Noisy radioactivity data analysis using parametric Poisson models

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Résumé. En métrologie, l'utilisation des concepts de seuil de décision et de limite de détection pose souvent de nombreux problèmes aux métrologues des laboratoires d'analyse de la radioactivité. Il est habituel de censurer les données lorsqu'il devient difficile de discerner la présence ou l'absence de l'activité, en raison du bruit dans les données de mesure. Cela signifie que si les résultats des mesures sont non significatifs (inférieurs au seuil de décision), l'analyse indique simplement que la valeur réelle (signal) de la radioactivité est inférieure à une certaine limite appelée limite de détection. Ces problèmes sont souvent liés à une mauvaise compréhension des formules de seuil de décision. En outre, la manière de générer un seuil de décision approprié et justifié n'est pas claire dans [ISO 11929(2020)]. Dans le présent document de recherche, nous élaborons une méthode statistique de détermination du seuil de décision le plus puissant. Ensuite, des méthodes d'approches statistiques sont adoptées pour estimer l'espérance et la variance de la radioactivité. L'efficacité et la faisabilité de ces approches sont corroborées par des applications sur des ensembles de données réelles de l'IRSN.

Mots-clés. Seuil de décision, Test du rapport de vraisemblance, Radioactivité, Données de bruit, Paramètres de nuisance.

Abstract. In metrology, the use of the concepts of decision limit and detection limit often poses many problems for metrologists in radioactivity analysis laboratories. It is usual to censor data when it becomes difficult to discern the presence or absence of activity, due to noise in the measurement data. This means that if the measurement results are insignificant (below the decision threshold), the analysis simply indicates that the actual value (signal) of radioactivity is below a certain limit called the detection limit. These problems are often due to a misunderstanding of the decision threshold formulas. In addition, it is not clear how to generate an appropriate and justified decision threshold in [ISO 11929(2020)]. In this research paper, we develop a statistical method for determining the most powerful decision threshold. Next, methods of statistical approaches are adopted to estimate the expectation and the variance of radioactivity. The effectiveness and feasibility of these approaches are corroborated by applications on IRSN data sets.

Keywords. Decision threshold, Likelihood ratio test, Predictive density, Radioactivity, Noise data, Nuisance parameters.

1 Introduction

Radioactivity measurement relies on characteristic limits (decision thresholds and detection limits). This commonly indicates that measurement results below the decision threshold (DT) are left censored and are considered as useless. Due to ever-lower levels of environmental activity, the number of radiological analyses for which metrologists are unable to provide significant results is increasing. Current standards steadily propose uniquely DT formulas without much justification. For instance, the DT, as defined by the standard [ISO 11929(2020)], is calculated as

$$y^\star = k_{1-\alpha} w u(n_n = 0),$$

where $k_{1-\alpha}$ denotes the quantile of the probability density of the measurement results y with a null parameter that exceeds the DT with the probability α, w refers to the value of a conversion factor and $u(n_n = 0)$ stands for the null measurement uncertainty of the net indication n_n. It is unclear in the standard [ISO 11929(2020)] how the DT is calculated and under which statistical test it is obtained. Currently, there is no clear method for the determination of DT of the radioactivity in literature. There is equally an imperious need in metrology for a clear method to determine an optimal DT due to ever-lower levels of environmental activity. The Institute for Radiological Protection and Nuclear Safety (IRSN) is thus highly interested in improving DT for radioactivity analyses. The aim of this short communication is to present an efficient statistical method capable of providing an optimal DT and study the statistical properties of the radioactivity.

2 Model and notations

In the field of metrology, in order to quantify the activity contained in various samples, and before the actual measurement of a specific sample is undertaken a calibration procedure is done. In measurement technology and metrology, calibration is the comparison of measurement values delivered by a device under test with those of a calibration standard of known accuracy. The formal definition of calibration by the International Bureau of Weights and Measures (BIPM) is the following: "Operation that, under specified conditions, in a first step, establishes a relation between the quantity values with measurement uncertainties provided by measurement standards and corresponding indications with associated measurement uncertainties (of the calibrated instrument or secondary standard) and, in a second step, uses this information to establish a relation for obtaining a measurement result from an indication."

For the measurements considered here, the calibration process leads us to the following counting procedure: for each sample A, two measurements of equal duration are taken: namely a blank measurement C_{blank} and a gross measurement C_{gross}, where C_{blank} is as similar as possible to C_{gross} but in condition that ensures the absence of the signal S. According to [ISO 11929(2020)], the Noisy radioactivity data are defined as follows

$$A = Y\left(C_{gross} - C_{blank}\right),$$

where

- $C_{blank} := C_{bl}$: is a random variable that models the noise of the counter: $C_{bl} \sim \mathcal{P}(\lambda)$.
- $C_{gross} := C_{gr} = S + B'$: is a random variable that is equal to the sum of two independent random variables, measured by counter, with S being the random variable of interest that models the signal and B' being a random variable that models the noise of the measurement of C_{gr}. $S \sim \mathcal{P}(\theta)$, $B' \sim \mathcal{P}(\lambda')$, then $C_{gr} \sim \mathcal{P}(\mu := \theta + \lambda')$.
- Y: is a Gaussian random variable that models the conventional calibration factor used in metrology, which maps the linear relation between the measured activity from a reference measurement system and the the measured activity coming from the counter and depends on the time duration and the sample volume.

