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DETECTION LIMITS FOR RADIOACTIVITY COUNTING

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Abstract. *In this paper, the detection limits for radioactivity counting are determined. Several decision rules are presented and applied to calculate the critical value. The value obtained by the best approximation for the alpha particles counting was used to evaluate the minimum detectable concentration in water, soil and air.*

Key words: *detection limits, radioactivity counting*

1. INTRODUCTION

Environmental radioactivity measurements may involve a material containing very small amounts of the radionuclide of interest. Measurement uncertainty often makes it difficult to distinguish such small amounts of radionuclides. Thus, an important characteristic of a measurement process is its detection capability, which is usually expressed as the smallest concentration of the radionuclide that can be at a certain confidence level distinguished from zero.

Effective experiment planning requires an accurate insight into the detection capabilities of the measurement procedures. The measure used to describe radioactivity detection capabilities is the minimum detectable concentration (MDC). The minimum detectable concentration is defined as an estimate of the true concentration of an analyte required to give a specified high probability that the measured response will be greater than the critical value [1].

There are still disagreements about the proper uses of the MDC concept. Some authors define the MDC strictly as an estimate of the nominal detection capability of a measurement process [2]. The opposing view is that the "sample specific" MDC is a useful measure for the detection capability of the measurement process, not just in theory, but in practice as well. For instance, NUREG/CR-4007 states plainly that "the critical (decision) level and detection limit (MDC) do vary with the nature of the sample" and that "proper assessment of these quantities demands relevant information on each sample, unless the variations among samples are quite trivial" [3].

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Neither version of the MDC can legitimately be used as a treshold value for a detection decision. The definition of the MDC presupposes that an appropriate detection threshold (i.e. the critical value) has already been defined [1].

THEORY

To detect an analyte in an environmental sample is to determine, to measure or to calculate its measurable quantity. The detection involves a choice between the "null hypothesis" H_0: the sample doesn't contain the analyte, and the "alternative hypothesis" H_1: the sample is not analyte-free. While testing the hypothesis there are two possible types of errors. Error type I (*false positive*) occurs if one rejects the H_0 when it is true, and error type II (*false negative*) occurs if one fails to reject the H_0 when it is false [4].

In radioactivity counting, the response variable of radionuclide (analyte) concentration is an instrument signal, usually the net number of counts observed (N_s). It is obtained by subtracting the estimated background, i.e. the blank signal (N_b), from the gross number of counts (N_g). To justify the rejection of the H_0, the value of N_s must exceed a certain treshold value, L_c, called the critical value of net counts, or decision treshold [5].

The critical value L_c is defined symbolically by the relation:

$$P[N_s > L_c \mid \mu = 0] \leq \alpha \tag{1}$$

where P denotes the probability, μ is the true value of the net signal, and α denotes the significance level, or the specified probability of a type I error (typically, $\alpha = 0{,}05$) [6].

The minimum detectable concentration (MDC), x_D, has been usually [1], [4] estimated by calculating the minimum detectable value of the net instrument signal, L_D, i.e. detection limit. It is defined by the relation [6]:

$$P[N_s \leq L_c \mid \mu = L_D] = \beta \tag{2}$$

where β is the probability of an error type II, usually chosen to be 0,05. Then, the MDC is simply obtained by dividing L_D with the sensitivity constant: $x_D = L_D / A$ [1].

To determine the value of L_c that satisfies the definition (1), one has to know the distribution of the counting data (N_s) under the H_0. If that distribution is approximately normal, within the well-known standard deviation σ_0, estimated by replicate blank measurements, the critical value of net counts is:

$$L_c = z_{1-\alpha}\sigma_0 \tag{3}$$

where $z_{1-\alpha}$ denotes the $(1-\alpha)$-quantile of the standard normal distribution.

If the mean blank count is extremely low, radiation counting data approximately follow the *Poisson* model. Then, whenever the blank count rate R_b is well known, the critical gross count, y_c, equals the smallest nonnegative integer n, such that:

$$e^{-R_b t_s} \sum_{k=0}^{n} \frac{(R_b t_s)^k}{k!} \geq 1 - \alpha \tag{4}$$

where t_s is the count time for the tested sample. The decision treshold, L_c, is simply obtained as $y_c - R_b\, t_s$ [1].

