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## Noisy radioactivity data analysis using parametric Poisson models

Guillaume MANIFICAT<sup>1</sup><sup>\*</sup>, Salima HELALI<sup>1</sup><sup>†</sup>, Kévin GALLIEZ, Miriam BASSO, Maxime MORIN, Raja BEN HAJRIA

<sup>1</sup> Institute for Radiation Protection and Nuclear Safety (IRSN) Paris, France

#### Abstract

In metrology, the use of decision threshold and detection limit concepts often creates 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 the activity, due to noise in the measurement data. This implies that if the measurement results are non significant which are below critical value of the test statistic which is called the decision threshold (DT) in metrology. The analysis simply states that the true value (signal) of the radioactivity is below a certain limit called the detection limit (DL). These problems are frequently related to the incorrect understanding of the DT formulas or the wrong choice between several formulas whose numerical results are significantly different. Moreover, it is often unclear how to generate an appropriate and justified DT. In the current research paper, we elaborate a statistical method of DT determination, capable of providing DT with a high statistical power, using a smaller number of repeated measurements. The method is then applied to a real test case. Next, statistical approaches methods are adopted to estimate the density, the expectation and the variance of the radioactivity. Some of its asymptotic properties are also discussed. Efficiency and feasibility of these approaches are corroborated through applications on simulated real data sets.

**Key Words**: Decision threshold, Likelihood ratio test, Radioactivity, Noise data, Nuisance parameters.

## 1 Introduction

The central aim of radiological environmental monitoring is to contribute to the knowledge of the radiological state of the environment as well as spatial and temporal evolution, in order to reduce radiation exposure from contaminated soil, waste storage facilities or other contaminated infrastructure, groundwater or surface water. Radioactivity measurement rests on characteristic limits (decision thresholds and detection limits). The decision thresholds is the name given in physics to the critical value of the hypothesis test (Currie (1968); Lehmann et al (2005)). The detection limit is the lowest possible value of the parameter that will on average be declared significant. When an observation is below the decision threshold, the estimation is usually left censored and this observation is 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

<sup>\*</sup>e-mail adress: guillaume.manificat@irsn.fr

<sup>&</sup>lt;sup>†</sup>e-mail adress: helali.salima@gmail.com

and without taking profit of the statistical literature. 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 result y, for a null parameter that exceeds the DT with the probability  $\alpha$ , 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 metreology 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 basic aim of this contribution is to present an efficient statistical method of DT determination, capable of providing an optimal DT in a particular measurement of the radioactivity, using a smaller number of repeated measurements. The perfect method of DT determination would involve an infinite number of real-world measurements, but such a method is in practice impossible, and some realistic methods need to be used so as to approximate the infinite-measurement cases. As such, large numbers of repeated measurements will increase the statistical confidence in the determined DT, but there will always be a trade-off between the number of measurements and the time allocated to acquire the data, with excessive repetitions of measurements being unnecessarily expensive and slow. Therefore, determining the DT with minimal repeated measurements is desirable and requires significant statistical analysis.

The paper is organized as follows. In the next section, we recall the main concepts and definitions considered throughout this work. In Section 3, we identify the model and the notations. In section 4, we display the optimal threshold of the radioactivity signal. In section 5, we propose an estimation of the radioactivity signal. In section 6 we present a non parametric estimator of the radioactivity and parametric estimators of its expectation and variance. The asymptotic normality properties of these estimators and their associated confidence intervals are determined. Sections 7 and 8 are devoted to the application results obtained through simulations and real data analysis. Several pertinent concluding remarks and potential future perspectives are provided in Section 9. Section 10 exhibits the proofs of the theoretical results.

## 2 Background

#### 2.1 Statistical test, decision threshold and confidence interval:

A statistical test corresponds to a procedure for deciding whether a hypothesis about a quantitative feature of a sample is true or false. The decision threshold of the hypothesis test represents the fixed value of the radioactivity such that, when the measurement result is greater than it, it is decided that the radioactivity is present. Grounded on Lovric (2011), a  $100(1 - \alpha)\%$  confidence interval is an interval estimate around a population parameter  $\vartheta$  that, under repeated random samples of size N, is expected to include  $\vartheta$ 's true value  $100(1 - \alpha)\%$  of time. The detection limit is the largest true value that would have a non-negligible probability of being considered insignificant by the measurement. It can therefore give rise to observations equal to the decision threshold.

#### 2.2 Distribution estimation:

A set of repeated measurement results represents a sample of the radioactivity that contains all possible measurement results, with their distribution being a radioactivity distribution. A sum-

mary statistics of the sample (e.g. mean, maximum or variance) stands for an estimator of the radioactivity summary statistics. The distribution of the values of estimators calculated on repeated samples is a sampling distribution. Due to the central limit theorem (JCGM 100 (2008)), it is possible for the sampling distribution to be considered normal even if the radioactivity distribution is not. When using an estimator, there are three main properties that describe their behaviour: bias, consistency and uncertainty (see Dodge (2003)). Bias corresponds to the difference between an estimator's expected value and the true value of the parameter being estimated. Consistency is the tendency, as the number of sampled data points increases, for an estimator to converge to the true value of the parameter being estimated. A stated measurement uncertainty is a non-negative parameter characterising the dispersion of the quantity values being attributed to a measurement, based on the used information (see De Bièvre (2012)).

#### 2.3 Re-sampling and Bootstrapping:

When attempting to improve the confidence in a measurement, taking more measurements is the obvious first step. However, there are practical and economic limits on the possible number of repeated measurements. Therefore, when data have been collected, and it would still be desirable to reduce the error on the estimator, re-sampling the data can be a solution. Re-sampling is a method of statistical analysis that uses a fixed number of measurements in order to simulate what would be expected to happen, if more measurements had been taken. Bootstrapping is a common re-sampling technique that has been applied to settle metrology problems in literature (for e.g. see Hiller et al (2014) and Ciarlini and Regoliosi (2000)).

#### 2.4 Uncertainty

Consider a measurement system used to quantify the activity contained in various samples. The counting procedure is as follows: two measurements of two samples are taken: a 'blank' measurement (no radioactivity) and a 'gross' (possibility of radioctivity). Departing from the value of the net count: **net** = **gross** – **blank**, we calculate the measured activity by: **A** = **net**/**c**, where *c* is a calibration factor. As for any measurement, it is necessary to estimate the uncertainty of the activity based on the measured activity *A*. If we assume that nuclear counting has a Poisson behaviour, we can write the uncertainties of gross and blank as  $\mu_{gross} = \sqrt{gross}$  and  $\mu_{blank} = \sqrt{blank}$ , respectively. If **gross** and **blank** are two independent variables, then the uncertainly of the net count is provided by  $\mu_{net} = \sqrt{\mu_{blank}^2 + \mu_{gross}^2}$ . If we neglect the uncertainty of the calibration factor *c*, then the final uncertainty on the measured activity is equal to:  $\mu_A = \mu_{net}/c$ . The relative uncertainty on the activity is equal to  $\frac{\mu_A}{A}$ .

