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Adaptive-network-based fuzzy inference system (ANFIS) model-based prediction of the surface ozone concentration

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Abstract: This paper presents the results of modeling the tropospheric concentration of ozone in dependence on volatile organic compounds - VOCs (benzene, toluene, *m*- and *p*-xylene, *o*-xylene and ethylbenzene) and inorganic compounds – NO_x (NO and NO₂) CO, H₂S, SO₂, and particulate matter (PM₁₀) in the ambient air, in parallel with meteorological parameters, *i.e.*, temperature, solar radiation, relative humidity, and wind speed and direction. The modeling was based on measured results obtained during the year 2009. The measurements were performed at the measuring station located within an agricultural area, near the city of Zrenjanin (Serbian Banat, Serbia). Statistical analysis of obtained data, based on bivariate correlation analysis, indicated that accurate modeling could not be performed using the linear statistics approach. Moreover, considering that almost all the input variables have wide ranges of relative change (ratio of variance compared to range), the nonlinear statistic analysis method based on only one rule describing the behavior of the input variable most certainly would not present sufficiently accurate results. For these reason, the employed modeling approach was based on Adaptive-Network-Based Fuzzy Inference System (ANFIS). The model obtained using the ANFIS methodology resulted in high accuracy, with a prediction potential of above 80 %, considering that obtained determination coefficient for the final model was $R^2 = 0.802$.

Keywords: ANFIS; modeling; NO_x; ozone; VOCs.

INTRODUCTION

Ozone plays an important role in controlling the chemistry and chemical composition of the troposphere. On the other hand, tropospheric ozone is a unique pollutant in that it is not emitted directly into the ambient air. Ozone enters the troposphere from the stratosphere.¹ A major part of tropospheric

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ozone, however, is also produced and destroyed in the troposphere by chemical reactions with different organic and/or inorganic compounds, catalyzed by solar radiation.^{2–5}

Tropospheric ozone may have a negative impact on the environment and public health when present in the lower atmosphere in excess quantities. Human health, terrestrial ecosystems, and the degradation of materials are impacted by poor air quality resulting from high ozone levels caused by photochemical ozone production of human-emitted precursors. In establishing the quality of ambient air standards, regulations were introduced to set limits on the emissions of pollutants in such a way that they may not exceed the prescribed maximum values. Due to its harmful impact on human health and on vegetation in rural areas, the new European Directive 2008/50/EC limits the ozone concentration in ambient air according to the AOT40 index.^{6–8} The AOT40 index could be used to evaluate the potential risk that ozone could pose to the vegetation in an investigated area during the period of plant growth.

According to available literature, most authors calculate that the tropospheric production of ozone by photochemistry is much larger than the ozone influx from the stratosphere.¹ This was also confirmed by NASA's "Global Tropospheric Experiment", which was facilitated during February–March 1994.¹ As a result of this experiment, the column O₃ photochemical production rate at subtropical latitudes determined for the western Pacific was found to be nearly 12 times larger than the nominal average Northern Hemispheric flux of O₃ from the stratosphere.⁹

Local changes in tropospheric ozone concentrations, such as stagnation episodes or altered transport patterns, could also be the result of climate changes and *vice versa*. The potential influence on climatic changes, as well as the oxidizing impact of tropospheric ozone, is significant through the entire depth of the troposphere. On the other hand, near ground levels of the troposphere have an important influence on the air quality.¹⁰

A deficit of representative observation locations in some parts of the world, with observational records of 15 years or more, is hampering the determination of long-term changes in tropospheric ozone concentrations on a global scale. This is especially a case in under developed and transitional countries, where organized measurement of the tropospheric concentration only commenced during the first decade of the 21st century.

Accordingly, measurements of the ozone concentration in the ambient air started in Serbia a few years ago. However, a comprehensive study concerning its genesis, level of concentration and possible risks that it presents to human health has not yet been performed because information on ozone dependences in this region of Europe is limited. The aim of this study was to obtain an insight into ground-level ozone concentrations in the medium region of the Serbian Banat, a

major agro-industrial region in Serbia, and to explore the possibility of determining dependencies between ozone concentration and important predictors.

