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Modeling of methane emissions using the artificial neural network approach

LIDIJA J. STAMENKOVIĆ[1#], DAVOR Z. ANTANASIJEVIĆ[2*#], MIRJANA Đ. RISTIĆ[1#], ALEKSANDRA A. PERIĆ-GRUJIĆ[1#] and VIKTOR V. POCAJT[1#]

[1]University of Belgrade, Faculty of Technology and Metallurgy, Karnegijeva 4, 11120 Belgrade, Serbia and [2]University of Belgrade, Innovation Center of the Faculty of Technology and Metallurgy, Karnegijeva 4, 11120 Belgrade, Serbia

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Abstract: The aim of this study was to develop a model for forecasting CH_4 emissions at the national level, using artificial neural networks (ANN) with broadly available sustainability, economical and industrial indicators as their inputs. ANN modeling was performed using two different types of architecture; a backpropagation neural network (BPNN) and a general regression neural network (GRNN). A conventional multiple linear regression (MLR) model was also developed in order to compare the model performance and assess which model provides the best results. ANN and MLR models were developed and tested using the same annual data for 20 European countries. The ANN model demonstrated very good performance, significantly better than the MLR model. It was shown that a forecast of CH_4 emissions at the national level using the ANN model could be made successfully and accurately for a future period of up to two years, thereby opening the possibility to apply such a modeling technique, which could be used to support the implementation of sustainable development strategies and environmental management policies.

Keywords: national emission; general regression neural network; backpropagation neural network; multiple linear regression.

INTRODUCTION

Due to higher economic and industrial development, air pollution has become a serious problem in many countries and regions in recent years; therefore, it has become essential to track the emission of pollutants that have a detrimental effect on the environment and human health. As a part of a global effort to reduce emission of air pollutants, the Convention on Long Range Transboundary Air Pollution (LRTAP),[1] the United Nation Framework Convention on

* Corresponding author. E-mail: dantanasijevic@tmf.bg.ac.rs
# Serbian Chemical Society member.
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Climate Change (UNFCCC)[2] and their associated protocols, imply an obligation for all countries to submit reports on estimated current and future emissions of air pollutants. To achieve this commitment, it is necessary to implement the application of suitable methods and models for estimating and simulating a range of emission scenarios.

Methane is one of greenhouse gases that contribute to climate changes and it is emitted into the atmosphere from various sources: transport and natural gas exploitation, landfills, coalmines, wastewater treatment, rice fields, the petrochemical industry, composting, metallurgical industry, farms, *etc*.[3]

The main sources of emission data for air pollutants, including CH_4, are emission inventories, which currently makes them a key foundation for air quality modeling and analysis. The values of emissions in emission inventories are a compilation of a large number of input parameters (more than 300 emissions sources), in which the method of calculating the total emissions is dependent on the employed emission model. According to the methodology proposed in the EMEP/CORINAIR Emission Inventory Guidebook provided by the European Environmental Agency (EEA),[4,5] emissions are calculated by multiplying the emission factors (*EF*) with the activity rate (*A*), a statistical parameter for the respective source. In an emission inventory, the values of the parameters are determined as best estimates and therefore the input data to the inventories could be classified as uncertain, which in turn, means that the reported data could also be assumed uncertain.[6,7] Numerous studies have discussed uncertainties in emission inventories.[6–11]

There are different approaches to modeling emission estimates and estimating CH_4 emissions has also been the subject of many studies in which different modeling approaches were used.[5,12–17] In order to predict emission at the national level, all the aforementioned approaches require the availability of a large amount of country-specific information for calculating emission factors and activity data. Thus, an important limitation for the applicability of existing models could be a lack of specific input information necessary for the calculation of emission factors for a specific country. This lack of input information is especially typical when considering developing countries, which often have the most severe problem with pollution and emission control, and it could therefore result in particularly high uncertainties in the estimated emissions.

