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Analysis of ^{7}Be behaviour in the air by using a multilayer perceptron neural network

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ABSTRACT

A multilayer perceptron artificial neural network (ANN) model for the prediction of the ^{7}Be behaviour in the air as the function of meteorological parameters was developed. The model was optimized and tested using ^{7}Be activity concentrations obtained by standard gamma-ray spectrometric analysis of air samples collected in Belgrade (Serbia) during 2009–2011 and meteorological data for the same period. Good correlation ($r = 0.91$) between experimental values of ^{7}Be activity concentrations and those predicted by ANN was obtained. The good performance of the model in prediction of ^{7}Be activity concentrations could provide basis for construction of models which would forecast behaviour of other airborne radionuclides.

1. Introduction

Worldwide daily gamma-spectroscopic analysis of airborne radionuclides provides a large amount of data about their activity concentrations and spatio-temporal distributions. Regardless of the production path, their concentrations show high fluctuations caused by various physical and meteorological properties of atmosphere. However, the establishing of the number of influencing parameters requires methods of advanced statistics, such as principal and independent component analysis. Artificial neural networks (ANN) can detect patterns within input data even in cases of large measurement errors or the presence of pronounced non-linearity in data. (Massart et al., 1998). The fact that ANNs do not rely on any assumption of relations between input variables presents an advantage over methods of component analysis or multivariate analysis. A number of algorithms have been developed based on neural networks with different network architectures (Gevrey et al., 2003; Rajer-Kanduč et al., 2003; Raimundo et al., 2003; Bechtler et al., 2001; Maier, 2000). Common to all types of networks is that each network must have one input and one output layer and at least one hidden layer allowing the network to describe the non-linearity of a given system (Agatonovic-Kustrin et al., 1998).

In the last two decades, neural networks have been used in various radiological applications. These include automatic identification of radioactive isotopes in gamma spectroscopy (Olmos et al., 1991), determination of parameters in monitoring of uranium enrichment (Vigneron et al., 1996), portable systems for rapid identification of radionuclides (Kangas et al., 2008; Keller and Kouzes, 1994; Keller et al., 1995), experimental nuclear structure physics (for the gamma ray tracking technique) (Akkoyun and Yildiz, 2012), optimization of gamma-spectrometric analysis parameters and monitoring of radioactive contamination of the environment (Pilato et al., 1999; Dragović et al., 2005, 2006; Dragović and Onija, 2005; Yoshida et al., 2002; Medhat, 2012).

The radionuclide ^{7}Be ($t_{1/2} = 53.3$ d) forms in the stratosphere and upper atmosphere in reactions of spallation on nuclei of oxygen and nitrogen by beams of protons and neutrons from cosmic radiation. Almost immediately after creation, atoms of ^{7}Be are captured by aerosol and follow their paths through the air masses. Aerosol contains most of the air pollutant particles, so they can be used as a tracer for circulation pathways in the atmosphere, as well as for deposition kinetics of atmospheric macro and micro particles (Todorovic et al., 1999; C. Rodenas et al., 1997).

It is well known that the production of ^{7}Be depends on the intensity of cosmic radiation reaching the upper atmosphere (Kikuchi et al., 2009). Seasonal variation of ^{7}Be in the lower layers of the atmosphere depends on the vertical and horizontal flow of air masses (Todorovic et al., 1999). Continuous daily monitoring of ^{7}Be

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activity concentration in surface air can provide information on solar activity (Kikuchi et al., 2009), since the ^{7}Be activity concentration in the air is independent of human activities, i.e. nuclear tests and technological development. In this study, we tested the ability of ANNs to predict activity concentration of ^{7}Be in near-ground air by using meteorological parameters as input.

2. Experimental

2.1. Sampling

Air sampling was conducted in the period from March 2009 to December 2011 at Kumodraž (44°44′24″N, 20°30′40″E), on the outskirts of Belgrade, Serbia. Air sampling was performed with two digital samplers DH 604EV.2 (F&J Speciality Products, Inc., Ocala, USA) located at positions separated by one meter. Samplers provided the air flow in the range of 15–120 m^3/h, with an initial air flow rate of 50 ± 5 m^3/h. The air samples were taken at the height of 124 cm from the ground. Samples were collected weekly with sampling time of six days, with a total of 101 samples. Digital samplers were capable of measuring temperature, pressure and relative humidity. Values for wind speed, insolation (sunny hours per day), global Sun warming and precipitation were provided by the Republic Hydrometeorological Service of Serbia (http://www.hidmet.gov.rs/ciril/meteorologija/klimatologija_godisnjaci.php).

