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A Bayesian approach to assess data from radionuclide activity analyses in environmental samples

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ABSTRACT

A Bayesian statistical approach is introduced to assess experimental data from the analyses of radionuclide activity concentration in environmental samples (low activities). A theoretical model has been developed that allows the use of known prior information about the value of the measurand (activity), together with the experimental value determined through the measurement. The model has been applied to data of the Inter-laboratory Proficiency Test organised periodically among Spanish environmental radioactivity laboratories that are producing the radiochemical results for the Spanish radioactive monitoring network. A global improvement of laboratories performance is produced when this prior information is taken into account. The prior information used in this methodology is an interval within which the activity is known to be contained, but it could be extended to any other experimental quantity with a different type of prior information available.

1. Introduction

Bayesian methods are increasingly being used with success in experimental sciences. These methods are particularly well-suited to incorporate available prior information into the analysis of the experimental quantities, providing a powerful and flexible tool, essential in several scientific areas. Bayesian Statistics provides a more intuitive assessment procedure, closer to the thinking of the scientist than classical methodologies [1–4]. In this paper, a Bayesian approach is introduced to assess experimental data analysis from the measurement of radionuclide activity concentration[1] in environmental matrices.

Any measurement result is in general a point estimate of the measured quantity (measurand), the true value of which remains unknown. Therefore, a set of plausible values of the measurand around the estimate should be appraised [5]. Under symmetry assumptions, this may be described by the standard deviation of a probability density describing the remaining uncertainty with respect to the true (unknown) value of the measurand (uncertainty of measurement [6]).

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[1] The radioactive contents in a sample is usually expressed by unit of volume (activity concentration) or mass (specific activity). However, further on in the text, the generic term “activity” will be employed to refer any of the possible forms to express the activity (absolute, specific, concentration).

In the case of the determination of radionuclide activity concentration, the uncertainty has an intrinsic component due to the counting rate that becomes increasingly important as the activity is lower, and other components associated to the rest of magnitudes involved in the measurement, such as detector calibration, background effect, nuclear parameters and sample mass or volume.

The aim of this paper is to show how the knowledge of prior information on the quantity of interest (activity) may be combined with the experimental data to obtain a better estimator of the quantity of interest than that obtained from a classical use of the experimental data. This has been implemented through Bayesian methods. Specifically, the Bayesian Statistics has been applied to study how the experimental data are modified when the analyst has been informed about an activity interval in which the true value of the activity is delimited. The case of study is the Inter-laboratory Proficiency Test organised periodically among Spanish environmental radioactivity laboratories that are producing the radiochemical results for the Spanish radioactive monitoring network [7]. The framework of these exercises consists on the determination of different radionuclide activities in a test sample distributed to the laboratories of the network. Each laboratory reports the results of the analysis to the coordinator, that will assess the laboratory performance through statistical analysis. A value is assigned for each measurand that is disclosed to participants after the reporting deadline, but an information interval on the activity level is given when distributing the samples on a regular basis. The inclusion of the information interval in the basis of the Proficiency Test is a general policy in the Spanish laboratory intercomparisons, to ensure the participants that the sample activity conforms with their routine analysis (environmental activity levels). The activity level interval is informed to participants since, for the monitoring networks analyses, the laboratories always know something about the usual levels of activity in the environmental samples they are measuring.

The results produced by the laboratories can be modified (to improve its quality), when this prior information is incorporated into the statistical model for assessment. To study this effect, a theoretical model has been developed and its application to the results of an Inter-laboratory Proficiency Test organised in Spain [8] is presented and discussed.

2. Posterior distribution of the activity

Any statistical procedure requires a model which describes the probabilistic relationship beween the obervations and the quantity of interest (in this case, the true activity contained in the sample). To this model, the Bayesian approach adds a prior distribution which encapsulates whatever prior knowledge is available on the true value of the quantity of interest. In our case, as it is explained later, observations are assumed to have a normal distribution centered at true value of the quantity of interest, which is assumed to belong to a particular finite interval.

Let x denote the laboratory measurement result of the activity,[2] and let u be the deviation which models the corresponding measurement uncertainty,[3] which is assumed to be known [6]. The pair (x, u) is the information determined by the laboratory. Let $\theta \in \Theta$ be the actual (unknown) activity; this is the quantity of interest. From the Bayesian point of view, there exists a probability density $\pi(\theta)$ (which is a rational measure of uncertainty about the value of θ, not a description of any frequentist variation), which encapsulates all available information about its value before the measurements are made. After the observation of x this probability distribution is updated, via Bayes theorem to the posterior distribution $\pi(\theta|x)$, which combines the information about θ contained in the data with the prior information contained in $\pi(\theta)$.

