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Nuclear Instruments and Methods in Physics Research A 564 (2006) 308–314
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

Simplex optimization of artificial neural networks for the prediction of minimum detectable activity in gamma-ray spectrometry

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Received 7 November 2005; received in revised form 12 March 2006; accepted 28 March 2006
Available online 11 May 2006

Abstract

A three-layer feed-forward artificial neural network (ANN) with a back-propagation learning algorithm was used to predict the minimum detectable activity (A_D) of radionuclides (^{226}Ra, ^{238}U, ^{235}U, ^{40}K, ^{232}Th, ^{134}Cs, ^{137}Cs and ^{7}Be) in environmental soil samples as a function of measurement time. The ANN parameters (learning rate, momentum, number of epochs, and the number of nodes in the hidden layer) were optimized simultaneously employing a variable-size simplex method. The optimized ANN model revealed satisfactory predictions, with correlation coefficients between experimental and predicted values 0.9517 for ^{232}Th (sample with ^{238}U/^{232}Th ratio of 1.14) to 0.9995 for ^{40}K (sample with ^{238}U/^{232}Th ratio of 0.43). Neither the differences between the measured and the predicted A_D values nor the correlation coefficients were influenced by the absolute values of A_D for the investigated radionuclides.

PACS: 29.30−h; 29.30.Kv; 29.90.+r

Keywords: ANN; Radionuclides; Minimum detectable activity; Simplex; Soil

1. Introduction

A performance criterion of gamma-ray spectrometry is the minimum detectable activity (A_D), defined qualitatively as the smallest amount of radionuclide that can be reliably determined, given the prevailing conditions of the particular spectral measurement. According to Currie [1], the activity value for which the false negative error is β, giving such a critical activity value A_C that the probability of the estimated quantity exceeding A_C is not greater than α if analyte is absent ($A_C = 0$, null hypothesis; α is the false positive error), is an accurate quantitative definition. In another formulation, the A_D is the true net activity for which the probability that the estimated value does not exceed A_C is β, where A_C is the minimum significant value of an estimated activity for which the probability that A_C exceeds the null net activity is α. Values $\alpha = \beta = 0.05$ (recommended within the analytical chemistry community) have been adopted in gamma-ray spectrometry for the computation of A_D (95% confidence level) [1].

Recently, artificial neural networks (ANNs) have been used in gamma-ray spectrometry for automatic identification of radionuclides from their spectra [2], prediction of uncertainty [3] and peak-to-background ratio [4], as well as for quantitative spectrometric analysis [5–7]. In our work, the ANN model for the prediction of the minimum detectable activity of radionuclides in soil samples as a function of measurement time was optimized using the simplex method.

In our previous work [3,4], the network was optimized by a "trial and error" method. This procedure for the optimization of ANN parameters can be very slowly convergent since the variables are usually not fully independent [8]. By simultaneous optimization of the ANN parameters this problem can be avoided. In this study, the widely used simplex algorithm proposed by Nelder and Mead [9] was applied to optimize the ANN parameters, because of the algorithm's simplicity and efficiency. The ANN model optimized by the simplex method was employed to predict the minimum detectable

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doi:10.1016/j.nima.2006.03.047

activity of radionuclides in environmental soil samples of different compositions.

2. Experimental

2.1. Gamma-ray spectrometric measurements

The measurements were performed using an ORTEC-AMETEK gamma-ray spectrometer (34% relative efficiency and 1.65 keV FWHM for ^{60}Co at 1.33 MeV, 8192 channels) shielded by 100 mm low-level (less than 50 Bq kg^{-1}) lead, including an interior lining consisting of 2 mm Cu, 1 mm Cd, and 4 mm Perspex. A mixed calibration source (MBSS 2) from the Czech Metrological Institute was used for efficiency calibration using the same geometry as the soil samples. For the purpose of quality assurance, independent calibration checks were run using two standard reference materials from the International Atomic Energy Agency (IAEA-385) and Environmental Measurement Laboratory, United States Department of Energy (QAP-9803) for intercomparison. Soil samples were measured in 1 L Marinelli beakers for 17 h. After that, no significant increase in the minimum detectable activity was observed. The obtained spectra were stored after each hour of measurement. The spectra were processed by Gamma Vision 32 software, which performs a simultaneous fit to all significant photopeaks appearing in the spectra [10].

