Modelling and Simulation of NO₂ Dispersion Phenomenon in Atmosphere by Analytical-Experimental Methods

DELIA PERJU 1, HARIETA PÎRLEA 1,*, GABRIELA-ALINA BRUSTUREAN 1, DANA SILAGHI-PERJU 1, SORIN MARINESCU 2
1 Politehnica University of Timisoara, Faculty of Industrial Chemistry and Environmental Engineering, Timisoara, 2 Victoriei Square, 30006, Bucharest, Romania
2 Politehnica University of Timisoara, Mechanical Engineering Faculty, Timisoara, 1 Mihai Viteazu Blvd., 30022, Timisoara, Romania

The European laws and recently the Romanian ones impose more and more strict norms to the large nitrogen dioxide polluters. They are obligated to continuously improve the installations and products so that they limit and reduce the nitrogen dioxide pollution, because it has negative effects on the human health and environment. In this paper are presented these researches made within a case study for the Timișoara municipality, regarding the modeling and simulation of the nitrogen dioxide dispersion phenomenon coming from various sources in atmosphere with the help of analytical-experimental methods. The mathematical model resulting from these researches is accurately enough to describe the real situation. This was confirmed by comparing the results obtained based on the model with real experimental values.

Keywords: atmospheric pollution, modeling, simulation

Nitrogen dioxide is a pollutant that creates problems at local, regional and global level. Besides the noxious effects that it has on the population’s health, it contributes indirectly to the global warming as a result of the photochemical pollution that it causes [1]. As a result, the European Union took the guide values, for the nitrogen dioxide emissions in the atmosphere, from the Health World Organization (OMS) that it imposed as limit values. Romanian took over these values into the national laws.

In the current world context when the accent on environment pollution problems are more and more pressing, the study of nitrogen dioxide emission reduction possibilities as well as the modeling ways of the phenomenon that accompany the dispersion of this pollutant in its cycle in nature is current and of a particular importance. This fact leads to the development of some series of mathematical models that allow the estimation and study of the main parameters that influence the degree of pollution [2-10].

In the work was elaborated a mathematic model which returns the contribution of each pollution source with existing nitrogen dioxide, in a certain reference point. Model’s verification was made by comparing the results obtained based on the model with the measured values of the nitrogen dioxide pollution levels in that point. The main nitrogen dioxide pollution sources existing in Timișoara municipalities are: two thermal power stations, the road traffic and micro-plants – in this class are included: apartment heating system, the ones in the industrial sector and the gas heaters.

Experimental part

In the case study presented in the work the parameters taken into consideration are: nitrogen dioxide emissions coming from the two thermal power stations (Center CET and South CET), from the appartment heating systems within the city and the road traffic; wind speed and direction, Cartesian coordinates of the reference point related to the emission sources; height of the pollution sources.

Obtaining the experimental data was made based on the legal documents supplied by: Distrigaz Nord, Colterm Timișoara, Centrul Meteo Timișoara (Timișoara Weather Center), Direcția regim remise de conducere și înmatriculare a vehiculelor Timiș (Timiș Agency for Driver License and Vehicle Registration) and Agenția de protecția mediului Timișoara (Timișoara Environment Protection Agency).

Colterm Timișoara supplied the daily NO₂ emissions for the months January, April, July, October 2004, for all the boilers and CAFs that exist at Center CET and South CET. The value of these emissions was obtained with the help of the computer software EMPOL, elaborated according to the Government’s Decision 541/2003 and Government’s Emergency Ordinance 34/2002. In the calculus algorithm were introduced the following initial data: gas composition, black oil, coefficient according to the fuel, the emission factor for NO₂, sulfur retention degree, gas fuel humidity, general data on the boiler, CO₂ and O₂ concentration in the burning gases.

Distrigaz Nord supplied the daily consumption of natural gas in Timișoara locality for the months January, April, July, October 2004 and gas chromatographic analysis bulletins, made according to STAS 12001-81, for the same period. Based on this data it was calculated the nitrogen dioxide emission coming from the apartment heating systems in the city according to an algorithm developed based on the data from literature [9].

