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A method of rapid testing of radioactivity of different materials

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ABSTRACT

A new method for the detection of low-level ionising radiation in solid, liquid or loose materials, which is based on the use of the Bayesian approach for the estimation of probabilistic parameters and a special statistical criterion, is offered in the present paper. We describe the algorithm and show the advantages of the method. The approach can be effective even in the case of extremely low signals whose intensity is much less than the background radiation.

1. Introduction

Ionising radiation is one of the major natural and man-made factors affecting human life and health. Due to recent changes in the conceptual approach, the problem of radiation safety does not only apply to the control of a limited number of potentially dangerous objects (plants and laboratories of nuclear fuel cycle, research and defence facilities of the appropriate profile, etc.), but is becoming more global (Marhulys & Bregadze, 2000). In particular, in the case of the building industry, up to 70% of radiation is contributed by natural gamma-emitting radionuclides contained in materials used and, as a result, there is uncontrolled proliferation of these radionuclides in building construction, including walls and ceilings of residences.

The activity concentrations are determined by gamma-ray spectrometry using high-purity germanium detectors (HPGe) and a multichannel analyser. To reach the highest level of accuracy, some researchers (Al-Saleh and Al-Berzan, 2007) conduct the measurement of the samples studied with an accumulating time for about 80,000 s.

Measurements of low-level radioactivity often give results in the order of the detection limit. For many applications it is important to concentrate on multi-isotope analyses of samples with low-level radioactivity. How to measure such kinds of samples? This requires the development of a special analytical approach. To overcome difficulties associated with

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the detection limit, some researchers have appealed to Bayesian statistics, a method that allows statistical inference on nuclide ratios taking into account both prior knowledge and all information collected from the measurements (e.g. Kacker, 2006; Zähringer & Kirchner, 2008; Kirchner, Steiner, & Zähringer, 2009; Dalal & Han, 2010; Qingpeia et al., 2013). Methods based on Bayesian statistics allow quantitative conclusion regarding counts of single isotopes whose activity is low compared to the background radiation. The application of such new approach is illustrated by a number of examples of environmental low-level radioactivity measurements (Kirchner et al., 2009). Qingpeia et al. (2013) note that their sequential Bayesian approach offers the advantages of shorter verification time during the analysis of spectra that contain low total counts, especially in complex radionuclide components.

In particular, Kirchner et al. (2009) disclose details of their method based on the Bayes' theorem. The Bayes theorem is written for the given problem as $f_A(a|X=x) = c(x)f_A(a)f_X(x|A=a)$ where $f_A(a)$ denotes the probability of the unknown A based on information available before the measurement is performed (the prior), $f_A(a|X=x)$ is the conditional probability of A under the condition that event x has been measured (the posterior), $f_X(x|A=a)$ is the conditional probability of measuring x given A, which constitutes the information gained from the measurement (a nuclear disintegration counting), and $c(x)$ is a normalization function. A is conceptualized as a random variable (with realisations a), which is in contrast to the conventional approach. Then the following expression for the probability $P(S)$ of the activities is used, which is originated from a suspected radioactive source S,

$$\begin{aligned} P(S) = \int D(a_1, a_2, \ldots, a_N)^* f_A(a_1, a_2, \ldots, a_N)|X \\ = (x_1, x_2, \ldots, x_N) da_1 da_2 \ldots da_N \end{aligned} \tag{1}$$

where $f_A((a_1, a_2, \ldots, a_N)|X = (x_1, x_2, \ldots, x_N))$ denotes the joint probability density distribution of the posterior of the N isotopes established after a measurement, and $D(a_1,a_2,\ldots,a_N)$ is a decision criterion with $D = 1$ if the activity ratios are consistent with a suspected source S and zero elsewhere.

Thereby for calculation of the probability, the researchers who used the conventional Bayesian approach described above have to utilise a number of trial functions $f_A(a|X=x)$, which are integrands in expression (1). Each next calculation requires a set of new such trial functions.