Artificial neural networks (ANNs) are adaptable systems that can determine relationships between different sets of data.[18] ANNs can be described as multivariate non-linear statistical methods where selected input variables are presented to the network and output variables (one or more) are derived. ANNs learn from examples and this particular modeling approach has proved to be a useful tool for many problems related to air quality and forecasting of emissions.[19–24] In recent years, due to their flexibility, considerable progress has been made in the application of ANNs for predicting CH_4 for the chosen area. Some examples of studies include modeling and prediction of ventilation methane emissions of U.S. longwall mines,[18] prediction of methane productions in anaerobic treatment of molasses wastewater,[25] and prediction of methane emissions from wetland ecosystems.[26]

This paper presents the development and evaluation of an ANN model for the prediction of CH_4 emissions at national levels, using sustainability, economic and industrial indicators as inputs. Two different ANN architectures, back-propagation neural network (BPNN) and general regression neural network (GRNN), were evaluated and their results compared with each other, as well as with the results of a multiple linear regression (MLR) model.

The main difference between conventional inventory based models and the proposed ANN model is that the ANN model requires a substantially smaller number of input parameters, which have significantly broader availability. Due to this factor, the ANN model could be applied to the forecasting of CH_4 emissions when counties and regions do not have a complete set of input parameters available, which is usually essential for other models based on activity levels and emission factors.

MODEL DESCRIPTION AND TESTING

Input and output data

An important step in developing ANN models is the selection of the input variables that have the most significant impact on the output.[27] Economic activities largely contribute to the total emission of methane, most contributing sectors being industrial processes, agriculture, and production and energy consumption.[3]

Several studies have already shown that ANN models, based on economic and sustainability indicators, can achieve good accuracy in forecasting pollutant emissions at the national level, *e.g.*, for greenhouse gases and PM10 emissions.[28–30] The Gross Domestic Product (GDP) is a broadly available country-specific parameter that was chosen for an input parameter as a general indicator of the size of the economy. The GDP has already demonstrated very good applicability as an input parameter for ANN models.[29,30]

Municipal waste generation and its disposal onto or into land were also chosen as a model input, since methane is produced from landfills by the anaerobic biodegradation of wet waste.[31] Since rice paddies are characterized by a high moisture content and relatively high organic carbon levels, as well as prolonged anaerobic conditions during rice growth, they are one of the major anthropogenic sources of methane;[17,32] therefore, the land used for rice cultivation was also selected as an input.

Livestock is another important source of methane emissions, which originate mainly from enteric fermentation and manure management of ruminant production systems.[33] The contribution of livestock to methane emissions was factored by selecting the number of cattle as the input parameter. In addition, since methane is a component of natural gas, primary natural gas production is directly connected with the emissions of methane and was consequently included as an input parameter. Data for all of these input variables at the national level, for every country included in this study, were published on Eurostat.[34]

Annual methane emissions at the national level were taken as the output variable, and the data for every country included in this study was published on EDGAR (the Emissions Database for Global Atmospheric Research).[35] EDGAR is a joint project of the European Commission JRC (Joint Research Centre) and the Netherlands Environmental Assessment Agency (PBL) and it provides global, past and present day anthropogenic emissions of greenhouse gases and air pollutants by country and on a spatial grid.

The variables chosen for inputs and the related output of the models, along with their units after normalization and data sources, are presented in Table I. The selected input variables were normalized per capita and/or per average GDP value of the EU27, in order to enable comparison of countries of different sizes.

TABLE I. List of input and output variables

Input variable	Unit[a]	Source of data
Gross domestic product (*GDP*)	–[b]	Eurostat[34]
Waste deposit onto or into land (*WDL*)	kg pc[c]	
Municipal waste generation (*MWG*)	kg pc	
Land use – rice (*LR*)	ha pc	
Number of cattle (*NC*)	cattle pc	
Primary production of gas (*PPG*)	toe pc[d]	
Output variable		
CH_4 (emission)	kg pc	Edgar[35]

[a]After normalization; [b]unit less because of normalization per GDP value of EU27; [c]per capita; [d]tons of oil equivalents

The ANN CH_4 emission models were trained, validated and tested with available data for 20 European countries for the period ranging from the years 2000 to 2008, since data after 2008 was not available. Three types of datasets were used:

A training set. The group of data with which the ANN was trained. Data from the years 2000 to 2006 was used for training; an example of the training data set for the year 2006 is presented in Table S-I of the Supplementary material to this paper.

