



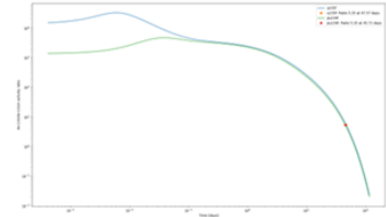
# Monte-Carlo Calculations of Isotopic Ratios of Fission Products Detected at IMS Radionuclide Stations

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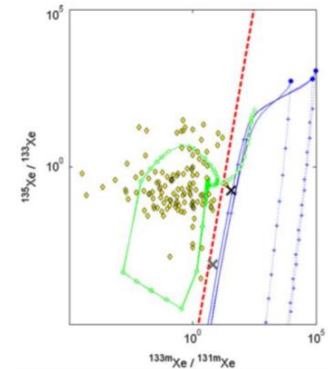
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- Event screening of a nuclear explosion from releases of civil facilities can be performed based on isotopic ratio analysis, e.g., four xenon relationship plot of  $\text{Xe}^{135}/^{133}$  vs  $\text{Xe}^{133\text{m}}/^{131\text{m}}$ .
- The explosion time of the nuclear event can be estimated by using a function of the isotopic ratio with time from the explosion time up to the stop of collection based on assumed scenarios, e.g., pairs of Zr-95/Nb-95, Ba-140/La-140, Xe-131m/Xe-133.
- This presentation demonstrates Monte-Carlo methods (MCM) estimating probability distributions of isotopic ratios, based on input distributions related to measurements and decay chains. Then the isotopic ratio, associated uncertainty and limits of the coverage interval can be estimated accordingly. Furthermore, the explosion time can also be estimated in the same way.



Ratio of  $\text{Xe}^{133}/\text{Xe}^{131\text{m}}$  determining explosion time

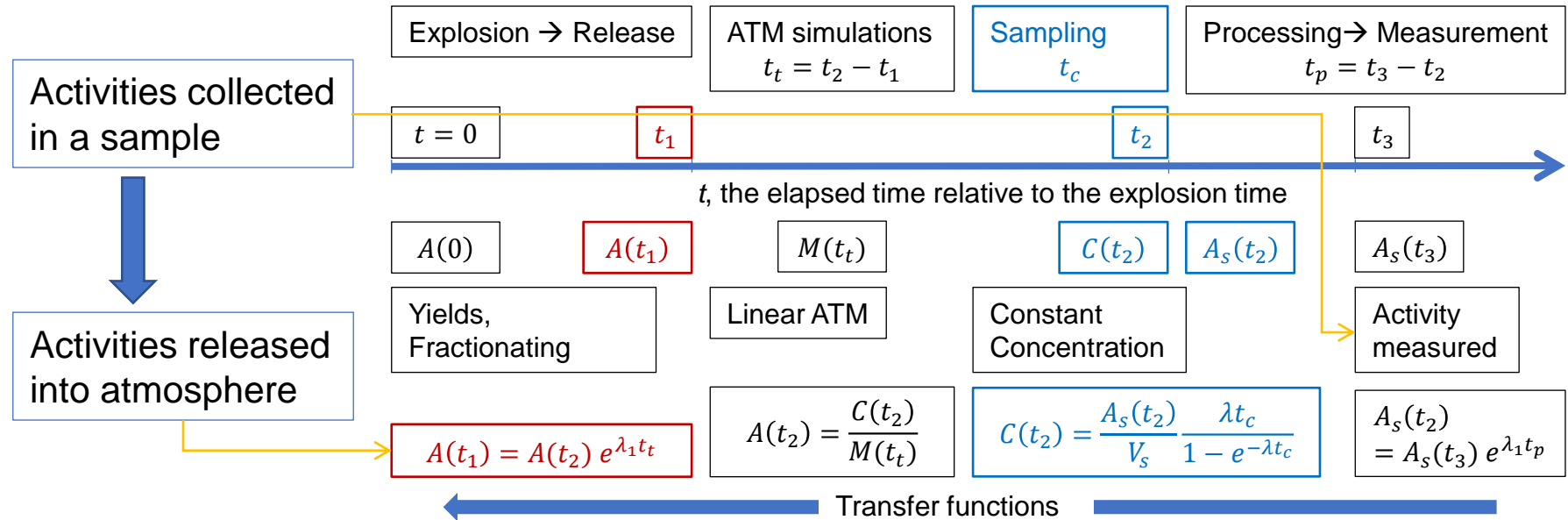


Plot of four xenon relationship discriminating nuclear explosions

# Schematic Time Series from a Nuclear Explosion to Sample Measurements

Procedures of isotopic ratio analysis by using transfer functions and ATM simulations

