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PREDICTABILITY OF LEAD-210 IN SURFACE AIR BASED ON MULTIVARIATE ANALYSIS

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Abstract*. Dependence of the lead-210 activity concentration in surface air on meteorological variables and teleconnection indices is investigated using multivariate analysis, which gives the Boosted Decision Trees method as the most suitable for variable analysis. A mapped functional behaviour of the lead-210 activity concentration is further obtained, and used to test predictability of lead-210 in surface air. The results show an agreement between the predicted and measured values. The temporal evolution of the measured activities is satisfactorily matched by the prediction. The largest qualitative differences are obtained for winter months.*

Key words*: lead-210, surface air, meteorological variables, teleconnection indices, multivariate analysis, wavelet transform analysis*

1. Introduction

Lead-210 is a naturally occurring radionuclide with a half-life of 22.23 years. The main source of ^{210}Pb in surface air is its radioactive parent radon-222 that emanates from the soil. After formation, ^{210}Pb attaches to aerosols whose fate is governed by atmospheric circulation and removal processes.

A number of studies that looked into the ^{210}Pb activity concentrations at different measuring sites have shown a uniform radionuclide distribution, both in the vertical and horizontal. For example, [1] showed no significant variations in the ^{210}Pb activity concentration at three locations over distance of approximately 200 km, and still noted a case in which the ^{210}Pb activity concentration rapidly changed due to a passage of a cold front. Similarly, [2] investigated differences in the ^{210}Pb activity concentration between two measurement sites at a distance of 12 km in the horizontal and of 800 m in the vertical. Their results also showed that over longer periods of time, the radionuclide was well mixed within the atmosphere. Further, [3] found good correlation between the ^{210}Pb activity concentrations across two sites 100 km apart, which were influenced by different local conditions, including different prevalent winds.

On the other hand, the vertical profile of the ^{210}Pb activity concentration in the atmosphere has been shown to reflect the fact that the radionuclide source is in the lowermost layer - surface air masses are richer in ^{210}Pb than air masses from higher altitudes, with the sharpest decrease in activity concentration between the ground and an altitude of 3 km [4, 5]. Further, the ^{210}Pb activity concentration is higher in continental air masses than in air masses originating over a body of water [2, 6, 7], which results in the radionuclide activity concentration variations in the horizontal, and also explains temporal differences in a location under influence of interchangeable winds of either continental or maritime origin [2].

The above mentioned studies, however, did not include an analysis of ^{210}Pb relation with large-scale atmospheric circulation. The North Atlantic Oscillation (NAO) index is one of the most commonly used teleconnection indices to describe a large-scale circulation pattern over the North Atlantic Ocean and surrounding land masses [8]. The two oscillation phases of NAO induce changes in large-scale circulation patterns [9], which further reflect on local weather conditions especially over eastern North America and across Europe, including Serbia [10]. The Polar/Eurasia pattern [8] is another teleconnection that has an impact on weather in Europe [11]. Further, the East Atlantic/Western Russia pattern [8] influences the amount of precipitation in southeast part of Europe [12], and can thus be a contributing factor in the amount of ^{210}Pb in surface air.

In contrast to a very limited number of studies looking into an influence of large-scale atmospheric transport on the ^{210}Pb activity concentrations, a link between local meteorological variables and the radionuclide activities has been extensively investigated [7, 13, 14, 15]. For example, to name only a few: [16] showed a strong positive relationship between the ^{210}Pb deposition and precipitation; [17] found a positive correlation of ^{210}Pb activity concentration with temperature, and negative correlation with precipitation, relative humidity and wind speed; [14] showed that washout is the most significant mechanism of ^{210}Pb removal from the atmosphere.

The goal of our analysis is to combine meteorological data and large-scale atmospheric transport patterns (quantified by teleconnection indices) and treat the ^{210}Pb concentration in surface air as a result of their interplay. Different statistical tools are employed to achieve this. On one hand, a set of multivariate methods incorporated in the Toolkit for Multivariate Analysis (TMVA) [18] is used. It is complemented by a wavelet transform spectral analysis [19]. A mapped functional behaviour which is obtained in the analysis is then used to test predictability of the ^{210}Pb activity concentration in surface air.