At somewhat higher count levels the *Poisson* distribution may be approximated by a normal distribution, and both models could be applied [7]. So, using the *Poisson* model in calculating the σ_0, the critical net count is given approximately by the equation:

$$L_c = z_{1-\alpha}\sqrt{\tilde{R}_b t_s\left(1+\frac{t_s}{t_b}\right)} \quad (5)$$

where t_b is the count time for the blank. The $\tilde{R}_b$ is an estimated value of the blank count rate (as, in practice, R_b is usually not well known), which is the weighted average of the measured blank and the gross count rates (because the gross one is the blank, under H_0).

Due to the corresponding statistical weights, w_b and w_g, three different decision rules have been derived. The combination: $w_b = 1$, $w_g = 0$, leads to the commonly used expression for the critical net count (formula A, in MARLAP [1] notation), equivalent to the equations published by several authors [2], [8], [9]:

$$\text{A:}\quad L_c = z_{1-\alpha}\sqrt{N_b\frac{t_s}{t_b}\left(1+\frac{t_s}{t_b}\right)} \quad (6)$$

Two other expressions, obtained for the other values of w_b and w_s, are formula B [10]:

$$\text{B:}\quad L_c = \frac{z_{1-\alpha}^2}{2} + z_{1-\alpha}\sqrt{\frac{z_{1-\alpha}^2}{4} + N_b\frac{t_s}{t_b}\left(1+\frac{t_s}{t_b}\right)} \quad (7)$$

and formula C [5], [11]:

$$\text{C:}\quad L_c = \frac{z_{1-\alpha}^2 t_s}{2t_b} + z_{1-\alpha}\sqrt{\frac{z_{1-\alpha}^2 t_s^2}{4t_b^2} + N_b\frac{t_s}{t_b}\left(1+\frac{t_s}{t_b}\right)} \quad (8)$$

For low mean blank counts, *Stapleton* aproximation [12] appears to outperform all of the above approaches. The critical value is:

$$L_c = d\left(\frac{t_s}{t_b}-1\right) + \frac{z_{1-\alpha}^2}{4}\left(1+\frac{t_s}{t_b}\right) + z_{1-\alpha}\sqrt{(N_b+d)\frac{t_s}{t_b}\left(1+\frac{t_s}{t_b}\right)} \quad (9)$$

where d is the parameter with a near-optimal choice $d = 0{,}4$, for $\alpha = 0{,}05$.

Poisson counting statistics also permits an "exact" test [11] for radionuclide detection, in which error type I rate is guaranteed to be no greater than the chosen value of α, although it may be less. In the nonrandomized version of the test [12], the H_0 is rejected if $N_g > y_c$, where y_c is equal to the smallest nonnegative integer n, such that:

$$\sum_{k=0}^{n}\binom{N_b+k}{N_b}\left(\frac{t_s}{t_s+t_b}\right)^k \geq (1-\alpha)\left(\frac{t_s+t_b}{t_b}\right)^{N_b+1} \quad (10)$$

Then, the critical net count is $y_c - N_b\,(t_s / t_b)$.

If the net signal N_s is normally distributed, the minimum detectable net signal, L_D, is determined implicitly by the equation:

$$L_D = L_c + z_{1-\beta}\sqrt{\sigma^2(N_s|\mu = L_D)} \qquad (11)$$

where $\sigma^2(N_s|\mu = L_D)$ denotes dispersion of the measured net signal, when the true mean signal equals L_D. This equation is the basis for the formulas that are commonly used for L_D, when the *Poisson*-normal approximation is assumed [1], [12]. If the only source of the signal variance considered is the *Poisson* counting statistics, the detection limit L_D is determined from the formula:

$$L_D = L_c + \frac{z_{1-\beta}^2}{2} + z_{1-\beta}\sqrt{\frac{z_{1-\beta}^2}{4} + L_c + N_b\frac{t_s}{t_b}\left(1 + \frac{t_s}{t_b}\right)} \qquad (12)$$

with L_c calculated using one of above presented formulas.