A validation dataset. The group of data provided to the ANN in the learning phase for the evaluation of modeling error, for effectively updating the best thresholds and weights,[36] and for preventing overtraining of the ANN. Approximately 15 % of data randomly selected from the training data set was used as the validation dataset.

A test dataset. A set of data newly presented to the ANN, thus used to evaluate the ANN's generalization capability. In this case, the data from years 2007 and 2008 was used as the test dataset.

ANN architecture

Two different ANN architectures were compared: the backpropagation neural network (BPNN), as the most frequently and widely applied ANN architecture, and the general regression neural network (GRNN), since it has already demonstrated good results in the modeling of emissions.[29,30]

Details of the two ANN architectures are described in Supplementary material to this paper.

Performance indicators

In order to compare the performance of the different models, in this study the following statistical parameters were used:

The root mean squared error (*RMSE*):

$$RMSE = \left[\overline{\left(C_\mathrm{P} - C_\mathrm{O} \right)^2} \right]^{1/2} \tag{1}$$

The mean absolute error (*MAE*):

$$MAE = \frac{1}{n} \sum \left| C_\mathrm{P} - C_\mathrm{O} \right| \tag{2}$$

The correlation coefficient (*R*):

$$R = \frac{\overline{(C_\mathrm{O} - \overline{C_\mathrm{O}})(C_\mathrm{P} - \overline{C_\mathrm{P}})}}{\sigma_{C_\mathrm{O}} \sigma_{C_\mathrm{P}}} \tag{3}$$

The index of agreement (*IA*):

$$IA = 1 - \frac{\overline{\left(C_\mathrm{P} - C_\mathrm{O} \right)^2}}{\left[\left| C_\mathrm{P} - \overline{C_\mathrm{O}} \right| + \left| C_\mathrm{O} - \overline{C_\mathrm{O}} \right| \right]^2} \tag{4}$$

The percentage of predictions within the factor of 1.25 from the observed values ($FA_{1.25}$):

$$0.8 < \frac{C_\mathrm{P}}{C_\mathrm{O}} < 1.25 \tag{5}$$

where C_P and C_O are the predicted and observed values, respectively; σ_O and σ_P are the standard deviation of observations and predictions, respectively; the over bars refer to the average of all values.

These statistical parameters were previously used for the evaluation of emission models and other modeling techniques.[18,29] *MAE* and *RMSE* measure the residual errors, thus providing a general understanding about the difference between the observed and modeled values. *R* provides a variability measure of the data reproduced in the model.[37] *IA* provides a correlation between the modeled and measured values and incorporates the error between those values. $FA_{1.25}$ represents the proportion of data for which the model results are in "proximity" with the measured values. The significance of these statistical metrics is not equal and in this study, *IA* and $FA_{1.25}$ were used as the key statistical parameters for evaluating the models.

RESULTS

In this study, two types of ANN architectures, BPNN and GRNN, were used to create models for the prediction of CH_4 emissions at the national level. Along with the development of the ANN models, for comparison purposes, a multiple linear regression (MLR) model was developed employing the same data as was used in the ANN models. The MLR model was created using SPSS 19 software[38] and the obtained MLR coefficients are presented in the following equation:

$$\begin{gathered} \mathrm{CH_4\ emission} = 0.0480 + 0.0001WDL - 0.0001MWG + \\ +0.0922NC - 6465.1928LR + 0.0008PPG + 0.0131GDP \end{gathered} \tag{6}$$

where *WDL* is waste deposit onto or into land, *MWG* is municipal waste generation, *NC* is the number of cattle, *LR* is land use – rice, *PPG* is primary production of gas and *GDP* is the gross domestic product.