2. Datasets

In Belgrade, Serbia, at the Vinča Institute of Nuclear Sciences, continual measurements of the ^{210}Pb activity concentrations in surface air started in 1985. The monthly mean activity concentrations in composite aerosol samples were determined on High Purity Germanium detectors by standard gamma spectrometry. The activity concentrations of ^{210}Pb were determined using the gamma energy of 46.5 keV. A detailed description of the measurement procedure is given in [15].

The meteorological daily data: minimum, maximum and mean temperature, atmospheric pressure, relative humidity, precipitation, sunshine hours and cloud cover data for Belgrade, were obtained from the European Climate Assessment & Dataset (ECA&D) [20] and the Republic Hydrometeorological Service of Serbia. In addition, a temperature variable, which does not have a local character, was included to investigate the extent to which the local meteorological variables influence the ^{210}Pb activity concentration in the air. The chosen variable was the Northern Hemispheric mean monthly temperature anomaly over land calculated from historical temperature records (http://www.cru.uea.ac.uk/cru/data/temperature/CRUTEM4-nh.dat visited on 10 March 2015). The temperature anomaly was derived as a deviation from a reference temperature value which, in this data set, was taken as the mean over a reference period 1961–1990. More details on the temperature anomaly calculations can be found in [21, 22].

The data for eight teleconnection indices of large-scale atmospheric circulation: North Atlantic Oscillation (NAO), East Atlantic (EA), East Atlantic/Western Russia, Scandinavia (SCAND), Polar/ Eurasia, Western Pacific (WP), East Pacific-North Pacific (EP-NP), and Pacific/North American (PNA) were obtained from the data archive of the United States National Oceanic and Atmospheric Administration's Climate Prediction Center (http://www.cpc.ncep.noaa.gov/data/teledoc/telecontents.shtml visited on 18 October 2013). A description of the procedure used to identify the Northern Hemisphere teleconnection patterns and indices is given in [8]. The monthly values of teleconnection indices since 1950 were available.

The temporal resolution of the input variables differed: the ^{210}Pb activity concentrations and the teleconnection indices, apart from NAO, were available as monthly mean values. This resolution implied a total number of data points that was insufficient for MVA which inherently requires a large number of points to determine the mapped behaviour. To overcome this drawback, an interpolation of the monthly measurements was performed using Fast Fourier Transform smoothing on monthly data (Fig. 1).

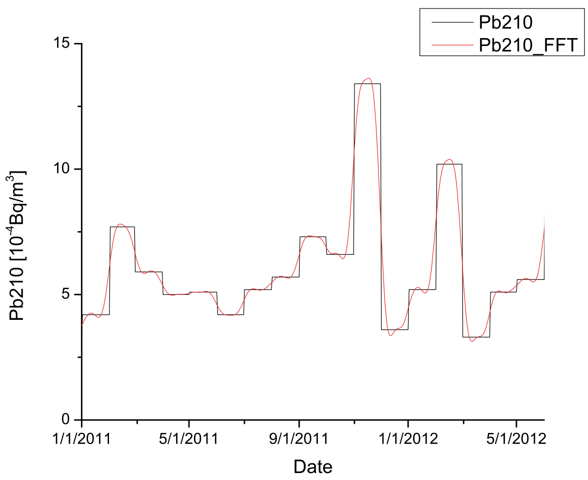

Fig. 1 Fast Fourier Transform of the monthly ^{210}Pb activity concentrations in surface air. The Fast Fourier Transform curve gives interpolated daily ^{210}Pb values which were used in the MVA classification and regression analysis.

3. Calculations

3.1. Multivariate Analysis

Many multivariate methods and algorithms for classification and regression are integrated in the analysis framework ROOT [23], and the Toolkit for Multivariate Analysis (TMVA) [18]. Multivariate analysis is used to create, test and apply all available classifiers and regression methods to single out one method that is the most appropriate and yields maximum information on the dependence of an investigated variable on a multitude of input variables. Thus, in TMVA there is no need to choose *a priori* a method for the data classification and regression – all of the techniques incorporated in TMVA are tested and the most suitable one is chosen for further analysis.

