Dispersion Modelling of Nitrogen Dioxide Coming from Timişoara Thermal Power Plants

D. Silaghi-Perju[*], D. Perju[**], H. Pîrlea[**], G. A. Dumitrel[**], L. Drăghici[**]

[*]POLITEHNICA University of Timisoara, Mechanical Engineering Faculty, 1 Blv. Mihai Viteazu, RO-300222 Timisoara, Romania

[**]POLITEHNICA University of Timisoara, Faculty of Industrial Chemistry and Environmental Engineering, 300006, Victoriei Square, nr. 2, Timisoara, Romania, harieta.pirlea@chim.upt.ro

Abstract: Nitrogen oxides, especially nitrogen dioxide, are pollutant substances that can affect the human health and the environment. Nitrogen dioxide emissions are important sources in the production of green house effect and photochemical smog. In the current global context when the accent on environmental pollution problems are more and more pressing, the reduction possibilities study of nitrogen dioxide emissions as well as the modelling ways of the phenomenon that accompany the dispersion of this pollutant in its cycle in nature is current and of a particular importance. In this paper the mathematical models that describe the dispersion of nitrogen dioxide emitted from the two thermal power stations (Center CET and South CET) of Timisoara city as a function of four meteorological parameters (temperature, humidity, pressure, wind speed) are presented. In order to develop and verify these models, the NO_2 daily average concentrations emitted from the thermal power stations as well as the daily average values of meteorological parameters for each day of the months: January, April, July and October 2004 were used. The testing of the mathematical models has been done by using the following performance criteria: average squared deviation (σ) and correlation coefficient (R). The models, as resulted from the values of the performance criteria prove to be enough accurate to describe the nitrogen dioxide dispersion in atmosphere.

Keywords: Nitrogen dioxide, dispersion

1. Introduction

Compared to the ordinary systems, in the case of the air pollution is almost impossible to bring the real system through different states with the planned changes of the input parameters (usually meteorological) in order to generate experimental data sets that should be used when designing the model [1,2]. Due to this reason, the design of such system is based upon certain experimental data taken under variable conditions – the meteorological parameters cannot be programmed. The dispersion of the polluting agents in the atmosphere is a dynamic process that takes place under relatively different conditions for every estimation, even if the pollution source may be constant. This is the reason why the design of such system may be carried out with less precision than in the case of the static systems.

The main factors that generally influence the dispersion of the polluting agents and particularly, the dispersion of the nitrogen dioxide are the following: the flow of the pollution source, the height of the pollution source, the Cartesian coordinates of the measuring point of the polluting agent concentration as well as the meteorological parameters. The previous studies have focused upon the influence of several meteorological parameters, such as the wind speed and direction [3-6], respectively: temperature, pressure, humidity and wind speed [7,8].

In this paper there will be presented the mathematical models that might define the influence which the simultaneous action of the 4 meteorological parameters has upon the dispersion phenomenon of the nitrogen dioxide in Timisoara City. Such models were obtained by means of the 2nd degree non-linear multiple regression, between the 5 parameters. The meteorological parameters herein considered are as follows: the temperature, the humidity, the pressure, the wind speed and direction.

2. Experimental

The most important stationary pollution source with nitrogen dioxide in Timisoara City is represented by the two thermal power stations that provides the city thermal heating: CET CENTRU and CET SUD. S.C. Colterm S.A manages two thermal power stations, respectively CET CENTRU and CET SUD. CET CENTRU has 6 boilers and 5 CAFs and the fuels used here are: black oil and natural gases. CET SUD manages two CAFs and two boilers and the fuels used here are: coal and natural gases.

