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1 Introduction

The SAUNA automatic atmospheric noble gas sampler (Ref. [1]) determines the amount of Xenon radioactivity in an atmospheric sample by a $\beta$-$\gamma$ coincidence measurement.

SAUNA produces full spectral information in one and two dimensions as well as other information necessary for detailed manual analysis and scrutiny when necessary. However, since the system is designed for continuous, fully automatic operation, a simple and robust algorithm suitable for automatic analysis of two-dimensional $\beta$-$\gamma$ coincidence data is also useful. Such an algorithm has been established as a part of the development of SAUNA, and has been implemented and tested with the SAUNA system in the framework of the International Noble Gas Experiment collaboration (Ref. [2]). The algorithm has been used to derive all numbers reported from SAUNA measurements so far. It may be considered a useful starting point for development of more precise analysis tools, manual or automatic, if such are required in the future. This report aims to present in some detail the SAUNA $\beta$-$\gamma$ coincidence data analysis algorithm.

The presentation in the following sections concentrates exclusively on the derivation of activity concentrations and minimum detectable concentration (MDC) estimates from raw numbers of counts in various Regions of Interest (ROIs). Little is said about the measurement itself, which is presented in detail in e.g. Ref. [1]. Here it is enough to note that the Xenon isotopes of interest all produce coincident electron (beta or conversion electrons) and photon (gamma or X-ray) radiation in the course of their decay. In SAUNA, photons are detected using a NaI scintillator and electrons are detected in the plastic scintillator material which also forms the walls of the measurement cell. Two measurements are performed to determine each activity concentration: a “gas background” measurement on the empty cell followed by a “sample” measurement on the cell after the introduction of an atmospheric sample. Simple rectangular ROIs are used to determine the numbers of counts associated with the decay of each isotope (or its background).

A main topic of this report is the treatment of the complications which arise due to factors such as

- the short half lives of the xenon isotopes of interest compared to the measurement times involved,

- the various sources of background present:

  - $\beta$-$\gamma$ decays from Radon daughters,
  - residual activity remaining in the measurement cell from the previous measurement (“gas memory background”) and
  - background arising from external sources (“ambient background”)

and

- the interference of the various spectral components of the xenon isotopes and background with each other.

The report is also meant to serve as a compilation of the formulae involved (mostly simple but some very tedious to write down), and therefore readability has occasionally been sacrificed for explicitness. In Section 2, the calculation of an atmospheric concentration of xenon radioactivity from a measured number of counts is reviewed, taking into account the finite decay times of the measured isotopes. In Sections 3 and 4, a method of calculating the net number of counts used to estimate the atmospheric concentration is presented, taking into account the various interference effects listed above.

The expressions given in Section 3 make use of known characteristics of detector response functions to estimate from each measured 2D histogram (sample or gas background) a “net number of counts” for each Region of Interest (ROI). Such known characteristics are ratios of counts in different regions stemming from the same source and “background regions” adjacent to the ROI, used to estimate the contribution of uncorrelated, approximately “flat” background (typically arising from external sources). The expressions
given in Section 4 combine the thus estimated net number of counts in the sample and background measurements to estimate a decay-corrected, background-subtracted number of counts. In all cases, formulae for the variances of the estimated quantities are also presented, on a form which allows division of the variance into parts contributed by counting statistics on the one hand and systematic uncertainties (i.e. calibration factors and counts ratios) on the other.

In Section 5, the method presented in the previous sections is extended to provide an estimate of the “zero-signal” variance used to estimate the Minimum Detectable Concentration (MDC) expected for a measurement.

The report concludes with an example of the analysis procedure, presented in Section 6.
2 Air concentration of xenon radioactivity

The xenon activity measurement is performed in three steps: collection, sample processing, and activity measurement. In addition, the activity measurement is preceded by a background measurement of the empty measuring cell. If constant atmospheric activity during sample collection is assumed, the air concentration of xenon radioactivity \( C_A \) is calculated using the formula

\[
C_A = \frac{c}{\epsilon_{\beta\gamma} \cdot \beta\gamma} \frac{\lambda^2 \cdot F_C \cdot F_P \cdot F_A}{V} \cdot t_{coll} \tag{1}
\]

where \( c \) is the background-corrected number of counts in the ROI corresponding to coincidences from the Xe isotope in question, \( \lambda \) is the decay constant of the isotope, \( \epsilon_{\beta\gamma} \) is the energy-integrated detection efficiency for the ROI in which the \( c \) counts were detected, \( \beta\gamma \) is the \( \beta - \gamma \) branching ratio to the decay yielding counts in this ROI, \( t_{coll} \) is the sample collection time and \( V \) is the sampled air volume. The factor \( F_C = 1 - e^{-\lambda t_{coll}} \) results from integration of a function expressing xenon activity collected by air sampling reduced by decay during the sampling. The activity function is displayed as the full blue curve in Fig. 1, which shows a schematic example of a sampling-processing-counting cycle in a generic air sampling system. The factor \( F_P = e^{-\lambda t_{proc}} \) accounts for decay during the processing of the sample (the short-dashed red curve). The factor \( F_A = 1 - e^{-\lambda t_{acq}} \) results from integration of the number of decays occurring during the measurement of the sample (the long-dashed green curve). Fig. 1 also shows how the number of counts accumulates, assuming 100 % detection efficiency and branching ratio 1 for the decay. If \( t_{coll} \) is expressed in seconds, \( \lambda \) in inverse seconds and \( V \) in \( m^3 \), the concentration \( C_A \) will be obtained in Bq/m\(^3\). In practice, the sampled air volume \( V \) is found by dividing the volume of xenon in the sample (determined accurately through gas chromatography) by the known proportion \( 8.7 \cdot 10^{-8} \) of stable xenon in air.

Considering only the uncertainties in the number of counts, the detection efficiency and the sample
volume, the relative uncertainty in the calculated concentration is given by

$$\left| \frac{\Delta C_A}{C_A} \right| = \sqrt{\left| \frac{\Delta c}{c} \right|^2 + \left| \frac{\Delta \epsilon_{\beta \gamma}}{\epsilon_{\beta \gamma}} \right|^2 + \left| \frac{\Delta V}{V} \right|^2}$$

(2)

For the purpose of determining the intrinsic quality of the system, and testing the agreement with other comparable sampling systems it is more useful to determine only the part of the relative uncertainty which stems exclusively from counting statistics:

$$\left| \frac{\Delta C_A}{C_A} \right| = \sqrt{\left| \frac{\Delta c}{c} \right|^2}$$

(3)

where $\Delta c$ should in this case also be calculated so that only statistical sources of uncertainty are considered.