The TMVA package includes various techniques, such as multi-dimensional likelihood estimation, linear and nonlinear discriminant analysis, artificial neural networks, support vector machine, and boosted/bagged decision trees. All the techniques in TMVA belong to the family of "supervised learning" algorithms. They make use of training events, for which the desired output is known, to determine the mapping function that either describes a decision boundary (classification) or an approximation of the underlying functional behaviour defining the target value (regression). All MVA methods see the same training and test data.

The multivariate methods are compared within the procedure in order to find one which, on the basis of input variables, gives a result satisfactorily close to the observed values of the output variable. More details on calculation procedure are given in [24].

In our analysis, the output variable was the activity concentration of ^{210}Pb in surface air, while the input variables were the nine meteorological variables, one derived variable, and eight teleconnection indices, adding to 18 input variables in total. A multivariate method that gave the best regression results in our study was the Boosted Decision Trees method.

3.1.1. Boosted Decision Trees

Boosted Decision Trees (BDT) is a method in which a decision is reached through a majority vote on a result of several decision trees. A decision tree consists of successive decision nodes which are used to categorise the events in a sample, while BDT represents a forest of such decision trees. The (final) classification for an event is based on a majority vote of the classifications done by each tree in the forest, which ultimately leads to a loss of the straightforward interpretation in a decision tree. More detailed information on training in BDT can be found in [24].

An importance of an input variable is measured by a “variable rank”. In BDT, this measure is derived by counting the number of times a specific variable is used to split decision tree nodes, and then weighting each split occurrence by the separation gain-squared it achieved and by the number of events in the node [18].

3.2. Wavelet Transform Analysis

Wavelet transform (WT) is a standard analytical tool in investigation of time series with nonstationarities at different frequencies [19]. In WT analysis, a calculated global wavelet power spectrum (which corresponds to Fourier power spectrum) is smooth and can therefore be used to estimate characteristic periods in the data sets. To detect these characteristic periods, a standard peak analysis was performed by searching the maximum and saddle (for hidden peaks) points in the global wavelet power spectra of the ^{210}Pb activities.

4. Results and Discussion

The WT analysis showed a number of characteristic periods in the ^{210}Pb activity concentration in surface air (Fig. 2). The periods are given by the time coordinates of the local maxima in the ^{210}Pb activity concentration power spectrum. Three short characteristic periods were found, with a seasonal one (at 2.6 months) most pronounced. The annual cycle (at 11.8 months) was also evident, as well as a longer period of approximately three years (at 36.5 months). Apart from the annual cycle [2, 14, 15], the other ^{210}Pb periodicities have not been studied in detail.

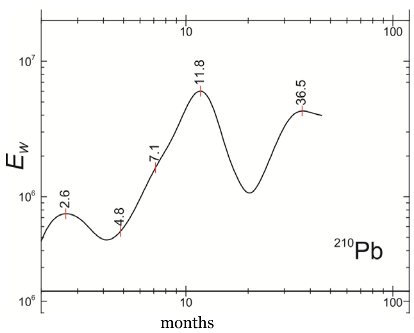

Fig. 2 Power spectrum of the ^{210}Pb activity concentration in surface air. Positions of the maximum and saddle points are marked with the assigned values of characteristic time in months.

To allow for the prominent annual cycle in the ^{210}Pb activity concentration in surface air, another input variable, called MonthDay, was introduced in the analysis. The MonthDay variable is purely mathematical – its values represent a sum of the month number (1 to 12) and the day number in a month divided by the total number of days in the given month. For example, a MonthDay value for 10 January is 1+10/31. This variable has an annual cycle and thus serves as a proxy of any contributing variable which is not specified in the analysis but also exhibits an annual cycle.

Prior to the multivariate analysis, the Pearson’s linear correlation coefficients for the input variables and ^{210}Pb activity concentrations were calculated (Tab. 1) using the monthly means. The strongest correlation was found with temperature anomaly. However, that correlation was not significant at the 0.05 level. Statistically significant linear correlation was obtained only for atmospheric pressure and three teleconnection indices: EP-NP, East Atlantic/Western Russia, and Polar/Eurasia.