On the other hand, Zabulonov and Burtniak (2008) argued that measurements of low-level radioactive samples of nonorganic and organic origin can reliably be performed only by special dosimetric and spectrometric instrumentation. They also mentioned that the detection of a low-level radioactive source is complicated by the presence of an existing background radiation, because the intensity of radiation of materials contaminated with radioisotopes is hidden in the natural background and the Compton scattering. These peculiarities make the timely detection of low-intensive radioactive sources unlikely.

Functional capabilities of specialised technical equipment which is now used for radiation monitoring of materials, also do not allow one to realise the problem of detection and control of unauthorized movement of low-level radioactive materials that are characterized by occasional, short and slight excess signals above the background. Therefore the solution of such problems rather requires a conceptually new approach. The new approach to the measurement of low-level radioactivity must appreciate not only technical and functional capabilities of the equipment, but also the algorithmic basis with appropriate software based on Bayesian statistics. Such approach is presented in the given work.

2. Methods

The most significant contribution to the realization of maximum sensitivity of the technical equipment can be reached by using both the efficiency of detectors that record the radiation as well as the algorithm that processes available statistical data.

In practice among the methods of analysis of radiation, most used spectrometric approaches allow the identification of sources of radiation. The spectrometric method is based on the measurement of the energy spectrum of radiation sources. As a result of the measurement one obtains not a true gamma spectrum, but the so-called discrete spectrum of radiation, which is a histogram of the distribution of pulses by energy channels of the analyser in accordance with the channels' amplitudes. Using this spectrometer one can determine both the number of pulses and the energy of each pulse.

In spectrometric devices primary information comes in the form of a random sequence of pulses from the detectors that record radiation. In addition to the registration of useful events, such information contains a number of obstacle signals caused by background radiation, electromagnetic fields, etc. leading to uncertainty. Thus the main task, which must be implemented through the technical facilities, is to detect slight increases in the radiation fields in places of observation and control, as well as the identification of the appropriate sources.

Note we are talking about a multichannel scaling data, which we use in our practice, and not the much more common "differential pulse height" spectrum.

Mathematically, the problem of detection and identification of radiation can be described as follows. Suppose, in a time $t \in [0, T]$ of continuous observation of a source of radiation we record n radioactive particles. The measurement forms a selection $x = (x_1,x_2,\ldots,x_n)$ of the general population and the allocation of each x_i are described by the Poisson distribution. The sample x is between fixed values X_0 and H_m. The chance of getting the measured value of x in the interval from X_0 to H_m is described by the distribution function (Janossy, 1965).

Let us denote the frequency of events of "getting radiation" in the bit interval $x_i \in (X_{j-1},X_j)$ as N_j. A statistical series grouped in such a way is the so-called histogram – a statistical analogue of the distribution curve. If each bit interval is plotted in correspondence with the energy of the registered particle, we obtain the spectral distribution of energy radiation.

While monitoring and controlling the source of radioactivity by the method of spectrometric analysis it is necessary to distinguish the background spectrum from the signal or spectrum that belongs to the radiation source. That is, one should identify the sudden appearance of radiation of a radioactive source by analysing spectra (the background and the source of radiation), which are obtained by observation. Or, on the basis of statistical data one has to reveal a jumpy change in the spectrum at low external influences. If a source of radiation of low activity (whose activity is at the level of the natural background) should be detected, the problem becomes so complicated that standard methods are unable to do this. To resolve the problem, i.e. to decipher complicate spectra, we propose a new approach based on the method of probabilistic analysis of histogram spectra of radiation below. The histogram spectra are constructed by using Bayesian statistics.

To control the sudden appearance of active radioactive sources, which is at or slightly higher than the background, we will use the Bayesian approach for the estimation of unknown parameters.

While doing this, we need to find the distribution of a parameter μ (considered as a random variable) with an available observation x. By Bayes' theorem, a posteriori distribution is calculated from the a priori probability distribution with density p (μ) and the likelihood function p $(x|\mu)$ by the formula (see, e.g. Chen, 2013):

$$p(\mu|x) = \frac{p(x|\mu)p(\mu)}{\int p(x|\mu)p(\mu)d\mu}. \tag{2}$$

If the posteriori distribution p $(x|\mu)$ belongs to the same family of probability distributions as the a priori distribution p (μ) (i.e., has the same form, but with different parameters), this family of distributions is called a paired family of likelihood functions p $(x|\mu)$. In Bayesian statistics a posteriori calculation of probabilities is greatly simplified for conjugate families of distributions.