The net number of counts $c$ to be used in the concentration equation (1) must be estimated from the two-dimensional $\beta - \gamma$ coincidence spectra collected during the background and sample measurements. The following sections present an algorithm to accomplish this.
3 Formulae for net numbers of counts in ROIs of interest

The $\beta - \gamma$ detector in SAUNA consists of a NaI(Tl) crystal surrounding a plastic scintillator cell, holding the gaseous xenon sample. A triple coincidence is created between the two photomultiplier tubes (PMTs) attached to the plastic scintillator and the NaI(Tl) PMT. A two-dimensional $\beta - \gamma$ energy spectrum is obtained by displaying the NaI(Tl) pulse height (corresponding to the photon energy) versus the summed pulse heights from the two scintillator cell PMTs (the electron energy).

All four xenon isotopes emit X-rays in the 30-keV region with a total branching ratio of about 50%, except for $^{135}$Xe, which has only a 5% X-ray branch. The associated conversion electron energies are 129, 45, 199, and 214 keV for $^{131m}$Xe, $^{133}$Xe, $^{133m}$Xe, and $^{135}$Xe, respectively. Other strong coincident decay modes are the 81-keV gamma ray in association with a beta decay with 346-keV endpoint energy in $^{133}$Xe, and the 249.8-keV gamma ray following the 910-keV endpoint energy beta decay in $^{135}$Xe. Based on a selection of these decays, a number of ROIs are defined in the $\beta - \gamma$ coincidence spectrum:

- ROI 1: The 81-keV gamma ray in association with a beta decay with 346-keV endpoint energy in $^{133}$Xe.
- ROI 2: The 30 keV X-ray associated with a beta decay with 346-keV endpoint energy in $^{133}$Xe. The 45-keV conversion electron energy is added in the beta pulse-height spectrum, resulting in a beta continuum shifted 45 keV towards higher energy.
- ROI 12: The 30 keV X-ray associated with the 129 keV conversion electron in $^{131m}$Xe
- ROI 13: The 30 keV X-ray associated with the 199 keV conversion electron in $^{133m}$Xe
- ROI 11: The 250-keV gamma ray in association with a 910-keV endpoint energy beta decay in $^{135}$Xe.

In addition, one ROI is defined to enable correction for any radon present in the sample:

- ROI 3: The 352-keV gamma ray in association with a 670-keV endpoint energy beta decay in $^{214}$Pb.

Finally, two ROIs are used in the background subtraction process:

- ROI 21: $^{135}$Xe 250-keV ROI background taken in the relatively flat region next to ROI 11.
- ROI 23: $^{214}$Pb 352-keV ROI background taken in the relatively flat region next to ROI 3.

The background subtraction for $^{133}$Xe, $^{131m}$Xe, and $^{133m}$Xe is performed using the decay-corrected gas background sample (see next section). The background treatment is different for $^{135}$Xe and $^{214}$Pb due to their short half-lives compared to the other isotopes. Also, a similar treatment of ROI 1 would not work, since the ambient background is relatively non-flat in that part of the spectrum. For these reasons, background ROIs (ROI 21 and 23) have been introduced for $^{135}$Xe and $^{214}$Pb but not for $^{133}$Xe, $^{131m}$Xe, and $^{133m}$Xe.

The labels $C_n$ used in the formulae presented in this section designate the number of counts found in ROI number $n$ in the $\beta - \gamma$ coincidence spectrum of the sample measurement. Fig. 2 shows the numbering of the ROIs. In the next section, labels $D_n$ will be used instead of $C_n$s to indicate counts in an ROI in the gas background measurement. Note that at this stage of the analysis, the treatment of the two measurements are exactly the same and independent of each other. Primed quantities $C'_n$ and $D'_n$ will indicate that some form of background subtraction has been carried out.

The $R_n$s in the formulae are the correction ratios used to estimate the amount of contamination from one ROI in another. The ratios used are:
• $R_0$: The net number of Rn-related counts in the $^{135}\text{Xe}$ ROI divided by the net number of Rn-related counts in the $^{214}\text{Pb}$ ROI.

• $R_1$: The net number of Rn-related counts in the $^{133}\text{gXe-80}$ ROI divided by the net number of Rn-related counts in the $^{214}\text{Pb}$ ROI.

• $R_2$: The net number of counts from the Compton background from $^{133}\text{gXe-80}$ in the $^{133}\text{gXe-30}$ ROI divided by the number of counts in the $^{133}\text{gXe-80}$ ROI.

• $R_5$: The net number of counts in the $^{131m}\text{Xe}$ ROI which originate from $^{133}\text{gXe}$ coincidences divided by the net number of counts in the $^{133}\text{gXe-80}$ ROI.

• $R_6$: The net number of counts in the $^{133m}\text{Xe}$ ROI which originate from $^{133}\text{gXe}$ coincidences divided by the net number of counts in the $^{133}\text{gXe-80}$ ROI.

These are deemed sufficient for a transparent presentation of the algorithm, additionally however, for a more complete treatment, counts originating from the decay of $^{135}\text{Xe}$, both Compton and associated X-rays, should be included by introducing two additional ratios.

Note that in all cases we have defined the net number of counts to enter the ratio, not the total number of counts in the ROI. Thus any constant background should not be included when the ratio is determined, nor when it is used to estimate the contamination in an ROI. Similarly, the Rn-related background in the $^{133}\text{gXe-80}$ ROI should not be included when determining the ratios $R_2$, $R_5$ and $R_6$, nor when they are used to estimate the background in the various affected ROIs. In the case of the ratios $R_5$ and $R_6$, however, the $^{133}\text{gXe-80}$ Compton background is defined to be included in the ratios, since the amount of such Compton background is a fixed feature of the detector response function.
Using the quantities thus defined we calculate the net numbers of counts and the associated variances according to the formulae given below.