In section 4, we consider the measurements of radionuclides in water samples as an example of this model of data. We will use the following notations:

- $X = C_{gr} - C_{bl}$: is the net count.
- DT: is the decision threshold: critical level of the hypothesis statistical test of θ: $H_0 : \theta = 0$ (no signal) / $H_1 : \theta > 0$ (signal).
- DL: is the detection limit: the largest true value θ that would have a non-negligible probability of being considered insignificant.
- $R := X\mathbb{1}_{\{X \geq DT\}} + DL\mathbb{1}_{\{X < DT\}}$.
- $D := YR$: models the radioactivity used by IRSN which has been so far based on [ISO 11929(2020)]. There is a loss of information because non significant results are left censored.
- n: is the size of data.
- ϕ: is the Gaussian distribution of $\mathcal{N}(0, 1)$.

Let $(A_1, \ldots, A_n)$ and $(D_1, \ldots, D_n)$ be two samples of independent identically distributed (i.i.d.) of random variables defined as follows:

$$\left(A_i = Y_i X_i = \frac{f_i}{\epsilon_i t_i V_i}(C_{gr,i} - C_{bl,i})\right)_{1 \leq i \leq n},$$

$$vs$$

$$\left(D_i = Y_i R_i = \frac{f_i}{\epsilon_i t_i}\left(X_i \mathbb{1}_{\{X_i > DT_i\}} + DL_i \mathbb{1}_{\{X_i \leq DT_i\}}\right)\right)_{1 \leq i \leq n},$$

where $X_i = \frac{1}{N_i}\sum_{k=1}^{N_i} X_i^k$, DT_i, DL_i are constructed by $X_i^1, \ldots X_i^{N_i}$ and N_i is the number of repetition of the measurements of the observed point data X_i. Noting that Y_i and $C_{bl,i}$

are constants for some i, all the parameters θ, λ, λ', μ are unknown and θ is the interest parameter. Throughout this research work, the following assumption will be considered:

$$\mathcal{A} : \lambda = \lambda'.$$

This assumption is usually justified by the fact that metrologists put a great emphasis on ensuring that the measurements are undertaken in very similar physical conditions (temperature, background radioactivity, apparatus, etc).

3 Main results

Our first result is the following proposition which gives the optimal DT_i for $i = 1 \ldots n$. A DT_i is constructed based on the repeated measurements $\left(C^j_{bl,i}, C^j_{gr,i}\right)_{1 \leq j \leq N_i}$ and the following pair of hypotheses test

$$H_0 : \theta = 0 \text{ vs } H_1 : \theta > 0,$$

with a fixed risk α.

Proposition 1 (DT of θ) *Under the hypothesis test $H_0 - H_1$ and the assumption $\mathcal{A}$, an optimal DT of the parameter θ for $i = 1 \ldots n$, denoted by DT_i, is generated by the resolution of the following equation:*

$$\alpha = I_{1/2}(DT_i + 1, \overline{C_{bl,i}} + 1),$$

where I is the regularized beta incomplete function. An approximation of DT_i is determined by

$$DT_i = \frac{k^2_{1-\alpha} + \sqrt{k^4_{1-\alpha} + 8k^2_{1-\alpha}\overline{C_{bl,i}}}}{2},$$

where $k_{1-\alpha} = \phi^{-1}(1 - \alpha)$.

The proof of Proposition 1 relies upon the predictive likelihood ratio test devoted to detect the presence of the signal. Noting that this test is uniformly the most powerful based on Neyman-Pearson Lemma. The following proposition specifies a decision threshold DT_i for $i = 1 \ldots n$ in the case where C_{bl} is large. We consider that C_{bl} is large when $C_{bl} > 10$.

Proposition 2 (DT when C_{bl} is large) *Consider that C_{bl} is large. Under the hypothesis test $H_0 - H_1$ and the assumption $\mathcal{A}$, an optimal DT of the parameter θ for $i = 1 \ldots n$, is obtained by the resolution of the following equation:*

$$\alpha = 1 - \frac{\Gamma(\lfloor DT_i + 1 \rfloor, 2\overline{C_{bl,i}})}{\lfloor DT_i \rfloor},$$

where $\Gamma(.,.)$ is the upper incomplete gamma function and $\lfloor . \rfloor$ is the floor function.