The values of the performance indicators of the developed models for the training and testing phase are given in Table II. A comparison of the actual and predicted values of CH_4 emission for the training data (years 2000–2006) is shown in Fig. 1.

TABLE II. Performance indicators values for the GRNN, BPNN and MLR models; pc – per capita

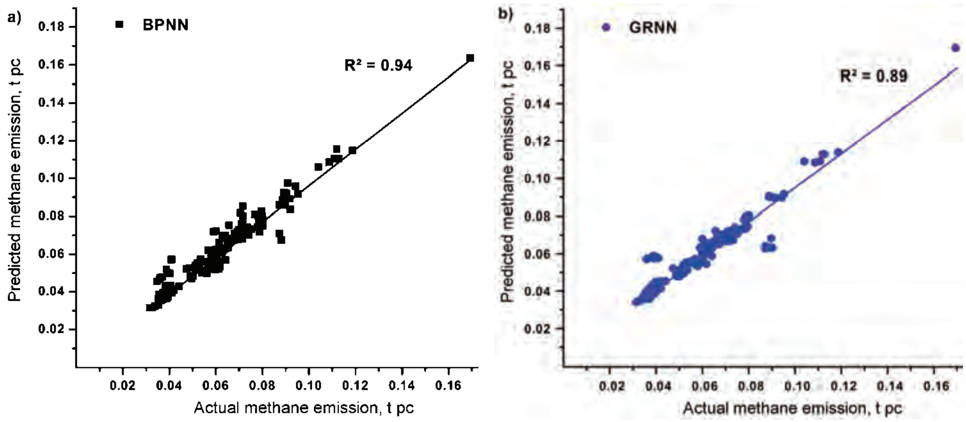
Model	Performance indicators for test set				
	IA	$FA_{1.25}$ / %	*MAE* / kg pc	*RMSE* / kg pc	*R*
Training data					
BPNN	1.00	94	3.4	5.0	0.97
GRNN	0.97	92	3.6	7.0	0.94
MLR	0.83	64	11.3	14.0	0.75
Test data					
BPNN	1.00	90	5.6	7.7	0.95
GRNN	0.98	93	4.8	7.3	0.96
MLR	0.90	58	11.9	252.5	0.83

The dataset for the years 2007–2008 is the test dataset, which is a set of "new" data, previously not presented to the models within the training phase. The test dataset, which is presented in Table S-II of the Supplementary material, was used to evaluate the capability of the models to make good predictions. The results of the BPNN, GRNN and MLR models applied on the test dataset are presented in Fig. 2 and the values of statistical performance indicators are presented in Table II. The actual CH_4 emissions, the GRNN predictions and the relative errors for the years 2007 and 2008 are presented in Table III.

DISCUSSION

Based on the results given in Table II and shown in Fig. 1 that were generated by the models in the training phase, the BPNN and GRNN led to much better results than the MLR. As can be seen, the results given by the ANN models could be regarded as very good.

The testing results showed that the use of the GRNN architecture led to slightly better results than the BPNN and considerably better results than the MLR (Fig. 2). The most important statistical performance indicators $FA_{1.25}$ and *IA* showed much better values for the test dataset in the case of the ANN models, *i.e.*, for the GRNN model, $FA_{1.25}$ was 93 % and for the BPNN, it was 90 %, while for the MLR, it was only 58 %. Moreover, for the GRNN model, *IA* was 0.98, for the BPNN, it was 1.00, while for the MLR, *IA* was 0.90. It could be concluded that the GRNN model was optimally trained since it demonstrated very good results with similar values of $FA_{1.25}$ and the error performance indicators derived for the training and test dataset. In contrast, the BPNN model had similar but slightly worse results for the test dataset compared to the training data (Table II), which means that the BPNN model may benefit from an adjustment in the design in order to optimize the prediction results.



