QUANTIFYING NONLINEARITIES IN GROUND LEVEL OZONE BEHAVIOR AT MOUNTAIN-VALLEY STATION AT OVNARSKO, BULGARIA BY USING NEURAL NETWORKSA.

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Abstract. Using data from a measuring station in Ovnarsko, Bulgaria, from May to September (1994-97), this paper shows that in order to accurately capture the ozone behavior, the statistical models of hourly surface ozone concentrations need to represent the non-linear relationship between predictor variables. Comparisons between multiple linear regression and multilayer perceptron neural network model of hourly surface ozone concentrations are made in order to reflect the quantity of these effects. The multilayer perceptron model captures the underlying function between the meteorological predictor variables and hourly ozone concentrations more accurately than the regression one. While the first one is not physically interpretable, the regression model gives much better glance on the physical background that controls these relationships. The predictor variables are meteorological parameters, measured at the station. No chemical predictand variables are used in the models, because there are no pollution sources in close distance around the study area.

Key words:: Ground level ozone, neural network, multiple regression

Introduction

During the past two decades, the amount of knowledge about the relationship between the meteorological elements and surface ozone concentrations has been described in many papers (Neu et al., 1993; Eder et al., 1994; Olszyna et al., 1997; Derwent et al., 1998; Gardner and Dorling, 2000), and statistical models of diurnal variations in surface ozone concentrations are made to quantify these relationship (Hubbard and Cobourn, 1998; Avramov et al., 1999; Gardner and Dorling, 2000). Working on the development of these models one encounters the problem of complicated non-linear relationships between predictor variables. Neural network (NN) models have the advantage to reflect non-linear interactions between modeled variables and its precursor, hence NN give better results (than linear regression) in the development of models of hourly surface ozone concentrations. Here a comparison between multilayer perceptron and linear regression model is made in order to quantify the complex relationship ozone - meteorological parameters of hourly data set from the mountain station in Ovnarsko, Bulgaria. The neural networks are capable of modeling the non-linear functional relationship between the predictor variables, but these relationships remain physically non interpretable. In contrast, the linear regression models are physically well explained.

Last ten years the neural networks, mostly the multilayer perceptron (MLP), have found application in the atmospheric sciences and particularity in the ozone studies (O. Pastor-Barcenas et al., 2005; E. Agirre-Basurko et al., 2004; Jeong-Sook Heo et al, 2004; Gardner and Dorling, 1998). One of their most important features is their ability to represent any smooth functional relationship between one or more predictor and predictant variables. If the results of a correctly trained MLP model almost match these from a linear regression model, then it can be confidently assumed that the functional relationship between the chosen input and output variables is linear. If the MLP models outperform the linear model, than in order to represent the relationship that is modeled it is necessary to include some form of nonlinearity. So the comparison between the linear model and the MLP model can be used to determine the complexity of the underlying function. Well-trained MLP models are at least as good as linear model and often outperform them.

Yi and Prybutok (1996) have created a neural network model for predicting daily maximum concentration of surface ozone in Central Europe, using the available meteorological variables from ground based stations as predictors. They show that NN outperform the regression models. On the other side Comrie (1997) comparing neural network and linear regression models for predicting daily maximum concentration of surface ozone in US urban regions, again using meteorological variables establishes only slight improvement when using NN. As mentioned by Gardner and Dorling (1998) this means that relationship between the analyzed meteorological variables and maximum daily concentrations of ozone can be well represented in terms of a linear model. If additional predictor variables are taken into account, probably (and most likely) the linear model will appear to be less effective than the NN one.

Although many factors influence ozone concentration, diurnal meteorological variations are the one that best explain diurnal variation of ozone concentrations. Understanding the relationship between meteorology surface layer parameters and ozone concentration is a key factor in choosing the right variables for ozone predicting models. Depending on the region the data are taken from, ozone precursors are different.