## **3** Model and notations

To report the method devised for DT determination and the proposed estimators of radioactivity, we need first to clarify a number of notations and assumptions considered throughout this work. These concepts are identified in this section. For each given point data of radioactivity 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 ensures the absence of the signal S. 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 calibration factor (measurement error).

Throughout this paper, we will equally use the following notations:

- $X := C_{gr} C_{bl}$ : is the net count.
- *DT*: is the decision threshold based on the hypothesis test of  $\theta$ :  $H_0: \theta = 0$  (no signal) /  $H_1: \theta > 0$ .
- *DL*: is the detection limit.
- $R := X \mathbb{1}_{\{X \ge DT\}} + DL \mathbb{1}_{\{X < DT\}}.$
- 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 have been left-censored.
- n: is the size of data.
- $\phi$ : 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 i.i.d. of random variables defined as follows:

$$(A_i = Y_i X_i = Y_i (C_{gr,i} - C_{bl,i}))_{1 \le i \le n},$$

$$vs$$

$$\left( D_i = Y_i R_i = Y_i \left( X_i \mathbb{1}_{\{X_i > DT_i\}} + DL_i \mathbb{1}_{\{X_i \le DT_i\}} \right) \right)_{1 \le i \le n},$$

where  $X_i = \frac{1}{N_i} \sum_{k=1}^{N_i} X_i^k$ ,  $DT_i$ ,  $DL_i$  are constructed by  $X_i^1, \dots, X_i^{N_i}$  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  $\theta$ ,  $\lambda$ ,  $\lambda'$ ,  $\mu$  are unknown and  $\theta$  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).

## 4 A method for the determination of a DT of $\theta$

The main target of this section is to provide an optimal DT of the parameter  $\theta$  for each point data  $X_i$  for  $i = 1 \dots n$  based on the likelihood ratio test, in order to make a decision about the presence of the signal. Let us consider the following data for  $i = 1 \dots n$ :

- Let  $C_{bl,i}^1, \ldots C_{bl,i}^{N_i}$  be an  $N_i$  i.i.d. sample drawn from the random variable  $C_{bl,i} \sim \mathcal{P}(\lambda_i)$ .
- Let  $C_{gr,i}^1, \ldots C_{gr,i}^{N_i}$  be an  $N_i$  i.i.d. sample drawn from the random variable  $C_{gr,i} \sim \mathcal{P}(\mu_i = \theta_i + \lambda_i)$ .

It is assumed that  $N_i \leq 5$  is insufficient to the asymptotic normality of an empirical estimator of  $\theta$ . In this section, in order to check the presence of the signal, we need to test the following pair of hypotheses

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

with a fixed risk  $\alpha$ . The following proposition provides the optimal  $DT_i$  for  $i = 1 \dots n$ . For a point data  $X_i$ , a  $DT_i$  is constructed based on the repeated measurements  $\left(C_{bl,i}^j, C_{gr,i}^j\right)_{1 \le j \le N_i}$ .

**Proposition 1** (**DT of**  $\theta$ ). Under the hypothesis test  $H_0 - H_1$  and the assumption  $\mathcal{A}$ , an optimal DT of the parameter  $\theta$  for  $i = 1 \dots n$ , 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

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

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

The proof of Proposition 1 relies upon the likelihood ratio test devoted to detect the presence of the signal in context, where the assumption  $\mathcal{A}$  is held. More details of this proof are supplied in section 10. The following proposition specifies a decision threshold  $DT_i$  for  $i = 1 \dots n$  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  $\theta$  for  $i = 1 \dots n$ , is obtained by the resolution of the following equation:

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

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

**Remark 1.** A confidence interval of the signal S is of the form:  $\left[x_i - DT_i \frac{\sqrt{X_i}}{N_i}, x_i + DT_i \frac{\sqrt{X_i}}{N_i}\right]$ .

## 5 Estimation of $\theta$ based on the count term X

Let us consider the following data

- $C_{gr,1} \dots, C_{gr,n} n$  iid sample drawn from the random variable  $C_{gr} \sim \mathcal{P}(\mu = \lambda + \theta)$ .
- $C_{bl,1}\ldots,C_{bl,n}$  n iid sample drawn from the random variable  $C_{bl} \sim \mathcal{P}(\lambda)$ .

**Proposition 3** (Estimator of  $\theta$ ). If  $C_{bl}$  and  $C_{gr}$  are large, an estimation of the parameter  $\theta$  is expressed in terms of

$$\theta_n = \frac{\sum_{i=1}^n C_{gr,i} - C_{bl,i}}{n}.$$

Based on the central limit theorem, the estimator  $\theta_n$  is asymptotically unbiased with a variance going to zero when  $n \to \infty$ .

### 6 Estimation of the radioactivity

Consider that A = XY, where  $Y \sim \mathcal{N}(a, b^2)$ . Noting that, in the count term  $X = C_{gr} - C_{bl}$ , we are basically interested in the law of  $C_{gr} = c_{gr} |\{C_{bl} = c_{bl}\}$ . Since X and Y are independent, then

$$\mathbb{E}(A) = \mathbb{E}(X)\mathbb{E}(Y) = \theta a.$$

 $\mathbb{VAR}(A) = \mathbb{VAR}(X)\mathbb{VAR}(Y) + \mathbb{VAR}(X)\mathbb{E}(Y)^2 + \mathbb{VAR}(Y)\mathbb{E}(X)^2 = b^2(\theta + 2\lambda) + a^2(\theta + 2\lambda) + b^2\theta^2.$ 

The following proposition provides estimators of the density of A;  $f_A$ ,  $\mathbb{E}(A)$  and  $\mathbb{VAR}(A)$  based on the iid data  $C_{gr,1}, \ldots, C_{gr,n}$  and  $C_{bl,1}, \ldots, C_{bl,n}$ .

<u>Non parametric estimation of  $f_A$ </u>: Let z be the observation of A = XY. We can consider the kernel non parametric estimator of the density  $f_A(z)$  defined by Rosenblatt (1956) as follows:

$$\widehat{f}_{A,n}(z) = \frac{1}{nh_n} \sum_{p=1}^n K\left(\frac{z-Z_p}{h_n}\right), \forall x \in \mathbb{R},$$

where  $(h_n)$  is a sequence of positive real numbers that goes to zero dependent, which is on the size n (smoothing parameter) called the bandwidth and K is the kernel function.