The data received from the Timișoara Weather Center found in the daily values of the weather parameters: pressure, temperature, humidity, wind speed and direction for the same period of the year 2004, in Timișoara municipality.

From the Timiș Agency for driver license and vehicle registration was obtained the dimension of the Timișoara municipality auto park in the year 2004. According to the European legislation – Directive 98/69/EC from 2000 – nitrogen dioxide emission was considered to be of 0,5 g/km [11].

* email: harieta.pirlea@chim.upt.ro; Tel: 0722877728
Based on the data received from the above mentioned institution were obtained by analytical-experimental methods the values for the NO$_2$ concentrations coming from two thermal power plants of the municipality, noted Center CET and South CET, from the micro-plants existing in the city, as well as from the road traffic. These concentrations have been calculated for a reference point located in the center of Timișoara municipality. The total concentration of NO$_2$ in this point was compared to the experimental values supplied by the Environment protection agency of Timișoara.

Results and discussions

At the base of analytical-experimental deduction of the mathematical model was the general equation of the balance of materials as well as a series of supplemental equations. In order to decrease the model's complexity were formulated the following simplifying hypotheses:

- all the apartment heating systems were considered as Hermann type, which is a medium class micro-plant, without filters and with a functioning efficiency of 90%;
- all the apartment heating systems have been distributed on the city’s surface, being assimilated to 5 stationary sources. This hypothesis is necessary because we don’t have official data regarding the number of apartment heating systems and their repartition on neighborhoods;
- Center CET and South CET have been considered two punctiform sources though each has several emission chimneys;
- the auto park of Timișoara municipality was considered to be equipped EURO 3.

Making the inventory of the variables that characterize the dispersion phenomena for the nitrogen dioxide, the following are identified:

- independent variables:
  - nitrogen dioxide emissions resulted from the two thermal power plants (Center CET and South CET), from the apartment heating systems from the city and from the road traffic;
  - wind’s speed and direction
  - Cartesian coordinates of the reference point in relation to the emission sources
  - height of pollution sources.
- dependent variables:
  - nitrogen dioxide concentrations resulting from the two thermal power plants (Center CET and South CET), from the apartment heating systems from the city and from the road traffic.

In order to determine the x and y Cartesian coordinates of the stationary pollution sources in relation to the reference point, it was taken into account the wind direction in each day from the studied months. The z Cartesian coordinate of the reference point was considered equal to 3 m which coincides to the height at which the NO$_2$ sensor was placed. Regarding the height of the pollution sources, in the case study the following values were considered, as presented in table 1.

In order to estimate the nitrogen dioxide concentration in the reference point was deduced the equation for the materials balance, for this pollutant, equation presented in relation (1):

$$C_{\text{ref}} = C_1 + C_2 + C_3 + C_4 + C_5 + C_6 + C_7$$

were:

- $C_{\text{ref}}$ – total concentration of nitrogen dioxide in the reference point, [µg/m$^3$];
- $C_1, C_2, C_3, C_4$ - NO$_2$ concentration in the reference point resulting from the northern, southern, eastern, western and center micro-plants [µg/m$^3$];
- $C_5, C_6$ - NO$_2$ concentration in the reference point resulting from the Center CET and South CET, [µg/m$^3$];
- $C_7$ - NO$_2$ concentration in the reference point resulting from the road traffic, [µg/m$^3$].

In order to estimate in the reference point the nitrogen dioxide concentrations, resulting from each of the 7 stationary sources it was used the Gaussian dispersion formula [4,12,13], relation (2):

$$C(x,y,z) = \frac{Q}{2\pi \sigma_x \sigma_y} \exp \left( -\frac{x^2}{2\sigma_x^2} \right) \left[ \exp \left( -\frac{y^2}{2\sigma_y^2} \right) \right] +$$

where:

- $C(x, y, z)$ – pollutant’s concentration in point (x, y, z) [µg/m$^3$];
- $u$ – wind speed, in wind’s direction [m/s];
- $Q$ – emitted debit of pollutant [g/s];
- $\sigma_x, \sigma_y$ - standard deviation of the concentration on the direction y and z, in the wind’s direction [m];
- $H$ – effective height from the ground level up to the center of the pollutant cloud [m].