Moreover, it is possible to specify the conditional probability density of the observation x, denoted by $p(x|\theta)$, by using a probability model which takes into account the physical process which generates the experimental data x for any given θ.

By Bayes theorem, the information about the value of θ after the experimental result has been obtained x is described by the corresponding posterior distribution $\pi(\theta|x)$ given by

$$\pi(\theta|x) = \frac{p(x|\theta)\,\pi(\theta)}{p(x)} = \frac{p(x|\theta)\,\pi(\theta)}{\int_{\Theta} p(x|\theta)\,\pi(\theta)\mathrm{d}\theta}. \tag{1}$$

This combines the information about θ contained in the data x with the information about θ contained in the prior distribution $\pi(\theta)$.

To obtain an explicit expression for $\pi(\theta|x)$, both $p(x|\theta)$ and $\pi(\theta)$ must be specified. In the problem we are interested, the variation of the measurement x around the true value of the activity θ is due to many, essentially independent causes; as a consequence, by the central limit theorem, it is reasonable to assume that x is normal distributed,[4] that is

$$p(x|\theta) = N(x|\theta, u) = \frac{1}{u\sqrt{2\pi}} \mathrm{e}^{-(1/2)\,(((x-\theta)^2)/(u^2))} \tag{2}$$

Thus, since u is assumed to be known, substituting into (1) the posterior distribution of the activity becomes

$$\pi(\theta|x) = \frac{N(x|\theta, u)\,\pi(\theta)}{\int_{\Theta} N(x|\theta, u)\,\pi(\theta)\,\mathrm{d}\theta}. \tag{3}$$

The prior information available by the laboratory about the activity θ is a reference interval $\Delta = [m, M]$ in which the activ-

[2] The result x may be obtained from a single measurement, or can be the mean value of a set of measurements.

[3] According to the GUM guide [6], the exact terminology of the measurement uncertainty of x is $u(x)$. This terminology will be simplified in the paper, and the uncertainty will be denoted by u. This simplification avoids the confusion that may arise of considering any functional dependence between u and x, which is not the case in the paper (there is no dependence in this context).

[4] It can be shown that this is a correct approximation when the uncertainties of input quantities used to determine the activity result x are small, that is, the same conditions that allow the (linear) uncertainties propagation rules of ISO GUM guide to be applied.

ity is included, so that it is known that $m \leq \theta \leq M$. This activity interval is informed by the organiser of the Proficiency Test to each laboratory, in order to indicate the activity level of the radionuclide contained in the sample. For example, the laboratory has to determine the activity of $^{239+240}$Pu contained in a water sample, and it is informed that the reference interval is $\Delta = [40, 100]\,\mathrm{Bq\,m^{-3}}$. The true value of the activity θ is known by the organiser with standard uncertainty according to the GUM,[5] $\theta = (49.8 \pm 1)\,\mathrm{Bq\,m^{-3}}$, whilst the laboratory has the mentioned prior information, plus the experimental information obtained from its own measurement [8].

Since there is no reason a priori to assume that some parts of Δ are more likely to contain θ than others, it seems reasonable to assume a uniform prior distribution for θ over this interval,[6] so that

$$\pi(\theta) = \begin{cases} \dfrac{1}{M-m} & \theta \in \Delta \\ 0 & \theta \notin \Delta \end{cases} \tag{4}$$

A far stronger argument may be given for the use of the prior (4). Indeed, it may be shown that this is the reference prior [2] for this model, that is the prior which precisely describes that the only available information about the value of θ is that it lies within the interval $\Delta = [m, M]$.

Substituting (4) into (3) the factors $1/(M-m)$ cancel out and one immediately gets

$$\pi(\theta|x) = \begin{cases} \dfrac{N(x|\theta, u)}{\displaystyle\int_m^M N(x|\theta, u)\,d\theta} & \theta \in \Delta \\ 0 & \theta \notin \Delta \end{cases} \tag{5}$$

Since for any normal density $N(x|\mu, u) = N(\mu|x, u)$, Eq. (5) may also be expressed as

$$\pi(\theta|x) = \begin{cases} \dfrac{N(\theta|x, u)}{\displaystyle\int_m^M N(\theta|x, u)\,d\theta} & \theta \in \Delta \\ 0 & \theta \notin \Delta \end{cases} \tag{6}$$