We equally find DT_i based on numerical Newton Raphson method in order to solve the previous equation using package *rootSolve* with the *R* software or by an approximation of the incomplete beta function. A confidence interval of the signal S is of the form: $\left[x_i - DT_i\frac{\sqrt{X_i}}{N_i}, x_i + DT_i\frac{\sqrt{X_i}}{N_i}\right]$. Finally, the following proposition provides global estimators of the expectation and the variance of the radioactivity.

Proposition 3 (Estimation of the radioactivity) *If X and Y are independent, then the considered estimators of the expectation and the variance are indicated by $\mu_{A,n}$ and $\sigma^2_{A,n}$ as follows*

$$\mu_{A,n} = \overline{X} \times \overline{Y},$$

$$\text{and } \sigma^2_{A,n} = \overline{Y}^2\sigma^2_{X,n} + \overline{X}^2\sigma^2_{Y,n},$$

$$\text{where } \overline{X} = \frac{1}{n}\sum_{i=1}^{n} X_i,\ \overline{Y} = \frac{1}{n}\sum_{i=1}^{n} Y_i,\ \sigma^2_{X,n} = \frac{1}{n-1}\sum_{i=1}^{n}(X_i - \overline{X})^2 \text{ and } \sigma^2_{Y,n} = \frac{1}{n-1}\sum_{i=1}^{n}(Y_i - \overline{Y})^2.$$

The proof of Proposition 3 is based on the bi-variate first order Taylor expansion of the function $g(X, Y) := XY$ at the point $a := (\mu_X, \mu_Y)$, where μ_X and μ_Y are the expectation of x and Y respectively.

4 Numerical studies

In our research work, in order to illustrate the proposed theoretical results, we used a data set of measurements of radionuclides released into water. In particular, we considered the data set consisting of $n = 360$ observations within the period $2019 - 2022$ of the following variables

- S: the measurement of tritium in environment (mainly rivers in France) by liquid scintillation counting by Bq/L.
- C_{bl}: pure water (deep water).
- $C_{gr} = S + B'$, where $B' \sim C_{bl}$.
- Y: calibration factor.

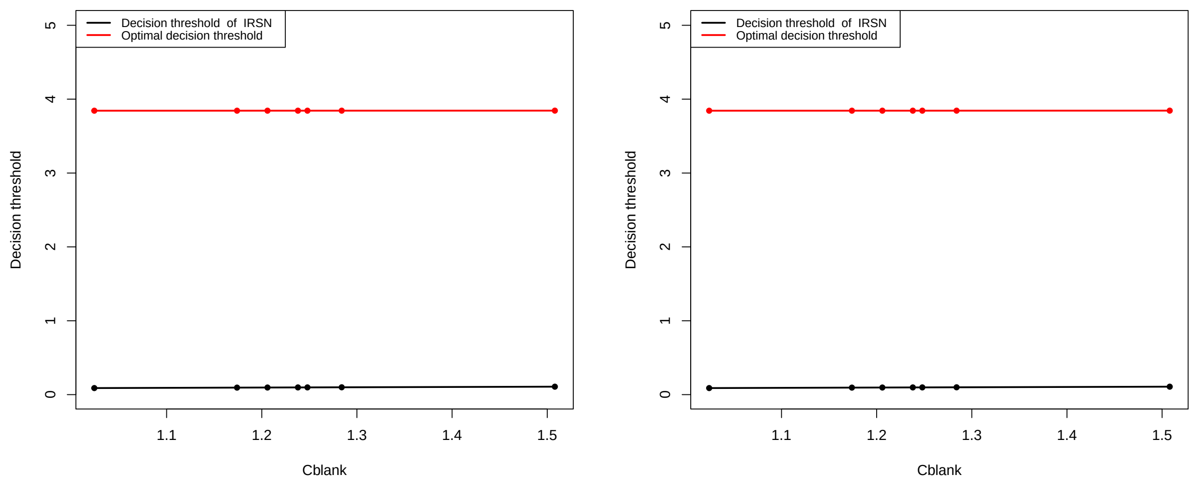

Figure 1: Results for DT of the count part for the radioactivity data in 2019 (left) and 2020 (right)

	Result of IRSN	Proposed method
Activity	6.08	6.08
DT	0.735	3.84
Confidence interval	$[6.08 \pm 0.94] = [5.13, 7.02]$	$[6.08 \pm 1.89] = [4.18, 7.97]$
Conclusion	$A > DT$	$A > DT$

Table 1: Example 1

	Result of IRSN	Proposed method
Activity	1.08	1.08
DT	0.737	3.32
Confidence interval	$[1.08 \pm 1.38] = [-0.30, 2.38]$	$[1.08 \pm 1.22] = [-0.14, 2.30]$
Conclusion	$A > DT$	$A < DT$

Table 2: Example 2

Figures 1 and 2 demonstrates that the proposed DT is higher than the DT defined in [ISO 11929(2020)], which implies that certain radioactivity values are declared significant with the standard when in fact they are not (see for instance Tables 1 and 2). With reference to Tables 1 and 2, we infer that the confidence interval of [ISO 11929(2020)] converge to the proposed confidence interval which is based on the proposed DT. Figure 3 shows that ratio of theoretical false positives and the observed false positives $\alpha_{thoeretical}/\alpha_{observed}$ is closer to 1 especially when C_{blank} is large, which implies the proposed decision threshold is optimal, as it is theoretically expected.