According to literature investigations, the ozone concentration is either NO_x or VOCs sensitive, where NO_x stands for inorganic components and VOCs stands for volatile organic components.^{11–14} For this reason, in parallel with the ozone concentration, for investigations presented in this paper, measurements of NO_x (NO , NO_2), SO_2 , CO , H_2S , particulate matter (PM_{10}) and VOCs (benzene, toluene, *m*- and *p*-xylene, *o*-xylene and ethylbenzene) was facilitated. In addition, it was decided to investigate the correlation of the concentration of each gaseous pollutant to meteorological parameters, as suggested by different authors.^{12,15–17} Accordingly, the meteorological parameters (wind direction, wind speed, air temperature, solar radiation and relative air humidity) were also measured.

EXPERIMENTAL

The locality where the measurements were facilitated, Banat, is a region in southeastern Europe divided among three countries: the eastern part belongs to Romania, the western part to Serbia (the Serbian Banat, mostly included in Vojvodina except for the small part, which is included in Central Serbia) and a small northern part belongs to Hungary.

Air quality monitoring and meteorological data

Continuous measurement of the air pollutants investigated in this study was facilitated using an automatic measuring station, located in the urban part of Zrenjanin city, center of the region. This station was originally assigned for acquisition of air pollution levels in the residential – business zone of the city, originating from exhaust gasses and other sources of pollution. The following air pollutants are continually measured at this location: BTEX (benzene, toluene, ethylbenzene and xylene) according to the EN 14662 method; Ozone according to the EN 14625 method, ISO 13964; carbon monoxide according to the EN 14626, ISO 4224:2000 method; PM_{10} (Particulate matter) according to the EN 12341 method; $\text{NO}/\text{NO}_2/\text{NO}_x$ (nitrogen oxides) according to the EN 14211 method and $\text{H}_2\text{S}/\text{SO}_2$ (sulfur compounds) according to the EN 14212, ISO 10498:2004 method.

Data collection

For modeling the dependence of ozone concentration on different predictors, the data obtained from the automated measuring station were used. The data were collected during the year 2009 in the period January–December. Measurement of the seventeen input parameters (X_i) and the one output (Y) parameter was enabled using the above-described measuring station, with data acquisition in the database at one-hour intervals. Before the model building phase, all the data points were examined for potential outliers. The measurement intervals, during which some of investigated input parameters were not recorded, for different reasons, were eliminated. After this, 1477 data sets remained for further analysis.

Specific details connected with Experimental are given in Supplementary material to this paper.

RESULTS AND DISCUSSION

The values of the measured input parameters (X_i) and the air quality indicator investigated in this work – output of the process (Y) in the form of des-

criptive statistics results – are presented in Table I. According to the results presented in Table I, potential risk of the ozone pollution in the air is obvious in this region, considering that measured hourly ozone concentration was in the range up to $162 \mu\text{g m}^{-3}$, which is above prescribed maximal value.

TABLE I. Values of the input (X_i) and the output (Y) variables of the model – descriptive statistics of 1477 data sets