In order to complete this study, we used the daily average values of the nitrogen dioxide concentrations emanated by CET CENTRU and CET SUD, as well as the daily average values of the meteorological parameters such as: temperature, pressure, humidity, wind speed, for January, April, July and October of 2004. Such data have

been legally provided by relevant institutions of Timisoara City:

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♦ The NO_x concentration resulted to every CAF and boiler of the two thermal heating stations: CET CENTRU and CET SUD, for every day in the 4 months subject herein: January, April, July and October in 2004.
♦ The features of all boilers and CAFs.
♦ The calorific power of the natural gases used, their chemical composition, the caloric power of the black oil as well as its chemical composition.
♦ The consumption of the natural gases for every boiler and CAFs, as well as the consumption of black oil and coal, the loading of the installation and the number of functioning hours per every day during the 4 months.

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♦ The daily average values of the meteorological parameters: temperature, pressure, humidity, wind speed for every day of the 4 months in 2004. These values have been measured at Pădurea Verde, the head quarter of this institution.

In order to obtain the daily average of the NO_x concentration derived from the two thermal heating stations, the EMPOL informatic system have been inserted the following initial data: the gas composition, the black oil composition, the coefficients of the combustion factors, the NO_x emission factor, the sulphur retaining degree, the humidity of the gas fuel, general data about the boiler structure, the CO_2 and O_2 concentration in the burning gases [9,10].

3. Results and discussion

Based on the data supplied by Colterm Timişoara and according to the values of the meteorological parameters there have been elaborated the mathematical parameters related to the nitrogen dioxide dispersion derived from the CET CENTRU and CET SUD, depending on the meteorological parameters.

Considering the multitude of the directly measured variables, respectively the measures calculated by means of certain dependency relations, in order to simplify the methodology of processing the experimental data, respectively to determine the mathematical models, there have been taken into consideration the following simplifying hypothesis:

⇒ both CET SUD, as well as CET CENTRU have been considered as two stationary punctiform emission sources of polluting gases. Thus, this simplifying hypothesis will have a great influence upon the criterium of performance of the established models.
⇒ The NO_x concentration for every thermal heating station has been obtained by summing up the concentrations from all the boilers and their afferent CAFs.
⇒ The NO_x concentration has been considered equal to the concentration of NO_2.

The ecosystem that has been studied is a non-linear and dynamic system, and this is the reason for which the mathematical modeling process is very complex.

In many practical cases, the relation between two or more parameters that occur within a certain process is sufficiently connected so that the variation of a parameter can be controlled and expressed based on the variation of the other parameters. Such functional relations are named as the stohastic or probabilistic relations. The study of these relations leads to the development of the multiple correlation theory [11-14].

From the mathematical point of view, the studied parameters are random variables, their variation within a limited field being set forth by random factors, and every parameter basically follows a well-determined distribution law [13].

The algorithm for elaborating the mathematical models is presented further on.

We assume that during the development of a process, there have been made *m* measurements of *n+1* variables. We wish to express one of these variables, *y*, depending on the other x_i variables, with $i = \overline{1, n}$, by means of a 2nd degree function, indicated in the relation (1):

$$Y(x_i) = a_0 + \sum_{i=1}^{n} a_{1i} x_i + \sum_{i=1}^{n} a_{2i} x_i^2 + \sum_{j=1}^{n-1}\sum_{i=1}^{n-j} b_{ji} x_j x_{i+j} \quad (1)$$

The coefficients a_0, a_{1i}, a_{2i}, …, b_{ji}, are referred to as the coefficient of the regression function. These are determined by the method of the smallest squares, so that the dispersion of the deviations of values experimentally set forth y in comparison to the values set forth based on the regression equation (1) to be minimum, respectively:

$$M(f) = M[y - Y(x_i)]^2 = \text{min imum}$$

where :

$$f(a_0, a_{1i}, a_{2i}, b_{ji}) = [y - a_0 - \sum_{i=1}^{n} a_{1i} x_i - \sum_{i=1}^{n} a_{2i} x_i^2 - \sum_{j=1}^{n-1}\sum_{i=1}^{n-j} b_{ji} x_j x_{i+j}]^2 \quad (2)$$