**Proposition 4** (Estimation of the radioactivity). The considered estimators of the expectation  $\mathbb{E}(A) := \mu_A$  and the variance  $\mathbb{VAR}(A) := \sigma_A^2$  are indicated by  $\mu_{A,n}$  and  $\sigma_{A,n}^2$  as follows

$$\mu_{A,n} = X \times Y,$$
  
and  $\sigma_{A,n}^2 = \overline{Y}^2 \sigma_{X,n}^2 + \overline{X}^2 \sigma_{Y,n}^2,$   
where  $\overline{X} = \frac{1}{n} \sum_{i=1}^n X_i, \, \overline{Y} = \frac{1}{n} \sum_{i=1}^n Y_i, \, \sigma_{X,n}^2 = \frac{1}{n-1} \sum_{i=1}^n (X_i - \overline{X}) \text{ and } \sigma_{Y,n}^2 = \frac{1}{n-1} \sum_{i=1}^n (Y_i - \overline{Y}).$ 

These approaches are asymptotically unbiased with a variance that goes to zero when  $n \to \infty$ . The following proposition demonstrates the asymptotic normality of the previous estimators. It is worth noting here that the mean estimator  $\mu_{A,n}$  is a function of the mean estimator of  $\mathbb{E}(X)$  and the one of  $\mathbb{E}(Y)$ . Likewise, the uncertainty estimator  $\sigma_{A,n}^2$  is a function of the uncertainty estimator of  $\sigma_X^2$  and the one of  $\sigma_Y^2$ , which is similar to the uncertainty approach in BIPM et al (2008) in Equation (10). The following proposition provides the asymptotic normality of the estimators in Proposition 4 and the associated confidence intervals. A confidence interval is constructed based on the repeated measurements  $(X_i, Y_i)_{1 \le i \le n}$ .

**Proposition 5** (Asymptotic normality and confidences intervals of the radioactivity:). The asymptotic normality of the estimators  $f_{A,n}(z)$  and  $\mu_{A,n}$  are expressed by

$$\sqrt{nh_n}(\widehat{f}_{A,n}(z) - f_A(z)) \xrightarrow[n \to +\infty]{\mathcal{L}} \mathcal{N}\left(0, f_A(z) \int K(u) du\right) \text{ and } \sqrt{n} \left(\mu_{A,n} - \mu_A\right) \xrightarrow[n \to +\infty]{\mathcal{L}} \mathcal{N}\left(0, \sigma^2\right),$$

where  $\sigma^2 = \mu_X^2 \sigma_X^2 + \mu_Y^2 \sigma_Y^2$ . Then the confidence intervals of  $f_A(z)$  and  $\mu_A$  are provided by

$$I_{f_A} = \left(\widehat{f}_{A,n}(z) \pm \frac{z_{1-\frac{\delta}{2}}\sigma_{f,n}(z)}{\sqrt{n}}\right) \text{ and } I_{\mu_A} = \left(\mu_{A,n} \pm \frac{z_{1-\frac{\delta}{2}}\sigma_n}{\sqrt{n}}\right),$$

where  $z_{1-\frac{\delta}{2}}$  is the normal  $\left(1-\frac{\delta}{2}\right)$  quantile,  $\sigma_{f,n}(x) = \sqrt{f_{n,A}(z) \int K(u) du}$  and  $\sigma_n = \sqrt{\mu_X^2 \sigma_{Y,n}^2 + \mu_Y^2 \sigma_{X,n}^2}$ .

## 7 Simulations

In this section, we investigate the performance of the theoretical results. In the simulation study, three sample sizes are considered, n = 50, n = 100, and n = 200, as well as the following models:

a) 
$$S \sim \mathcal{P}(n,2), B \sim \mathcal{P}(n,50), C_{blank} \sim \mathcal{P}(n,50)$$
 and  $N = 5$ .

b) 
$$S \sim \mathcal{P}(n, 15), B \sim \mathcal{P}(n, 10), C_{blank} \sim \mathcal{P}(n, 10)$$
 and  $N = 5$ .

c)  $S \sim \mathcal{P}(n, 15), B \sim \mathcal{P}(n, 9), C_{blank} \sim \mathcal{P}(n, 10)$  and N = 5.

#### 7.1 Comparison between the estimations of $\theta$ using X and R

In this subsection of the simulation study, three proposed estimators of  $\theta$  are considered, with reference to Proposition 3. They are defined as follows:

- Based on the sample  $X_1, \ldots, X_n$ , we consider  $\tilde{\theta}_n = \frac{1}{n} \sum_{i=1}^n X_i$ .
- Based on the sample  $R_{1,irsn}, \ldots, R_{n,irsn}$ , we consider  $\overline{\theta_n} = \frac{1}{n} \sum_{i=1}^n R_{i,irsn}$ , where for  $i = 1 \ldots n$ ,

$$R_{i,irsn} = X_i \mathbb{1}_{\{X_i \ge DT_{i,irsn}\}} + DL_{i,irsn} \mathbb{1}_{\{X_i < DT_{i,irsn}\}},$$

$$DT_{i,irsn} = k_{1-\alpha}\sqrt{2C_{bl,i}}, \text{ and } DL_{i,irsn} = DT_{i,irsn} + 0.5(k_{1-\alpha})^2 \left[1 + \sqrt{1 + \frac{4DT_{i,irsn}^2}{k_{1-\alpha}^4}}\right]$$

• Based on the sample  $R_{1,opt}, \ldots, R_{n,opt}$ , we consider  $\hat{\theta}_n = \frac{1}{n} \sum_{i=1}^n R_{i,opt}$ , where for  $i = 1 \ldots n$ ,

$$R_{i,opt} = X_i \mathbb{1}_{\{X_i \ge DT_{i,opt}\}} + DL_{i,opt} \mathbb{1}_{\{X_i < DT_{i,opt}\}}$$

$$\alpha = 1 - \frac{\Gamma(\lfloor DT_{i,opt} + 1 \rfloor, 2C_{bl,i})}{\lfloor DT_{i,opt} \rfloor}, \text{ and } DL_{i,opt} = DT_{i,opt} + 0.5(k_{1-\alpha})^2 \left[ 1 + \sqrt{1 + \frac{4DT_{i,opt}^2}{k_{1-\alpha}^4}} \right]$$

