

Assessment of air quality using diffusive samplers and ADMS-Urban

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The main sources of inorganic pollutants are emissions from facilities of the energy sector and transport exhaust emissions. Nitrogen dioxide (NO₂) as one of the most important inorganic air pollutants forms during the combustion process, especially in motor vehicles. Passive diffusive samplers used to measure NO₂ become more popular because of their simplicity, low cost and possibility to measure in large areas, including cities, regions or even different countries. The aim of this paper is to compare the data on air quality assessment obtained by means of indicative measurements and modelling based on the data from Žirmūnai district, Vilnius city, Lithuania.

Nitrogen dioxide was measured with diffusive samplers in 25 points in the district. Samplers were attached to the street light poles. Diffusive samplers consisted of stainless steel mesh discs coated with triethanolamine. Higher concentrations of nitrogen dioxide were measured near intensive traffic streets: Kareivių and Žirmūnų. The average NO₂ concentration was up to 39.0 µg/m³ at the measurement points located near these streets. 2.2 times lower concentrations (17.7 µg/m³) of nitrogen dioxide were measured at the measurement points located in the yards of apartment houses further from the heavy traffic streets. The air quality of Žirmūnai district was also assessed by modelling dispersion of nitrogen dioxide from motor exhaust emissions with the ADMS-Urban program. The highest concentrations of nitrogen dioxide calculated using simulation were in the north-western part of Žirmūnai district: intersection of Kareivių, Kalvarijų and Ozo streets. NO₂ concentration at this crossroad was up to 60.0 µg/m³. The lowest concentration of NO₂ (14.0–16.0 µg/m³) was recorded at the measurement points located further from road traffic as the main source of pollution. Nitrogen dioxide concentrations in ambient air of Žirmūnai district measured with diffusive samplers were compared with the results obtained using the ADMS-Urban program. The error between two methods ranged from 2.5 to 35.8%. The concentrations measured with diffusive samples differed by 13.9% on average from the concentrations modelled with ADMS-Urban. Simulation data was within the 30% uncertainty of nitrogen dioxide permitted in the Directive 2008/50/EC.

Key words: nitrogen dioxide, dispersion, diffusive sampler, modelling, ADMS-Urban

INTRODUCTION

One of the dominant sources of air pollution affecting environmental living quality in urban areas is road traffic-induced air pollution (Wang et al., 2008; Baltrėnas et al., 2008; Vaitiekūnas, Banaitytė, 2007; Fenger, 2009). Emissions from motor ve-

hicles influence the temporal and spatial patterns of regulated gases, particulate matter, and toxic air pollutant concentrations within urban areas (Venkatram et al., 2007; Vardoulakis et al., 2003). Air quality monitoring studies carried out near major roadways have detected enlarged concentrations, compared to overall urban background levels, of motor-vehicle emitted compounds, including carbon monoxide (CO), nitrogen oxides (NO_x),

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particulate matter (PM), polycyclic aromatic hydrocarbons (PAHs) and benzene (Vardoulakis et al., 2003; Venkatram et al., 2007).

Nitrogen dioxide (NO₂) is a pollutant of the urban atmosphere (Finlayson-Pitts, Pitts, 2000; Cape et al., 2004; Sujetovienė, 2010). NO₂ also impacts as atmospheric ozone-forming chemistry (Alvarez et al., 2008; Valuntaitė et al., 2009). Outdoor concentrations of NO₂ can vary widely and rapidly, ranging from a few micrograms per cubic meter to peaks of several hundreds of micrograms per cubic meter during particular episodes of high pollution (Afif et al., 2009). Nitrogen dioxide was selected for analysis as an indicator of traffic-related air pollution (Malinauskiene et al., 2011).

Diffusion tubes are simple passive samplers which collect gas by molecular diffusion (Palmer et al., 1976; Kot-Wasik, 2007; Campbell et al., 1994). Diffusion tubes have an advantage of being a low cost, convenient way of mapping spatial distributions of NO₂ (Baltrėnas et al., 2011). A disadvantage of the method is that it can only provide a concentration that is averaged over the period of exposure and it is not possible to measure short-term concentrations (Bush et al., 2001).