Below some of the most important meteorological parameters, which influence ozone concentration are mentioned. Having in mind the investigations concerning the relationship meteorology - troposphere ozone, one can mention the following meteorological parameters as important precursors of ozone: *solar irradiation*, influencing the speed and amount of photochemical production of ozone; *temperature*, controlling the rate of photochemical production; *vertical temperature gradient*, influencing the vertical mixing in the atmosphere and thereafter the ozone concentration near the ground; *surface* *winds*, controlling the formation of the diurnal pattern of surface ozone concentrations in mountain valleys and costal areas, when synoptic scale pressure gradients are small, and thermal local winds prevail (Brother et al., 1985; Prevot et al., 1993); *aloft winds*, responsible for the transport of ozone and its precursors; *precipitation*, decreasing the ozone concentration by means of wet deposition; and *relative humidity*, chemically controlling the ozone concentration.

In the present paper, the statistical relationships between ozone concentrations and its meteorological precursors are investigated by means of linear regression and neural network model (NN). The primary objective of this paper is to draw attention on the instrument of NN and its application in ozone studies, as well as to quantify the importance of the non-linear interactions between ozone and its precursors and also between the predictor variables.

Neural networks (The Multilayer Perceptron)

As mentioned in the introduction, the main advantage of multilayer perceptron is its ability to model any smooth functional relationship between one or more predictor and predictand variables. The nature of the functional relationship is learned directly from the data in the process of the supervised learning. In many cases, determining the relationship is difficult or even impossible by means of standard statistical techniques and then the neural network seems to be of much advantage.

The network could be considered as a model with input and output data, and the weights and thresholds playing the role of free parameters of the model. In this way it is possible to model a function of arbitrary complexity, and the number of neurons in each layer determines the complexity of the function. In each particular problem the exact number of hidden layers and the number of neurons in each layer has deep meaning.

Training the neural network is carried out by the training algorithms, which aim to set the weights and thresholds of each neuron, so as to minimize the prediction error made by the network. The available historical data is used to automatically adjust the weights and thresholds in order to minimize this error. In NN the price we pay to model very complex functions is the practical impossibility to absolutely minimize the network error. Although we can adjust the network to minimize its error, we can never be sure that it has reached the smallest possible value.

Although the best known and used training algorithm is back propagation it is not used in the present paper. Instead second order algorithms are used. Their choice is a consequence of many training procedures carried out in order to get the best network topology and training algorithm to fit the available data. Training of the multilayer perceptron was performed with the Levenberg - Marquardt training algorithm. It's based on a discreet Newton method for optimization. It is the fastest of all the training algorithms, and is the one to give best results on the data used in this paper.

The most desirable property of a network is its ability to generalize with unknown data. The network is trained to minimize the error on the error surface of the training set, but the possibility of an indefinite training set does not minimize the error on the real error surface - the one of the real unknown function, which we try to model. The most important

manifestation of this difference is the problem of over-learning. To overcome this problem a part of the training cases is set apart, to make independent check as the algorithm progresses - it is called verification set. The choice of the right structure of network is made by using this set. If in the process of training, the verification error stops to decrease and begins to increase, it is a sign that over-learning has began and training must be stopped.
To verify the ability of the model to generalize, after training is done, a third set of data - test set is set aside. The final model is tested on the test set in order to assure that results from the training and verification sets are correct.

Data

In the study, data from the experimental study site Ovnarsko is used. It is situated on the south slope of the Govedarci valley, located in the northwestern part of Rila Mountain, Bulgaria. The valley is surrounded by high ridges and is open to the northeast by a narrow mountain pass. The experimental site is situated at 1600 m above sea level. There are no anthropogenic sources for ozone precursors for at least 50 km in southern direction and about 30 km in western one. Besides, the mountain ridge is a high obstacle for the west to east transport of air pollutants. Diurnal cycle of ozone concentrations in Govedarci is highly influenced by the local winds (mountain - valley circulation).

Ground level ozone concentrations and meteorological measurements in Ovnarsko were carried out in the period 1994-97. The measurements were taken from spring to autumn.

Ozone was sampled through a Teflon filter at 2 m height. Due to the inherent stability of UV adsorption technique, the authors believe the measurement error of the ozone concentrations reported here to be within the limits of ± 5 ppb. The instrument Teflon filter was changed approximately every 15 days. The equipment is described in Table1.