The calculated correlation coefficients describe the measure of linear correlation between the ^{210}Pb activity concentrations and the input variables. However, apart from linear, other types of dependance between variables could exist. Each method incorporated in the MVA gives its own ranking (as one of the results), which does not necessarily coincide with a ranking of another method, or with the order given by the linear correlation coefficients.

Table 1. Pearson's linear correlation coefficients (r) of the input variables and the ^{210}Pb activity concentration in surface air, and the BDT variable ranking. The correlation coefficients significant at the 0.05 level are given in bold.

Variable	r	BDT rank
Temperature anomaly	-0.47	1
Precipitation	-0.22	18
Atmospheric pressure	**+0.21**	15
EP-NP	**+0.19**	6
Cloud cover	-0.16	17
East Atlantic/Western Russia	**+0.14**	8
Polar/Eurasia	**+0.12**	7
NAO	+0.10	4
SCAND	+0.08	9
Mean temperature	-0.08	11
Minimum temperature	-0.08	12
Maximum temperature	-0.07	14
Relative humidity	+0.07	16
Sunshine hours	-0.07	13
PNA	+0.04	5
WP	+0.03	3
EA	+0.008	10
MonthDay value	N/A	2

4.1. Regression Analysis

A result of MVA regression method training is an approximation of the underlying functional behaviour that defines the dependence of the target value, the ^{210}Pb activity concentrations in our analysis, on the input variables. This set of calculations was based on the measurements performed during the training period, which was from 1985 to 2010 in our case. Predictability of the ^{210}Pb activity concentration in surface air was tested in the ensuing calculations, in which the measurements for 2011 and 2012 were used.

The analysis indicated that the best regression method, in which the output values (evaluated, or predicted, ^{210}Pb activity concentrations) were closest to the measured concentrations, was BDT. In an ensemble of multivariate methods, the average quadratic deviation between the evaluated and measured values was the least for BDT (Fig. 3). The BDT output deviation from the measurements over the training period was close to zero for the majority of data points (Fig. 4), which confirmed the good quality of the regression method.

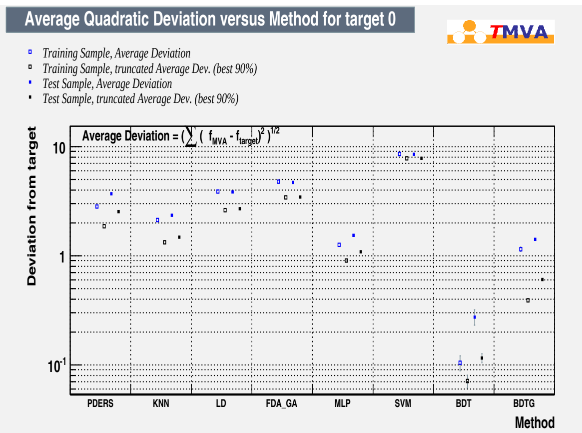

Fig. 3 Average quadratic deviation for multivariate regression methods. The deviation was the least for BDT.

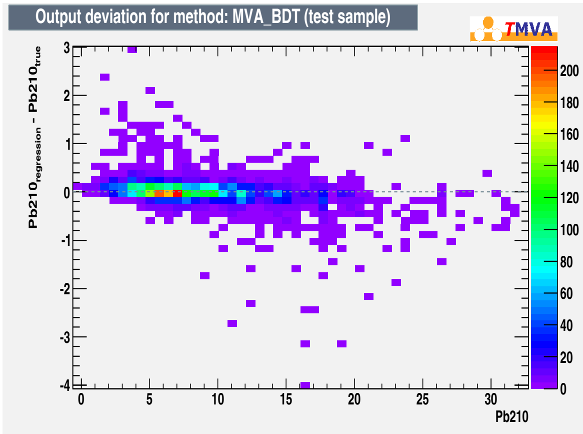

Fig. 4 Difference between the BDT evaluated and the measured ^{210}Pb activity concentrations. The colour bar on the right gives the number of data points.

One of the results given by the BDT method is a variable ranking (Table 1). Apart from temperature anomaly (rank 1), all other meteorological variables were ranked as less important than the teleconnection indices. The influence of the large-scale circulation is not well understood, but the local ^{210}Pb activity concentration could be significantly influenced by mesoscale and synoptic scale variations in atmospheric pressure (as quantified by the teleconnection indices). The local variations in atmospheric pressure, on the other hand, could play a minor role (low BDT rank in Table 1).