The minimum detectable activities for the following radionuclides detected in soil samples were analyzed: ^{226}Ra (351.9 keV emitted by ^{214}Pb), ^{238}U (92.6 keV emitted by ^{234}Th), ^{235}U (143.9 keV), ^{40}K (1460.8 keV), ^{232}Th (911.2 keV emitted by ^{228}Ac), ^{134}Cs (795.8 keV), ^{137}Cs (661.6 keV) and ^{7}Be (477.6 keV). The minimum detectable activity (A_D) was computed according to the following equation:

$$A_D = \frac{2(1+2B)^{1/2}+1}{\varepsilon I_a t m} \tag{1}$$

where B is the number of background baseline counts under the gamma-ray photopeak in the region of interest, ε is the detection efficiency at the photopeak energy in question, I_a is the absolute intensity of gamma-ray transition (%), t is the time of measurement, and m is the sample mass (kg) [10].

2.2. Artificial neural network

Since the three-layer feed-forward neural network with a back propagation algorithm had proved satisfactory in our previous work [3,4], it was also used in current study.

Minimum detectable activities for eight radionuclides from three different measurement times were used to train the network (Table 1). A training set, which adequately represents the whole data set, is imperative. The "leave-10%-out" method was applied for the cross-validation—an indication of whether or not memorization was taking place. This method was shown to be the most suitable for the cross-validation in our previous work [3,4]. When the ANN memorizes the training data, it produces acceptable results for current data, but poor results when tested on unseen ones [11,12]. The neural network model was tested to predict the A_D for radionuclides in samples with different ^{238}U/^{232}Th ratios, not used in the training phase.

Table 1
ANN training data set

t (h)	A_D (Bq kg^{-1})							
	^{226}Ra	^{238}U	^{235}U	^{40}K	^{232}Th	^{134}Cs	^{137}Cs	^{7}Be
1	10.6	0.97	2.60	5.07	1.82	0.62	0.37	4.41
9	3.41	0.37	1.02	1.90	0.67	0.18	0.12	1.42
17	2.54	0.27	0.64	1.34	0.48	0.13	0.10	1.08

The root mean square error (RMSE) was used as the error function for finalizing the training and the testing process. It was computed from the following equation:

$$\mathrm{RMSE} = \frac{\sqrt{\sum_{i=1}^{n}(d_i - a_i)^2/n}}{x} \tag{2}$$

where d_i is the desired output (experimental values) in the training or testing set, a_i is the actual output (ANN's predicted values) in the training or testing set, n is the number of data in the training or testing set, and x is the average value of the desired output in the training or testing set.

2.3. Simplex algorithm

The Nelder and Mead simplex method [9] is implemented for solving n-dimensional unconstrained minimization problem to find $\boldsymbol{X} = \{x_1, x_2, \ldots, x_n\}$ which minimizes $f(\boldsymbol{X})$.

The geometric figure formed by a set of $(n+1)$ points in an n-dimensional space is called a simplex. The basic idea in the simplex method is to compare the value of the objective function f at the $(n+1)$ vertices $\{\boldsymbol{X}_i\}$ of a simplex and move the simplex gradually toward the optimum point during the iterative process. Starting from an initial simplex with $(n+1)$ known vertices $\{\boldsymbol{X}_i\}$, a new vertex will be computed to define a new simplex using reflection, expansion, or contraction. The flow chart is shown in Fig. 1.

If $\boldsymbol{X}_h$ is the vertex corresponding to the highest value of the objective function among the vertices of a simplex, we can expect the point $\boldsymbol{X}_r$ obtained by reflecting the point $\boldsymbol{X}_h$ in the opposite face of a simplex to have a smaller value. Mathematically, the reflected point $\boldsymbol{X}_r$ is given by

$$\boldsymbol{X}_r = a(\boldsymbol{X}_{os} - \boldsymbol{X}_h) + \boldsymbol{X}_{os} \tag{3}$$

where $\boldsymbol{X}_{os}$ is the centroid of all the points $\boldsymbol{X}_i$ except $\boldsymbol{X}_h$

$$\boldsymbol{X}_{os} = \frac{1}{n}\sum_{i=1, i\neq h}^{n+1} \boldsymbol{X}_i \tag{4}$$

and a is the reflection coefficient ($a>0$), defined as

$$a = \frac{||\boldsymbol{X}_r - \boldsymbol{X}_{os}||}{||\boldsymbol{X}_h - \boldsymbol{X}_{os}||} \tag{5}$$

where $\|\cdot\|$ denotes the Euclidian norm.