Table 1 Measured variables and the apparatuses used

Measured variables in the station of Ovnarsko	**apparatuses**	**Period**	**Height**	**Units**
O_3	TECO 49	1 hour	1m	ppb
Wind speed	VAISALA WAA12	"	10 m	m/s
Wind direction	VAISALA WAA12	"	10 m	degree
Vertical speed		"	10 m	cm/s
Summed solar radiation	Eppley	"	2 m	W/m^2
Precipitation	MRI	"	1 m	mm
Air temperature	Vaisala HMP35C	"	4 m	°C
Relative humidity	Vaisala HMP35C	"	4 m	%
Air temperature	Vaisala HMP35C	"	0.5 m	°C
Relative humidity	Vaisala HMP35C	"	0.5 m	%
Wetness	Campbell Scientific	"	2 m	wet/dry
Soil heat flux	Rad Energy	"	-0.05 m	W/m^2
Soil temperature	thermocouples	"	-0.05 m	°C

Based on earlier publications, and taking into account the measurements at Ovnarsko, in this work the following parameters are taken as precursors for ozone concentrations:

Solar irradiation	at 2 m
Precipitation	at 1 m
Wind direction	at 10 m
Wind speed	at 10 m
Temperature	at 4 m
Relative humidity	at 4 m

The models and plots in this work are created over the hour mean values, except if mentioned others.

Results and discussion

Multiple regression analysis

Despite the nonlinear relationship between ozone and its precursors, regression has been commonly used to study this relationship. In this paper such analysis is done in order to establish the relationship between ozone and its precursors, and to compare the results with those obtained by neural networks.

Using the parameters mentioned above as predictors, the following linear regression equation 1) is obtained:

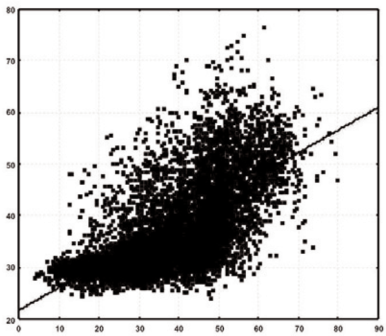

Figure1. Scatter plot of observed vs. predicted ozone concentrations for multiple regression analysis. Coefficient of correlation r = 0.66053

Table2 Multiple regression coefficients. "Beta" coefficients are the regression coefficients with standardised independent variables. "B" is the coefficients of the regression equation

	Beta	B
Free term		41.545
Solar radiation (Rad)	0.154	0.008
Precipitation (Prec)	0.072	1.832
Wind speed (WSp)	0.186	2.811
Wind direction (WDir)	0.056	0.009
Temperature (T)	0.151	0.380
Relative humidity (RH)	-0.31	0.380

$$O3 = 41.454+0.008Rad+1.832Prec+2.811WSp+0.009WDir+0.38T-0.198RH \quad (1)$$

For the abbreviations see table 2. In table 2 the coefficients in equation 1 (B) are presented together with the regression coefficients with standardised independent variables (Beta)

Figure 1 compares the measured ozone values and those obtained by means of linear regression. The best fit linear regression line from equation 1 is shown. Correlation coefficient is r = 0.66. Figure 2 shows the diurnal variation in ozone concentration for measured and regression modeled data. It is easy to see the characteristic diurnal variation of ozone at a "valley slope" site. The formation and breakup of the nocturnal boundary layer and the nocturnal katabatic wind have a major impact on diurnal variation. Ozone concentration decreases from the beginning of the nocturnal katabatic down slope wind (S - SW direction) and throughout the night, due to the surface deposition in the thin surface layer which is almost completely isolated from the upper air layers. The upper air layer - residual layer is not affected by the dry deposition mechanism, and there ozone concentrations stay higher. The concentration quickly rises during the morning, when solar heating of the surface results in intensification of the vertical mixing, especially after daily upslope and up valley winds are formed. The vertical instability during the day results in mixing the higher and ozone richer air masses with those from below (where the measurements are taken). That is the reason of the relatively high wind speed coefficient in the regression equation. Precisely the vertical mixing must be attributed to the wind shear, but as long as we do not have this information, it is represented in the term of the wind speed.