Hence, the resulting posterior distribution $\pi(\theta|x)$ is a truncated normal, that vanishes outside the interval Δ, and is normalised inside the interval, so that $\int_\Delta \pi(\theta|x)\,d\theta = 1$ (Fig. 1). A limit case of this distribution is the situation $\Delta = (-\infty, \infty)$, that

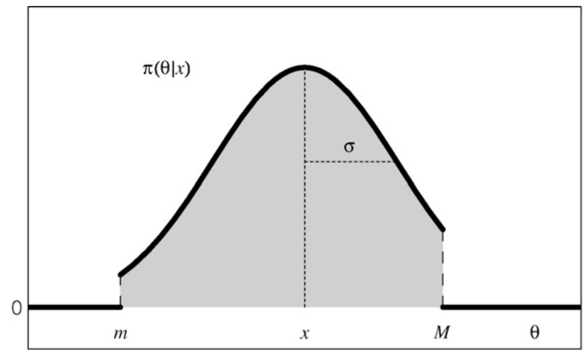

Fig. 1 – Probability density $\pi(\theta|x)$ of the true activity θ when it is considered both experimental and prior information.

is, when no prior knowledge about the activity value is considered. Some information is however always available about the value of the measurand (since the activity is a positive quantity, $a \geq 0$, and the number of radioactive atoms in a sample cannot be infinite), though this information is (almost) never utilised.[7] This corresponds to the classical treatment of the experimental data, that implicitly ignores the additional knowledge about the limited value of the activity, what in turn directly leads to the classical result, the normal probability distribution $\pi(\theta|x) = N(\theta|x, u)$. This may occur even when prior information is considered, if the interval is large enough.[8] The difference between $\pi(\theta|x)$ and the normal (not truncated) distribution becomes relevant when the interval is small enough, so that it provides significant information not contained in the normal distribution itself. This is the case that will be studied, when the effects of prior information available about the true value of activity θ are noticeable.

3. Posterior expectation of the activity

The posterior density, $\pi(\theta|x)$, describes all available information about the value of the true activity θ, combining prior knowledge and experimental information. However, it is required to obtain a point estimator of θ, which act as summary of the information contained in $\pi(\theta|x)$. In other words, a single value must be obtained as a result that combines the experimental data and our prior knowledge. Being the posterior distribution $\pi(\theta|x)$, the result is simply the expected value of θ. Since the posterior expectation of θ is Bayesian modification of the initial experimental value x, we will denote this expectation as x_B. This may be computed as the ratio of two

[5] The test sample was prepared by the CIEMAT Metrologic Laboratory for the Measurement of Ionising Radiations.

[6] This rectangular distribution represents the prior knowledge of the quantity of interest, the real activity. In the GUM guide, rectangular and other types of distributions are used to describe the incomplete knowledge about an input quantity in order to propagate the uncertainty (type B uncertainties). The central idea is the same in both cases: the distribution represents the (always incomplete in metrology) knowledge of the experimental quantity. Rectangular and other distributions used to describe the incomplete knowledge of input quantities in the GUM guide can be seen in fact as prior distributions. In order to use them, a prior knowledge about the input quantity must be available. Otherwise, the shape of the distribution (rectangular, triangular) could not be decided.

[7] A particular and very interesting limit case can be studied when the only knowledge available is that the activity is a positive quantity, that is, $\Delta = [0, \infty)$. It should be noted that this prior information is used in the recent revision of ISO 11929-7 standard for the determination of characteristic limits for ionising radiation measurements [9].

[8] That is, if $M > x + 3u$ and $m < x - 3u$ then $\pi(\theta|x) \simeq N(\theta|x, u)$, since the difference between distribution $\pi(\theta|x)$ and $N(\theta|x, u)$ is the probability in the tails, that in such case is not relevant.

integrals,

$$x_B = E[\theta|x] = \int_m^M \theta\,\pi(\theta|x)\,d\theta = \frac{\int_m^M \theta\,N(\theta|x,u)\,d\theta}{\int_m^M N(\theta|x,u)\,d\theta}, \tag{7}$$

both of which may be expressed in terms of simple definite integrals. Moreover, with an appropriate change of variables, the integral in the denominator of (7) may be written in terms of the error function as

$$I = \int_m^M N(\theta|x,u)\,d\theta = \frac{1}{2}\left[\mathrm{erf}\left(\frac{1}{\sqrt{2}}\frac{M-x}{u}\right) - \mathrm{erf}\left(\frac{1}{\sqrt{2}}\frac{m-x}{u}\right)\right] \tag{8}$$

where erf(z) is the error function defined as

$$\mathrm{erf}(z) = \frac{2}{\sqrt{\pi}}\int_0^z e^{-\omega^2}\,d\omega. \tag{9}$$