EXPERIMENT

The alpha particle counting was performed by the gas-flow proportional α / β counter LB 5100, series III, *Tennelec*, USA, with the $\alpha-$ counting efficiency $\varepsilon = 0{,}41$. The gas-flow detector uses P-10 mixture of 10% methane and 90% argon. The count times t_s and t_b in all experiments were 3600 s.

The detection limits were determined for drinking water, soil and air. After sampling, the tap water sample was preserved by acidification to pH 2 by nitric acid. The 500 ml aliquot was extracted and evaporated to constant mass, according to the procedures described in reference [13]. The soil sample was heated at 480°C for 3 h. The air sample was obtained by the air sampler operating at airflow rate of 100 l/min, for 50 h (the fractional efficiency of the filter was 90%). After the sampling, ^{222}Rn and ^{220}Rn short-lived progenies were allowed to decay for 5 days. Mass was determined on Scaltec SBC 31, with the accuracy of 1 mg.

RESULTS

In order to estimate the mean blank count rate R_b for $\alpha-$particle counting, 20 measurements of the blank sample have been performed. The obtained results were the following:

24 13 27 21 19 15 13 17 13 20 25 16 17 17 22 11 10 17 20 26

The mean value of the blank counts, calculated from the obtained data was $\overline{N}_b = 18{,}15$, and the corresponding variance $S_b^2 = 24{,}66$. The χ^2 – test showed that $S_b^2/\overline{N}_b$, for 19 degrees of freedom, was not significant ($0{,}3 > p > 0{,}1$). This means that the above set of data can be approximated by the *Poisson* distribution, therefore the formulas (3)-(12) for L_c and L_D could be applied.

As stressed previously, if the mean blank count rate, R_b, is well defined by the calculated experimental mean value of N_b ($\widetilde{R}_b = 5\cdot10^{-3}$ counts/s), the pure *Poisson* model, i.e. inequation (4), could be applied. Using the Fortran program, we have found that the condition (4) (with $\alpha = 0{,}05$) is fulfilled for $n \geq 26$. The critical net value is then: $L_c = 26 - 18{,}15 = 7{,}85$ counts.

Applying the formulas (6), (7) and (8), with $z_{1-\alpha} = 1{,}645$, we have calculated $L_c = 9{,}91$, $L_c = 11{,}36$ and $L_c = 11{,}36$ counts, respectively. The *Stapleton* approximation (the equation (9)), gives the critical value $L_c = 11{,}38$ counts. Using the exact test (the inequation (10)), we have obtained the result $y_c = 30$, which implies that $L_c = 11{,}85$ counts.

The value of L_c determined either by formula C or *Stapleton* approximation appears to be the most correct for our experiment, where neither too high nor too low value of $\overline{N}_b$ is observed. Namely, formula A is recommended for higher values of $R_b t_s$, where error type I rate closely approaches the chosen value of α [12]. Also, we have rejected the pure *Poisson* counting result, $L_c = 7{,}85$ counts, since it is only applicable for a well defined true R_b [1], which is not guaranteed in our case of only 20 measurements.

In further calculations, the decision treshold value $L_c = 11{,}36$ counts is substituted in equation (12), giving the detection limit $L_D = 24{,}15$ counts.

The MDC, actually representing the minimum detectable activity concentration [4] (in Bq/l, Bq/kg or Bq/m^3) is then determined as: MDC = L_D/A, where sensitivity A in our case is the product of: the detector efficiency ε, the sample counting time t_s and the analysed fraction of volume (or mass) of the sample.

CALCULATION OF MDC FOR WATER, SOIL AND AIR

Water – After evaporating the volume $V = 0{,}5$ l of drinking water, we obtained $m_1 = 0{,}155$ g for the solid residue, and separated $m_2 = 0{,}118$ g for further analysis. We observed 24 gross counts, for $t_s = 3600$ s, which implies that $N_s = 24 - 18{,}15 = 5{,}85$ counts. Comparing this result with the critical value, it can be concluded that the signal is not detected, as $N_s < L_c$. The MDC for the drinking water is:

$$\text{MDC} = \frac{L_D}{\varepsilon \cdot t_s \cdot V \cdot m_2/m_1} = 0{,}043 \text{ Bq/l}.$$

This value meets the requirement of American *Federal Register* [14] for drinking water MDC, which is 0,111 Bq/l, and it is close to the value of MDC = 0,037 Bq/l, of *Brookhaven laboratory* [13].