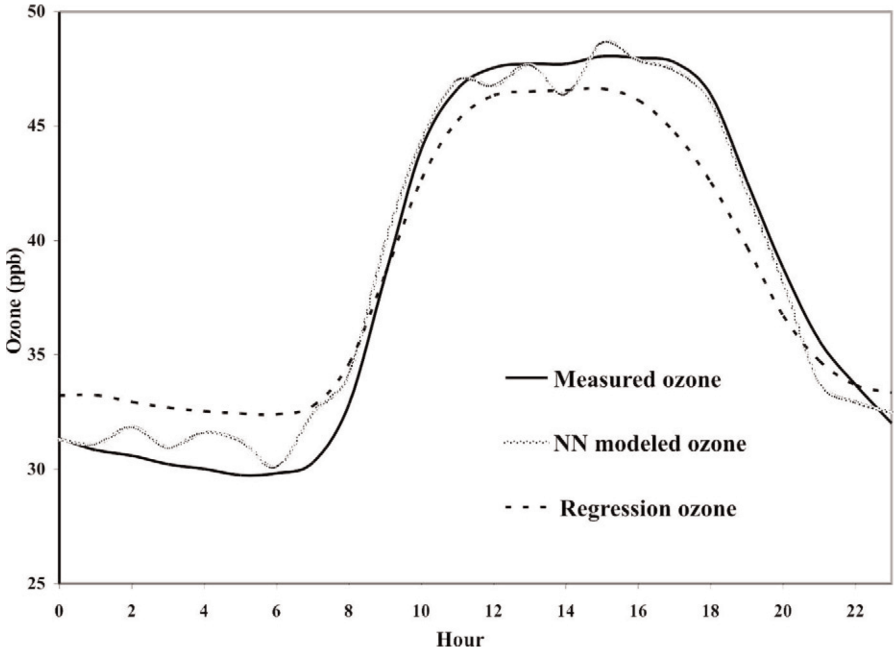

Figure2. Diurnal variations of ozone over the experimental period for measured values, perceptron modeled values and regression analysis values.