With a similar change of variables, the integral in the numerator of (7) may be written in terms of the last result as

$$\int_m^M \theta\,N(\theta|x,u)\,d\theta = x\,I + \frac{2u}{\sqrt{\pi}}\int_{(1/\sqrt{2})((m-x)/u)}^{(1/\sqrt{2})((M-x)/u)} \omega\,e^{-\omega^2}\,d\omega$$
$$= x\,I + u^2[N(m|x,u) - N(M|x,u)]. \tag{10}$$

and, therefore, substituting into (7) one finally has

$$x_B = x + \varepsilon, \tag{11}$$

$$\varepsilon = 2u^2\,\frac{N(m|x,u) - N(M|x,u)}{\mathrm{erf}((1/\sqrt{2})\,((M-x)/u)) + \mathrm{erf}((1/\sqrt{2})\,((x-m)/u))}. \tag{12}$$

The expression (11), $x_B = x + \varepsilon$, can be interpreted as follows; with quadratic loss, the best point estimator x_B of the activity θ when the prior information is known, is equal to the the initial experimental value x, modified by a perturbation ε that contains the prior information available on the true value of the activity.

It can be shown that, as one would expect, the correction term ε tends to zero when the interval is big enough, that is, if $M \gg x + 3u$ and $m \ll x - 3u$. If any of these conditions is not satisfied (the interval Δ is small enough) the value of the correction is not negligible. Moreover, it is possible to study the tendency of the correction, from the asymmetric shape of the posterior probability density, that is, from the relative position of the data x inside the interval Δ (see Fig. 1). As the sign of the denominator in (12) is positive, the sign of ε is determined by the sign of the numerator. Thus, if $N(m|x,u) > N(M|x,u)$ then $\varepsilon > 0$, while if $N(m|x,u) < N(M|x,u)$ then $\varepsilon < 0$. This last case corresponds to Fig. 1, and therefore the correction tends to place x_B to the left side of the original value x. In general, the term ε tends to correct the asymmetric shape of the distribution, allowing to determine a value x_B better centered in the interval Δ of possible θ values than the conventional estimator, the initial data x. It can be demonstrated that this correction will improve the estimate x directly obtained from the data, giving a value x_B which in most cases will be closer to θ than x. Only when x lies between θ and the middle point of the interval, the value x_B will not improve, most probably the result will stay essentially unaltered, as the correction is zero for values x placed in the middle of the interval. The general improvement of the data will be shown using real experimental data in next section.

4. Application to Proficiency Test data CSN/CIEMAT-04

In order to examine what variation produces the described Bayesian approach in the results of the activity measurements, the data of the Inter-laboratory Proficiency Test of environmental radioactivity laboratories CSN/CIEMAT-2004 have been used [8]. This test consisted on the measurement of a synthetically prepared water sample, containing the radionuclides listed in Table 1, whose reference activities x_{Ref} (metrologically certified) are in the characteristic range of environmental samples.

The total number of participant laboratories in the Proficiency Test was 35, but since all of them do not have capacity to analyse all the radionuclides, the number of participants in the measurement of each radionuclide (N in the table) is smaller. In the participation basis of the test, the coordinator informed to each laboratory the interval $\Delta = [m, M]$ in which the activity of each radionuclide is limited. Once the measure-

Table 1 – Radionuclides determined in the Inter-laboratory Proficiency Test of environmental radioactivity laboratories CSN/CIEMAT-2004

Radionuclide	N	x_{Ref} (Bq m^{-3})	σ_p (% of x_{Ref})	m (Bq m^{-3})	M (Bq m^{-3})
^{3}H	17	10136	12	8000	15000
^{90}Sr	27	187	20	100	400
^{137}Cs	35	499	8	200	900
$^{239+240}$Pu	10	49.8	14	40	100
^{241}Am	13	50.2	16	30	100
α_T	33	90.8	22	50	150
β_T	33	1333	17	800	1800
β_P	23	808	13	600	1200

α_T, β_T, β_R, are, respectively, gross alpha, gross beta and residual beta index. The number of participants in the analysis of each radionuclide is N, being x_{Ref} the reference activity, σ_p the precision established for each radionuclide (expressed as a percentage of x_{Ref}) and m, M the minimum and maximum activity values of the interval Δ, informed by the test organiser to the laboratories.