The prediction of pollutant concentrations with aid of regulatory air quality models is an essential part for air quality management strategies (Mohan et al., 2011; Szyda et al., 2009; Kryza et al., 2010; Januševičienė, Venckus, 2011). Air quality modelling was conducted using ADMS-Urban, the most comprehensive version of the Atmospheric Dispersion Modelling System (ADMS) version 2.3 developed by Cambridge Environmental Research Consultants Ltd. (CERC). ADMS-Urban is a PC-based model of dispersion in the atmosphere of pollutants released from multiple industrial, domestic and road sources in urban areas. ADMS-Urban can take account of chemical reactions, non-Gaussian distributions of concentrations, diffusion in street canyons or around buildings. Meteorological inputs are treated by an advanced pre-processor (Leuzzi, 2002). Meteorological conditions have a significant influence upon the composition of atmosphere aerosol and over pollutant dispersion (Veriankaitė et al., 2011).

A significant difference between ADMS-Urban and other models used for air dispersion model-

ling in urban areas is that ADMS-Urban applies up-to-date physics using parameterisations of the boundary layer structure based on the Monin-Obukhov length and the boundary layer height (Silva, Mendes, 2011; Arciszewska et al., 2001). Other models characterise the boundary layer imprecisely in terms of the Pasquill stability parameter. In the up-to-date approach, the boundary layer structure is defined in terms of measurable physical parameters, which allow for a realistic representation of the changing characteristic of dispersion with height. The result is generally a more accurate and soundly based prediction of the concentrations of pollutants (CERC, 2006; Arciszewska et al., 2001).

The aim of this paper is to compare the data on the air quality assessment obtained by means of indicative measurements and modelling based on the data in Žirmūnai district of Vilnius city.

MATERIALS AND METHODS

Measuring NO₂ with diffusive samplers

Nitrogen dioxide measurements were carried out in 25 points (Fig. 1) in Žirmūnai district of Vilnius city over a two-week period in October–November 2011.

The diffusive tube samplers applied in this study consisted of a polypropylene tube 34 mm long and 21 mm inner diameter and a closely fitting cap. In one end of the diffusive tube, one stainless steel mesh was placed. For the preparation of diffusive tubes stainless steel meshes were impregnated with 20% aqueous solutions of TEA. The analysis after exposure of samplers was done by spectrophotometric determination of nitrite, using the Saltzman method. The accuracy was $\pm 10\%$.

The amount of nitrite ions in a sample was obtained with the help of calibration plot derived from standard nitrite solutions. The amount of extracted nitrite for samplers was used to calculate ambient NO₂ concentrations.

During field measurements all samplers were placed in special shelters to protect them from rain and minimize the wind influence during exposure. Three diffusive samplers of the same type were placed in each measurement point. Special care was taken at all times when handling the passive