The net number of counts from $^{214}$Pb-352 coincidences:

$$C'_3 = C_3 - C_{23}$$

$$\left(\Delta C'_3\right)^2 = \left(\Delta C_3\right)^2 + \left(\Delta C_{23}\right)^2 = C_3 + C_{23}$$

(4) \hspace{1cm} (5)

The net number of counts from $^{135}$Xe-250 coincidences:

$$C'_{11} = C_{11} - R_0 \cdot C'_3 - C_{21}$$

$$\left(\Delta C'_{11}\right)^2 = \left(\Delta C_{11}\right)^2 + \left(\Delta (R_0 C'_3)\right)^2 + \left(\Delta C_{21}\right)^2$$

$$= \left(\Delta C_{11}\right)^2 + R_0^2 \left(\Delta C'_3\right)^2 + C'_3\left(\Delta R_0\right)^2$$

$$= C_{11} + C_{21} + R_0^2 C_3 + R_0^2 C_{23} + C'_3\left(\Delta R_0\right)^2$$

(7)

where the variance can be split into statistical and systematic contributions:

$$\left(\Delta C'_{11}\right)^2_{\text{stat}} = C_{11} + C_{21} + R_0^2 C_3 + R_0^2 C_{23}$$

$$\left(\Delta C'_{11}\right)^2_{\text{syst}} = C'_3\left(\Delta R_0\right)^2$$

(8) \hspace{1cm} (9)

The net number of counts from $^{133}$Xe-80 coincidences:

$$C'_1 = C_1 - R_1 \cdot C'_3$$

$$\left(\Delta C'_1\right)^2 = \left(\Delta C_1\right)^2 + \left(\Delta (R_1 C'_3)\right)^2$$

$$= \left(\Delta C_{1}\right)^2 + R_1^2 \left(\Delta C'_3\right)^2 + C'_3\left(\Delta R_1\right)^2$$

$$= C_1 + R_1^2 C_3 + R_1^2 C_{23} + C'_3\left(\Delta R_1\right)^2$$

(11)

$$\left(\Delta C'_1\right)^2_{\text{stat}} = C_1 + R_1^2 C_3 + R_1^2 C_{23}$$

$$\left(\Delta C'_1\right)^2_{\text{syst}} = C'_3\left(\Delta R_1\right)^2$$

(12) \hspace{1cm} (13)

The net number of counts from $^{131m}$Xe coincidences:

$$C'_{12} = C_{12} - R_5 \cdot C'_1$$

$$\left(\Delta C'_{12}\right)^2 = \left(\Delta C_{12}\right)^2 + \left(\Delta (R_5 C'_1)\right)^2$$

$$= \left(\Delta C_{12}\right)^2 + R_5^2 \left(\Delta C'_1\right)^2 + C'_1\left(\Delta R_5\right)^2$$

$$= \left(\Delta C_{12}\right)^2 + R_5^2 \left(\Delta C_1\right)^2 + R_5^2 R_1^2 \left(\Delta C'_3\right)^2 + R_5^2 C_3\left(\Delta R_1\right)^2 + C'_1\left(\Delta R_5\right)^2$$

$$= C_{12} + R_5^2 C_1 + R_5^2 R_1^2 C_3 + R_5^2 R_1^2 C_{23} + R_5^2 C'_3\left(\Delta R_1\right)^2 + C'_1\left(\Delta R_5\right)^2$$

(15)

$$\left(\Delta C'_{12}\right)^2_{\text{stat}} = C_{12} + R_5^2 C_1 + R_5^2 R_1^2 C_3 + R_5^2 R_1^2 C_{23}$$

$$\left(\Delta C'_{12}\right)^2_{\text{syst}} = R_5^2 C'_3\left(\Delta R_1\right)^2 + C'_1\left(\Delta R_5\right)^2$$

(16) \hspace{1cm} (17)

The net number of counts from $^{133m}$Xe coincidences:

$$C'_{13} = C_{13} - R_6 \cdot C'_1$$

$$\left(\Delta C'_{13}\right)^2 = \left(\Delta C_{13}\right)^2 + \left(\Delta (R_6 C'_1)\right)^2$$

(18)
\[ \begin{align*}
= (\Delta C_{13})^2 + R_6^2(\Delta C_1')^2 + C_1' \, (\Delta R_6)^2 \\
= (\Delta C_{13})^2 + R_6^2(\Delta C_1')^2 + R_6^2 R_1^2(\Delta C_3')^2 + R_6^2 C_3' \, (\Delta R_1)^2 + C_1' \, (\Delta R_6)^2 \\
= C_{13} + R_6^2 C_1 + R_6^2 R_1^2 C_3 + R_6^2 R_1^2 C_{23} + R_6^2 C_3' \, (\Delta R_1)^2 + C_1' \, (\Delta R_6)^2 \\
(\Delta C_{13}')^2_{\text{stat}} &= C_{13} + R_6^2 C_1 + R_6^2 R_1^2 C_3 + R_6^2 R_1^2 C_{23} \\
(\Delta C_{13}')^2_{\text{syst}} &= R_6^2 C_3' \, (\Delta R_1)^2 + C_1' \, (\Delta R_6)^2 
\end{align*} \]

The net number of counts from $^{133}\text{Xe-30}$ coincidences:

\[ \begin{align*}
C_2' &= C_2 - C_1'_{12} - C_1'_{13} - R_2 \cdot C_1' \\
(\Delta C_2')^2 &= (\Delta C_2)^2 + (\Delta C_1')^2 + R_2^2(\Delta C_1')^2 + C_1' \, (\Delta R_2)^2 \\
&= (\Delta C_2)^2 + (\Delta C_1')^2 + R_2^2(\Delta C_1')^2 + C_1' \, (\Delta R_2)^2 \\
&+ R_2^2(\Delta C_1')^2 + R_2^2 R_1^2(\Delta C_3')^2 + R_2^2 C_3' \, (\Delta R_1)^2 + C_1' \, (\Delta R_2)^2 \\
&= (\Delta C_2)^2 + R_2^2(\Delta C_1')^2 + R_2^2 R_1^2(\Delta C_3')^2 + R_2^2 C_3' \, (\Delta R_1)^2 + C_1' \, (\Delta R_2)^2 \\
&= C_{13} + R_6^2 C_1 + R_6^2 R_1^2 C_3 + R_6^2 R_1^2 C_{23} + R_6^2 C_3' \, (\Delta R_1)^2 + C_1' \, (\Delta R_2)^2 \\
&= C_2 + C_{12} + C_{13} \\
&+ (R_2^2 + R_3^2 + R_6^2) C_1 + (R_2^2 + R_3^2 + R_6^2) R_1^2 C_3 + (R_2^2 + R_3^2 + R_6^2) R_1^2 C_{23} \\
&+ (R_2^2 + R_3^2 + R_6^2) R_1^2 C_{23} + (R_2^2 + R_3^2 + R_6^2) C_3' \, (\Delta R_1)^2 \\
&+ C_1' \, (\Delta R_2)^2 + C_1' \, (\Delta R_3)^2 + C_1' \, (\Delta R_6)^2 
\end{align*} \]