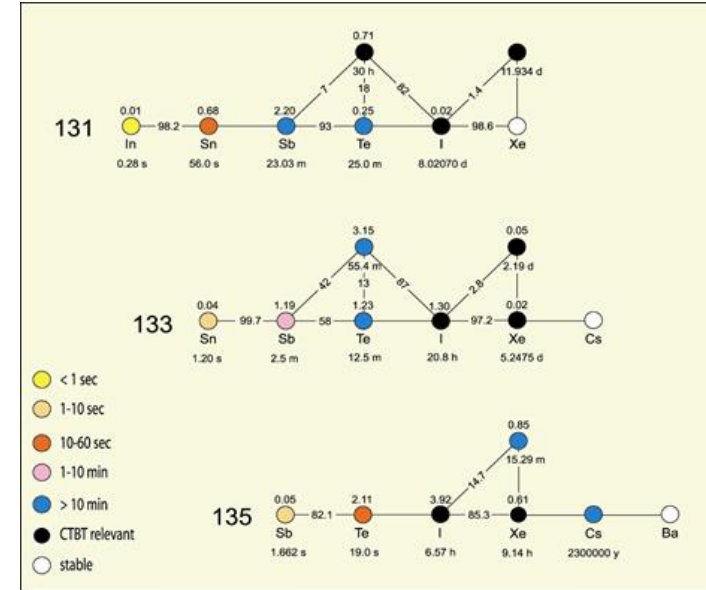
- Forward: Activities released into atmosphere are derived from UNE scenarios.
- Backward: Released activities are estimated by activities measured in samples.



- Activity evolution ( $A_1(t)$ ,  $A_2(t)$ ) in mass decay chains is estimated by Bateman equations under independent yields of fission products and full- or part-ingrowth.
- Activity ratios ( $R_{21}(t)$ ) with explosion yields are independent in batch reactor model but dependent in convection-driven leakage model.

$$R_{21}(t) = \frac{A_2(t)}{A_1(t)};$$

$$N_{m,n}(t) = \sum_{k=1}^n [N_{m,k}^0 \cdot \prod_{l=k}^{n-1} b_{m,l} \cdot \sum_{j=1}^n (\frac{T_{m,n}}{T_{m,j}} \cdot \prod_{i=1}^{n-1} \frac{1}{1 - T_{m,i}/T_{m,j}} e^{-\ln 2 \cdot t/T_{m,j}})];$$





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 the IMS station.
- Released activities are estimated based on activities measured in samples at IMS stations.

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



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

Linear SRS



$$M(t_t)$$

$$R(t_2) = \frac{AC_2(t_2)}{AC_1(t_2)}$$

Simplest case:  
independent decay



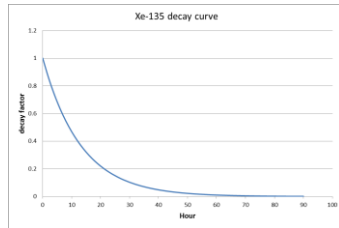
$$R(t_2) = R(t_1)e^{-(\lambda_2 - \lambda_1)t_t}$$

# The ratios of activity concentrations in the plume related to the isotopic ratios of source terms

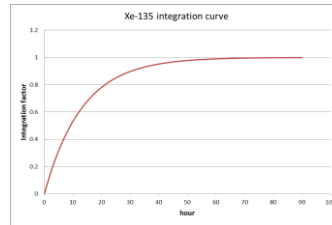
Decay chains are interrupted due to activity accumulation during sampling.

- Ratios of concentrations in the plume ( $R(t_2)$ ) is only dependent on decay constants.
- Ratios of activities collected in the sample ( $R_s(t_2)$ ) is dependent on not only decay constants ( $\lambda_1, \lambda_2$ ) but also collection time ( $t_c$ ).