Let the random selection x be as described by a distribution with unknown mean μ and known variance σ^2 (according to the central limit theorem, when $n \to \infty$ the Poisson distribution passes into the normal distribution). The a priori distribution of the parameter μ describes the normal distribution with expectation μ_0 and variance σ_0. Then for conjugate families of distributions a posteriori distribution of the parameter μ is normal with an average

$$\alpha = \left(\frac{\mu_0}{\sigma_0^2} + \frac{\sum_{i=1}^{n} x_i}{\sigma^2}\right) \Big/ \left(\frac{1}{\sigma_0^2} + \frac{n}{\sigma^2}\right), \tag{3}$$

and the dispersion

$$\beta = \left(\frac{1}{\sigma_0^2} + \frac{n}{\sigma^2}\right)^{-1}. \tag{4}$$

How is eq. (3) derived using eq. (2)? This is known information available in the literature (e.g. DeGroot, 1970, 2004; Sorenson, 1980). Relation (4) is one of the parameters of the a posteriori distribution.

Expressions (3) and (4) are very important for the processing of data obtained at measurements. First, we get a spectrum from the scintillation detector and this spectrum becomes our primary data for further processing. Second, these types of data arrive continuously (e.g. every second) and are a characteristic of an objective process that we investigate. Third, in the spectrum the background component is constantly present, as the background uninterruptedly fluctuates, and at the same time the information on a radioactive source may also be present, which we wish to measure. Fourth (this is important!!), a sequence of spectra coming from the detector is a united family of distributions (this state is one of the majors in the Bayesian statistics). These remarks allow us to calculate the parameters of the posteriori distribution using relations (3) and (4).

In radiation control and monitoring of sources of radiation by the method of spectrometric analysis, the information from detectors arrives as a series of spectra $S = \{S_1, S_2, \ldots, S_i\}$ $i = \overline{1, \infty}$. Under S_i we understand a histogram (spectrum) composed of data x_i during a specified time, i.e. a (given) discrete time interval $\Delta t = t_i - t_{i-1}$ during which pulses are accumulated (for instance, this can be 1 min). Each spectrum S_i arrives at the processing system in the said time interval $\Delta t = t_i - t_{i-1}$ that determines the time within which the presence of a source of radiation has to be detected. To determine parameters of the a priori distribution of a signal $x \in S_i$, we will use the prehistoric information. Let us consider the set of measurements for the previous period $x \in S_{i-1}$. The size of selections (spectra S_i), which are considered, is independent of the accumulation time and the intensity of radiation and depends only on the detection device used. For modern HPGe detectors the selection size (or the number of channels) is constant and can be equal to 1024, 2048, 4096, 8192 or even more values.

To identify and determine the source of radiation in the field of the detector, the intensity of which is slightly higher than the background level (or even slightly less), it is necessary to formulate a statistical criterion (see, e.g. Wit, van den Heuvel, & Romeyn, 2012).

If a Bayesian average μ of pulses x of radiation in the S_i spectrum deviates from the mean value μ_0 in the S_{i-1} spectrum by three standard deviation values for the Bayesian average, then a source of radiation is present:

$$|\alpha - \mu_0| \geq K \cdot \beta, \quad K = \overline{1,\ 3} \tag{5}$$

where β is defined in relation (4). The criterion (5) has allowed us to develop a method and algorithm for detecting the sudden appearance of radioactive sources. Namely, K can vary from 1 to 3 depending on the required accuracy or the probability of detection of a radioactive source. K is the number equal to the quantity of mean-square deviations, which is set by the operator and stored in memory. In other words, the threshold, which identifies the source of radiation (it is given in the criteria (5) as $K \cdot \beta$), is a quantile of the normal distribution: we consider a family of conjugate distributions for Bayesian statistics with the function of plausibility for the normal distribution.