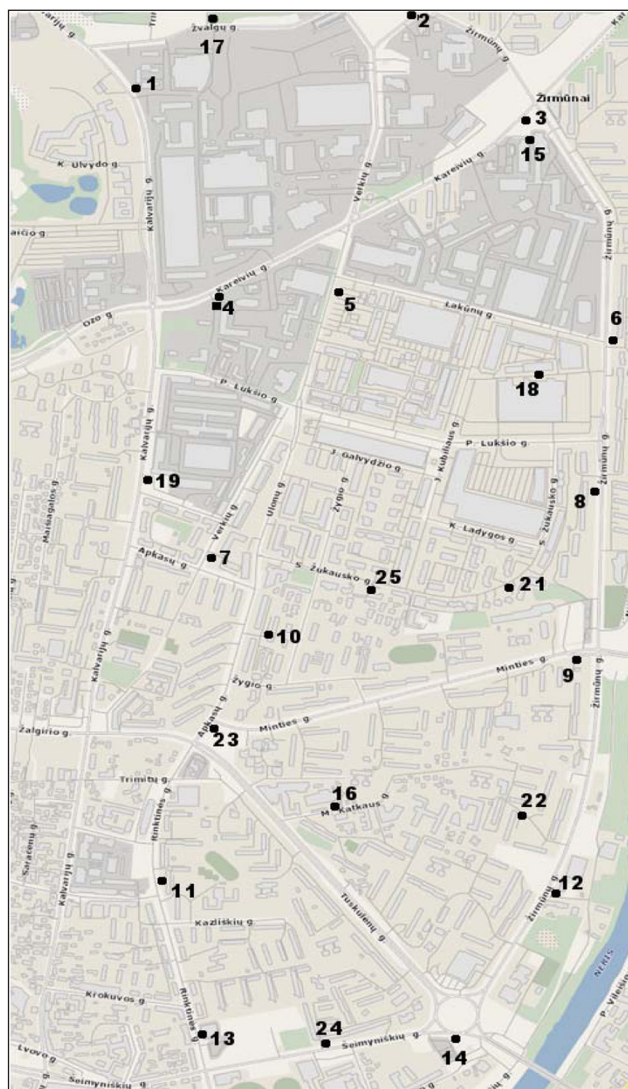


Fig. 1. Diffusive samplers' location in Žirmūnai district
 ■ – air monitoring station

samplers. All samplers were kept in airtight bags during transportation to and from field. After exposure samplers were kept in the refrigerator until preparation for analysis.

Modelling NO₂ concentration

Pollutant concentrations calculated by ADMS-Urban were compared with concentrations measured with diffusive samplers and recorded at one monitoring station.

ADMS-Urban version 2.3 and Surfer 10 programmes were employed for the study. The numerical outputs were compared with monitored two weeks averages of nitrogen dioxide in order to validate the model.

Road source emission rates were calculated from traffic flow data by using the in-built database of traffic emission factors. The 2003 Design Manual for Roads and Bridges (DMRB 2003) database of traffic emissions contains emission factors depending on vehicle category (light or heavy vehicles), average speed (from 5.0 to 130.0 km/h) and traffic count (from 0 to 100 000 vehicle/h), for NO_x, CO, PM₁₀ and VOC (CERC, 2006). The data entered for each road were: elevation of road, road width, canyon height, road geometry, emissions (g/km/s) calculated within ADMS-Urban from vehicle count per hour, average speed (km/h). Žirmūnai district roads were divided into 70 sections with different vehicle flow to represent real traffic information.

The program calculates only NO_x concentration from vehicle flow. NO_x-NO₂ correlation is used to calculate NO₂ concentration. This chemistry option uses a relatively simple function, the Derwent-Middleton Correlation, to estimate the concentration of NO₂ from a given concentration of NO_x (Owen et al., 2000).

Meteorological data for the study were obtained from the Vilnius Meteorological Station. The following hourly meteorological data were employed for modelling: temperature near surface (°C), relative humidity (%), wind speed (m/s), wind direction (degree clockwise from north), precipitation rate (mm/h) and cloud cover (oktas).

RESULTS AND DISCUSSION

With an annually growing number of vehicles in Vilnius, the air pollution level increases every year. The present situation can be defined by measuring ambient air concentration with diffusive samplers or with the help of pollutant dispersion modelling programs.

Meteorology plays an important role in air pollutant formation, dispersion, transport and dilution (Bimbaitė, Girgžienė, 2007; Valuntaitė et al., 2009). The measurement of meteorological parameters (temperature, relative humidity, wind speed, wind direction, precipitation and cloud cover) was performed for assessment of nitrogen dioxide dispersion peculiarities in Žirmūnai district.

During the experiment the temperature changed from 0 to 11 °C, the relative humidity changed from 41 to 100%. The wind speed and direction were changeable during the period of measurement.

The measured average wind speed was 2.3 m/s (8.0 m/s at the maximum) (Fig. 2) during the measurement period. The prevailing wind direction was south-east.

The investigation of nitrogen dioxide concentrations was carried out using diffusive samplers. Nitrogen dioxide samplers were exposed in Žirmūnai district for the duration of two weeks in the autumn season of 2011. Collected NO_2 was determined using a spectrophotometer in the laboratory. The average nitrogen dioxide concentration was $27.3 \mu\text{g}/\text{m}^3$ (varied from 16.9 to $39.2 \mu\text{g}/\text{m}^3$) (Fig. 3). Similar measurements were carried out in another Lithuania city, Kaunas. Laurinavičienė (2010) measured nitrogen dioxide in Kaunas in 2003–2007. NO_2 concentration ranged from 11.4 to $26.3 \mu\text{g}/\text{m}^3$. Similar results were obtained by Lozano (2010) during the sampling campaign with passive diffusion samplers in Spain. The average NO_2 concentration for Seville area in 2000 was $23.7 \mu\text{g}/\text{m}^3$. The obtained average concentrations were similar, but minimum and maximum values were slightly different, 7.6 and $52.1 \mu\text{g}/\text{m}^3$, respectively. During the Lozano campaign, measurements were carried out in 139 sites, meanwhile in our campaign the investigated area was smaller and included only 25 measurement points.