During  
Sampling

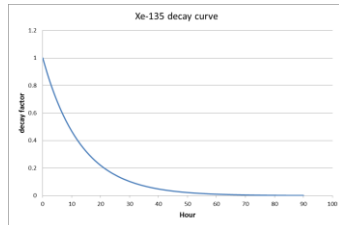


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$$R_s(t_2) = R(t_2) \frac{\lambda_1}{\lambda_2} \frac{1 - e^{-\lambda_2 t_c}}{1 - e^{-\lambda_1 t_c}}$$

Before and  
after sampling



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

A function of the isotopic ratio with time from the explosion time up to the stop of collection 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:  $R(t_2) = \frac{AC_2(t_2)}{AC_1(t_2)}$

$$N_{m,n}(t) = \sum_{k=1}^n [N_{m,k}^0 \cdot \prod_{l=k}^{n-1} b_{m,l} \cdot \sum_{j=1}^n \left( \frac{T_{m,n}}{T_{m,j}} \cdot \prod_{\substack{i=1 \\ l > j}}^{n-1} \frac{1}{1 - T_{m,i}/T_{m,j}} e^{-\ln 2 \cdot t/T_{m,j}} \right)]$$

The simplest case:  
Independent decay

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

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

Analytically

Numerically

$t_2$

Isotopic ratios and associated uncertainties are dependent on not only concentrations but also their uncertainties and covariances. (see the next two slides and examples behind)

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



## 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:

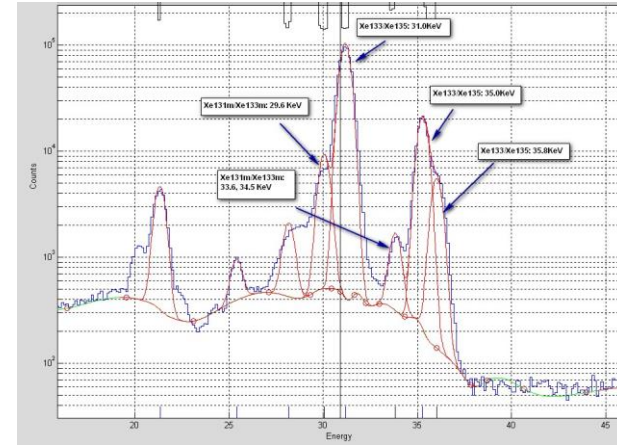
- Cs-134/Cs-137,
- There is no correlation caused by either decay chains or peak areas.

### 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.

## 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 fitting.
- 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

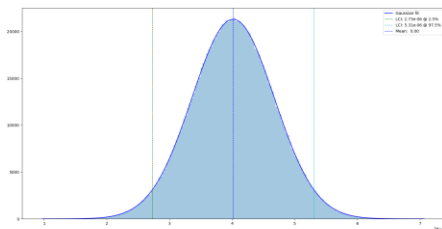
## Correlations in the NCC method are due to three ways,

- 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.

Zr/Nb-95 were not detected at SEP63, but detected in the lab analysis at ITL10.

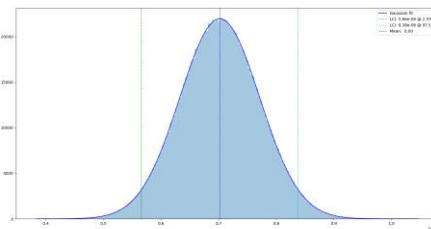
- In this preliminary study, Gaussian distributions of concentrations were used and the correlation was ignored. The distribution of the ratios was estimated by MCM, then the mean and Limits of the coverage interval (LCI) (95%) were estimated accordingly.

$C_1(t_2)$  : Gaussian  
Zr-95:  $7.02 \pm 9.87(\%)$  uBq/m<sup>3</sup>



$C_2(t_2)$  : Gaussian  
Nb-95:  $4.02 \pm 16.4 (\%)$  uBq/m<sup>3</sup>

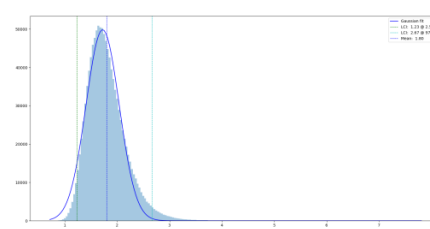
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$$R(t_2) = \frac{C_2(t_2)}{C_1(t_2)}$$

Non-linear  
model

$R(t_2)$  : Not Gaussian  
Nominal: 1.75  
Mean: 1.80; LCI: (1.23, 2.67)