Measured parameter	Unit	Model symbol	Range	Min.	Max.	Mean		SD	Var.
						Statistic	SE		
SO ₂	$\mu\text{g m}^{-3}$	$X_{1,1}$	220.4	0.0	220.4	17.651	0.5962	22.9147	525.082
CO	$\mu\text{g m}^{-3}$	$X_{1,2}$	3937	0	3937	738.89	12.928	496.835	246845.423
H ₂ S	$\mu\text{g m}^{-3}$	$X_{1,3}$	73.91	0.00	73.91	1.9636	0.16270	6.25268	39.096
NO	$\mu\text{g m}^{-3}$	$X_{1,4}$	232.4	0.7	233.1	28.495	0.7208	27.7018	767.388
NO ₂	$\mu\text{g m}^{-3}$	$X_{1,5}$	125.8	4.0	129.8	32.967	0.5473	21.0322	442.354
NO _x	$\mu\text{g m}^{-3}$	$X_{1,6}$	446.3	5.5	451.8	76.516	1.5230	58.5321	3426.008
PM ₁₀	$\mu\text{g m}^{-3}$	$X_{1,7}$	378.9	0.0	378.9	42.078	0.9161	35.2083	1239.627
Benzene	$\mu\text{g m}^{-3}$	$X_{2,1}$	14.40	0.00	14.40	1.6015	0.05703	2.19180	4.804
Toluene	$\mu\text{g m}^{-3}$	$X_{2,2}$	29.33	0.00	29.33	2.4257	0.07458	2.86618	8.215
<i>m</i> -, <i>p</i> -Xylene	$\mu\text{g m}^{-3}$	$X_{2,3}$	20	0	20	1.47	0.058	2.233	4.987
<i>o</i> -Xylene	$\mu\text{g m}^{-3}$	$X_{2,4}$	9.55	0.00	9.55	0.4682	0.03126	1.20120	1.443
Ethylbenzene	$\mu\text{g m}^{-3}$	$X_{2,5}$	10	0	10	0.42	0.030	1.143	1.306
Wind direction	°	$X_{3,1}$	344	10	354	188.11	1.837	70.605	4985.082
Wind speed	m s^{-1}	$X_{3,2}$	5.39	0.18	5.57	1.6843	0.02270	.87241	0.761
Air temperature	°C	$X_{3,3}$	47.6	-12.5	35.1	15.136	0.2498	9.5989	92.140
Solar radiation	W m^{-2}	$X_{3,4}$	844	4	848	136.36	5.452	209.518	43897.918
Relative humidity	%	$X_{3,5}$	75	17	92	64.96	0.434	16.688	278.491
Ozone	$\mu\text{g m}^{-3}$	Y	160.7	1.3	162.0	70.111	0.8850	34.0110	1156.750

Defining the linear correlation dependence between the output and the input parameters with a significant value of the coefficient of correlation (R^2) provides the possibility of predicting a potential excess O₃ concentration in the air in the investigated area using linear statistical analysis methods, such as multiple linear regression analysis (MLRA). MLRA is one of the most widely used methodologies for expressing the dependence of a response variable on several independent variables.²¹ For defining the linear correlation dependence in the form: output of the model $Y = f(\text{input})$ of the model ($X_{1,1}$ – $X_{3,5}$), a bivariate correlation analysis was performed. As the result of this analysis, the Pearson correlation (PC) coefficients with the corresponding statistical significance were calculated (Table S-I). In cases where the values of the PC coefficients of the output and most of the input variables are above a value of 0.5 with a high statistical significance ($p < 0.05$), the linear modeling approach should be taken into consideration.

However, according to values presented in Table S-I of the Supplementary material to this paper, it could be concluded that there was not a high linear dependence between the ozone concentration in the air (Y) and the input variables, with the exceptions of the correlations $Y-X_{3,3}$ ($r = 0.647$; $p < 0.01$) and $Y-X_{3,5}$ ($r = -0.496$; $p < 0.01$), although statistical significance was recorded for most of the correlated pairs. According to these values, it was decided that using MLRA to obtain dependence between the ozone concentration and the investigated predictors would not result in a high accuracy.

A low value of correlation between two variables does not automatically mean that interdependence of their behavior does not exist. This is only an indicator that the linear modeling approach cannot describe their intercorrelation. This is usually good indicator that further modeling should be based on the dynamic behavior of the variables.¹⁹ In such cases, modeling could be facilitated using a nonlinear statistic approach, such as Artificial Neural Networks (ANNs) – in cases where the input variables do not have wide range during the complete time interval of observation,^{15,18,20} or an Adaptive-Network-Based Fuzzy Inference System for variables with a wide range of change.^{21,22}

Modeling approach based on an adaptive-network-based fuzzy inference system

In recent years, artificial intelligence (AI) based methods have been proposed as alternatives to traditional linear statistical ones in many scientific disciplines. The literature demonstrates that AI models such as ANN and neuro-fuzzy techniques are successfully used for air pollution modeling and forecasting.^{22–28}

According to the measurement series for the variables presented in Table I, it can be concluded that almost all have a wide range of relative change (ratio of variance compared to range). For example, the relative change of variables ranges from 37.64 for H_2S to 5.32 in case of CO . Accordingly, a modeling approach based on one rule describing the dynamic changes of the input variables, belonging to a group of nonlinear statistic analysis methods (such as ANNs), probably would not result with a sufficiently accurate prediction.²⁵ For this reason, the further modeling approach was based on an Adaptive-Network-Based Fuzzy Inference System (ANFIS).