Soil – After heating the initial mass $m_1 = 17{,}419$ g of the soil, the mass $m_2 = 12{,}225$ g remained, and $m_3 = 0{,}975$ g was separated for α – counting. The number of observed gross α – counts was $N_g = 56$, leading to the value $N_s = 37{,}85 > L_c$, and the net signal was then considered as detected. The MDC for the soil was calculated as:

$$\text{MDC} = \frac{L_D}{\varepsilon \cdot t_s \cdot m_1 \cdot m_3/m_2} = 11{,}7 \text{ Bq/kg}.$$

Air – Five days after the sampling, the air filter was analysed for total α activity. The activity concentration of $C = 1{,}28 \cdot 10^{-4}$ Bq/m^3 was detected. The minimum detectable concentration of the air particulate was then calculated as [15]:

$$\text{MDC} = \frac{L_D}{\varepsilon t_s K F t_f} = 6{,}06 \cdot 10^{-5}\ \text{Bq/m}^3,$$

where F denotes the airflow rate, K is the fractional filter efficiency and t_f denotes the duration of the air sampling.

CONCLUSION

In the performed α – counting experiment, the observed blank counts follow the *Poisson* statistics. The most applicable *Poisson*-normal approximations for detection limits evaluation, for the obtained experimental mean value of the blank counts, appeared to be those of *Nicholson* (formula C) and *Stapleton*. Comparing the measured net signal with the calculated critical value, it was concluded that there are no α –emmiters in drinking water, contrary to the detected presence of the α –emmiting radionuclides in the soil and in the air.

The presented results indicate that the minimum detectable concentration (MDC) is not only instrumentaly influenced by the blank signal, the counting efficiency and the counting time, but it also depends on the characteristics of the sample (the fraction of volume or mass, etc). For three samples, drinking water, soil and air, analysed in this paper, the calculated values of MDC are: 0,043 Bq / l, 11,7 Bq / kg and $6{,}06 \times 10^{-5}$ Bq/m^3, respectively. The differences between the obtained results originate from different amounts of the analysed samples that were used to cover the detector surface in a thin slice geometry.

REFERENCES

1. Multi-Agency Radiological Laboratory Analytical Protocols (MARLAP) draft (2003)
2. American National Standards Institute (ANSI), N13.30 (1996)
3. U. S. Nuclear Regulatory Commision (NRC), NUREG/CR-4007, Washington (1984)
4. U. S. Nuclear Regulatory Commision (NRC), NUREG-1507, Washington (1998)
5. International Organization for Standardization (ISO), ISO 11929, Part 1-2, ISO, Geneva, Switzerland (2000)
6. L. A. Currie, *in* International Atomic Energy Agency IAEA-TECDOC-1401, pp. 9-34 (2004)
7. G. F. Knoll, Radiation detection and measurement, New York, John Wiley & Sons (1979)
8. L. A. Currie, *Analytical Chem.* **40** (1968) pp. 586-593
9. D. J. Strom, P. S. Stansbury, *Health Phys.* **63** (1992) pp. 360-361
10. J. E. Turner, Atoms, radiation and radiation protection, New York, John Wiley & Sons (1995)
11. W. L. Nicholson, AEC Research and Development Report HW-76279 (1963)
12. D. J. Strom, J. A. MacLellan, *Health Phys.* **81** (1) (2001) pp. 27-34
13. Brookhaven National Laboratory, 1998 Site environmental report, Appendix B (1998)
14. Federal Register, **65** (78) (2000) p. 21618
15. U. S. Nuclear Regulatory Commision (NRC), NUREG-1400, Washington (1993)

GRANICE DETEKCIJE ZA BROJANJE ALFA ČESTICA

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U radu su određene granice detekcije za merenje radioaktivnosti pomoću brojača α / β *čestica. Prikazana su različita pravila odluke i primenjena pri izračunavanju kritične vrednosti. Vrednost dobijena korišćenjem najbolje aproksimacije za obavljeni eksperiment brojanja* α *čestica, primenjena je u proračunu minimalne merljive koncentracije za: vodu, zemlju i vazduh.*