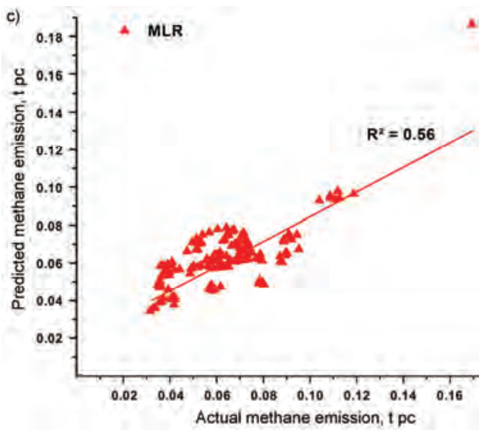
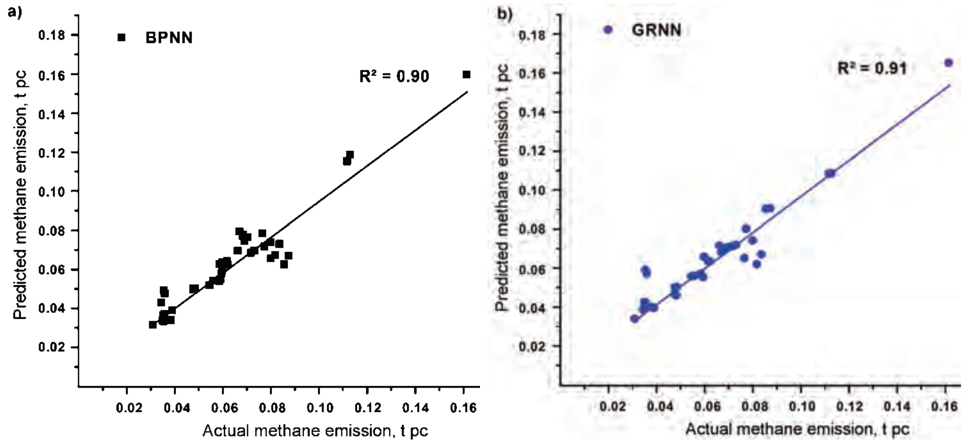


Fig. 1. Comparisons of actual and predicted values of the CH_4 emission for the training data set (years 2000–2006). a) BPNN, b) GRNN and c) MLR.

The GRNN model made 75 % of its predictions successfully, with the relative error of less than 10 %, another 10 % of its predictions had a relative error within the range of 10–20 %, while only 15 % of its predictions had a relative error higher than 20 %. In the case of Hungary, Bulgaria and The Netherlands, the predictions of the GRNN model had a relative error within the range of 10–20 %, while in the case of Slovakia, Poland and Greece, the relative error was higher than 20 % for both test years (Table III).

Uncertainties related to the input parameters used in the emission inventory approach (the activity statistics and emission factors) are considerable, ranging from 20–30 to up to 50 % in the case of Austria, which leads to the uncertainties



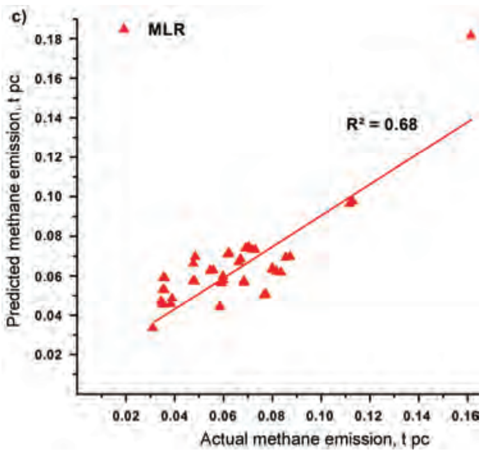


Fig. 2. Comparisons of actual and predicted values of CH_4 emission for the test data set (years 2007–2008). a) BPNN, b) GRNN and c) MLR.