\[ \begin{align*}
(\Delta C_2')^2_{\text{stat}} &= C_2 + C_{12} + C_{13} \\
&+ (R_2^2 + R_3^2 + R_6^2) C_1 + (R_2^2 + R_3^2 + R_6^2) R_1^2 C_3 + (R_2^2 + R_3^2 + R_6^2) R_1^2 C_{23} \\
(\Delta C_2')^2_{\text{syst}} &= (R_2^2 + R_3^2 + R_6^2) C_3' \, (\Delta R_1)^2 \\
&+ C_1' \, (\Delta R_2)^2 + C_1' \, (\Delta R_3)^2 + C_1' \, (\Delta R_6)^2 
\end{align*} \]
4 Formulae for background-subtracted numbers of counts

The quantities $C_n$ and $D_n$, calculated as above are used to find the background-subtracted number of counts to be used in the concentration equation. These corrected values are labelled $c_{133}^{80}$, $c_{133}^{30}$, $c_{135}$, $c_{131m}$ and $c_{133m}$. In the expressions below, the factor $F$ takes into account the radioactive decay of the "memory" background and the different measurement times in the sample and background measurements. As Fig. 3 attempts to explain, it expresses the transition from a number of counts in an ROI in the gas background measurement to an expected contribution to the background in the same ROI in the sample measurement. The factor $F$ is given by:

$$F = \frac{t_{\text{real}}}{t_{\text{bgr}}} \frac{t_{\text{live}}}{t_{\text{real}}} e^{-\lambda \cdot \tau} (1 - e^{-\lambda \cdot t_{\text{real}}}) / (1 - e^{-\lambda \cdot t_{\text{bgr}}})$$

where $\lambda$ is the decay constant of the isotope in question, $\tau$ is the time elapsed between the start of the background measurement and the start of the sample measurement and the various $t:s$ represent live and real acquisition times for the sample and background measurements. Using the thus defined quantities, the values of $c_{133}^{80}$, $c_{133}^{30}$, $c_{135}$, $c_{131m}$ and $c_{133m}$ and their variances are calculated as follows:

$$c_{133}^{80} = C_1' - F \cdot D_1'$$

$$\left(\Delta c_{133}^{80}\right)^2 = \left(\Delta C_1\right)^2 + F^2 \left(\Delta D_1\right)^2$$

$$= \left(\Delta C_1\right)^2 + R_1^2 \left(\Delta C_3\right)^2 + C_3^2 \left(\Delta R_1\right)^2$$

$$+ F^2 \left(\Delta D_1\right)^2 + R_1^2 \left(\Delta D_3\right)^2 + D_3^2 \left(\Delta R_1\right)^2$$