The calculus expressions of the standard deviations are presented in the relations 3, 4 and 5 [13]:

$$\sigma_x = ax^b$$

$$\sigma_y = 465.11628x(tan(\Theta))$$

$$\Theta = 0.017453293c - d\ln(x)$$

where:

- x – distance between the pollution source and reference point, [km];
- a, b – constants, whose value depends on x and on the atmospheric stability class.
- c, d – constants whose value depends on the atmospheric stability class.

Effective height from the ground level until the center of the plume rise is deduced with the help of the relation (6):

$$H = h + \Delta h$$

where:

- $h$ – height of the chimney [m];
- $\Delta h$ – height of the pollutant cloud [m].

Height of the plume rise is deduced based on the algorithm presented in figure 1 [14,15]:

<table>
<thead>
<tr>
<th>Zone</th>
<th>North</th>
<th>South</th>
<th>Est</th>
<th>West</th>
<th>Center CET</th>
<th>South CET</th>
</tr>
</thead>
<tbody>
<tr>
<td>h [m]</td>
<td>10</td>
<td>14.5</td>
<td>8</td>
<td>4</td>
<td>11.5</td>
<td>54.2</td>
</tr>
</tbody>
</table>
were:
F – buoyancy factor \([\text{m}^4/\text{s}^3]\);
x_f – downwind distance from plume source to point of maximum plume rise \([\text{m}]\);
u – windspeed at actual stack height \([\text{m/s}]\);
s – stability parameter \([\text{s}^{-2}]\).

The above mentioned calculus method was applied for each day from the four considered months: January, April, July and October. From the daily values of the nitrogen dioxide concentrations in the reference point, resulted from the 7 stationary sources were made the monthly averages, these being presented in the table 2.

In order to estimate the reference point of the nitrogen dioxide concentrations resulting from the mobile pollution sources (road traffic) relation (7) was used:

\[
C_r = \left( \frac{N \cdot 10^4}{V} \right) \cdot \frac{1}{n} \tag{7}
\]

- \(N\) – European emission norm [11], \([\text{g/km}]\), \(N=0.5 \text{ g/km}\);
- \(V\) – volume of the dispersion area, \([\text{m}^3]\), considered 45000 \(\text{m}^3\);
- \(N\) – number of mobile sources.

Monthly average values of the nitrogen dioxide concentrations resulting from mobile pollution sources (road traffic) are presented in the table 3. At the calculation of these values, the number of mobile sources was chosen according to the traffic specific for each month.

After applying the developed mathematical model, the total concentration of nitrogen dioxide in the reference point for the months considered, is presented in figure 3.

From the figure 2 is found that, in the reference point, the total concentration of nitrogen dioxide does not surpass the limit value admitted by the law for none of the studied months. For the month of January we find a higher concentration fact that may be explained by the relatively low temperatures for this period. This reflects a higher consumption of fuel and natural gases. In opposition the degree of pollution is at the lowest in the month of July, period destined for vacations.

Comparing the influence of the stationary and mobile sources on the pollution with nitrogen dioxide in the considered reference point, is found that the mobile sources have an overwhelming weight in relation to the stationary ones (>90%).

The weight of stationary nitrogen dioxide pollution sources in relation to the total pollution from the reference point is presented in figure 3.

From the graphic we can see that, during the months April and July the stationary sources have an insignificant influence on nitrogen dioxide pollution in the center of the city.

From the stationary sources, the biggest influence on the pollution at the studied reference point belongs to Center CET. The weather conditions from January are reflected in the induced pollution degree by this source: 4.4%.

Apartment heating systems with the biggest influence on nitrogen dioxide pollution from the center of the city are the ones located in the north, west and center of the city.

The test of the mathematical model was made by comparing the calculated results based on the mathematical model with the experimental ones, supplied by the Timișoara environment protection Agency. Correlation of this data is presented in the figure 4.