For each model of a), b) and c), we compute the Integrated Squared Error (ISE) and the Integrated Absolute Error (IAE) of  $\theta_n$ ,  $\overline{\theta_n}$  and  $\theta_n$ :

$$ISE(\theta_n) = (\theta_n - \theta)^2$$
 and  $IAE(\theta_n) = |\theta_n - \theta|$ ,

where  $\theta_n \in \{\widetilde{\theta_n}, \overline{\theta_n}, \widehat{\theta_n}\}.$ 

			n		
		Model	50	100	1000
		X	0.0196	0.0067	0.0002
a)	$ISE(\theta_n)$	$R_{irsn}$	55.587	0.1799	1.1104
		$R_{opt}$	0.2979	0.0599	0.1015
		X	0.1400	0.0820	0.0148
	$IAE(\theta_n)$	$R_{irsn}$	7.4556	0.4242	1.0537
		$R_{opt}$	0.5458	0.2449	0.3186
		X	0.0023	0.0032	0.0014
b)	$ISE(\theta_n)$	$R_{irsn}$	0.04812	0.0055	0.0082
		$R_{opt}$	0.0023	0.0032	0.0014
		X	0.0485	0.0566	0.0378
	$IAE(\theta_n)$	$R_{irsn}$	0.2193	0.0744	0.0910
		$R_{opt}$	0.0485	0.0566	0.0378
		X	1.7742	2.0563	0.8273
c)	$ISE(\theta_n)$	$R_{irsn}$	1.7742	2.0563	0.9367
		$R_{opt}$	1.7742	2.0563	0.8273
		X	1.3320	1.4340	0.9096
	$IAE(\theta_n)$	$R_{irsn}$	1.3320	1.4340	0.9678
		$R_{opt}$	1.3320	1.4340	0.9096
-		-			

Table 1: Results for ISE and IAE for N = 5 trials of the proposed estimators of  $\theta$   $(\tilde{\theta_n}, \overline{\theta_n}, \hat{\theta_n})$  for n = 50, n = 100, n = 1000.

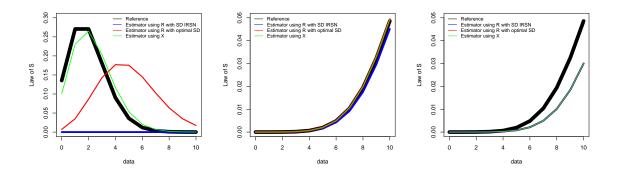


Figure 1: Results for the Signal distribution estimators of  $\mathcal{P}(\theta)$  using X,  $R_{irsn}$  and  $R_{opt}$  for n = 50, N = 5 for high values of  $C_{bl}$  and  $C_{gr}$  for model a) (left) b) (center) and c) (right).

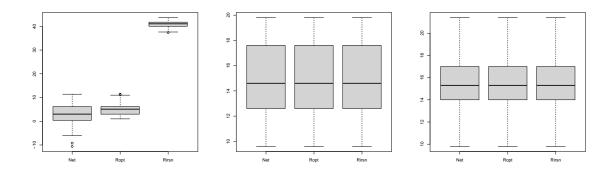


Figure 2: Results for the boxplots using X,  $R_{irsn}$  and  $R_{opt}$  for n = 50, N = 5 for high values of  $C_{bl}$  and  $C_{gr}$  for model a) (left) b) (center) and c) (right).

Based on Table 1 and Figures 1 and 2, we infer that:

- For all cases,  $ISE(\theta_n)$  and  $IAE(\theta_n)$  based on the model X and  $R_{opt}$  are smaller than those obtained by the model  $R_{irsn}$ .
- The *ISE* and the *IAE* decrease as the sample size increases.
- The Poisson law estimators  $\mathcal{P}(\tilde{\theta}_n)$  and  $\mathcal{P}(\hat{\theta}_n)$  based on the models X and  $R_{opt}$  respectively, can yield better results compared to the Poisson law estimator  $\mathcal{P}(\bar{\theta}_n)$  grounded on the  $R_{irsn}$  model.
- The average of of  $R_{opt}$  is closer to the average of X compared to that of  $R_{irsn}$ .

In addition, the performed simulations reveal that the proposed DT defined in Proposition 1 can entail satisfactory estimates of the signal  $\theta$ , better than the *DT* defined in ISO 11929 (2020).

#### 7.2 Performance of the Kernel density estimator

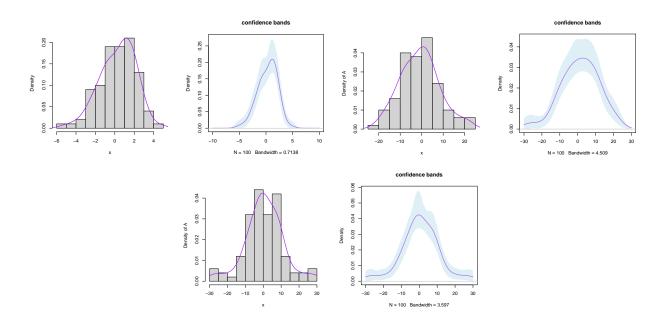


Figure 3: Results of the non parametric kernel density estimator of the  $f_A$  density and its confidence interval for n = 50, N = 100 for high values of  $C_{bl}$  for model a) (left up) b) (right up) and c) (down).

			n	
		50	100	1000
a)	$MSE(f_{A,n}(z))$	0.003040	0.001463	0.000131
b)	$MSE(f_{A,n}(z))$	0.000561	0.000287	$2.237 { m e}^{-05}$
c)	$MSE(f_{A,n}(z))$	0.000277	0.000230	$2.415\mathrm{e}^{-05}$

Table 2: Results for ISE and IAE for N = 5 trials of the kernel denity estimator of  $f_A(z)$  for n = 50, n = 100, n = 1000.

The density curves in Figure 3 are closer to the histogram of data. This figure shows that kernel estimator is a good approach of radioactivity density. Based on Table 7.2, we deduce that MSE of  $f_{A,n}(z)$  are closer to zero and decrease as the sample size increases.

## 8 Real data analysis

The monitoring of the environment is carried out by measuring the main radionuclides released into water, air and soil. 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 water by Bq/L.
- $C_{bl}$ : pure water (deep water)

- $C_{gr} = S + B'$ , where  $B' \sim C_{bl}$ .
- Y: calibration factor.