The measurement point number 4 was located near the Žirmūnai air monitoring station. Nitrogen dioxide concentration obtained with a diffusive sampler was compared with the measurements taken at this station situated 10 m away. The measurement results obtained by two methods during the two weeks campaign were in good agreement. The two weeks average NO_2 concentration recorded in the station was $37.5 \mu\text{g}/\text{m}^3$, meanwhile diffusive samplers showed the average $39.2 \mu\text{g}/\text{m}^3$ NO_2 concentration. The standard error between two different nitrogen dioxide measurement methods was only 4.3%.

Higher nitrogen dioxide concentrations were recorded in the measurement points near four-lane streets and larger intersections, where traffic volume was higher. NO_2 concentrations from 35.0 to $40.0 \mu\text{g}/\text{m}^3$ were measured in three measurement points in Žirmūnai district. These measurement points were located near the heavy traffic streets (location 3, 4 and 8 on the map) (Fig. 3). Traffic jams formed every day in these measurement points, when people were going to and from work. 30.0 – $35.0 \mu\text{g}/\text{m}^3$ concentration of nitrogen dioxide was recorded in five measurement points (location 6, 9, 19, 23 and 24 on the map) (Fig. 3). Lower nitrogen dioxide concentrations (25.0 – $30.0 \mu\text{g}/\text{m}^3$)