$$= \left(\Delta C_1\right)^2 + R_1^2 \left(\Delta C_3\right)^2 + F^2 \left(\Delta D_1\right)^2 + R_1^2 R_1^2 \left(\Delta D_3\right)^2$$
\[ c_{133} = C_2 - F \cdot D'_2 \]
\[ (\Delta c_{133}^{30})^2 = (\Delta C_2)^2 + F^2(\Delta D'_2)^2 \]
\[ = (\Delta C_2)^2 + (\Delta C_{12})^2 + (\Delta C_{13})^2 \]
\[ + (R_2^2 + R_3^2 + R_6^2)(\Delta C_1)^2 + (R_2^2 + R_5^2 + R_6^2)R_1^2(\Delta C_3')^2 \]
\[ + F^2(\Delta D_2)^2 + F^2(\Delta D_{12})^2 + F^2(\Delta D_{13})^2 \]
\[ + F^2(R_2^2 + R_3^2 + R_6^2)(\Delta D_1)^2 + F^2(R_2^2 + R_5^2 + R_6^2)R_1^2(\Delta D_3)^2 \]
\[ + (R_2^2 + R_3^2 + R_6^2)C_3^2(\Delta R_1)^2 + C_1^2(\Delta R_2)^2 + C_1^2(\Delta R_5)^2 + C_1^2(\Delta R_6)^2 \]
\[ + F^2(R_2^2 + R_3^2 + R_6^2)D_3^2(\Delta R_1)^2 + F^2D_1^2(\Delta R_2)^2 \]
\[ + F^2D_1^2(\Delta R_5)^2 + F^2D_1^2(\Delta R_6)^2 \]
\[ = C_2 + C_{12} + C_{13} + (R_2^2 + R_3^2 + R_6^2)C_1 \]
\[ + (R_2^2 + R_3^2 + R_6^2)R_1^2C_3 + (R_2^2 + R_5^2 + R_6^2)R_1^2C_{23} \]
\[ + F^2D_2 + F^2D_{12} + F^2D_{13} + F^2(R_2^2 + R_5^2 + R_6^2)D_1 \]
\[ + F^2(R_2^2 + R_3^2 + R_6^2)R_1^2D_3 + F^2(R_2^2 + R_3^2 + R_6^2)R_1^2D_{23} \]
\[ + (R_2^2 + R_3^2 + R_6^2)C_3^2(\Delta R_1)^2 + C_1^2(\Delta R_3)^2 + C_1^2(\Delta R_5)^2 + C_1^2(\Delta R_6)^2 \]
\[ + F^2(R_2^2 + R_3^2 + R_6^2)D_3^2(\Delta R_1)^2 \]
\[ + F^2D_1^2(\Delta R_2)^2 + F^2D_1^2(\Delta R_5)^2 + F^2D_1^2(\Delta R_6)^2 \]
\[ = (\Delta C_{133})_{\text{stat}}^2 \]
\[ = C_2 + C_{12} + C_{13} + (R_2^2 + R_3^2 + R_6^2)C_1 \]
\[ + (R_2^2 + R_3^2 + R_6^2)R_1^2C_3 + (R_2^2 + R_5^2 + R_6^2)R_1^2C_{23} \]
\[ + F^2(D_2 + D_{12} + D_{13} + (R_2^2 + R_3^2 + R_6^2)D_1 \]
\[ + (R_2^2 + R_3^2 + R_6^2)R_1^2D_3 + (R_2^2 + R_3^2 + R_6^2)R_1^2D_{23} \]
\[ + (C_1^2 + F^2D_1^2)(\Delta R_2)^2 + (C_1^2 + F^2D_1^2)(\Delta R_5)^2 \]
\[ + (C_1^2 + F^2D_1^2)(\Delta R_6)^2 \]
\[ = (\Delta c_{131m})^2 = (\Delta C_{12})^2 + F^2(\Delta D_{12})^2 \]
\[ = (\Delta C_{12})^2 + R_3^2(\Delta C_1)^2 + R_5^2R_1^2(\Delta C_3)^2 \]
\[ + F^2(\Delta D_{12})^2 + F^2R_3^2(\Delta D_1)^2 + F^2R_5^2R_1^2(\Delta D_3)^2 \]
\[\begin{align*}
+ R_3^2 C_3^2 (\Delta R_1)^2 + C_1' (\Delta R_5)^2 + F^2 R_2^2 D_3' (\Delta R_1)^2 + F^2 D_1' (\Delta R_5)^2 \\
= C_{12} + R_2^2 C_1 + R_3^2 R_1^2 (C_3 + C_{23}) \\
+ F^2 D_{12} + F^2 R_2^2 D_1 + F^2 R_3^2 R_1^2 (D_3 + D_{23}) \\
+ R_3^2 C_3^2 (\Delta R_1)^2 + C_1' (\Delta R_5)^2 + F^2 R_2^2 D_3' (\Delta R_1)^2 + F^2 D_1' (\Delta R_5)^2 \\
(\Delta c_{131m})^2_{\text{stat}} = C_{12} + R_2^2 C_1 + R_3^2 R_1^2 (C_3 + C_{23}) \\
+ F^2 (D_{12} + R_2^2 D_1 + F^2 R_3^2 R_1^2 (D_3 + D_{23})) \\
(\Delta c_{131m})^2_{\text{syst}} = R_3^2 C_3^2 + F^2 R_2^2 D_3' (\Delta R_1)^2 + (C_1' + F^2 D_1') (\Delta R_5)^2 \\
(\Delta c_{131m})^2 = (\Delta c_{131m})^2_{\text{stat}} + (\Delta c_{131m})^2_{\text{syst}}
\end{align*}\]
5 Calculation of minimum detectable concentrations (MDCs)

The minimum detectable concentration (MDC) for a measurement can be estimated using the concentration equation, with the corrected number of counts \( c \) replaced by an estimate of the number of counts \( n_{LD} \) which constitutes the lower limit of detection. If \( \sigma_0 \) is the standard deviation of the background, i.e. the standard deviation of the number of counts in the ROI expected when no true signal is present, setting \( n_{LD} = 2.71 + 4.65 \cdot \sigma_0 \) (48) yields a limit such that an activity resulting on the average in this number of counts detected will in 95 % of all measurements result in a number of counts greater than the level attained by the background in 5 % of all measurements (see e.g. [3]). Thus, the MDC is given by

\[
MDC = \frac{2.71 + 4.65 \cdot \sigma_0}{\epsilon_{\beta', \beta} \cdot \beta' \cdot \lambda^2 \cdot F_C \cdot F_P \cdot F_A \cdot \lambda} \quad (49)
\]

where the remaining problem is how to correctly estimate \( \sigma_0 \).

If we consider the example of \(^{133}\text{Xe}\) (81 keV counts distribution), the formula for the corrected number of counts can be re-written in a slightly expanded form:

\[
e_{133}^{80} = C'_1 - F \cdot D'_1 = C_1 - R_1 \cdot C'_3 - F \cdot (D_1 - R_1 \cdot D'_3) \quad (50)
\]

The term \( C_1 \) contains the true signal as well as four main components of background and contamination:

- The ”signal” of \(^{214}\text{Pb}\) coincidences due to the decay of Rn introduced into the measurement cell together with the Xe sample.
- The ”memory” background of \(^{214}\text{Pb}\) coincidences due to the decay of Rn remaining in the cell from previous measurements.
- The ”memory” background of \(^{133}\text{Xe}\) coincidences due Xe gas remaining in the cell from previous measurements.
- An ambient background, assumed constant in time, from cosmic rays, \(^{40}\text{K}\) and other well-known sources of background.

The second term on the right-hand side of (50) is an estimate of the first two background sources, the Rn ”signal” and the Rn ”memory”, which both contribute to the count rate in the \(^{214}\text{Pb}\) ROI used to correct for Rn contamination in the \(^{133}\text{Xe}\) (81 keV) ROI through the ratio \( R_1 \). The third term, in parentheses, estimates the Xe ”memory” and the constant background, since the counts are extracted from the gas background measurement immediately preceding the sample measurement and corrected so that there is no net contribution from the Rn memory. There is a minor source of error in the fact that the decay correction factor \( F \) also operates on what is supposed to be constant background, but it turns out that below approximately 100 keV in the gamma spectrum, this part of the background is in any case very small. Also, for all Xe isotopes except \(^{135}\text{Xe}\), the decay correction part of \( F \) is quite close to one. In the case of \(^{135}\text{Xe}\), an extra correction term is needed as can be seen in the correction formulae in the previous sections.