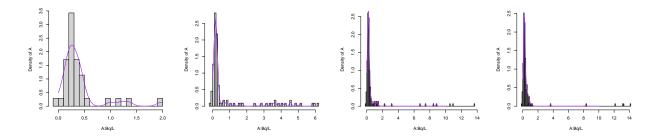


Figure 4: Results for the non parametric kernel density estimator of the density of  $f_A$  for the radioactivity data within 2019-2022 (left  $\rightarrow$  right)

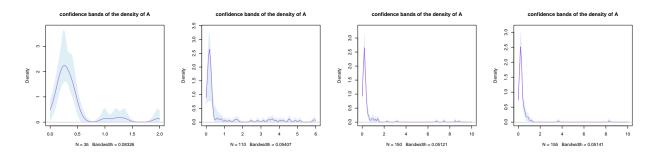


Figure 5: Results for the non parametric confidence bands of the density  $f_A$ , based on the kernel density estimator for the radioactivity data within 2019-2022 (left  $\rightarrow$  right)

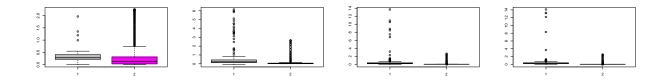


Figure 6: Results of boxplot for the radioactivity data within 2019-2022 (left  $\rightarrow$  right)

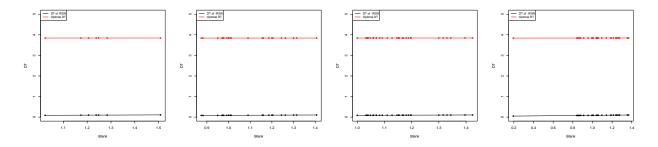


Figure 7: Results for DT of the count part for the radioactivity data within 2019-2022 (left  $\rightarrow$  right)

	2019	2020	2021	2022
$\text{ISE}(\mu_{A,n})$	0.021049	0.121666	0.092424	0.105236
$IAE(\mu_{A,n})$	0.145083	0.348807	0.304014	0.324401

Table 3: Results for ISE and IAE of the proposed estimator of  $\mathbb{E}(A)$  (Taylor method) for the radioactivity data (reference:  $\overline{A}$ ).

	2019	2020	2021	2022
$\operatorname{ISE}(\widehat{f}_{A,n}(z))$	0.000910	0.001543	0.000146	$7.175e^{-06}$
$\operatorname{IAE}(\widehat{f}_{A,n}(z))$	0.026128	0.034026	0.010487	0.002319

Table 4: Results for ISE and IAE of the kernel estimator  $\widehat{f}_{A,n}(z)$  for the radioactivity data (reference: histogram).

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

Table 5: Example 1

	Result of ISO	Proposed method
Activity	1.08	1.08
DT	0.737	3.32
CI	$[1.08 \pm 0.77] = [0.30, 1.85]$	$[1.08 \pm 1.22] = [-0.14, 2.30]$
Conclusion	A > DT	A < DT

Table 6: Example 2

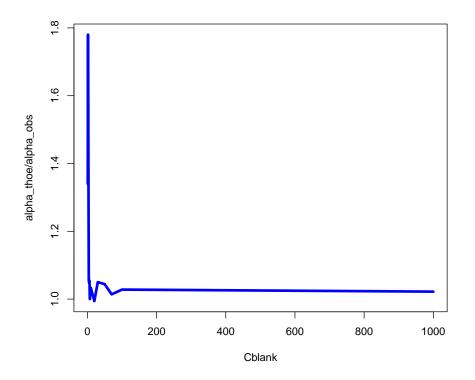


Figure 8: Comparison of the ratio of real/theoretical false positives..

Figure 4 displays the results of density kernel estimators within 2019 - 2022. The density curves in Figure 4 are closer to the histogram of data. This figure shows that kernel estimator is a good approach of radioactivity density. Grounded on Table 4, we deduce that ISE and IAE of  $f_{A,n}(z)$ are closer to zero and decrease as the sample size increases. Hence, it can be concluded that the kernel estimators are robust and perform well in terms of estimating the density of the radioactivity, as it is theoretically expected. Table 3 shows that ISE and IAE of the proposed estimator of  $\mathbb{E}(A)$ are closer to zero and decrease as the sample size increases. Figure 7 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 5 and 6). With reference to Tables 5 and 6, we infer that the confidence interval of ISO 11929 (2020) converge to the proposed confidence interval which is based on the proposed DT. In figure 8, we tested the DT determined in Proposition 1. We determined the ratio of the false positive rate between the observed risk  $\alpha_{practice}$  and the theoretical risk test  $\alpha_{theoritical} = 0.05$  for a given value of parameter  $\lambda$ . We have made 1000 draws of the random variable Y. For each draw, we determine the DT, then we make a new draw which we compare to the previously DT calculated. Figure 8 shows that ratio of real/theoretical false positives  $\alpha_{thoeretical}/\alpha_{practice}$  is closer to 1 especially when  $C_{blank}$  is large, which implies the Decision threshold is optimal, as it is theoretically expected.

## 9 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. Several theoretical results related to the model of the radioactivity; in particular to the density estimator 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.

## 10 Proofs

**Proof of Proposition 1**: For  $i = 1 \dots n$ , the predictive likelihood ratio for testing  $H_0$  vs  $H_1$  is determined by

$$\frac{l(\theta_1, (C_{gr,i}|C_{bl,i}))}{l(\theta_0, (C_{gr,i}, |C_{bl,i}))} = \frac{l(\theta, (C_{gr,i}|C_{bl,i}))}{l(0, (C_{gr,i}|C_{bl,i}))} \\
= \frac{\frac{\lambda^{C_{bl,i}} \exp(-\lambda)}{C_{bl,i}!} \times \frac{(\lambda + \theta)^{C_{gr,i}} \exp(-\lambda - \theta)}{C_{gr,i}!}}{\frac{\lambda^{C_{bl,i}} \exp(-\lambda)}{C_{bl,i}!} \times \frac{\lambda^{C_{gr,i}} \exp(-\lambda)}{C_{gr,i}!}}{\frac{C_{gr,i}!}{C_{gr,i}!}} \\
= \exp(\theta) \left(1 + \frac{\theta}{\lambda}\right)^{C_{gr,i}}.$$

Since  $\theta > 0$ , the likelihood ratio is increasing in a monotonic way with  $C_{gr,i}$ . Then based on Lemma of Neyman-Pearson in Lehmann et al (2005), the test using variable  $C_{gr,i}$  will be the uniformly most powerful test, and the set of  $C_{gr,i}$  where  $\frac{l(\theta_1, (C_{gr,i}|C_{bl,i}))}{l(\theta_0, (C_{gr,i}|C_{bl,i}))} > k$  is equivalent to the set of  $C_{gr,i}$  where  $C_{gr,i} > DT_i$ . The best test to determine whether  $\theta > 0$  will be in the form of  $C_{gr,i} > DT_i$ .