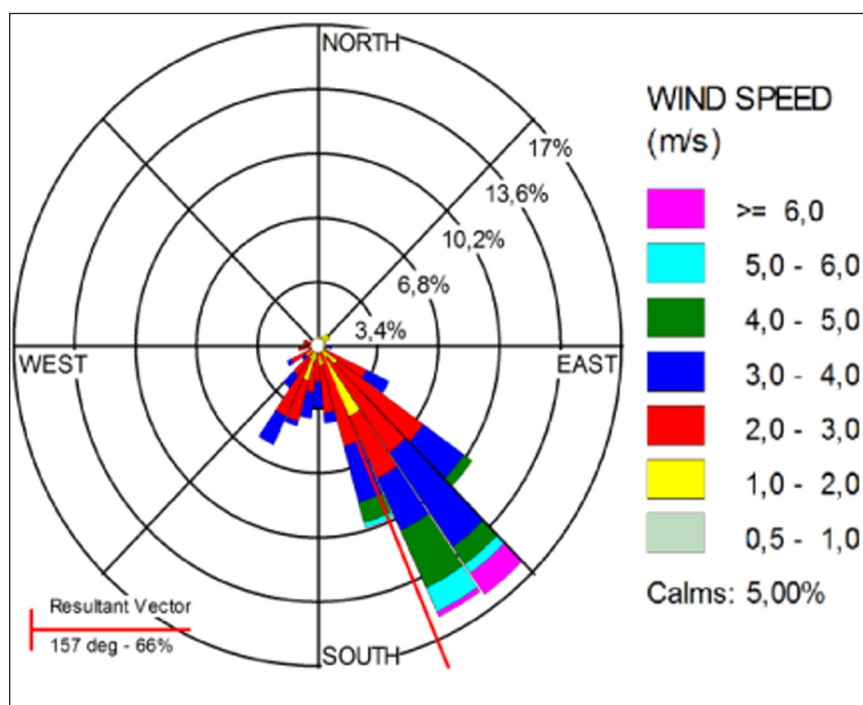


Fig. 2. Wind speed and direction measured at the Vilnius Meteorological Station

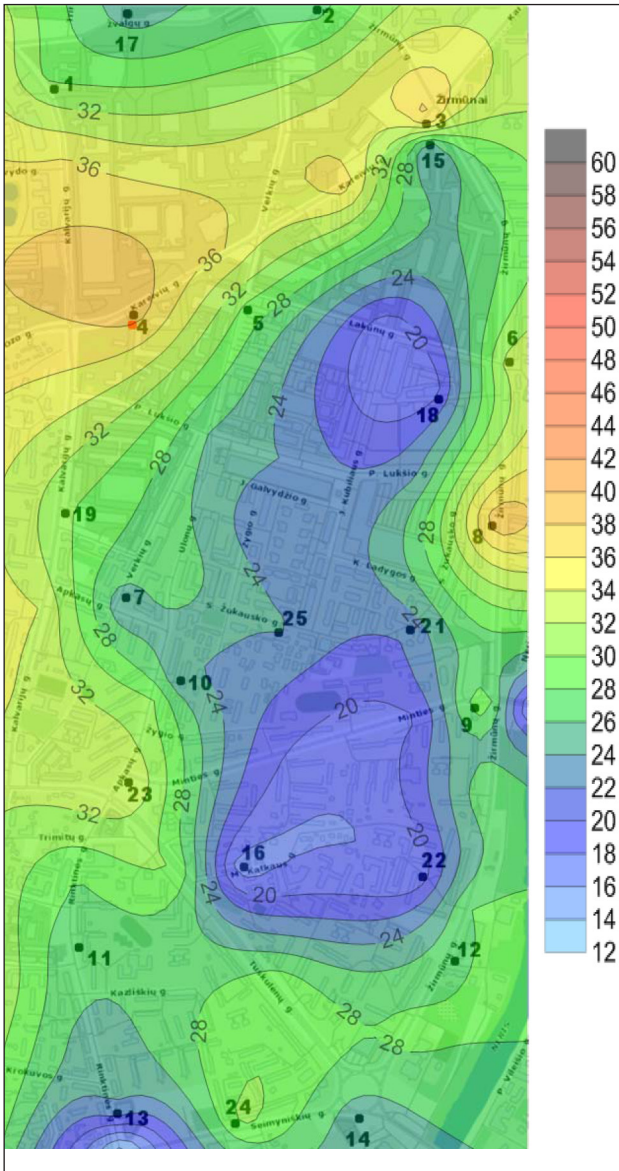


Fig. 3. Concentrations of nitrogen dioxide measured using diffusive samplers in Žirmūnai district

were detected in five points located near the streets with lower intensity vehicle flows (two-line streets) (location 1, 2, 5, 11 and 12 on the map) (Fig. 3). 20.0–25.0 $\mu\text{g}/\text{m}^3$ concentrations were recorded in nine measurement points in Žirmūnai district (location 7, 10, 13, 14, 15, 17, 20, 21 and 25 on the map) (Fig. 3). The measured nitrogen dioxide concentrations were lower than 20.0 $\mu\text{g}/\text{m}^3$ at the pints further from intensive traffic streets (Fig. 3).

The modelled dispersion of NO_2 is shown in Fig. 4. Simulation results were obtained by modelling fluxes of motor vehicle traffic and evaluation of background emissions. Background concentrations of nitrogen dioxide were from the air mo-

nitoring station in Lazdynai district. ADMS-Urban is the most widely used advanced dispersion model for urban areas, being used extensively in China, United Kingdom and other countries and providing a practical tool for assessing and managing urban air quality (Williams, Girnary, 2002; Blair et al., 2003; Lad, 2006).

Nitrogen dioxide dispersion from the maximum concentrations near the most intensive streets (Kalvarijų Str., Ozo Str., Kareivių Str.) dissipated to background levels. Dispersion depended on meteorological conditions (wind strength and direction, rainfall, air temperature), as well as buildings, especially those close to the carriageway (Baltrėnas et al., 2008).

The maximum modelled NO_2 concentration arising from traffic emissions in an open road cal-

