamma}$ – decay - corr. ^{133m}Xe; $V_0[\mathrm{m}^3] = \frac{V_{Xe}\,[\mathrm{cm}^3]}{0.086\,[\mathrm{cm}^3/\mathrm{m}^3]}$;

The WLS method allows use of the matrix form for solution and covariance matrix for uncertainties

$$\mathbf{K}_\theta = \left(\mathbf{R}_F^{\mathrm{T}}\mathbf{W}\mathbf{R}_F\right)^{-1}, \quad \mathbf{K}_\theta = \begin{bmatrix} \mathbf{K}_\beta & \mathbf{K}_{\beta\gamma} \\ \mathbf{K}_{\gamma\beta} & \mathbf{K}_\gamma \end{bmatrix}$$

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Uncertainties (WLS matrix approach)

Covariance matrix for activity

$$\mathbf{K}_A = \mathbf{H}\mathbf{K}_\beta\mathbf{H} + \mathbf{S}_1$$

statistical error — systematic error

Covariance matrix for activity concentration

$$\mathbf{K}_{Ac} = \frac{1}{V_0^2}\mathbf{G}_s\mathbf{G}_p\mathbf{\Gamma}\mathbf{K}_A\mathbf{\Gamma}^{\mathrm{T}}\mathbf{G}_s\mathbf{G}_p + \mathbf{S}_2$$

systematic error

SD and Pearson's r

$$\sigma_A^i = \sqrt{K_A^{ii}},\ r_A^{ij} = K_A^{ij} \Big/ \sqrt{K_A^{ii}K_A^{jj}}$$

$$\sigma_{Ac}^i = \sqrt{K_{Ac}^{ii}},\ r_{Ac}^{ij} = K_{Ac}^{ij} \Big/ \sqrt{K_{Ac}^{ii}K_{Ac}^{jj}}$$

Confidence region

A confidence k-dimensional ellipsoid around an activity vector with confidence probability p can be represented as

$$\left(\vec{A} - \vec{A}^*\right)^{\mathrm{T}} \mathbf{K}_A^{-1} \left(\vec{A} - \vec{A}^*\right) = F_k^{-1}(p),$$

$F_k^{-1}(p)$, - quantile chi-squared with k degrees of freedom

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Methods/data

It is known that the markers of the nature of a nuclear event are the activity ratios of ^{133}Xe and ^{135}Xe, ^{131m}Xe and ^{133m}Xe

First-order uncertainty propagation formula for ratios

$$\mathbf{K}_\rho = \mathbf{J}_\rho \mathbf{K}_A \mathbf{J}_\rho^{\mathrm{T}} \qquad \boldsymbol{\rho}(\mathbf{a}) = \begin{bmatrix} \rho_1 & \rho_2 & \rho_3 \end{bmatrix}^T = \left(\frac{A_2}{A_3} \quad \frac{A_5}{A_4} \quad \frac{A_2/A_3}{(A_5/A_4)^m} \right)^{\mathrm{T}}, \qquad \mathbf{J}_\rho = \begin{bmatrix} 0 & A_3^{-1} & -A_2 A_3^{-2} & 0 & 0 \\ 0 & 0 & 0 & -A_5 A_4^{-2} & A_4^{-1} \\ 0 & \frac{1}{A_3}\left(\frac{A_5}{A_4}\right)^{-m} & -\frac{A_2}{A_3^2}\left(\frac{A_5}{A_4}\right)^{-m} & \frac{A_2 m}{A_3 A_4}\left(\frac{A_5}{A_4}\right)^{-m} & -\frac{A_2\, m}{A_3\, A_5}\left(\frac{A_5}{A_4}\right)^{-m} \end{bmatrix}$$

Using the obtained SD estimates for ratios, we can use the z-test $z_1 = \frac{\hat{\rho}_1 - k_1}{\sigma_{\rho_1}}, z_2 = \frac{\hat{\rho}_2 - k_2}{\sigma_{\rho_2}}, z_2 = \frac{\hat{\rho}_3 - 1}{\sigma_{\rho_3}} \sim N(0,1)$