Table 2 – Data from $^{239+240}$Pu analysis in the CSN/CIEMAT-2004 Proficiency Test

Lab code	x (Bq m^{-3})	u (Bq m^{-3})	x_B (Bq m^{-3})	$\lvert x_B\rvert/\lvert x\rvert$	$\lvert z\rvert$	$\lvert z_B\rvert$	$\lvert z_B\rvert/\lvert z\rvert$
1	47.60	1.10	47.60	1.000	0.316	0.316	1.000
2	34.90	1.00	40.18	1.151	2.138	1.380	0.645
3	41.20	4.25	43.87	1.065	1.234	0.851	0.690
4	40.70	1.62	41.58	1.022	1.306	1.179	0.903
5	53.40	1.10	53.40	1.000	0.516	0.516	1.000
6	43.05	1.49	43.12	1.002	0.968	0.958	0.989
7	43.50	1.75	43.60	1.002	0.904	0.890	0.985
8	42.00	2.50	42.92	1.022	1.119	0.987	0.882
9	53.60	4.50	53.62	1.000	0.545	0.548	1.005
10	62.00	1.50	62.00	1.000	1.750	1.750	1.000

$\Delta = [40, 100]$ Bq m^{-3}.

ments are performed, each laboratory reported the results of activity and its uncertainty, (x, u) respectively, for each analyzed radionuclide. The evaluation of the Proficiency Test was performed following the international protocols [10–12]. The assessment of performance is calculated for each laboratory using the statistical scheme z-score,

$$z = \frac{x - x_{Ref}}{\sigma_p} \tag{13}$$

where σ_p is the fitness for purpose based "standard deviation for proficiency assessment", which is established by the coordinator for each radionuclide, and whose value range between 8% of the reference value (^{137}Cs) and 22% (gross alpha activity α_T). The z-score is therefore an indication of the deviation of the measurement x with respect to the reference value x_{Ref} expressed in terms of the established deviation σ_p. Interpretation of z-score is based on the normal distribution $N(x|_{Ref}, \sigma_p)$ assuming that x_{Ref} is very close to θ, so that a value $|z| \le 2$ is designated satisfactory, $|z| > 3$ is considered not-satisfactory and $2 < |z| < 3$ is designated acceptable.[9]

To compare the effects of the Bayesian approach, two z-scores will be calculated: using the activity value x reported by the laboratory, and using the modified Bayesian value $x_B = \theta^*$, and in this case we will denote z-score as z_B, that is

- Result without prior information; $x \to z = ((x - x_{Ref})/(\sigma_p))$
- Result with prior information; $x_B \to z_B = ((x_B - x_{Ref})/(\sigma_p))$

Since the Bayesian approach allows to determine a value x_B, that should be more accurate than the value x initially determined experimentally, this would have to be translated into a z-score with a smaller absolute value in the Bayesian case, that is, it would have to be obtained $|z_B| \le |z|$, or equivalently $|z_B|/|z| \le 1$

In Table 2 the data relative to $^{239+240}$Pu analysis are presented: The laboratory code, the reported values of activity and its uncertainty, (x, u) respectively, the Bayesian activity x_B, the quotient $|x_B|/|x|$, that shows the variation experienced by the Bayesian value x_B with respect to the classic value x, the absolute values of z-score: classic $|z|$ and Bayesian $|z_B|$, and the quotient $|z_B|/|z|$, which describes the variation of the Bayesian score with respect to the classic one.

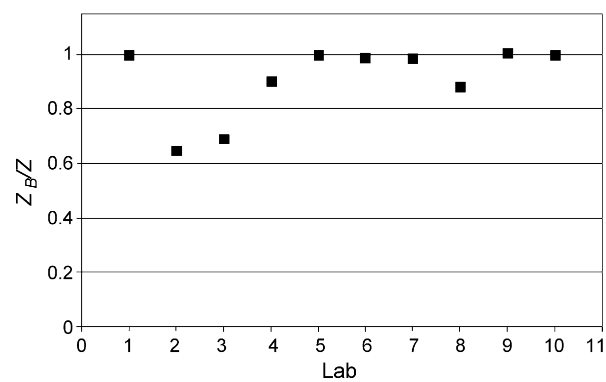

Fig. 2 – Quotient $|z_B|/|z|$ (Bayesian z-score with respect to the classic one) for the $^{239+240}$Pu analysis in the INTER/CSN-2004 Proficiency Test.