### Table 2

<table>
<thead>
<tr>
<th>Month</th>
<th>North</th>
<th>South</th>
<th>Est</th>
<th>West</th>
<th>Center CET &amp; South CET, ([\mu g/\text{m}^3])</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jan</td>
<td>1.023E+00</td>
<td>1.172E-02</td>
<td>1.833E-02</td>
<td>1.027E+00</td>
<td>1.447E+00</td>
</tr>
<tr>
<td>Apr</td>
<td>7.223E-10</td>
<td>6.958E-02</td>
<td>2.883E-02</td>
<td>9.329E-02</td>
<td>4.999E-05</td>
</tr>
<tr>
<td>July</td>
<td>1.748E-09</td>
<td>1.273E-02</td>
<td>1.791E-03</td>
<td>1.082E-01</td>
<td>5.387E-06</td>
</tr>
<tr>
<td>Oct</td>
<td>5.506E-09</td>
<td>4.606E-01</td>
<td>2.654E-01</td>
<td>2.441E-01</td>
<td>1.119E+00</td>
</tr>
</tbody>
</table>

**Fig.1.** Logical diagram with the calculus algorithm of height of the plume rise.
Table 3
NITROGEN DIOXIDE CONCENTRATIONS IN THE REFERENCE POINT, RESULTING FROM THE ROAD TRAFFIC

<table>
<thead>
<tr>
<th>Month</th>
<th>Number of mobile sources /analysis interval [buc]</th>
<th>Concentrations in the reference point, resulted from the mobile sources [µg/m³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>January</td>
<td>6.5</td>
<td>72.2222</td>
</tr>
<tr>
<td>April</td>
<td>4</td>
<td>44.4444</td>
</tr>
<tr>
<td>July</td>
<td>2.25</td>
<td>25.0000</td>
</tr>
<tr>
<td>October</td>
<td>3</td>
<td>33.3333</td>
</tr>
</tbody>
</table>

Fig. 2. Total concentration of nitrogen dioxide in the reference point for the considered months

Fig. 3. The weight of stationary nitrogen dioxide pollution sources in relation to the total pollution from the reference point

Fig. 4. Correlation between the experimental data and the ones calculated based on the mathematical model
Almost symmetrical disposition of the experimental results in relation to the ones calculated based on the model confirm the accuracy of the generated mathematical model. This is also confirmed by the relative values of the errors [16], presented in table 4.

Relative errors may result from the following causes:
- the measurement of the weather parameters was not made in the same location with the measurements of the total concentration of nitrogen dioxide;
- apartment heating systems are randomly distributed in the city, they are of different types and located at various heights;
- mobile sources are of small and medium tonnage, are equipped with motors with ignition by spark or compression and equipped from NON EURO to EURO 4;
- the number of mobile sources in the analysis interval considered resulted after a statistic and probabilistic research.

Conclusions
The mathematical model developed for the simulation of the dispersion phenomenon of NO₂ in the atmosphere allowed to establish the influence of each pollution source with nitrogen dioxide in the studied reference point.

In the studied reference point, where the Timișoara Environment Protection Agency performed analysis, it is found that the major influence of nitrogen dioxide pollution belongs to the means of transportation. They have an influence of 90-95% at pollution of the center of the city with the nitrogen dioxide.

By comparing the results of the mathematical model with the experimental data it is found a good concordance between them, fact confirmed by relative errors. This proves that the model is accurate enough to simulate the dispersion phenomenon of the nitrogen dioxide in the atmosphere.

References
1. GLAVAN, Ș., Circulația rutieră și protecția mediului, Miron, Timișoara, 1999
2. SAVII, G., LUCHIN, M., Modelare și simulare, Eurostampa, Timișoara, 2000, p. 261
5. STERN R., YAMARTINO, R.J., GRAFF, A., 26th ITM on Air Pollution Modelling and its Application, May 26-30, 2003, Istanbul, Turkey
16. PETRIÒR, E., Probabilități și statistică, Editura Politehnica, Timișoara, 2001