For this, we cancel the partial derivates of the function (2) in relation with the coefficients a_0, a_{1i}, a_{2i}, b_{ji}:

$$\frac{\partial f}{\partial a_0} = -2\sum_{k=1}^{m}[y_k - a_0 - \sum_{i=1}^{n} a_{1i} x_{ik} - \sum_{i=1}^{n} a_{2i} x_{ik}^2 - \sum_{j=1}^{n-1}\sum_{i=1}^{n-j} b_{ji} x_{jk} x_{i+j,k}] = 0$$

$$\frac{\partial f}{\partial a_{1i}} = -2\sum_{k=1}^{m}[y_k x_{ik} - a_0 x_{ik} - \sum_{i=1}^{n}a_{1i}x_{ik}^2 - \sum_{i=1}^{n}a_{2i}x_{ik}^3 - \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}x_{jk}x_{i+j,k}x_{ik}] = 0 \qquad i=\overline{1,n}$$

$$\frac{\partial f}{\partial a_{2i}} = -2\sum_{k=1}^{m}\Big[y_k x_{ik}^2 - a_0 x_{ik}^2 - \sum_{i=1}^{n}a_{1i}x_{ik}^3 - \sum_{i=1}^{n}a_{2i}x_{ik}^4 - \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}x_{jk}x_{i+j,k}x_{ik}^2\Big] = 0 \quad, \; i=\overline{1,n}$$

$$\frac{\partial f}{\partial b_{1i}} = -2\sum_{k=1}^{m}[y_k x_{1k}x_{i+1,k} - a_0 x_{1k}x_{i+1,k} - \sum_{i=1}^{n}a_{1i}x_{ik}x_{1k}x_{i+1,k} - \sum_{i=1}^{n}a_{2i}x_{ik}^2 x_{1k}x_{i+1,k} - \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}x_{jk}x_{i+j,k}x_{1k}x_{i+1,k}] = 0 \qquad , \; i=\overline{1,n-1}$$

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$$\frac{\partial f}{\partial b_{ji}} = -2\sum_{k=1}^{m}\Big[y_k x_{jk}x_{i+j,k} - a_0 x_{jk}x_{i+j,k} - \sum_{i=1}^{n}a_{1i}x_{ik}x_{jk}x_{i+j,k} - \sum_{i=1}^{n}a_{2i}x_{ik}^2 x_{jk}x_{i+j,k} - \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}x_{jk}^2 x_{i+j,k}^2\Big] = 0 \qquad , \; i=\overline{1,n-j}$$

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$$\frac{\partial f}{\partial b_{n-1,n}} = -2\sum_{k=1}^{m}[y_k x_{n-1,k}x_{nk} - a_0 x_{n-1,k}x_{nk} - \sum_{i=1}^{n}a_{1i}x_{ik}x_{n-1,k} - \sum_{i=1}^{n}a_{2i}x_{ik}^2 x_{n-1,k}x_{nk} - \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}x_{jk}x_{i+j,k}x_{n-1,k}x_{nk}] = 0$$

Therefore, we obtain the system (3) of $(1+2n+C_n^2)$ equations with unknown values a_0, a_{1i}, a_{2i}, b_{ji}, with $i=\overline{1,n}; j=\overline{1,n}; i\neq j$:

$$ma_0 + \sum_{i=1}^{n}a_{1i}\sum_{k=1}^{m}x_{ik} + \sum_{i=1}^{n}a_{2i}\sum_{k=1}^{m}x_{ik}^2 + \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}\sum_{k=1}^{m}x_{jk}x_{i+j,k} = \sum_{k=1}^{m}y_k$$

$$a_0\sum_{k=1}^{m}x_{ik} + \sum_{i=1}^{n}a_{1i}\sum_{k=1}^{m}x_{ik}^2 + \sum_{i=1}^{n}a_{2i}\sum_{k=1}^{m}x_{ik}^3 + \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}\sum_{k=1}^{m}x_{jk}x_{i+j,k}x_{ik} = \sum_{k=1}^{m}y_k x_{ik} \qquad i=\overline{1,n}$$