Once again, we introduce the criterion (5) to compare the spectra and substitute the calculation of cumbersome integrals in the Bayes' expression for simple algebraic manipulations. We do not investigate parameters of the Bayesian statistics as such; we propose a simple algorithm on the basis of this statistics, which also takes into account possible statistical errors. Such an algorithm can be implemented in controllers and due to recurrence it is able to work in real time.

Almost all values of the normally distributed random variable are in a range $[x - 3\sigma, x + 3\sigma]$; more strictly: starting from the probability of about 0.9973, values of the normally distributed random variable fall within this range. Also the probability is known for values of $K = 1$ and $K = 2$. The method is written, for example, in the monograph (Shmoylova, Minashkin, & Sadovnikova, 2011).

Our approach is associated with the introduction of the algorithm described below that collects the information step-by-step and which generates a reliable result.

The algorithm includes the following steps:

Step 1. We expect a priori parameters of the distribution (a priori mean value μ_0 and the mean square deviation σ_0) on the basis of spectra $S_1, S_2, \ldots, S_{i-1}$ by the formulae

$$\mu_0 = \frac{1}{N}\sum\nolimits_{i=1}^{N} S_i, \tag{6}$$

$$\sigma_0 = \sqrt{\frac{1}{N-1}\sum\nolimits_{i=1}^{N}(S_i - \mu_0)^2}, \tag{7}$$

where S_i is the integral value of the spectrum, N is the dimension of the preview window $(N << (i-1), \quad i = \overline{0,\ \infty})$.

Step 2. On the basis of the $S_1, S_2, \ldots, S_{k-1}, S_i$ spectra, we can calculate parameters $\overline{x}$ and σ by using eq. (3). Namely, let us determine

$$\overline{x} = \sum_{i=1}^{N} S_i; \tag{8}$$

then eq. (3) can be rewritten as follows

$$\alpha = \left(\frac{\mu_0}{\sigma_0^2} + \frac{\overline{x}}{\sigma^2}\right) \Big/ \left(\frac{1}{\sigma_0^2} + \frac{n}{\sigma^2}\right) \tag{9}$$

where the parameters μ_0 and σ_0 are calculated by expressions (6) and (7), respectively; σ is calculated by the recurrent algorithm.

The algorithm works with a sliding window, i.e. indexes can run inside the window, but the width of the window does not change during operation (it is a constant). The window is given at the beginning of the work when the operator sets data. In step 2 one does not see the index K, as the value of K has been set in step 1.

Step 3. Determine the values of the α and β parameters by relations (3) and (4) of the a posteriori distribution.

Step 4. Use the statistical criterion of identification of the emergence of a source of radiation. If $|\alpha - \mu_0| \geq K\beta,\ \ K = \overline{1,\ 3},$ then we can determine the presence of a source of radiation. Otherwise, no source.

Step 5. Increment the index $i \rightarrow i+1$. Wait for the arrival of a new spectrum, S_{i+1}, and go to Step 1 for the study of data of the new spectrum.

Thus, spectra are coming from the sensor. We may assume that when the device is switched off, only background radiation is coming during some time (for example, from 5 s to 5 min). During this interval we operate with a priori distribution. After that a posteriori distribution accounts for the relevant evidence related to the particular case being examined. It is this spectral distribution in which we have to determine whether a radioactive source is present or not. If the radioactive source has not been revealed, the distribution is attributed to a priori distribution and the next one will be treated as a posteriori distribution. The algorithm is based on this the principle.

3. Results

While studying the proposed method, a series of test experiments has been conducted. Measurements have been performed by using a spectrometer with the BDEG type, 4–31 scintillation detector (normally used for detecting gamma-ray radiation). The system used to carry out measurements is of course typical for that used by other researchers. The difference is only in the use of the algorithm for detecting a source of radioactivity.

One can ask, how is the background taken into account? The algorithm works as follows: For some time we assume that we measure only the background and that at any moment a radioactive source may appear. That is, all the time we compare two spectra that are neighbours through the time of accumulation.

Results obtained at measurements in times t_i and t_{i+1} of histograms of energy radiation with the same area (the values obtained during the 1-min measurement) are compared with the criterion (5) for $K = 3$ in the above algorithm. The value of criterion A_i for each moment of time t_i of measurements is calculated. Initial spectra and trends are depicted in Figs. 1–3, right and left, respectively. In the experiment 1 (Fig. 1), we calculate the criterion for the background radiation.