As a basis for the construction of a set of fuzzy if–then rules, the ANFIS system based on selected membership functions can be used. The ANFIS structure is obtained by embedding the fuzzy interference system into the framework of adaptive networks.²⁹ An adaptive network is a network structure consisting of a number of nodes connected through directional links. The outputs of these adaptive nodes depend on modifiable parameters pertaining to these nodes.³⁰ The pattern in which these parameters should be iteratively varied, aimed at minimizing the final error, is specified by the learning rule. Moreover, according to Takagi

and Sugeno, the fuzzy inference system (FIS) is a framework based on fuzzy set theory and fuzzy if–then rules.³¹ The three main components of a FIS structure are: a rule base, a database and a reasoning mechanism. The appropriate number of if – then rules for levels of ranges of the input variables is located in the rule base. An example of a rule used in the investigations presented in this paper might be “registered ozone concentration in the air will be high if the wind speed is low”, where items such as low and high represent linguistic variables. The database defines the membership functions applied in the fuzzy rules and the reasoning mechanism performs the inference procedure.³²

In this way, for example, if there are two input variables (X_1 and X_2), and assuming that their ranges can be divided into two levels, there would be the rule base with two rules for modeling the value of the output variable Y :

Rule 1. If X_1 is in the range A_1 and X_2 is in the range B_1 , then:

$$f_1 = p_1x_1 + q_1x_2 + r_1;$$

Rule 2. If X_1 is in the range A_2 and X_2 is in the range B_2 , then:

$$f_2 = p_2x_1 + q_2x_2 + r_2.$$

In the case when $f(x_1, x_2)$ is a first-order polynomial, the model is called a first-order Sugeno fuzzy model.

The graphical presentation of a general ANFIS network is presented in Fig. 1. The procedure for the construction of such an ANFIS structure is described in details in the literature,²⁵ where a similar modeling approach was used to predict the potential increase in the SO_2 concentration in the ambient air near a copper smelter. The ANFIS architecture can be presented with five layers, in which X_1 and X_2 are inputs to the nodes in layer 1, A_i and B_i are the linguistic labels of the ranges of the input variables (small, large, *etc.*) associated with the node function. Membership functions of the nodes located in layer 1 ($O_i^1 = \mu_{A_i}(X_i)$ or $O_i^2 = \mu_{B_i}(X_i)$) specify the degree to which the given X_i satisfies the quantifier A_i , B_i , *etc.* Usually, membership functions are either bell-shaped with a maximum equal to 1 and a minimum equal to 0, or a Gaussian function. Nodes located in layer 2 are multipliers, which are multiplying the signals exiting the layer 1 nodes. For example $O_i^2 = W_i = \mu_{A_i}(X_i) \times \mu_{B_i}(X_i)$, $i = 1, 2$, *etc.* The output of each node represents the firing strength of a rule. The i -th node of layer 3 calculates the ratio of the firing strength of the i -th rule to the sum of the firing strengths of all rules. In this way, $O_i^3 = W_i = W_i / (W_1 + W_2 + \dots)$, $i = 1, 2, \dots$. Every node i in layer 4 has a node function of following type: $O_i^4 = \bar{W}_i \cdot f_1 = \bar{W}_i \cdot (p_i x_1 + q_i x_2 + r_i)$, where p_i , q_i and r_i will be referred to as consequent parameters. The single node of layer 5 is the node that computes the overall output as the summation of all incoming signals, *i.e.*:

$$O_i^5 = \sum_i \overline{W_i} f_i = \frac{\sum_i W_i f_i}{\sum_i W_i}$$

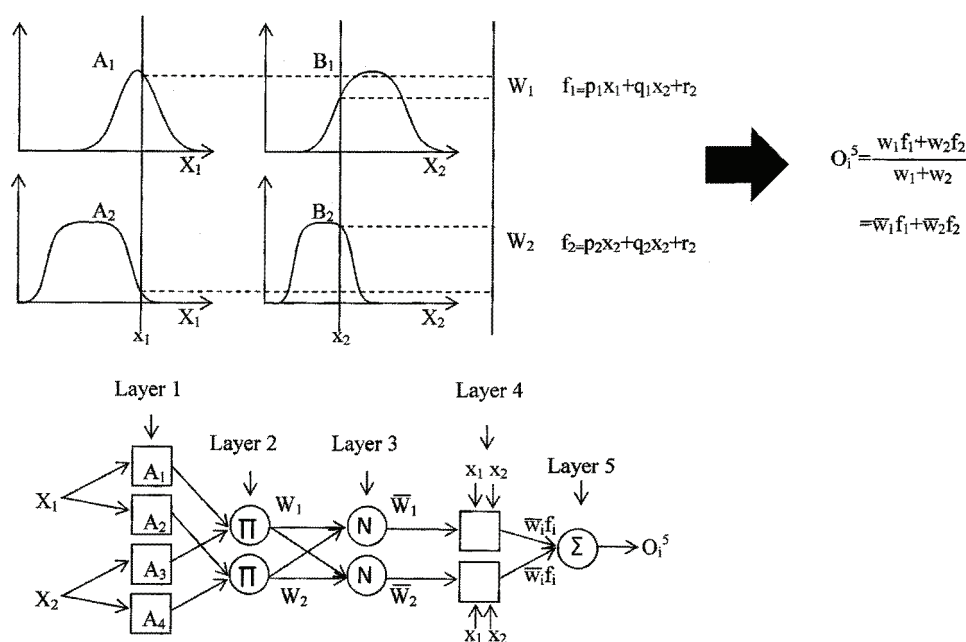


Fig. 1. Graphical presentation of the ANFIS.

The training of the parameters in the ANFIS structure is accommodated according to the hybrid learning rule algorithm, which is an integration of the gradient descent method and least square methods. In the forward pass of the algorithm, the functional signals advance until layer 4 and the consequent parameters are identified by the least squares method to minimize the measured error. In the back propagation pass, the premise parameters are updated by the gradient descent method.³²

According to the number of input variables, their ranges and the variations, presented in Table I, it was decided that a two-rule ANFIS network should be applied. A Gaussian function was selected as the membership function. There were 17 input variables ($X_{1,1}$ to $X_{3,5}$) with one output variable (Y).

To apply the ANFIS methodology, the assembly of 1477 input and output samples was divided into two groups. The first group consisted of 1067 ($\approx 70\%$) randomly selected samples, and this group was used to train the model, whereas the second group consisted of the 410 ($\approx 30\%$) remaining samples from the starting data set, and this group was used to test the model. The selection of the

variables for these two stages was realized using a random number generator, which was based on a Bernoulli distribution. During the training phase, correction of the weighted parameters (p_i , q_i , r_i , *etc.*) of the connections was achieved through the necessary number of iterations, until the mean squared error between the calculated and measured outputs of the ANFIS network was minimal. During the second phase, the remaining 30 % of the data was used for testing the “trained” network. In this phase, the network used the weighted parameters determined during the first phase. These new data, excluded during the network training stage, were incorporated as new input values (X_i), which were then transformed into a new output (Y). Matlab ANFIS editor was used for the calculations realized in this study.

Accordingly, the network-training phase was performed iteratively until the moment when the error between measured and calculated values of output variable (the O_3 concentration in the air – Y) was not minimized and remained constant. In the case of the investigation presented in this paper, the optimal number of iterations (epochs) was 10. The obtained results from the training stage could be evaluated by comparison of the calculated values of Y with the measured ones (Fig. 2).

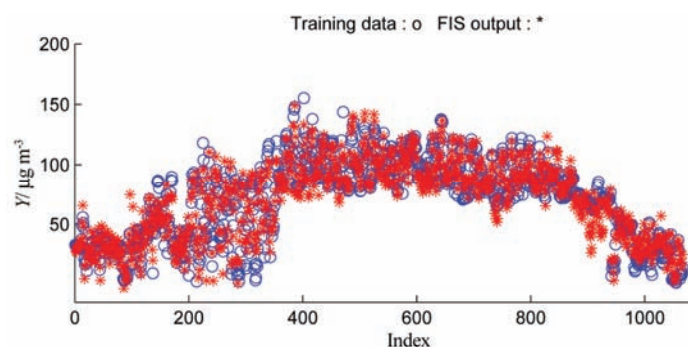


Fig. 2. Dependences between the calculated and measured values for the ozone concentration in the training stage (measured – $\circ$; model predicted – $*$).