$\mathrm{H0_i}$: $A_2/A_3 = k_1, A_5/A_4 = k_2, \frac{A_2/A_3}{(A_5/A_4)^m} = 1.$

We need methods of combining the achieved levels of significance p-value (multiple comparisons problem). To test the joint hypothesis H0: against the one-sided alternative, you can for example use the statistic , which takes into account the correlation relationships of the parameters

$$z_\Sigma = (\vec{1}^T \mathbf{K}_\rho^{-1} \vec{1})^{-1/2} \vec{1}^T \mathbf{K}_\rho^{-1} \left(\vec{\rho}^* - \vec{\mathbf{h}} \right) \sim N(0,1)$$

A more accurate calculation uncertainty of ratio

- Fieller's method for testing the hypothesis for a ratio of normal random variables, e.g. H0: $\frac{A_2}{A_3} = k$ is based on statistics $z_F = \frac{\hat{A}_2 - k\hat{A}_3}{\sqrt{D_{A2} + k^2 D_{A3} - 2kK_A^{23}}} \sim N(0,1);$
- Monte-Carlo and bootstrap.

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XeMat software

XeMatrix is a software tool developed by VNIIA for MIKS. It utilizes the standard-spectrum and simultaneous-fitting methods for the purpose of analyzing β-γ coincidence radioxenon spectra. Spectra are read in the International Monitoring System (IMS) pulse height data (PHD) format. The final result is the activity and activity concentration for ^{135}Xe, ^{133}Xe, ^{131m}Xe, and ^{133m}Xe.

All of the above formulas are implemented in the XeMat algorithm:

- the library of reference β-γ spectra saved in the R matrix in view of the efficiency normalization;
- unmixing calculation for the sample and gas-background spectra, which allows one to control the goodness of fit and blunders of background variations (estimates should not differ widely from each other);
- the closed form of the LS approach and the covariance matrix are used to calculate the errors;
- the ability to set a priori thresholds for detection of each nuclide: the MDA and MDC depend on the priori knowledge of the presence of nuclides.