In the experiment 2 (Fig. 2), we introduce a testing point source of gamma radiation (^{137}Cs, 661.7 keV) in the field of vision of the detector for 10 s. The criterion is calculated for this case as well. The additional radiation source is introduced at 31, 91 and 121 min; the time of accumulation of the spectrum is 1 min. Here, the additional radioactivity is far above the background level; hence any method can succeed in detecting it. The example is chosen simply for the demonstration of our approach.

In the experiment 3 (Fig. 3) we calculate the criterion at a short (less than 5 s) and longer (over 10 s) insertion of the same gamma source, though its intensity was very low, at the level of the background. The adding radiation source is run at 11 and 51 min. To calculate the criterion, we use the viewport range $N = 5$ (the number of histograms), which correspond to the 5-min time.

For the experiments, we have chosen a window length of 5 min. This value is set at the beginning. The selected value of $N = 5$ depends only on the time required for training of the system. During this time we cannot identify the source of radioactivity; we can only measure the background. After 5 min we may start to analyse the presence of the source.

When processing the background, i.e. the experiment 1 (Fig. 1), it is not clear whether the source of radioactivity is present or not.

In processing data of the experiment 2 (Fig. 2), a source of radiation has been in sight of the detector 3 times and the

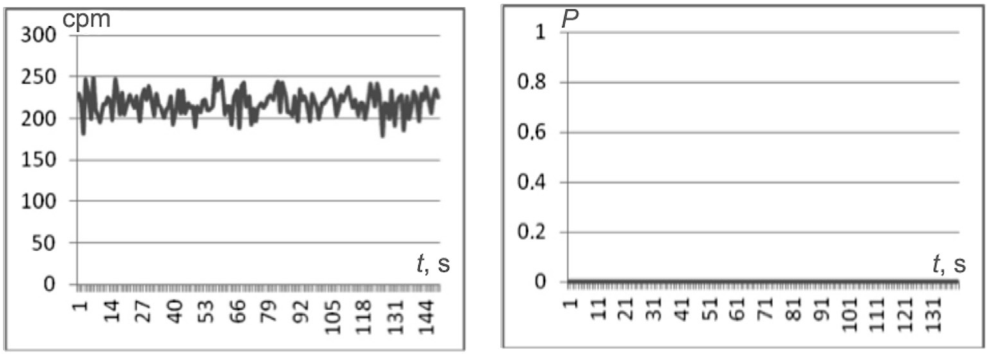

Fig. 1 – Experiment 1. Left: Data of background radiation; the units on the *x*- and *y*-axis are time (in seconds) and the counts per minute, respectively. Right: Trend values of the criterion (5) for background radiation; the units on the *x*- and *y*-axis are time (in seconds) and the probability, respectively. *P* is the feature whether have we revealed a radioactive source ($P = 1$) or not revealed ($P = 0$). This is checked by the criterion (5). The left chart shows how the spectrum changes over time, and the right chart shows how the algorithm is working (analogously for Figs. 2 and 3).

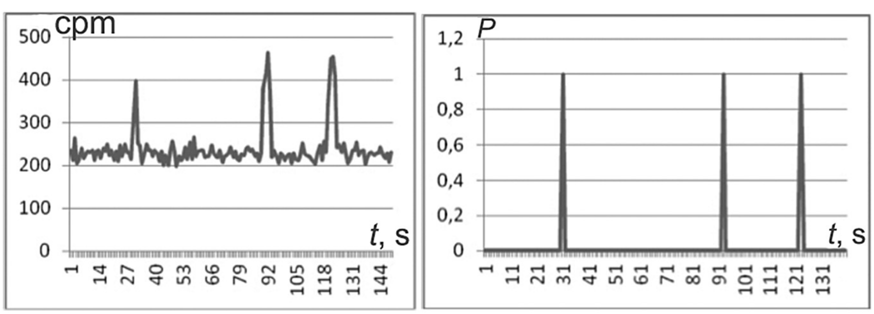

Fig. 2 – Experiment 2. Left: Data of background radiation; the units on the *x*- and *y*-axis are time (in seconds) and the counts per minute, respectively. Right: Trend values of the criterion (5) for background radiation; the units on the *x*- and *y*-axis are time (in seconds) and the probability, respectively.