It thus appears that all the important sources of background and contamination are accounted for by the last two terms. The variance of the null-signal background can then be estimated as

\[
\sigma_0^2 = (\Delta(R_1 \cdot C'_3))^2 + (\Delta(F \cdot D'_1))^2 + (\Delta(F \cdot R_1 \cdot D'_3))^2
= (C'_3 \Delta R_1)^2 + (R_1 \Delta C'_3)^2 + F^2 \cdot (\Delta D_1)^2 + F^2 \cdot (\Delta D'_3)^2 + (R_1 \Delta D'_3)^2
= R_1^2(C_3 + C_{23}) + F^2(D_1 + R_1^2(D_3 + D_{23})) + C'_3^2(\Delta R_1)^2 + F^2 \cdot D'_3(\Delta R_1)^2 \quad (51)
\]
or, if only statistical sources of uncertainty are included

\[ \sigma_{0,\text{stat}}^2 = R_1^2(\Delta C_1^2)^2 + F^2((\Delta D_1)^2 + R_1^2(\Delta D_3)^2) = R_1^2(C_3 + C_{23}) + F^2(D_1 + R_1^2(D_3 + D_{23})) \] (52)

For the other isotopes, the MDC is estimated in an analogous manner. The expressions necessary to assemble the formulae estimating \( \sigma_0 \) for any of the four relevant Xe isotopes (\( C_{n,s}, D_{n,s} \) and their variances) are all found in the previous two sections. In the case of the \( {}^{133}\text{Xe} \) concentration from the 30-keV region, explicitly

\[ c_{133}^{30} = C_2' - F \cdot D_2' = C_2 - C'_1 - R_2 \cdot C_1' - F \cdot (D_2 - D_1' - D'_{13} - R_2 \cdot D_1') \] (53)

which yields the following estimate for the zero-signal background variance:

\[ \sigma_0^2 = (\Delta C_1')^2 + (\Delta C_1)^2 + (\Delta (R_2 \cdot C_1'))^2 + F^2(\Delta D_1')^2 + (\Delta D_{13})^2 + (\Delta (R_2 \cdot D_1'))^2 \]

\[ = C_{12} + R_6^2C_1 + R_6^2R_1^2C_3 + R_6^2R_1^2C_{23} + R_6^2C_3^2(\Delta R_1)^2 + C_1^2(\Delta R_2)^2 \]

\[ + C_{13} + R_6^2C_1 + R_6^2R_1^2C_3 + R_6^2R_1^2C_{23} + R_6^2C_3^2(\Delta R_1)^2 + C_1^2(\Delta R_2)^2 \]

\[ + R_2^2(C_1 + R_1^2C_3 + R_1^2C_{23}) + R_2^2C_3^2(\Delta R_1)^2 + C_1^2(\Delta R_2)^2 \]

\[ + F^2D_2 \]

\[ + F^2(D_{12} + R_6^2D_1 + R_6^2R_1^2D_3 + R_6^2R_1^2D_{23} + R_6^2D_3^2(\Delta R_1)^2 + D_1^2(\Delta R_6)^2) \]

\[ + F^2(D_{13} + R_6^2D_1 + R_6^2R_1^2D_3 + R_6^2R_1^2D_{23} + R_6^2D_3^2(\Delta R_1)^2 + D_1^2(\Delta R_6)^2) \]

\[ + F^2(R_2^2(D_1 + R_1^2D_3 + R_1^2D_{23}) + R_2^2D_3^2(\Delta R_1)^2 + D_1^2(\Delta R_2)^2) \] (54)

Keeping only the statistical sources of uncertainty in this case yields

\[ \sigma_{0,\text{stat}}^2 = C_{12} + R_6^2C_1 + R_6^2R_1^2C_3 + R_6^2R_1^2C_{23} + C_{13} \]

\[ + R_6^2C_1 + R_6^2R_1^2C_3 + R_6^2R_1^2C_{23} + R_6^2(C_1 + R_1^2C_3 + R_1^2C_{23}) \]

\[ + F^2D_2 \]

\[ + F^2(D_{12} + R_6^2D_1 + R_6^2R_1^2D_3 + R_6^2R_1^2D_{23}) + D_{13} + R_6^2D_1 + R_6^2R_1^2D_3 + R_6^2R_1^2D_{23} \]

\[ + F^2(R_2^2(D_1 + R_1^2D_3 + R_1^2D_{23}) \] (55)
Figure 4. $\beta - \gamma$ spectrum from atmospheric sample containing 50 mBq/m$^3$ $^{133}$Xe (upper left) and corresponding empty-cell background (upper right). The lower panes show the gamma (left) and beta (right) projections, with the empty-cell background filled.

6 Example of the analysis procedure: 50 mBq/m$^3$ $^{133}$Xe atmospheric sample

The SAUNA air sample collected between 22:00 UTC December 27 2000 and 10:00 UTC December 28 2000 at the Bundesamt für Strahlenschutz Institute of Atmospheric Radioactivity (IAR) in Freiburg, Germany, contained 52 mBq/m$^3$ $^{133}$Xe. The left part of figure 4 shows the resulting $\beta - \gamma$ spectrum. The right part of the figure shows the background measurement done during the 12-hour period preceding the introduction of the air sample into the measurement cell. The sample of December 27–28 2000 will be used as an example of the analysis procedure described in the previous sections.

Tables 1 and 2 display the contents (raw numbers of counts) in the various ROIs involved in the extraction of the net numbers of counts $c_{^{133}}^{30}$, $c_{^{133}}^{80}$ in the sample from $^{133}$Xe decay, as well as the interference-corrected numbers of counts resulting from applying the formulae of Section 3. Table 1 shows the ROI contents and corrections for the sample measurement and Table 2 holds the same information for the background measurement immediately preceding it.