The test set (total 410 vectors), which examined the fidelity of the model, showed that the model could be used to estimate the O_3 concentration quite satisfactorily. A comparison of the measured and ANFIS model calculated values for the testing stage are presented in Fig. 3. It could be concluded that excellent fitting was obtained.

In the training stage, the ANFIS modeling approach predicted the ozone concentration in the air with a determination coefficient $R^2 = 0.92$ (Fig. 4), which does represent a large significance. The value of the determination coefficient (R^2) for the test set was smaller to some extent 0.802 (Fig. 5), however, the results showed that the ANFIS modeling methodology led to acceptable funct-

ional dependencies between the selected variables and the O_3 concentration. Accordingly, using the model described in this paper, the O_3 concentration in the air could be predicted as the function of investigated input variables, with an accuracy of above 80 %.

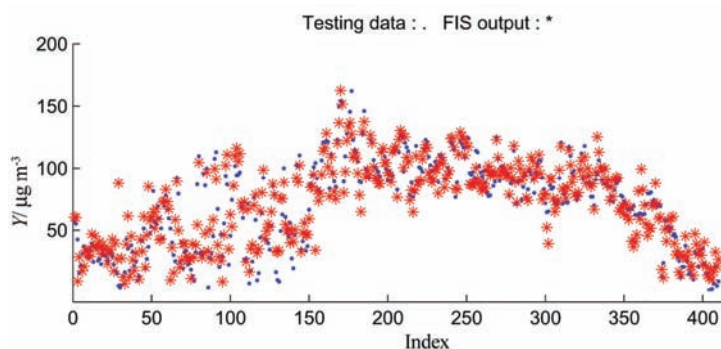


Fig. 3. Dependences between the calculated and measured values for the ozone concentration in the testing stage (measured – $\circ$; model predicted – $*$).

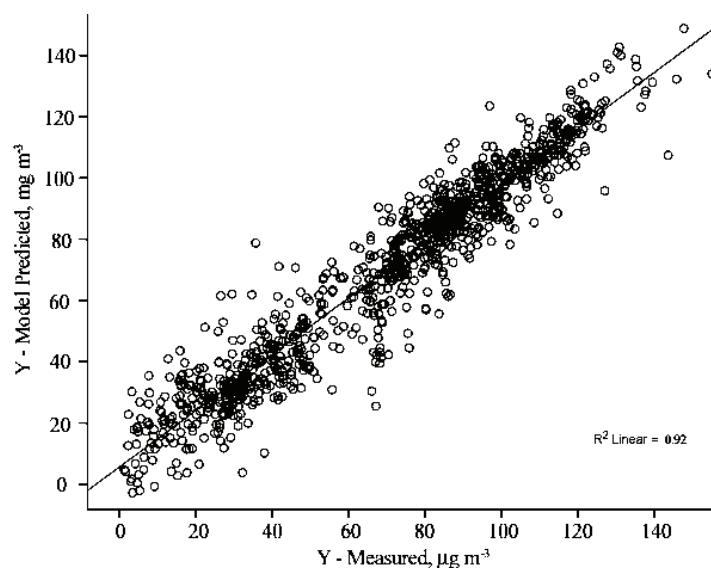


Fig. 4. Coefficient of determination between the measured and model predicted O_3 concentration in the training stage.

Final validation of the model accuracy was performed on data collected during two months of 2012. The data were collected during August and December to assess predictability of the model in different seasons. The obtained coefficients of determination were $R^2 = 0.782$ and 0.764 for August and December, respectively. These values indicated that the developed ANFIS model could be

used for a sufficiently accurate prediction of the dependence of the ozone concentration on the investigated input parameters in the investigated region.