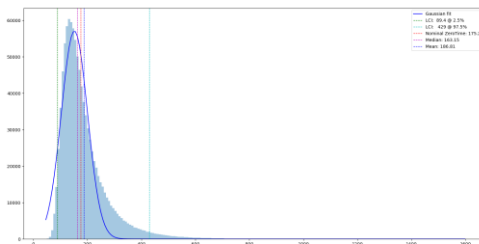
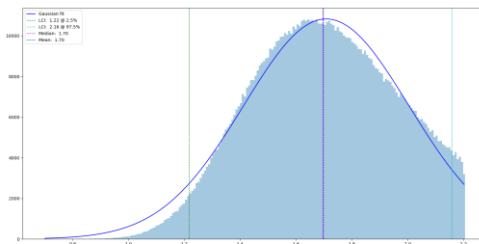
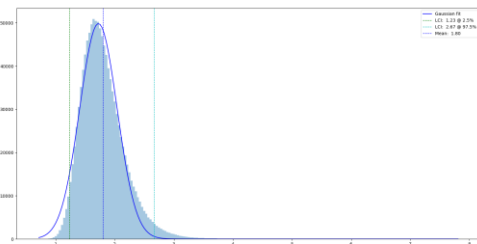


- The distribution of the explosion times was estimated simply by the parent-daughter decay of Zr-95 to Nb-95, and the mean and LCIs were estimated accordingly.
- The nominal value of 175.3 days is different from the mean value of 186.8 days. It is caused by non-Gaussian distribution of the ratios as well as the truncated distribution due to the limit of the equilibrium ratio of 2.205 for Zr-95 to Nb-95 decay.

$R(t_2)$ : Not Gaussian  
Nominal: 1.75  
Mean: 1.80;

Zr/Nb-95 decay:  
Equilibrium ratio of 2.205

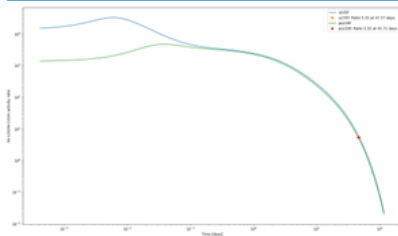
$t_2$ : Not Gaussian  
Nominal: 175.3 days  
Mean: 186.8 days  
LCI: (89.4, 429) days





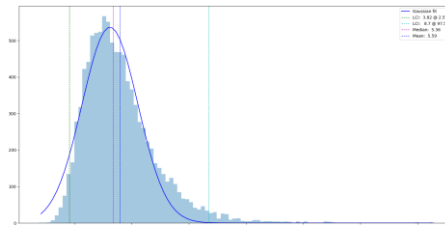
- The sample at JPX38 at 19:00 on 8 April 2013 (the stop of collection)
  - Xe-133:  $3.05 \pm 0.14$ ; Xe-131m:  $0.57 \pm 0.11$  (mBq/m<sup>3</sup>);
- The explosion time was estimated by the Monte-Carlo method.
  - Gaussian distributions of concentrations were used simply in this preliminary analysis.
  - The mean of 47.3 days with the LCI (41.7, 51.7) for U235 full-ingrowth model.
  - The estimation could be improved further by more detailed activity evolution models, Poisson distributions of gross counts in ROIs measured and calibration data.

Numeric solution of explosion time with two boundaries for scenarios.

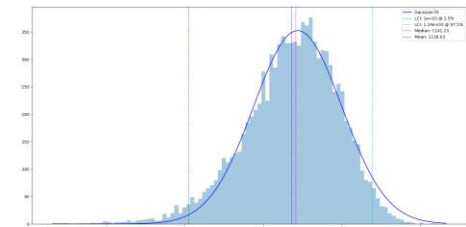


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$$R(t_2) = \frac{C_{133}(t_2)}{C_{131m}(t_2)}$$



Distribution of explosion times  
Nominal: 47.6 days  
Mean:: 47.3 days



- In the preliminary studies, the distributions of concentrations were supposed as Gaussian and the correlations were ignored as well.
- Generally, the peak counts follow Poisson distribution, especially for low level counts. And it is independent between gross numbers of counts.
- Probability distributions of measurands, e.g. activity, concentration and isotopic ratio, can be obtained by the Monte-Carlo method in further studies, directly based on distributions of peak counts in the measurement spectra, calibration data and related parameters, resulting in realistic estimates for measurands, their uncertainties and associated limits of the coverage interval.

- Isotopic ratios and associated uncertainties are dependent on concentrations as well as associated uncertainties and covariances. It might be complicated in analytical analysis procedure.
- In contrast, it is a relatively simple procedure to derive the probability distribution of isotopic ratios by using the MC method, based on distributions of activities or peak counts measured. The estimated mean of the ratios and associated limits of the coverage interval can improve the event screening.
- Using the MC method for event timing based on isotopic ratios assures more accurate results than simply using the concentration ratio.

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