In order to determine  $DT_i$ , the level constraint

$$P_{H_0}(C_{gr,i} > DT_i | C_{bl,i} = c_{bl,i}) = \alpha$$

must be satisfied. Or, knowing the measure of  $C_{bl,i}$ , based on Manificat (2024), the probability of having  $C_{bl,i}$  in  $C_{gr,i}$  is a negative binomial  $\mathcal{NB}(c_{gr,i} + 1, 1/2, 1/2)$ . Then

$$\mathbb{P}_{H_0}(C_{gr,i} = c_{gr,i} | C_{bl,i} = c_{bl,i}) = \mathbb{P}(C_{gr,i} = c_{gr,i} | C_{bl,i} = c_{bl,i}, \theta = 0) = \frac{(c_{gr,i} + c_{bl,i})!}{c_{gr,i}! c_{bl,i}!} \left(\frac{1}{2}\right)^{c_{gr,i} + c_{bl,i}+1}$$

Since we have  $N_i$  observation of  $C_{gr,i}$  and  $C_{bl,i}$ , then  $DT_i$  is defined as follows:

$$\mathbb{P}\left(\overline{C_{gr,i}} > DT_i | \overline{C_{bl,i}} = \overline{c_{bl,i}}\right) = \alpha$$

$$= \mathbb{P}\left(\overline{C_{gr,i}} > DT_i | \overline{C_{bl,i}} = \overline{c_{bl,i}}\right)$$

$$= 1 - \mathbb{P}\left(\overline{C_{gr,i}} \le DT_i | \overline{C_{bl,i}} = \overline{c_{bl,i}}\right)$$

$$= 1 - F(DT_i)$$

$$= 1 - \left[1 - I_{1/2}\left(\overline{C_{bl,i}} + 1, DT_i + 1\right)\right]$$

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

where I is the regularized beta incomplete function and F is the cumulative distribution of the negative binomial. Based on Abramowitz and Stegum (1988), the approximation of I for large  $C_{gr,i}$  and  $C_{bl,i}$ , is expressed as

$$I_x(a,b) = \phi \left[ 3 \frac{(bx)^{1/3} (1 - \frac{1}{9b}) - (a(1-x))^{1/3} (1 - \frac{1}{9a})}{\sqrt{\frac{(a(1-x))^{2/3}}{a} + \frac{(bx)^{2/3}}{b}}} \right] + O\left(\frac{1}{\min(a,b)}\right)$$

for a, b > 0 and  $x \in [0, 1]$ . Therefore, in our context, we obtain

$$1 - \alpha \sim \phi \left( \frac{DT_i - \overline{C_{bl,i}}}{\sqrt{\overline{C_{bl,i}} + DT_i}} \right).$$

Hence,

$$\phi^{-1}(1-\alpha)\sqrt{\overline{C_{bl,i}} + DT_i} = DT_i - \overline{C_{bl,i}}.$$

Through solving the second degree equation,

$$-DT_i^2 + DT_i \left(k_{1-\alpha}^2 + 2\overline{C_{bl,i}}\right) + k_{1-\alpha}^2 \overline{C_{bl,i}} - \overline{C_{bl,i}}^2,$$

where  $k_{1-\alpha} = \phi^{-1}(1-\alpha)$ , we obtain  $\Delta > 0$  and we can choose

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

We equally find  $DT_i$  based on numerical Newton Raphson method using package *rootSolve* with the R software.

**Proof of proposition 2**: It is be noted, knowing the measure of  $C_{bl,i}$ , the probability of having  $C_{bl,i}$  in  $C_{gr,i}$  is a negative binomial  $\mathcal{NB}(c_{gr,i}+1,1/2,1/2)$ .

$$\mathbb{P}(C_{gr,i} = c_{gr,i} | C_{bl,i} = c_{bl,i}, \theta = 0) = \frac{(c_{gr,i} + c_{bl,i})!}{c_{gr,i}! c_{bl,i}!} \left(\frac{1}{2}\right)^{c_{gr,i} + c_{bl,i} + 1}$$

If  $C_{bl,i} \to \infty$ , then

$$\mathbb{P}(C_{gr,i} = c_{gr,i} | C_{bl,i} = c_{bl,i}, \theta = 0) \to \frac{1}{\sqrt{4\pi c_{bl,i}}} \exp\left[-\frac{(c_{gr,i} - c_{bl,i})^2}{4c_{bl,i}}\right] = \frac{1}{\sqrt{4\pi c_{bl,i}}} \exp\left[-\frac{(c_{gr,i} + c_{bl,i} - 2c_{bl,i})^2}{4c_{bl,i}}\right]$$

In addition, we have

$$\lim_{n \to \infty} \mathcal{P}(\alpha) = \mathcal{N}(\alpha, \alpha)$$

Thus, when  $C_{bl} \to \infty$ , we get

$$\frac{\alpha^x \exp(-x)}{x!} \sim \frac{1}{\sqrt{2\pi\alpha}} \frac{\exp(-(x-\alpha)^2)}{2\alpha}$$

Then, we have in our context,

$$\frac{1}{\sqrt{4\pi c_{bl,i}}} \exp\left[-\frac{(c_{gr,i}+c_{bl,i}-2c_{bl,i})^2}{4c_{bl,i}}\right] \sim \frac{(2c_{bl,i})^{c_{gr}+c_{bl,i}}\exp(-2c_{bl,i})}{(c_{gr,i}+c_{bl,i})!}$$

which is a Poisson distribution of parameter  $2c_{bl,i}$ . Then  $DT_i$  is defined as follows:

$$\mathbb{P}\left(\overline{C_{gr,i}} > DT_i | \overline{C_{bl,i}}\right) = \alpha 
= \mathbb{P}\left(\overline{C_{gr,i}} > DT_i | \overline{C_{bl,i}}\right) 
= 1 - \mathbb{P}\left(\overline{C_{gr,i}} \le DT_i | \overline{C_{bl,i}}\right) 
= 1 - F(DT_i) 
= 1 - \text{cumulative distribution of } \mathcal{P}(2c_{bl}) 
= 1 - \frac{\Gamma(\lfloor DT_i + 1 \rfloor, 2\overline{C_{bl,i}})}{\lfloor DT_i \rfloor},$$

where  $\Gamma(.,.)$  is the upper incomplete gamma function and  $\lfloor . \rfloor$  is the floor function (see Barbour et al (1992)). We equally find  $DT_i$  based on numerical methods using package *rootSolve* with the R software.