Similarly, a high rank obtained for temperature anomaly (Table 1) may imply that the ^{210}Pb activity concentration in surface air is to a certain extent insensitive to relatively fast variations in local temperature.

The high rank of the MonthDay variable could indicate an existence of another contributing factor with a strong annual cycle.

4.2. ^{210}Pb activity concentration prediction

The final step in our analysis was an evaluation of the ^{210}Pb activity concentration outside the training period for which the mapped functional behaviour was obtained. In other words, the input variables for 2011 and the first half of 2012 were used to calculate the output variable which was then compared to the measured ^{210}Pb activity concentration over that period (Fig. 5).

The standard deviation of all the absolute and relative differences between the BDT evaluated and measured ^{210}Pb values were $0.64 \cdot 10^{-4}$ Bq/m^3 and 0.12, respectively.

The temporal evolution of the ^{210}Pb activity concentration was satisfactorily captured by the regression method (Fig. 5). However, the regression was not able to quantitatively predict the observed values in winter periods, when the radionuclide activity concentration reached maximum values. Thus, during January and February 2011, the evaluated values were conspicuously higher than the measured ones, while in November and December of the same year, the evaluation first underestimated and then overestimated the ^{210}Pb

activity concentration. It can, however, be argued that the sum of the predicted ^{210}Pb activity concentrations for these two months matched the sum of the measured values. Furthermore, a local maximum seen in February 2012 was satisfactorily reproduced by the method.

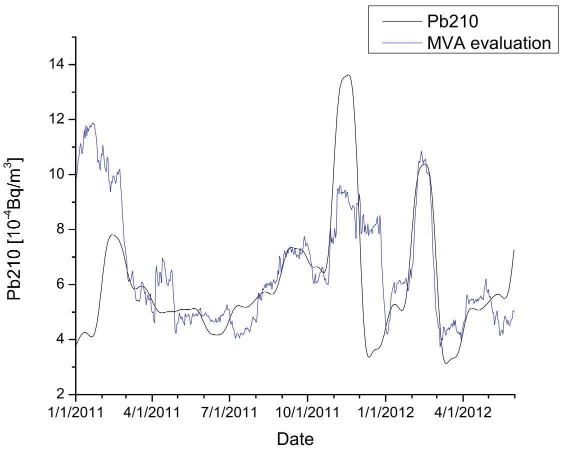

Fig. 5 ^{210}Pb activity concentration measured in surface air (black line) and evaluated by MVA (blue line) during 2011 and the first half of 2012.

The above results may indicate that in winter some additional processes play a key role in the ^{210}Pb activity concentration in surface air. Emanation of radon-222, the parent radionuclide, could be affected by soil conditions, such as snow cover, soil temperature and moisture, and it could thus induce changes in the ^{210}Pb source abundance. Future choice of input variables should include some soil parameters in an attempt to increase the accuracy of the ^{210}Pb prediction in winter months.

Further assessments and refinements of the prediction should also address the limitation given by the temporal resolution of the measured ^{210}Pb activity concentration in the air. The interpolation of the monthly data performed to obtain daily values (Fig. 1) could give rise to spurious relations between the ^{210}Pb activity concentration and input variables, which could reflect on the mapped functional behaviour, and in turn, on the prediction of ^{210}Pb in surface air. A possible way to check the validity of our method is to run the prediction using only the measured monthly values, while still employing the daily data in the training period. However, a preferred approach to our method refinement would be to use a more comprehensive database with the measured ^{210}Pb activity concentration of higher temporal resolution.

5. Conclusions

The dependence of the ^{210}Pb activity concentration in surface air on different meteorological variables and indices of large-scale circulation was investigated using multivariate analysis.

Boosted Decision Trees was singled out as the best regression method with the least average quadratic deviation between the evaluated and measured activity concentrations. The importance variable ranking given by the BDT method implied a greater influence of large-scale transport than of the local meteorological variables.

The prediction of the ^{210}Pb activity concentrations showed an agreement with the measurements, except in winter months when the largest differences were obtained.

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