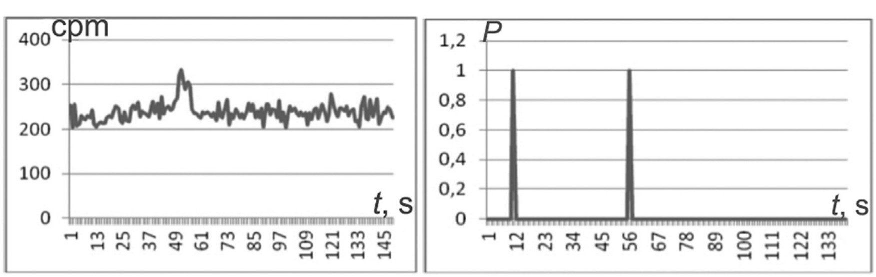

Fig. 3 – Experiment 3. Left: Data of background radiation; the units on the *x*- and *y*-axis are time (in seconds) and the counts per minute, respectively. Right: Trend values of the criterion (5) for background radiation; the units on the *x*- and *y*-axis are time (in seconds) and the probability, respectively.

source has been visible by the detector in total for more than 10 s.

In processing data of experiment 3 (Fig. 3), the radiation source has been in sight of the detector only once (during at least 3 s). The next time when the source got in sight the detector for at least 3 s, nothing is happening.

In the experiment 3 shown in Fig. 3 we have used the same source of radioactivity, as was the case in experiment 2 (Fig. 2). However, in experiment 3 (Fig. 3), the source of radioactivity has significantly been screened by a metal plate and the intensity of the source did not exceed the background of gamma-ray radioactivity.

4. Conclusions

We have proposed the probabilistic method and the algorithm for detecting the sudden appearance of radioactive sources in sight of the detector, which are based on the use of both the Bayesian approach for the estimation of parameters as well as the use of a special statistical criterion. The method allows us to detect abrupt changes in the integral value of the background radiation intensity (of the total size of selection, or the area of interest) when the value exceeds the allowable limit.

Our R&D team has successfully been using the described method for about 30 years applying it even at the most difficult situations when any other approach becomes inapplicable. For example, in the beginning of 1990s, using this method we created a detailed map of radioactive contamination of a 2500 km^2 area around the destroed Chornobyl nuclear power plant (the measuring equipment was installed on board of a drone). A number of devices were designed: a special chair, the device "Screener" designed for measuring human radioactivity, which won a golden medal at the medical exhibition in Brussels in 1993; a portable workstation, the device "Vector" designed for integrated environmental radiation monitoring, which was recognized as the best by the United Nations in the beginning of 2000s; the device "Food Light" for measuring of radioactive isotopes in food, which won a tender in Japan after the Fukushima nuclear desastar in 2011, and is used now there; etc.

The proposed probabilistic method for detecting a low-level radioactive source has a number of advantages. The method: i) evaluate the degree of inconsistency of histograms in real time; ii) is characterised by high efficiency, allowing one to record the appearance of any source of radiation, even when the source is moving; iii) does not require additional technical equipment; a special analytical software is enough; iv) can be applicable to any other kinds of measurements of low-intensive signals of any nature (satellite antennas, spaceship communications, inerton field measurements, etc.).

Besides, the described method is the most sensitive one, which makes it possible to reveal a source of radiation even when the signal to noise ratio is 1:1000 (the simple sensitivity criterion is given by the coefficient K (5) and the measuring time is a few seconds), though all other best methods are able to distinguish a signal only at the signal to noise ratio equal to 1.5:1 (which also requires a long sighting time, up to 10 min).

Moreover, the proposed method can be used in a number of applications dealing with other kinds of signals, such as infrared, radio, microwave and terahertz including signals associated with an inerton field (Krasnoholovets, 2014). This method is indispensable for the use at artificial satellites when the detector's sighting time is only 2–3 s.

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