**Proof of Proposition 3** : It is to be noted that the Delaporte law is a convolution between the negative binomial law and the Poisson.

$$\mathbb{P}(C_{gr} = d | C_{bl} = c, \theta) = \sum_{c' \in \mathbb{N}} \frac{\theta^{d-c'} \exp(-\theta)}{(d-c')!} \frac{(c'+c)!}{c'!c!} \left(\frac{1}{2}\right)^{c'+c}$$
$$= \sum_{c'=0}^{d} \mathbb{P}(N_1 = d - c') \mathbb{P}(N_2 = c')$$
$$= \mathbb{P}(N_1 + N_2 = d)$$
$$= g_1 \star g_2(d),$$

where  $N_1 \sim \mathcal{P}(\theta)$ ,  $N_2 \sim \mathcal{NB}(c, \frac{1}{2}, \frac{1}{2})$  and  $g_1$  stands for the Poisson law and  $g_2$  corresponds to the negative binomial law.

The negative binomial law is a sum of geometric laws. Therefore according to the TLC theorem, the negative binomial law converges to the normal distribution of mean c and variance 2c, when  $C_{blank}$  (observation c)  $\rightarrow \infty$ :

$$g_2(c') = \frac{(c'+c)!}{c'!c!} \left(\frac{1}{2}\right)^{c'+c} \to \frac{\exp\left(\frac{-(c'-c)^2}{4c}\right)}{\sqrt{4\pi c}} = \frac{\exp\left(\frac{-(c'+c-2c)^2}{4c}\right)}{\sqrt{4\pi c}}.$$

It is noteworthy that, if a Gaussian distribution has a large variance, it can be considered equivalent to a Poisson distribution  $\mathcal{P}(2c)$ :

$$\frac{\exp\left(\frac{-(c'+c-2c)^2}{4c}\right)}{\sqrt{4\pi c}} \sim \frac{(2c)^{c'+c}\exp(-2c)}{(c'+c)!}.$$

As a matter of fact, we can write the Delaporte law as a convolution of two Poisson distributions with a parameter given by the sum of the Poisson parameters. Thus, we obtain

$$\mathbb{P}(C_{gr} = d | C_{bl} = c, \theta) = \sum_{c'=0}^{d} \mathbb{P}(N_1 = d - c') \mathbb{P}(N_2 = c' + c)$$
$$= \frac{(\theta + 2c)^{d+c}}{(d+c)!} \exp(-\theta - 2c).$$

In order to determine the estimator of  $\theta$ , we calculate the predictive likelihood function

$$l(C_{gr,1},\ldots,C_{gr,n}|C_{bl,1},\ldots,C_{bl,n},\theta) = \prod_{i=1}^{n} \frac{(\theta+2c_i)^{d_i+c_i}}{(d_i+c_i)!} \exp(-\theta-2c_i).$$

Since we need to use the logarithm in order to turn multiplication into addition, the log-likelihood L is provided by

$$L(C_{gr,1}, \dots, C_{gr,n} | C_{bl,1}, \dots, C_{bl,n}, \theta) = \log l(C_{gr,1}, \dots, C_{gr,n} | C_{bl,1}, \dots, C_{bl,n}, \theta)$$
  
= 
$$\sum_{i=1}^{n} \log \left( \frac{(\theta + 2c_i)^{d_i + c_i}}{(d_i + c_i)!} \exp(-\theta - 2c_i) \right).$$

Therefore, we need to resolve the following equation

$$\frac{\partial L(C_{gr,1},\ldots,C_{gr,n}|C_{bl,1},\ldots,C_{bl,n},\theta)}{\partial \theta} = 0.$$

Hence,

$$\sum_{i=1}^{n} \frac{d_i - c_i - \theta}{\theta + 2C_{bl,i}} = 0.$$
(10.1)

Since  $C_{gr,i}$  is large, then (10.1) implies  $\sum_{i=1}^{n} C_{bl,i} - \sum_{i=1}^{n} C_{gr,i} + n\theta = 0$ . Thus,

$$\theta_n = \frac{\sum_{i=1}^n C_{gr,i} - C_{bl,i}}{n}.$$

**Proof of Proposition 4 :** We first state the radioactivity as follows:

$$A = \frac{f}{\epsilon t V} (C_{gross} - C_{blank})$$
  
= YX  
=  $g(Y, X)$ 

where  $X = C_{gr} - C_{bl}$ ,  $Y = \frac{f}{\epsilon t V} \sim \mathcal{N}(a, b^2)$  and g(u, v) = uv. For any g(x; y), the bi-variate first order Taylor expansion about any  $a = (a_x, a_y)$  is

$$g(x,y) = g(a) + \frac{\partial g}{\partial x}(a)(x - a_x) + \frac{\partial g}{\partial y}(a)(y - a_y) + Rest.$$

Switching to random variables with finite means  $\mathbb{E}(X) := \mu_x$  and  $\mathbb{E}(Y) := \mu_y$ , we can choose the expansion point to be  $a = (\mu_x, \mu_y)$ . In that case, the first order Taylor series approximation for g(X, Y) is

$$g(X,Y) = g(a) + \frac{\partial g}{\partial x}(a)(X - \mu_x) + \frac{\partial g}{\partial y}(a)(Y - \mu_y) + Rest.$$

The approximation for  $\mathbb{E}(g(X, Y))$  is therefore

$$\mathbb{E}(g(X,Y)) \sim \mathbb{E}(g(a)) + \frac{\partial g}{\partial x}(a)\mathbb{E}((X-\mu_x)) + \frac{\partial g}{\partial y}(a)\mathbb{E}((Y-\mu_y))$$
  
=  $g(\mu_x,\mu_y) + 0 + 0.$ 

Relying on the definition of variance, the variance of g(X, Y) is

$$\mathbb{VAR}(g(X,Y)) = \mathbb{E}\left[\left(g(X,Y) - \mathbb{E}(g(X,Y))\right)^2\right] \sim \mathbb{E}\left[\left(g(X,Y) - g(\mu_x,\mu_y)\right)^2\right].$$

In this respect, using the first order Taylor expansion for g(X, Y) expanded around a:

$$\begin{split} \mathbb{VAR}(g(X,Y)) &\sim \mathbb{E}\left[\left(g(\mu_x,\mu_y) + \frac{\partial g}{\partial x}(\mu_x,\mu_y)(X-\mu_x) + \frac{\partial g}{\partial y}(\mu_x,\mu_y)(Y-\mu_y) - g(\mu_x,\mu_y)\right)^2\right] \\ &= \mathbb{E}\left[\left(\frac{\partial g}{\partial x}(\mu_x,\mu_y)(X-\mu_x) + \frac{\partial g}{\partial y}(\mu_x,\mu_y)(Y-\mu_y)\right)^2\right] \\ &= \mathbb{E}\left[\left(\frac{\partial g}{\partial x}(\mu_x,\mu_y)\right)^2(X-\mu_x)^2 + 2\frac{\partial g}{\partial x}(\mu_x,\mu_y)(X-\mu_x)\frac{\partial g}{\partial y}(\mu_x,\mu_y)(Y-\mu_y) + \left(\frac{\partial g}{\partial y}(\mu_x,\mu_y)\right)^2(Y-\mu_y)^2\right] \\ &= \left(\frac{\partial g}{\partial x}(\mu_x,\mu_y)\right)^2\mathbb{VAR}(X) + 2\frac{\partial g}{\partial x}(\mu_x,\mu_y)\frac{\partial g}{\partial y}(\mu_x,\mu_y)\mathbb{COV}(X,Y) + \left(\frac{\partial g}{\partial y}(\mu_x,\mu_y)\right)^2\mathbb{VAR}(Y). \end{split}$$