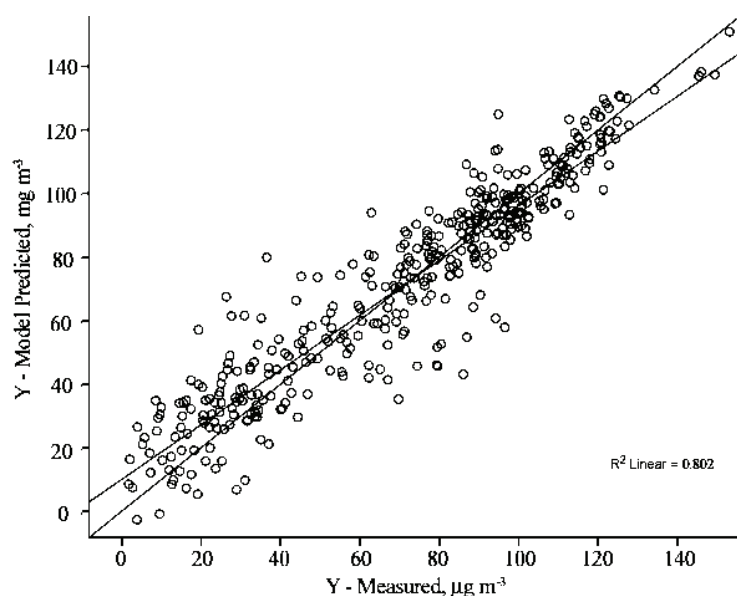


Fig. 5. Coefficient of determination between the measured and model predicted O_3 concentration in the testing stage.

CONCLUSIONS

Considering the importance of the daily O_3 concentrations in the atmosphere of urban regions, this research was aimed at developing proper prediction models using the ANFIS model. Since input selection is a significant step in modeling, it was decided to measure both VOCs and NO_x as potential predictors. Additionally, meteorological parameters were recorded. The goodness of final model fit was evaluated using R^2 values. The obtained values of 0.92 and 0.802 in training and testing stage, respectively, demonstrated that an accurate prediction model for ozone could be obtained using the ANFIS model. The obtained results could be used for further analysis of investigated problem. Further analysis would include the sensitivity of the model to the separate influence of VOCs and NO_x . In this way, it is planned to obtain a model that would be able to determine the origin of daily O_3 concentration changes, *e.g.*, is the ambient O_3 concentration VOCs or NO_x sensitive. This is of importance for determining the reasons for O_3 concentrations in excess of the limiting values in this region.

SUPPLEMENTARY MATERIAL

Details of geography, air quality monitoring and meteorological data as well as correlation matrix for the input and the output variables are available electronically from <http://www.shd.org.rs/JSCS/>, or from the corresponding author on request.

ИЗВОД
ПРЕДВИЂАЊЕ КОНЦЕНТРАЦИЈЕ ПОВРШИНСКОГ ОЗОНА НА ОСНОВУ ANFIS
МОДЕЛА

МАРИЈА САВИЋ, ИВАН МИХАЈЛОВИЋ, МИЛИЦА АРСИЋ И ЖИВАН ЖИВКОВИЋ

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У раду су приказани резултати моделовања концентрације тропосферског озона као зависност од испарљивих органских једињења – VOCs (бензен, толуен, *m*-, *p*-ксилен, *o*-ксилен и етилбензен); неорганских једињења – NO_x (NO и NO₂), CO, H₂S, SO₂ и PM₁₀ (*particulate matter*)) у ваздуху паралелно са метеоролошким параметрима: температура, сунчево зрачење, релативна влажност, брзина и правац ветра. Моделовање се заснива на измереним резултатима добијеним у току 2009. године. Мерења су обављена на мерној станици која се налази у пољопривредном подручју, у близини града Зрењанина. Статистичка анализа добијених података, на основу биваријантне корелационе анализе, показује да прецизно моделовање не може бити изведено помоћу приступа линеарне статистике. Такође, с обзиром на то да скоро све улазне варијабле имају широк спектар релативне промене (однос варијансе у односу на опсег), метод нелинеарне статистичке анализе заснован на само једном правилу за описивање понашања улазне варијабле, највероватније не би могао да представи довољно прецизне резултате. Из тог разлога, моделовање је засновано на ANFIS приступу. Модел добијен коришћењем методологије ANFIS резултирао је високом прецизношћу, уз потенцијално предвиђање изнад 80 %, с обзиром на то да је добијени коефицијент детерминације за коначни модел био $R^2 = 0,802$.

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