TABLE III. Actual and CH_4 emission predicted by the GRNN model with relative errors (RE) for the years 2007 and 2008; pc – per capita

Country	Year 2007			Year 2008		
	Actual emission kg pc	*GRNN* kg pc	*RE* %	Actual emission kg pc	*GRNN* kg pc	*RE* %
Bulgaria	77.28	80.17	3.74	76.54	65.12	–14.92
Czech Republic	55.80	55.94	0.28	54.45	55.73	2.37
Denmark	68.33	69.30	1.42	68.39	69.32	1.36
Estonia	79.95	74.12	–7.29	79.97	74.22	–7.19
Greece	35.44	42.54	20.02	34.87	42.49	21.85
Spain	38.87	39.53	1.71	38.37	39.96	4.15
France	62.16	63.34	1.90	61.67	63.32	2.67
Latvia	66.19	71.49	8.01	66.93	68.16	1.84
Lithuania	71.69	71.22	–0.65	72.98	71.92	1.46
Luxembourg	111.64	108.51	–2.81	112.85	108.51	–3.84

TABLE III. Continued

Country	Year 2007			Year 2008		
	Actual emission kg pc	*GRNN* kg pc	*RE* %	Actual emission kg pc	*GRNN* kg pc	*RE* %
Hungary	35.73	39.35	10.13	35.12	40.18	14.39
Netherlands	59.80	65.75	9.95	59.77	65.87	10.21
Austria	48.20	45.97	–4.64	47.54	46.72	–1.72
Poland	83.57	67.05	–19.77	81.71	62.03	–24.09
Portugal	58.45	56.48	–3.39	58.44	57.12	–2.25
Romania	58.91	55.93	–5.06	59.35	55.34	–6.75
Slovenia	70.33	70.88	0.78	69.00	70.43	2.07
Slovakia	35.86	57.23	59.60	35.40	59.08	66.90
Finland	87.25	90.57	3.80	85.49	90.24	5.55
UK	48.39	50.26	3.86	47.61	50.12	5.27

in the estimated methane emissions of about 20–40 %.[9] Since the GRNN model uses significantly fewer input variables and provides predictions with the relative error lower than 20 % in case of most countries, the ANN approach could be considered as a viable alternative for the prediction of annual methane emissions on the national level, especially in cases when the emission inventory approach cannot be applied because of lack of data.

The obtained deviation between the actual data and predicted values for all countries for which the obtained relative error was higher than 10 % could be related to the quality of the input and output data used for the creation and evaluation of the model.

In the case of Slovakia, the data for waste deposit onto or into land (WDL) for the year 2001 was estimated. In addition, the data for municipal waste generation (MWG) was collected from the National Waste Catalogue until 2001 but from 2002 onwards from The European List of Waste.[34] The usage of estimated and non-standard training data could result in the large relative error in the year 2007 and 2008 (Table III). The relative error for The Netherlands for the year 2008 was 10.21 %, and this model error could be related to the ban of the direct disposal of mixed municipal waste in 2003, which led to a decrease in *WDL* from 50 kg pc in 2002 to only 11 kg pc in 2004.[39]

In the case of Greece, the training data of the input parameter *GDP* for the year 2005 was uncertain because of the inaccurate reporting of the country's economic performance by the government. The influence of this parameter was probably the reason for higher attained values of the relative error for both test years.

The *MWG* data for Poland and Hungary were estimated, for the year 2003 in the case of Hungary and from 2005 to 2008 in the case of Poland, which could explain the higher relative error values obtained (14.39 % for Hungary and –24.09 % for Poland).

In the case of Bulgaria, the GRNN model showed a relative error of –14.92 % for 2008. In fact, the landfill site in Sofia was closed between October 2005 and December 2007, which led to a temporary storage of 2.5 % of the collected municipal waste in 2005 and 10 % in 2006 and 2007. These amounts were not included in the figures for municipal waste generated and also caused a drop in the municipal waste landfilled in 2006 and 2007.[34] Considering all these factors, changes in MWG for the training years 2005 and 2006 could be a reasonable explanation for the higher relative error for the year 2008.