The information in tables 1 – 2 must then be inserted into the formulae of Section 4 to yield background-subtracted number of counts $c_{^{133}}^{30}$ and $c_{^{133}}^{81}$. The decay constant of $^{133}$Xe is $\lambda = 1.53 \cdot 10^{-6} \text{s}^{-1}$, the real times of the sample and background measurements were 10.9 hours and 11.0 hours, respectively, and the corresponding live times were 9.4 hours and 7.9 hours, respectively. With a time of 12.1 hours elapsed between the start of the background measurement and the start of the sample measurement, equation 27
<table>
<thead>
<tr>
<th>ROI</th>
<th>sample counts</th>
<th>interference-corrected</th>
<th>uncertainty σ</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{133}\text{Xe}$ 81 keV</td>
<td>1742</td>
<td>$1742 - 0 \times 27 = 1742$</td>
<td>43</td>
</tr>
<tr>
<td></td>
<td>$C_1$</td>
<td>$C_1 - R_1 \times C_3' = C_1'$</td>
<td></td>
</tr>
<tr>
<td>$^{133}\text{Xe}$ 30 keV</td>
<td>2221</td>
<td>$2221 - (-73.958) - 34.402 - 0.162 \times 1742 = 1978.352$</td>
<td>61</td>
</tr>
<tr>
<td></td>
<td>$C_2$</td>
<td>$C_2 - C_{12}' - C_{13}' - R_2 \times C_1' = C_2'$</td>
<td></td>
</tr>
<tr>
<td>$^{131m}\text{Xe}$</td>
<td>534</td>
<td>$534 - 0.349 \times 1742 = -73.958$</td>
<td>28</td>
</tr>
<tr>
<td></td>
<td>$C_{12}$</td>
<td>$C_{12} - R_5 \times C_1' = C_{12}'$</td>
<td></td>
</tr>
<tr>
<td>$^{133m}\text{Xe}$</td>
<td>503</td>
<td>$503 - 0.269 \times 1742 = 34.402$</td>
<td>26</td>
</tr>
<tr>
<td></td>
<td>$C_{13}$</td>
<td>$C_{13} - R_6 \times C_1' = C_{13}'$</td>
<td></td>
</tr>
<tr>
<td>$^{214}\text{Pb}$</td>
<td>108</td>
<td>$108 - 81 = 27$</td>
<td>14</td>
</tr>
<tr>
<td></td>
<td>$C_3$</td>
<td>$C_3 - C_{23} = C_3'$</td>
<td></td>
</tr>
<tr>
<td>$^{214}\text{Pb}$ bgr</td>
<td>81</td>
<td>$-4 = -$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$C_{23}$</td>
<td>$-4 = -$</td>
<td></td>
</tr>
</tbody>
</table>

Table 1. ROI contents from sample measurement yielding 52 mBq/m$^3$ $^{133}\text{Xe}$. The uncertainties given are the standard deviations $\sigma = \sqrt{\langle (\Delta C_n')^2 \rangle_{stat}}$ calculated using the expressions for the variances given in Section 3, including statistical sources of error only.

<table>
<thead>
<tr>
<th>ROI</th>
<th>background counts</th>
<th>interference-corrected</th>
<th>uncertainty σ</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{133}\text{Xe}$ 81 keV</td>
<td>28</td>
<td>$28 - 0 \times 10 = 28$</td>
<td>6</td>
</tr>
<tr>
<td></td>
<td>$D_1$</td>
<td>$D_1 - R_1 \times D_3' = D_1'$</td>
<td></td>
</tr>
<tr>
<td>$^{133}\text{Xe}$ 30 keV</td>
<td>32</td>
<td>$32 - (-0.772) - (-4.532) - 0.162 \times 28 = 32.768$</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>$D_2$</td>
<td>$D_2 - D'<em>{12} - D'</em>{13} - R_2 \times D_1' = D_2'$</td>
<td></td>
</tr>
<tr>
<td>$^{131m}\text{Xe}$</td>
<td>9</td>
<td>$9 - 0.349 \times 28 = -0.772$</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>$D_{12}$</td>
<td>$D_{12} - R_5 \times D_1' = D'_{12}$</td>
<td></td>
</tr>
<tr>
<td>$^{133m}\text{Xe}$</td>
<td>3</td>
<td>$3 - 0.269 \times 28 = -4.532$</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>$D_{13}$</td>
<td>$D_{13} - R_6 \times D_1' = D'_{13}$</td>
<td></td>
</tr>
<tr>
<td>$^{214}\text{Pb}$</td>
<td>94</td>
<td>$94 - 84 = 10$</td>
<td>14</td>
</tr>
<tr>
<td></td>
<td>$D_3$</td>
<td>$D_3 - D_{23} = D_3'$</td>
<td></td>
</tr>
<tr>
<td>$^{214}\text{Pb}$ bgr</td>
<td>84</td>
<td>$-4 = -$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$D_{23}$</td>
<td>$-4 = -$</td>
<td></td>
</tr>
</tbody>
</table>

Table 2. ROI contents from background measurement preceding the sample measurement yielding 52 mBq/m$^3$ $^{133}\text{Xe}$. The uncertainties given are the standard deviations $\sigma = \sqrt{\langle (\Delta D_n')^2 \rangle_{stat}}$ calculated using the expressions for the variances given in Section 3, including statistical sources of error only.
energy-integrated detection efficiency $\epsilon_{\beta\gamma_{81}}$ for 81 keV ROI | 0.5
branching ratio $\beta\gamma_{30}$ for 30 keV ROI | 0.4
$\beta\gamma_{81}$ for decay yielding 81 keV photons | 0.373
$\beta\gamma_{30}$ for decay yielding 30 keV X-rays | 0.489

$^{133}\text{Xe}$ decay constant $\lambda$ | $1.53 \cdot 10^{-6}$ s$^{-1}$
sampling time $t_{coll}$ | 12.0 h (43333 s)
sampling/decay integral $F_C$ | 0.0641
preparation time $t_{proc}$ | 6.8 h (24529 s)
presentation decay factor $F_P$ | 0.9632
counting time $t_{acq}$ | 10.9 h (39082 s)
counting integral $F_A$ | 0.0580
sampled air volume $V$ | 6.22 m$^3$

Table 3. System- and measurement specific parameters needed to calculate the atmospheric radioactivity concentration from the net number of counts (Equation 1).

yields the decay correction factor $F$ as follows (10 hours = 36000 seconds):

$$F = \frac{t_{\text{bgr}}^{\text{real}}}{t_{\text{bgr}}^{\text{live}}} \frac{t_{\text{live}}}{t_{\text{real}}} e^{-\lambda t_{\text{real}}} (1 - e^{-\lambda t_{\text{real}}})/(1 - e^{-\lambda t_{\text{bgr}}^{\text{real}}})$$