For our case g(x, y) = xy, the Taylor approximation is indicated as follows:

$$A = g(X, Y)$$
  
=  $\mathbb{E}(X)\mathbb{E}(Y) + \frac{\partial g}{\partial x}(\mathbb{E}(X), \mathbb{E}(Y))[X - \mathbb{E}(X)] + \frac{\partial g}{\partial y}(\mathbb{E}(X), \mathbb{E}(Y))[Y - \mathbb{E}(Y)] + Rest.$ 

The approximation of  $\mathbb{E}(A) = \mathbb{E}(g(X, Y))$  is:

$$\mathbb{E}(A) \sim \mathbb{E}(X)\mathbb{E}(Y) \sim \overline{XY}.$$

We therefore have

$$\begin{split} \frac{\partial g}{\partial X} &= Y, \ \frac{\partial g}{\partial Y} = X, \\ & \left(\frac{\partial g}{\partial X}(\mathbb{E}(X), \mathbb{E}(Y))\right)^2 = \mathbb{E}(Y)^2, \\ \frac{\partial g}{\partial X}(\mathbb{E}(X), \mathbb{E}(Y)) \frac{\partial g}{\partial Y}(\mathbb{E}(X), \mathbb{E}(Y)) = \mathbb{E}(X)\mathbb{E}(Y) \\ & \left(\frac{\partial g}{\partial Y}(\mathbb{E}(X), \mathbb{E}(Y)\right)^2 = \mathbb{E}(X)^2. \end{split}$$

Then, as X and Y are independent, the approximation of  $\mathbb{VAR}(A) = \mathbb{VAR}[g(X, Y)]$  is:

$$\mathbb{VAR}(A) \sim \mathbb{E}(Y)^2 \mathbb{VAR}(X) + \mathbb{E}(X)^2 \mathbb{VAR}(Y).$$

Hence,

$$\mu_{A,n} = \overline{XY} = \frac{1}{n} \sum_{i=1}^{n} X_i \frac{1}{n} \sum_{i=1}^{n} Y_i.$$
$$\sigma_{A,n}^2 = \left(\frac{1}{n} \sum_{i=1}^{n} Y_i\right)^2 \frac{1}{n-1} \sum_{i=1}^{n} (X_i - \mu_{X,n})^2 + \left(\frac{1}{n} \sum_{i=1}^{n} X_i\right)^2 \frac{1}{n-1} \sum_{i=1}^{n} (Y_i - \mu_{Y,n})^2.$$

**Proof of Proposition 5** : Asymptotic properties and confidence interval of  $\mu_{A,n}$ : Based on TLC Theorem, we get

$$\sqrt{n}(\overline{X} - \mu_X) \xrightarrow[n \to +\infty]{\mathcal{L}} \mathcal{N}(0, \sigma_X^2) \text{ and } \sqrt{n}(\overline{Y} - \mu_Y) \xrightarrow[n \to +\infty]{\mathcal{L}} \mathcal{N}(0, \sigma_Y^2).$$

Since  $\overline{X}$  and  $\overline{Y}$  are independent, then

$$\sqrt{n} \left[ \begin{pmatrix} \overline{X} \\ \overline{Y} \end{pmatrix} - \begin{pmatrix} \mu_X \\ \mu_Y \end{pmatrix} \right] \xrightarrow[n \to +\infty]{\mathcal{N}} \mathcal{N} \left( \begin{pmatrix} 0 \\ 0 \end{pmatrix}, \Sigma \right),$$

where  $\Sigma$  the variance covariance matrix is

$$\Sigma = \begin{pmatrix} \sigma_X^2 & 0\\ 0 & \sigma_Y^2 \end{pmatrix}.$$

Based on Delta method, we obtain

$$\sqrt{n}\left(\mu_{A,n}-\mu_{A}\right)=\sqrt{n}\left(g(\overline{X},\overline{Y})-g(\mu_{X},\mu_{Y})\right)\underset{n\to+\infty}{\overset{\mathcal{L}}{\longrightarrow}}\mathcal{N}\left(0,Dg(\mu_{X},\mu_{Y})\Sigma Dg(\mu_{X},\mu_{Y})^{t}\right),$$

where  $Dg(\mu_X, \mu_Y) = \left(\frac{\partial g}{\partial \mu_X}, \frac{\partial g}{\partial \mu_Y}\right)^t = (\mu_Y, \mu_X)^t$  stands for the Jacobian matrix of  $g(\mu_X, \mu_Y)$ . Since  $Dg(\mu_X, \mu_Y) \Sigma Dg(\mu_X, \mu_Y)^t = \mu_X^2 \sigma_Y^2 + \mu_Y^2 \sigma_X^2,$  then the previous result allows us to build up the confidence interval of  $\mathbb{E}(A) = \mu_A$  which has the following form

$$I_{\mu_A} = \left(\overline{XY} \pm \frac{z_{1-\frac{\delta}{2}}\sigma_n(x)}{\sqrt{n}}\right),\,$$

where  $z_{1-\frac{\delta}{2}}$  is the normal  $(1-\frac{\delta}{2})$  quantile and  $\sigma_n(x) = \sqrt{\overline{X}^2 \sigma_{Y,n}^2 + \overline{Y}^2 \sigma_{X,n}^2}$ . Based on Rosenblatt (1956), it follows that

$$\sqrt{nh_n}(\widehat{f}_{A,n}(z) - f_A(z)) \xrightarrow[n \to +\infty]{\mathcal{L}} \mathcal{N}\left(0, f_A(z) \int K(u) du\right).$$

Then, the confidence interval based on kernel density estimator is given by

$$I_{f_A(z)} = \left(\widehat{f}_{A,n}(z) \pm \frac{z_{1-\frac{\delta}{2}}\sigma_f(z)}{\sqrt{n}}\right),$$

where  $z_{1-\frac{\delta}{2}}$  is the normal  $\left(1-\frac{\delta}{2}\right)$  quantile and  $\sigma_f(z) = \sqrt{\widehat{f}_{n,A}(z)} \int K(u) du$ .

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