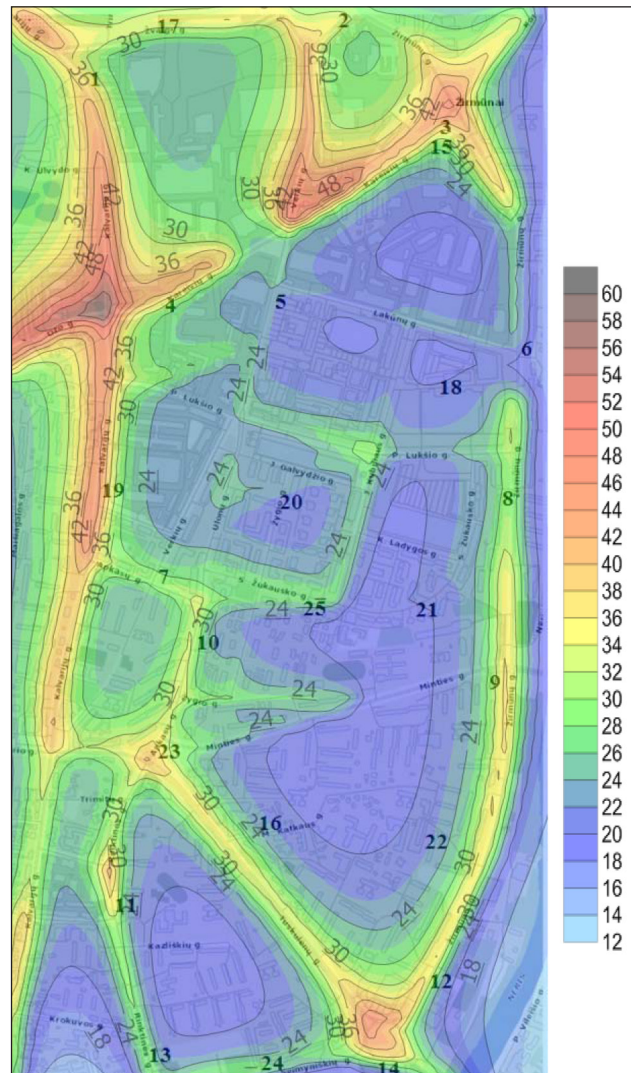


Fig. 4. Modelled concentrations of nitrogen dioxide in Žirmūnai district

culated with ADMS-Urban was up to $60.0 \mu\text{g}/\text{m}^3$ in the north-western part of Žirmūnai district, intersection of Kareivių Str., Kalvarijų Str. and Ozo Str (Fig. 4). Emitted pollutants were transported from the crossroad in the northwest direction when southeast wind was blowing. NO_2 concentrations of up to $50.0 \mu\text{g}/\text{m}^3$ appeared near lower intensity intersections (Kareivių and Verkių Str., Kareivių and Žirmūnų Str.). An average of 1 700 light vehicles per hour passed Kareivių Street.

Nitrogen dioxide concentrations obtained by measuring with diffusive samplers were compared with the results obtained in the simulation with the ADMS-Urban software. Diffusive samplers were taken as the basis for comparison.

Nitrogen dioxide concentrations in ambient air obtained by numerical simulation were presented in a range of values. The lower and upper range values varied within $2.0 \mu\text{g}/\text{m}^3$. In order to compare numerical simulation with experimental results, the average values of nitrogen dioxide in the same 25 measurement points were calculated.

The measured and computed concentrations (average values over the two weeks experimental period) for different measurement points are shown in Fig. 5. The difference of nitrogen dioxide concentrations measured by diffusive samplers and calculated by modelling was up to $12.0 \mu\text{g}/\text{m}^3$.

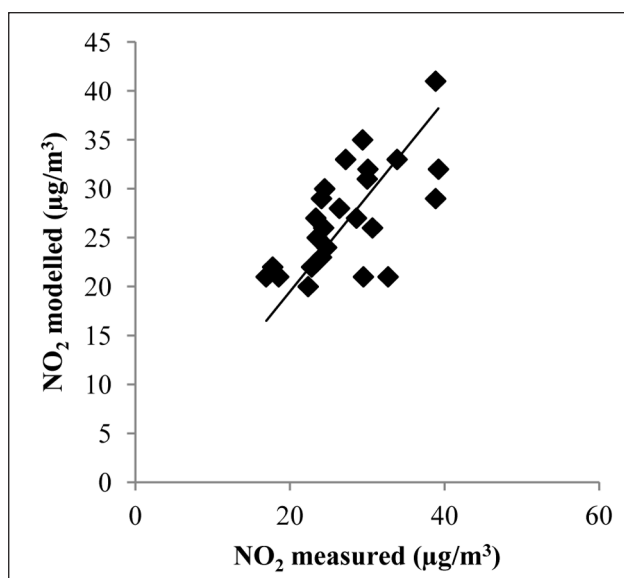


Fig. 5. Comparison between NO_2 concentrations obtained with diffusive samplers and modelled with ADMS-Urban

The standard error between NO_2 concentration measurements and modelling varied from 2.5 to 35.8%. The measured nitrogen dioxide concentration differed from the modelled concentration by 13.9% on average. This difference could be caused by automobile traffic imbalance. There were used average vehicle flows of the last couple of years, but not the exact quantity of cars that passed during that two-week measuring campaign. Simulation results were within the 30.0% permitted modelling uncertainty of nitrogen dioxide indicated in the Directive 2008/50/EC.