Results

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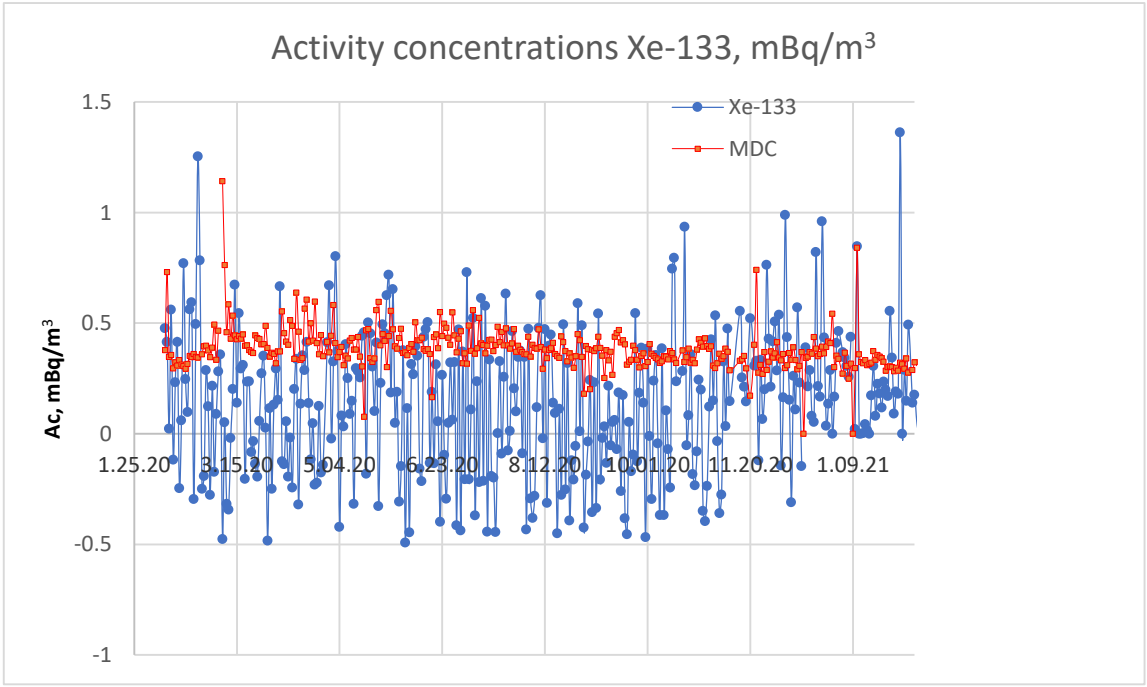
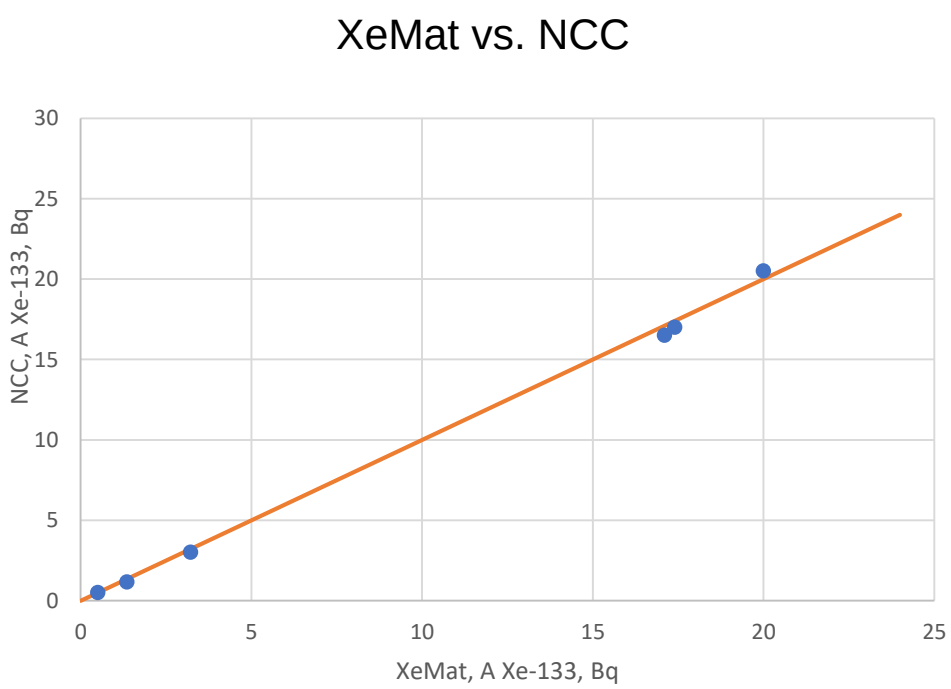
Activity concentration calculated using XeMat (MIKS data)

Some results are presented. Comparison between XeMat and NCC shows good agreement.





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Conclusion

- We studied possible approaches to estimating the activity using unmixing beta-gamma spectra: frequentist and bayesian.
- The maximum likelihood (Poisson) approach will have the advantage over the least squares method when the counting rate is very low and the background is well known. But only the LS method has a closed-form and covariance matrix. Matrix methods (LS) are very convenient for calculating errors and the MDC.
- Regions of interest should be set based on the efficiency vs. robustness tradeoff. We recommend using methods with ROIs, since they are much more resistant to perturbations.
- The Bayesian approach allows us to use a priori knowledge about the background activities of isotopes, which potentially provides the best result. It is worth noting that the choice of a positive flat a priori distribution is not quite realistic such a priori does not take into account the probability of the absence of any component in the mixture.
- Comparison between XeMat and the NCC shows good agreement.