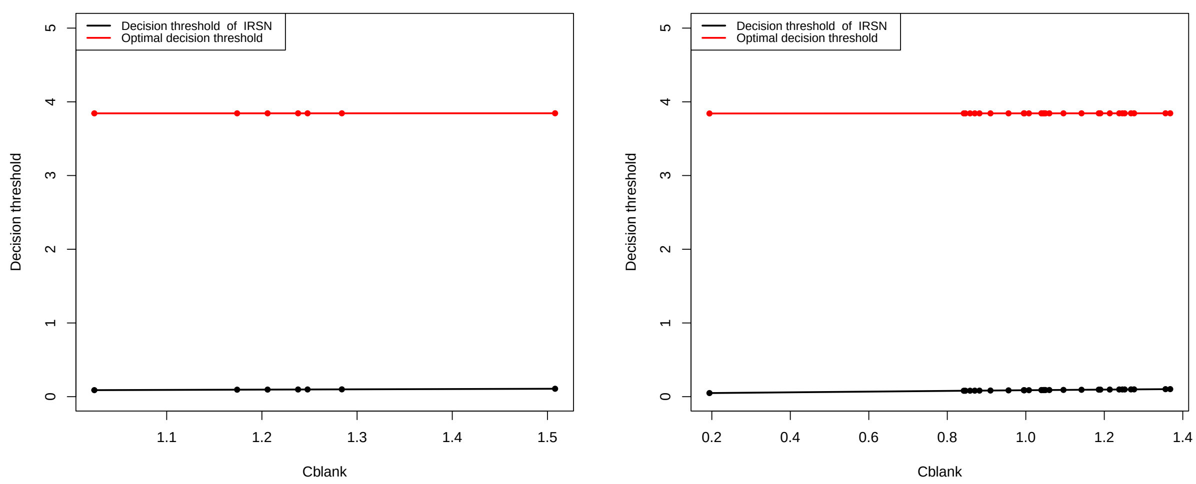

Figure 2: Results for DT of the count part for the radioactivity data in 2021 (left) and 2022 (right)

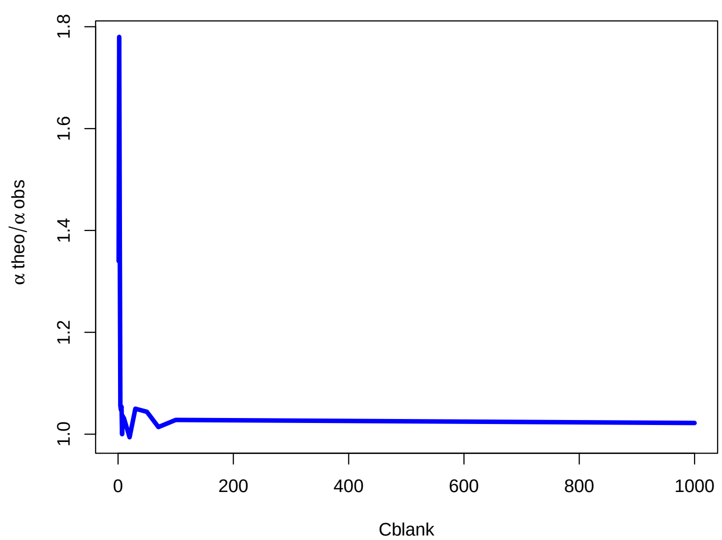

Figure 3: The ratio of theoretical false positives and the observed false positives $\dfrac{\alpha_{theo}}{\alpha_{obs}}$.

5 Conclusion

In this research paper, we proposed an optimal decision threshold based on Neyman-Pearson Lemma in order to detect the signal of the radioactivity. Theoretical results related to the model of the radioactivity; in particular to the expectation, the variance and the confidence interval of the activity signal, are presented. The application of our method was illustrated with the study on real datasets of tritium in water. In this respect, we would assert that this synthesis can be regarded as a preliminary study for further investigations on the decision threshold of the activity signal. Indeed, our work provides a theoretical foundation of the optimal decision theshold in the case of the alpha/beta/gamma spectrometry.

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