$$a_0\sum_{k=1}^{m}x_{ik}^2 + \sum_{i=1}^{n}a_{1i}\sum_{k=1}^{m}x_{ik}^3 + \sum_{i=1}^{n}a_{2i}\sum_{k=1}^{m}x_{ik}^4 + \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}\sum_{k=1}^{m}x_{jk}x_{i+j,k}x_{ik}^2 = \sum_{k=1}^{m}y_k x_{ik}^2 \qquad i=\overline{1,n} \qquad (3)$$

$$a_0\sum_{k=1}^{m}x_{1k}x_{i+1,k} + \sum_{i=1}^{n}a_{1i}\sum_{k=1}^{m}x_{ik}x_{1k}x_{i+1,k} + \sum_{i=1}^{n}a_{2i}\sum_{k=1}^{m}x_{ik}^2 x_{1k}x_{i+1,k} + + \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}\sum_{k=1}^{m}x_{jk}x_{i+j,k}x_{1k}x_{i+1,k} = \sum_{k=1}^{m}y_k x_{1k}x_{i+1,k} \qquad , \; i=\overline{1,n-1}$$

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$$a_0\sum_{k=1}^{m}x_{jk}x_{i+j,k} + \sum_{i=1}^{n}a_{1i}\sum_{k=1}^{m}x_{ik}x_{jk}x_{i+j,k} + \sum_{i=1}^{n}a_{2i}\sum_{k=1}^{m}x_{ik}^2 x_{jk}x_{i+j,k} + \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}\sum_{k=1}^{m}x_{jk}^2 x_{i+j,k}^2 = \sum_{k=1}^{m}y_k x_{jk}x_{i+j,k} \qquad , \; i=\overline{1,n-j}$$

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$$a_0\sum_{k=1}^{m}x_{n-1,k}x_{nk} + \sum_{i=1}^{n}a_{1i}\sum_{k=1}^{m}x_{ik}x_{n-1,k}x_{nk} + \sum_{i=1}^{n}a_{2i}\sum_{k=1}^{m}x_{ik}x_{n-1,k}x_{nk} + \sum_{j=1}^{n-1}\sum_{i=1}^{n-j}b_{ji}\sum_{k=1}^{m}x_{jk}x_{i+j,k}x_{n-1,k}x_{nk} = \sum_{k=1}^{m}y_k x_{n-1,k}x_{nk}$$

By solving the equation system (3) we obtain the values of the coefficients of the regression function (1).

Based on this mathematic algorithm we have presented above, by particularizing the n=5, we attempted to pursuit the dependency of the nitrogen dioxide concentration upon the simultaneous action of 4 meteorological parameters: pressure, temperature, humidity and wind speed. In relation (4), we emphasized the mathematic model obtained for the dispersion of the nitrogen dioxide generated by CET CENTRU and in relation (5), the mathematic model obtained for the dispersion of the nitrogen dioxide emanated from CET SUD. For the both mathematical models already set forth, we calculated: the correlation multiple coefficients and the square average deviation.

$$Y = 1.367\cdot10^3 - 0.882x_1 + 77.066x_2 + 25.954x_3 - 17.308x_4 - 1.318\cdot10^{-4}x_1^2 + 0.015x_2^2 - 0.199x_3^2 + 52.538x_4^2 - 0.077x_1x_2 - 0.287x_2x_3 + 6.836x_3x_4 - 0.651x_4x_1 + 4.44\cdot10^{-3}x_1x_3 - 3.847x_2x_4 \qquad (4)$$

- ◆ Square average deviation $\sigma = 263{,}607$
- ◆ Correlation coefficient $R = 0.863$

$$Y = 1.061\cdot10^3 - 0.337x_1 - 19.793x_2 - 30.766x_3 + 195.867x_4 - 8.409\cdot10^{-4}x_1^2 + 2.198x_2^2 + 0.264x_3^2 - 99.572x_4^2 - 0.137x_1x_2 + 1.33x_2x_3 - 6.927x_3x_4 + 0.811x_4x_1 - 7.674\cdot10^{-3}x_1x_3 - 6.235x_2x_4 \qquad (5)$$