CONCLUSIONS

This study shows the development of a model for predicting emissions of CH_4 at the national level, using artificial neural networks (ANNs) and widely available sustainability, economic and industrial indicators as their inputs. Sustainability and the economic parameters considered to contribute the most to CH_4 emissions were used as input parameters for the models: gross domestic product (GDP) as a measure of industrial activity, waste generation and disposal, surface of land under fields of rice, number of cattle and the primary production of gas. Two ANN architectures were used, a back propagation neural network (BPNN) and a general regression neural network (GRNN), and both models were developed and tested using annual data for European countries for the period from the year 2000 to 2008, and subsequently compared with a corresponding multiple linear regression (MLR) model.

The performance evaluation of the models was realized using multiple statistical performance indicators. Based on the results of the evaluation, it could be concluded that the ANN models provide good and reliable predictions of CH_4 emissions, which were significantly better than the conventional MLR model. It was shown that the GRNN model demonstrates similar, but somewhat better predictions in comparison to the BPNN model.

The key advantage of the ANN approach in comparison with conventional emission inventory-based models is that the ANN models use a drastically smaller number of input parameters. In addition, emission inventory-based models use strictly defined input parameters the determination of which requires substantial field studies, while the proposed ANN approach uses widely available sustainability, economic and industrial parameters as its inputs. Furthermore, the ANN models allow simulation of different CH_4 emission scenarios by changing the values of input variables, for example to gain foresight because of proposed regulatory emission reduction actions. A developed ANN model could be applied for the prediction of emissions of CH_4, which could be very helpful in the implementation of sustainable development strategies and environmental management policies.

Further research is planned to expand the model to include other environmental quality indicators, such as emission of ammonia, ozone precursors, and in the application of new techniques for the optimization of the inputs, such as principal component analysis, correlation analysis and genetic algorithms.

SUPPLEMENTARY MATERIAL

Details of the two ANN architectures, s well as training and test datasets, Tables S-I and S-II, are available electronically from http://www.shd.org.rs/JSCS/, or from the corresponding author on request.

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ИЗВОД

МОДЕЛОВАЊЕ ЕМИСИЈЕ МЕТАНА ПРИМЕНОМ ВЕШТАЧКИХ НЕУРОНСКИХ МРЕЖА

LIDIJA J. STAMENKOVIĆ[1], DAVOR Z. ANTANASIJEVIĆ[2], MIRJANA Đ. RISTIĆ[1], ALEKSANDRA A. PERIĆ-GRUJIĆ[1] и VIKTOR V. POCAJT[1]

[1]*Универзитет у Београду, Технолошко–металуршки факултет, Карнегијева 4, 11120 Београд* и [2]*Универзитет у Београду, Иновациони центар Технолошко–металуршког факултета, Карнегијева 4, 11120 Београд*

У овом раду приказан је развој модела, заснованог на вештачким неуронским мрежама (*Artificial Neural Network* – ANN), за предвиђање вредности емисије метана на националном нивоу, при чему су као улазне променљиве коришћени економски и индустријски индикатори и индикатори одрживог развоја. Тестиране су две различите ANN архитектуре: неуронска мрежа са пропагацијом грешке уназад (*Backpropagation Neural Network* – BPNN) и неуронска мрежа са општом регресијом (*General Regression Neural Network* – GRNN). Развијен је и линеарни модел заснован на вишеструкој линеарној регресији (*Multiple Linear Regression* – MLR) са којим су упоређене перформансе наведених ANN модела. ANN и MLR модели су развијени и тестирани коришћењем података за 20 земаља Европске уније за период од 2000. до 2008. године. Поређење предвиђања ANN модела са актуелним вредностима CH_4 емисије је показало да се овом методологијом емисије метана на националном нивоу могу успешно и прецизно одредити до две године унапред, при чему је ANN модел знатно прецизнији у поређењу са MLR моделом. Наведени ANN модел се може користити као подршка при доношењу националних стратегија одрживог развоја.

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