Table 2 shows that the values $|z_B|/|z|$ are practically equal or smaller than the unit, what confirms the general improvement produced by the Bayesian treatment of the experimental data using the prior knowledge of the interval $\Delta = [m, M]$, even in this case in which θ is near one border of the interval. Fig. 2 represents $|z_B|/|z|$ for each laboratory, showing clearly that $|z_B|/|z| \le 1$. It can be observed that, from 10 data, 4 of them have improved their quality, whereas the rest remains essentially unaltered. The biggest change in z is for laboratory 2 ($|z_B|/|z| = 0.645$), from $z = 2.14$ to $z_B = 1.38$, changing noticeably the quality of the result, that in fact produces a shift on the scoring, from acceptable ($2 < |z| < 3$) to satisfactory ($|z| \le 2$). With respect to the relative modification of the analysis results $|x_B|/|x|$, the greatest change, 15%, is for the same laboratory,[10] being the rest below the 6%. Indeed, these modifications of

[9] It must be mentioned that the International Standard Protocols commonly used to perform and evaluate Proficiency Tests [10–12] use the classical (not Bayesian) decision theory. The Proficiency Test described in this paper [8] was designed and evaluated following these protocols, and therefore classical z-score was used. A very interesting study on how to compare measurement results using the Bayesian Decision theory can be found in [13].

[10] The laboratory 2 reports an activity value (34.90) that is outside the provided information interval (40–100). The Bayesian transformation is specially significant in this case, as it moves the activity value into the information interval (40.18).

the activity values are significant with respect to the standard deviation for proficiency assessment ($\sigma_p = 14\%$), and therefore produce the mentioned variation in the scoring.

Summarising, the decreasing trend of z-score values observed in Fig. 2 points to a global improvement in performance through Bayesian treatment. This can be confirmed by means of the sum of quadratic deviations, being smaller in the Bayesian approach, in particular $S = \sum_{i=1}^{N} |z|^2 = 14.5$, whereas $S_B = \sum_{i=1}^{N} |z_B|^2 = 10.4$. With respect to the individual laboratory results, Fig. 2 demonstrates that each data improves, or remains unchanged. Regarding the decission of a laboratory when reporting a result, concerning to the application of the Bayesian approach, it has been clearly demonstrated that the results most probably will improve. Therefore, it is always worth to apply the Bayesian transformation. In other words, each laboratory can decide whether to apply or not the Bayesian approach, but it must at least know that the fact of not applying it can be disadvantageous for the quality of the results emitted by the laboratory, as it has been shown in the case of laboratory 2.

It could be objected that the Bayesian method is artificial, since if the interval Δ is unknown, the quality of the measurement would be z (not z_B) and, therefore the performance of the laboratory is the initial value of z-score. Nevertheless, radiochemical analysis involves experimental information and parametric (not measured) information. In particular, the experimental information is the counting rate of the sample, the measurement of the background, the calibration of the detector, the chemical recovery, the mass of the sample, the time of counting, and in general any directly measurable information. The parametric (which is equivalent to prior) information necessary to determine the final result of the activity are the decay constants, the half-life periods, the emission intensities, etc. of the radiation. The final quality of the result of the analysis will depend therefore on the quality of all the information used in the determination of the mesurand. In consequence, the laboratory should incorporate suitably all the available prior information, in order to report the best quality result. In particular, the prior knowledge of the interval in which the activity is limited is an information that should not be to be ignored, being comparable to any other prior information used in the determination of the mesurand, such as the tabulated constants, or any other used parameter not measured.

It may then be stated that the use of the Bayesian estimate $x_B = E[\theta|x]$ improves the quality of the analysis. This result is a direct consequence of using all the available information, both experimental and prior information.

5. Conclusions

The developed model has shown that the quality of the results produced by a laboratory can be improved, by incorporating the prior information known by the analyst, through Bayesian statistics. This model has been applied to the scope of radioactive analyses and the utilised prior information is an information interval on the activity level, but it could be extended to any experimental quantity under different prior available information. The analyst is always acquainted on some prior information on the measurand, and can apply the Bayesian approach, but only when the information is significant compared to the experimental data, the modification can be noticed. In the case of environmental radioactivity measurements, when the information interval is small enough, compared to the confidence limits of the uncertainty, the change would be appreciated. Moreover, the modification increases as the value of the measurement is far from the middle point of the interval.

Proficiency Tests constitute a suitable application field to essay and validate this Bayesian methodology, where a deeper study for quantitative assessment of the laboratory results can be achieved, and the z scoring of participants can be improved.

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