(56)

$$= \frac{39683.00 \text{ s} \ 33732.26 \text{ s}}{28565.21 \text{ s} \ 39082.00 \text{ s}} e^{-0.0000153 \text{ s}^{-1} \ 43459.00 \text{ s}} \frac{1 - e^{-0.0000153 \text{ s}^{-1} \ 39082.00 \text{ s}}}{1 - e^{-0.0000153 \text{ s}^{-1} \ 39683.00 \text{ s}}}$$

$$= 1.389 \times 0.863 \times 0.936 \times \frac{0.0580}{0.0589}$$

$$= 1.105$$

The decay-corrected background term can now be subtracted from the sample number of counts (equations 28 and 32):

$$c_{133}^{81} = C_1' - F \cdot D_1' = 1742 - 1.105 \cdot 28 = 1711 \pm 43$$

(57)

$$c_{133}^{30} = C_2' - F \cdot D_2' = 1978.352 - 1.105 \cdot 32.768 = 1942 \pm 61$$

where the statistical uncertainties have been calculated using equations 30 and 34, using $R_2 = 0.162$, $R_3 = 0.349$, $R_6 = 0.269$ and $R_1 = 0$.

These numbers can be compared to the raw numbers of counts from tables 1 – 2: 1742 and 2221 counts for the 81 keV and the 30 keV ROI, respectively. The conclusion is encouraging: for a sizeable Xenon plume coming under circumstances with normal background, the various interference and background effects and the manipulations necessary to account for them in the analysis clearly influence the end result in a comparatively minor way.

The numbers (57) must also be corrected for the counting dead-time, in this case the correction factor is $t_{\text{real}}/t_{\text{live}} = 39082.00 \text{ s} / 33732.26 \text{ s} = 1.159$, and the final numbers of counts become

$$c_{133}^{81} = 1981 \pm 50$$

(58)

$$c_{133}^{30} = 2250 \pm 71$$

Equation 1 can be used to calculate the atmospheric concentrations from the net numbers of counts extracted above. The parameters needed aside from the numbers of counts are listed in Table 3.
equation yields the following atmospheric concentration of $^{133}$Xe, extracted from the two ROIs:

$$
C_{A_{81}} = 1981 \cdot \frac{1}{0.5 \cdot 0.373 \cdot 0.0641 \cdot 0.9632 \cdot 0.0580} \cdot 6.22 \cdot (1.53 \cdot 10^{-6})^2 \cdot 43333 \text{ mBq/m}^3 \\
= 1981 \cdot 5.362 \cdot 6.537 \cdot 10^{-10} \cdot 6966.72 \text{ mBq/m}^3 \\
= 1981 \cdot 2.442 \cdot 10^{-5} \\
= 48 \pm 2 \text{ mBq/m}^3 \\

C_{A_{30}} = 2252 \cdot \frac{1}{0.4 \cdot 0.489 \cdot 0.0641 \cdot 0.9632 \cdot 0.0580} \cdot 6.22 \cdot (1.53 \cdot 10^{-6})^2 \cdot 43333 \text{ mBq/m}^3 \\
= 2252 \cdot 5.112 \cdot 6.537 \cdot 10^{-10} \cdot 6966.72 \text{ mBq/m}^3 \\
= 2252 \cdot 2.328 \cdot 10^{-5} \\
= 52 \pm 2 \text{ mBq/m}^3 \\
$$

where the statistical uncertainties have been calculated simply by propagating the uncertainties in $C_{30,81}$, i.e. assuming no (statistical) uncertainties in the other parameters entering the concentration equation.

To estimate the Minimum Detectable Concentrations (MDCs) (equation 49) we use the number of counts $n_{LD}^{30,80} = 2.71 + 4.65 \cdot \sigma_0^{30,80}$ corresponding to the relevant lower limit of detection (see section 5) instead of the corrected number of counts in an actual spectrum (e.g. $C_{133}$) in the expressions 59. By equation 52, the statistical variance in the null-signal background for the 80 keV ROI can be estimated by

$$
(\sigma_0^{80})^2 = R_1^2(C_5 + C_{23}) + F^2(D_1 + R_2^2(D_3 + D_{23})) \\
= 0 \cdot (108 + 81) + (1.105)^2 \cdot (28 + 0 \cdot (94 + 84)) \\
= 34.189 \\
$$

and the slightly more complicated expression 55 yields for the case of the 30 keV ROI (using $R_2 = 0.162$, $R_5 = 0.349$, $R_6 = 0.269$ and omitting terms containing $R_1 = 0$):

$$
(\sigma_0^{30})^2 = C_{12} + C_{13} + C_1(R_2^2 + R_5^2 + R_6^2) + \\
F^2(D_2 + D_{12} + D_{13} + D_1(R_2^2 + R_5^2 + R_6^2)) \\
= 534 + 503 + 1742 \cdot ((0.162)^2 + (0.349)^2 + (0.269)^2) + \\
(1.105)^2 \cdot (32 + 9 + 3 + 28 \cdot ((0.162)^2 + (0.349)^2 + (0.269)^2)) \\
= 1482.208 \\
$$

Thus, $n_{LD}^{80} = 2.71 + 4.65 \cdot \sqrt{34.189} = 29.9$ and $n_{LD}^{30} = 2.71 + 4.65 \cdot \sqrt{1482.208} = 181.7$ and the MDCs become

$$
MDC_{81} = 29.9 \cdot 2.42 \cdot 10^{-5} \text{ mBq/m}^3 = 0.7 \text{ mBq/m}^3 \\
MDC_{30} = 181.7 \cdot 2.42 \cdot 10^{-5} \text{ mBq/m}^3 = 4.2 \text{ mBq/m}^3 \\
$$
References


[2] The International Noble Gas Experiment, to be published

Xenon air activity concentration analysis from coincidence data

The report contains a short summary of the formulae, definitions, quantities and methods used to calculate concentrations and minimum detectable concentrations of atmospheric radioxenon samples collected by the Swedish automatic Xenon sampling system (SAUNA).
Analys av xenonaktivitetskoncentrationer i luft ur koincidensdata

Rapporten innehåller en kort sammanfattning av de formler, definitioner, storheter och metoder som används för att beräkna koncentrationer och detektionsgränser för atmosfäriska xenonprov från det svenska automatiska xenoninsamlingssystemet (SAUNA).