Cellulose filter paper FJ213340 1.770 mm thick with an efficiency of 65% on the DOP (dioctyl phthalate) test was used. The DOP test was used to determine the efficiency of the filter with an area of 100 cm^2 for air or gas filtration with the flow rate of 32 L/min containing dioctyl phthalate particles at a concentration of 100 mg/L (Lazarević et al., 2009).

2.2. Gamma-ray spectrometric analysis

The measurements were performed on a gamma-ray spectrometric system AMETEK-AMT (ORTEC, USA) with a p-type coaxial high-purity germanium (HPGe) detector with relative efficiency of 59.2% measured on the line 1.33 MeV ^{60}Co. Resolution of the device was 1.78 keV on the line 1.33 MeV ^{60}Co. The detector was housed in home-made lead casing without cadmium layer. Lead protection was 11 mm thick and covered with a copper sheet 5 mm thick.

The gamma-ray spectrometer was calibrated for the filter geometry. The solution used for calibration was obtained by dilution of the reference IAEA material (Cert. No: 9031-OL-159/08), which had certified values for ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y and ^{60}Co. The obtained activity values for listed nuclides were in agreement with the certified corrected for dilution. The standard was made by dripping the radioactive solution on the circular filter paper in the hexagonal network. Fifty-five points were applied, each with a volume of 10 μL. Using the Gamma Vision 32 ver. 6.01 software package (ORTEC, Oak Ridge, USA, 2001), ^{7}Be activity concentration in air samples were evaluated from its line at 477.6 keV, with a relative standard deviation that ranged from 0.62 to 7.61%, for 250 000 s by.

2.3. Data analysis

2.3.1. Principal component analysis (PCA)

This is a method originating from multivariate statistical analysis, which allows for the identification of major factors within a multidimensional dataset. The PCA was performed using the SPSS v 13 software package (SPSS Inc., Chicago, USA, 2003) on the set of gamma spectra acquired during 2010. The extracted sources of spectral variance were tested on the correlation with meteorological parameters; the parameters corresponding to the highest correlation coefficients were selected and used as input for the NN analysis.

2.3.2. Neural network (NN)

The neural network is applied to problems where the relationships are complex or unknown. The most common type of NN used in the analysis of environmental samples is the multilayer perceptron. This type of NNs consisted of input, output and at least one hidden layer. They are easy for use and with low number of parameters. Three-layer and four-layer perceptron networks have proven to be the most optimal for analysis of environmental samples (Pilato et al., 1999; Dragović et al., 2005, 2006; Dragović and Onija, 2005; Videnova et al., 2006; Kumar Gautam et al., 2008, Gryning, 2001). In our case, the three-layer feed-forward network with the back-propagation algorithm was selected, based on previous experience in neural network application in gamma-ray spectrometry (Pilato et al., 1999; Dragović et al., 2005, 2006; Dragović and Onija, 2005).

The training process of this network was performed in two phases. In the first phase, the data contained in the input layer were sent to the hidden layer through the input nodes. Nodes in the hidden layer calculate the weight sums of input data. All these sums were then sent to the output layer through the activation functions as single end data.

In our case, we used the logistic function as an activation function which is a representative of sigmoid activation functions:

$$f_j = \frac{1}{1 + \exp\left(-\sum w_{ji} o_i + b\right)} \tag{1}$$

where w_{ji} is the weight factor whose value connects the lower layer node i to the upper layer node j, o_i is the output value of the node i, while b represents a bias. The bias (limit of neural activation) is used to calculate the total numerical value of all nodes located in the single layer.

In the second phase of network training, the error was calculated between the calculated and experimentally obtained values in the output layer using the so-called general delta rule (Ljn and Lee, 1996). Based on this rule, nodes in the output layer are adjusted according to the values of nodes in the input layer based on the following equation:

$$w_{ji}^{n+1} = w_{ji}^{n} + \eta \delta_j o_j + \alpha w_{ji}^{n} \tag{2}$$

where δ_j is the signal error at node j, o_j is the output value of node j, n is the number of iterations, η is the learning rate, and α is the momentum. The learning rate affects the step size in the space of weight coefficients and controls the speed at which the network learns. Momentum is included in the calculation to add the previous changes of weight coefficients to the current change during the training process. In determining the value of the learning rate and momentum, we used previous experience in the application of neural networks in gamma-spectrometry and determined that both these values are 0.1 (Dragović et al., 2005).

The key part of the calculation in artificial neural networks is forming the sets for training and testing the network. The training set is used to optimize the model characteristics, while the testing set is used to assess the generalization capabilities of a given network. The training set contained logarithmic values of meteorological parameters as input data and those of ^{7}Be activity concentrations from 2009 as output data. The same parameters from 2010 were used as input and output data in the testing set.

The optimal number of nodes in the hidden layer was found by calculating the RMSE (root mean square error) for different values