CONCLUSIONS

1. The highest concentrations of NO_2 (from 30.0 to $40.0 \mu\text{g}/\text{m}^3$) in Žirmūnai district measured with diffusive samplers were obtained near intensive traffic streets: Kalvarijų, Žirmūnų and Kareivių.

2. NO_2 concentration obtained with diffusive samplers located near the air monitoring station was compared with the average concentration recorded in the station. The standard error between two different nitrogen dioxide measurement methods was only 4.3%.

3. The maximum NO_2 concentrations up to $60.0 \mu\text{g}/\text{m}^3$ were obtained in the Ozo and Kalvarijų Streets intersection, the most intensive crossroad of Žirmūnai district.

4. Concentrations of nitrogen dioxide in ambient air in 25 measurement points in Žirmūnai district measured by diffusive samplers and modelled with ADMS-Urban were in good agreement; the results obtained by two methods differed by 13.9% only and did not exceed the permissible 30.0% modelling uncertainty under the Directive 2008/50/EC.

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ORO KOKYBĖS VERTINIMAS NAUDOJANT DIFUZINIUS ĖMIKLIUS IR ADMS-URBAN MODELIAVIMO PROGRAMĄ

S a n t r a u k a

Didžiąją dalį neorganinių teršalų į aplinką išmeta energetikos sektoriaus objektai ir transporto priemonės. Vienas svarbiausių neorganinių oro teršalų – azoto dioksidas (NO₂) – susidaro vykstant degimo procesams, jo pagrindinis šaltinis – autotransportas. Azoto oksidų ir kitų oro teršalų nustatymui naudojami difuziniai ėmikliai, kurie vis labiau populiarėja dėl savo paprastumo, pigumo ir galimybės išdėstyti daugelyje vietų apimant miestus, regionus ar net skirtingas šalis. Šio darbo tikslas buvo palyginti indikatorinio ir modeliavimo metodų rezultatus remiantis Vilniaus miesto Žirmūnų mikrorajono oro kokybės vertinimu. Azoto dioksidas matuotas difuziniais ėmikliais, kurie dvi savaites 25-iose rajono vietose buvo pritvirtinti ant gatvių apšvietimo stulpų. Difuziniuose ėmikliuose naudotas trietanolamino (TEA) tirpalas ir nerūdijančio plieno tinklelis. Didesnės azoto dioksido koncentracijos buvo nustatytos prie intensyvaus eismo Kareivių ir Žirmūnų gatvių – vidutiniškai 39,0 μg/m³. Atokiau nuo šių gatvių, daugiabučių kiemuose, šios koncentracijos buvo 2,2 karto mažesnės (17,7 μg/m³). Žirmūnų mikrorajono oro kokybė buvo įvertinta ir modeliuojant iš autotransporto išsiskiriančio azoto dioksido sklaidą ADMS-Urban programa. Didžiausia azoto dioksido koncentracija, kuri apskaičiuota taikant transporto srautus įvertinančią modeliavimą, nustatyta Žirmūnų šiaurės vakarinėje dalyje – Kareivių, Kalvarijų ir Ozo gatvių sankryžoje: ties šia sankryža NO₂ koncentracija siekė 60 μg/m³. Mažiausios NO₂ koncentracijos (14,0–16,0 μg/m³) nustatytos toliau nuo pagrindinio taršos šaltinio – autotransporto eismo – esančiuose matavimo taškuose. Palyginus azoto dioksido koncentracijas Žirmūnų mikrorajono aplinkos ore, gautas matuojant difuziniais ėmikliais, ir rezultatus taikant ADMS-Urban programinį modeliavimo paketą, nustatyta paklaida svyravo nuo 2,5 iki 35,8 %. Matavimų metu gautos koncentracijos vidutiniškai 13,9 % skyrėsi nuo ADMS-Urban modeliavimo programa gautų rezultatų. Modeliavimo duomenys neviršija 2008/50/EB direktyvoje nurodytos neapibrėžties nustatant azoto dioksido koncentraciją modeliavimo būdu.

Raktažodžiai: azoto dioksidas, sklaida, difuzinis ėmiklis, modeliavimas, ADMS-Urban