- ◆ Square average deviation $\sigma = 245{,}93$
- ◆ Correlation coefficient $R = 0.817$

Based on the value of the square average deviations underlined in table 1, we noticed that in the case of the simultaneous action of the 4 meteorological parameters, their values are greater, therefore we may sum up that this result is owed to the simplification of the model by means of which all the exhaustion funnels of every thermal heating station were considered a single source of emission. The correlation multiple coefficient has a better value which acknowledges the accuracy of the models.

TABLE 1. Values of the square average deviations and value of the correlation multiple coefficients

No.	Thermal heating station	Square average deviation	Correlation multiple coefficient
1	CET CENTRU	263.607	0.863
2	CET SUD	245.93	0.817

4. Conclusions

In case of the correlation of the nitrogen dioxide by the simultaneous action of 4 meteorological parameters, there have been obtained better values for the markers of the model accuracy. This fact suggests that the dispersion phenomenon and respectively the prediction of the nitrogen dioxide concentration were well described by the simultaneous action of all the studied meteorological parameters.

The correlation coefficients of the models do not reach the level of the coefficients afferent to the static processes, due to different reasons: the process is dynamic, for the accomplishment of modelling there have been made a series of simplifications; the nature undergoes a continuous evolution and the meteorological parameters depend on a series of uncontrollable factors. Also the sources of azote dioxide emissions are not constant.

At the same time, the smaller values of the correlation multiple coefficients are due to a series of other causes:

- The humidity has an antagonic influence compared to the other meteorological parameters towards the dispersion of the nitrogen dioxide.

- The air humidity determines the transformation of the nitrogen dioxide in azotic acid, and consequently, its concentration is diminished.

REFERENCES

1. Savii G., Luchin M., Modelare şi simulare, Ed. Eurostampa, Timişoara, **2000**.
2. Todinca T., Geantă M., Modelarea şi simularea proceselor chimice, Ed. Politehnica, **1999**.
3. Cheremisinoff N., Handbook of air pollution, prevention and control, PhD, N&P Limited, **2002**.
4. Sawyer C. N., Perrz L. McCarty, Parkin G.F., Chemistry for Environmental Engineering and Science, McGraw Hill, **2003**.
5. Dorai R., Modeling of plasma remediation of nox using global kinetic models accounting for hydrocarbons, http://uigelz.ece.uiuc.edu/pub/theses/rajesh_ms_thesis.pdf.
6. Schnelle Karl B.jr., Partha R. Dey, Atmospheric Dispersion Modeling Compliance Guide, McGraw-Hill, **1999**.
7. Perju D., Pîrlea H., Brusturean G.-A., Silaghi-Perju D., Marinescu S., *Rev. Chim.* (Bucureşti), vol. 59, Nr.10, **2008**.
8. Pîrlea H., Silaghi-Perju D., Perju D., Rusnac C., *Rev. Chim.*, vol. 57, nr.7, p. 743-748, **2006**.
9. ***, H.G. 541/2003 privind stabilirea unor măsuri pentru limitarea emisiilor în aer ale anumitor poluanţi proveniţi din instalaţii mari de ardere, **2003**.
10. ***, OUG 34/2002 privind prevenirea, reducerea şi controlul integrat al poluării, **2002**.
11. Aloman A., Statistică şi probabilitate în experimentul ştiinţific, Ed. Matrix Rom, Bucureşti, **1998**.
12. Petrişor E., Probabilităţi şi statistică, Ed. Politehnica, Timişoara, **2001**.
13. Jalobeanu C., Raşa I., Probleme de calcul numeric şi statistic MATHCAD, Ed. Albastră, Cluj-Napoca, **1995**.
14. Dumitrescu M., Florea D., Tudor C., Probleme de teoria probabilităţilor şi statistică matematică, Editura Tehnică, **1985**.