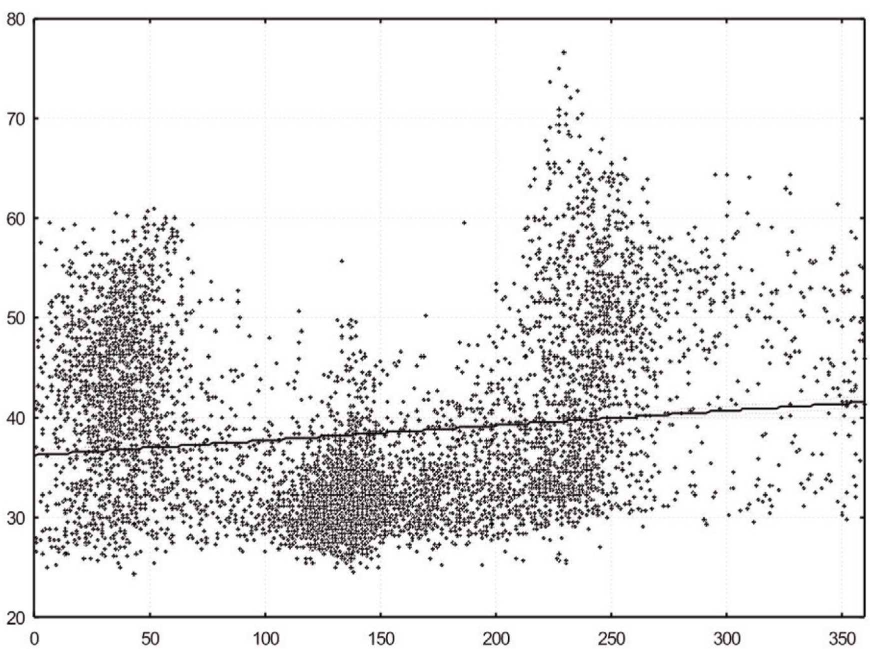

Figure3. Scatter plot of ozone concentration vs. wind direction

Relationship between wind direction and ozone concentration is shown in Figure 3. It is easy to see the nonlinearities between ozone concentration and wind direction as well as wind directions related to high ozone concentrations. The first maximum of ozone is related to wind coming from N - NE (0 - 45°). That is the daily up valley wind, which causes vertical mixing of the surface air with higher and ozone-richer air from the residual layer. The other maximum is in agreement with the synoptic scale western winds (27°). Here we find the highest ozone concentrations referred to intrusion of ozone rich air masses, originating from other places. The minimum of the ozone concentrations is connected with the nocturnal katabatic S - SW (180 - 225°) winds, which lead to strengthening the nocturnal inversion and depletion of ozone due to dry deposition.

Neural networks

Building a good NN model for predicting ozone concentrations requires the proper precursors to be chosen, and a good choice of three sets of data to be done. In the present work we have a set of 6877 cases. For training 3000 cases are randomly chosen. Verification set also contains 3000 cases that are randomly chosen, and the rest 877 cases are left for the test set. Experiments are carried out with similar initial conditions for different net topologies.

The coding of input and output data is defined in accordance with the computing units we use. In the experiments neurons with bipolar activation function - hyperbolic tangent are used. The simulations are carried out with Matlab. The precursor parameters are the same as in the linear regression analysis: solar irradiation, temperature, precipitation, wind direction, wind speed, relative humidity.

Having in mind the non-linear nature of the examined process, and the low correlation coefficient obtained by the multiple regression analysis we expected that using a two- or three- layer neural network would give better results. This is in agreement with many investigations (O. Pastor-Barcenas et al., 2005; E. Agirre-Basurko et al., 2004; Jeong-Sook Heo et al, 2004; Gardner and Dorling, 2000) We investigate variant network topologies. The choice of topology is connected with the amount of training cases the training dataset consists of. The number of neurons in the hidden layer is also important, because too many neurons will lead to exact remembering of the training dataset and to decrease of the generalization ability of the network. On the other hand if the number of neurons in the network is low, it will not be powerful enough to model the underlying non-linear relationship. Experiments were carried out with two-layer network with 5, 10, 15, 20, 25, 30, 35, 40, 45, 50 neurons in the hidden layer and with three-layer network with 10 neurons in the first hidden layer and from 5 to 50 neurons in the second hidden layer. The comparatively large number of cases in the training set (3000) guarantees that the high number of neurons will not lead to over-learning and loosing generalization abilities. Here the Levenberg-Marquardt training algorithm is used, which appeared to be the best one in this study. The number of cases in each subset is chosen on the basis of many experiments with different neural networks and training algorithms.

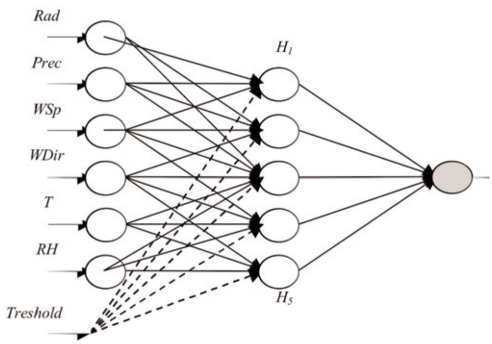

Figure4. Simplified structure of a neural network for predicting surface ozone concentration with one hidden layer and 5 neurons in it. Abbreviations are the same as in table 2.

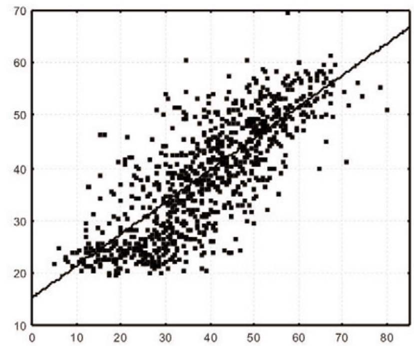

Figure5. Scatter plot of observed vs. predicted ozone concentrations for neural network. Coefficient of correlation r = 0.76681

The scheme of the model with one hidden layer is represented in Figure 4, where for convenience only part of the connections between neurons is shown. Table 3 represents the correlation coefficients between calculated and measured values for each topology of the network for the three subsets - training, validation and test set. The best topology of the network is with 45 neurons in the hidden layer - it gives the highest correlation coefficient in the test set - 0.77. Experiments with different combinations of the data in the three data sets confirm that this is really the best topology. Results, obtained with three layer network are not shown here, because they do not show better results.

Table 3: Correlation coefficients for different topologies of the network for the three subsets - training, validation and test set (with bold is shown the best topology)

Topology	*Training set*	*Verification set*	*Test set*
6 - 5 - 1	0.7793	0.76444	0.76527
6 - 10 - 1	0.79687	0.76849	0.76694
6 - 15 - 1	0.7873	0.75972	0.75814
6 - 20 - 1	0.79465	0.77263	0.76239
6 - 25 - 1	0.80203	0.76698	0.76681
6 - 30 - 1	0.79701	0.76268	0.77503
6 - 35 - 1	0.79138	0.76419	0.76369
6 - 40 - 1	0.80819	0.77547	0.7618
6 - 45 - 1	**0.79674**	**0.77058**	**0.77612**
6 - 50 - 1	0.80523	0.76503	0.75557

The linear correlation between the neural network ozone values and the measured ones is shown in Figure 5. The diurnal variation of ozone concentration obtained by the neural network model with topology 6-45-1 is shown in Figure 2 (gray line). It can be clearly seen that the curve giving the diurnal ozone variation obtained by neural network, is closer to the curve of the measured values and represents the diurnal ozone changes better than the one representing linear regression model.

The curve in Figure 2 is not as smooth as the other two curves, because it is a result of averaging over 877 cases, while the curves for the measured and calculated by the regression equations concentrations result from averaging over all 6877 cases. This reason eliminates any doubt that the network might be over-learned, which may rise given the fact that the NN curve is not smooth. The use of verification set is another reason to eliminate the doubt about over-learning.

Although outperforming the regression modeling technique, neural networks has one big offset - it is hard to draw any physical information out of it, i.e. no information from the neurons' weights and thresholds can be drawn about the weights of each predictor in the final score. Nevertheless, because of their better results, neural networks are commonly used during the past 10 years to solve non-linear problems of high complexity.

Conclusion

As can be seen from the regression analyses, ozone concentrations in the

Govedarci valley are strongly correlated with the thermal driven local winds system. During the night, the low level ozone concentration is reduced, because the down-slope wind separates the thin surface layer from the higher ozone rich levels in the boundary layer. During the day, up-valley wind supports the vertical mixing and entrainment of ozone rich air from above. The synoptic scale winds from west (bringing ozone rich air generated at other places) are responsible for the highest concentration of ozone in the valley.

In agreement with some former experiments (Gardner and Dorling, 2000), these results shows that significant increases in the evaluation and prediction of ozone concentrations are possible when Multilayer perceptron neural network is used. NN performed better, since they are unconstrained and allowed arbitrary interactions and nonlinear relationships between predictor variables. The diurnal ozone variation confirms that statement. Of course regression models are physically interpretable, while the MLP models cannot be easily interpreted, although various sensitivity test and model comparisons may provide insight into their physical meanings.

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Количествени оценки на нелинейните взаимодействия между предикторите на концентрацията на приземния озон в Овнарско, посредством използване на невронни мрежи.

В. Александров, С. Великов, Е. Донев, Д. Иванов

Резюме: Сравнявайки регресионен модел с многослойни невронни мрежи (многослоен перцептрон) върху данни от озон - метеорологична станция в т.б. Овнарско (Рила планина) е направена количествена оценка на влиянието на нелинейните взаимодействия между метеорологичните предиктори на приземния озон върху концентрациите му. Макар и невронните мрежи да не могат да обяснят физически влиянието на метеорологичните елементи върху концентрацията на озон, те се оказват по-добър инструмент (от регресионния анализ) в определянето на концентрацията на приземния озон при дадени метеорологични параметри. Така те се явяват надежден и евтин начин за предсказване на приземните озонови концентрации за разглежданата станция. В работата са използвани данни за вегетативния период на растенията (от Май да Септември), през който високите концентрации на озон оказват силно негативно действие върху растежа на горите.