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Indoor Nitrogen Oxides

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1. Introduction

Nitrogen monoxide (NO) and nitrogen dioxide (NO₂) are gases produced by high-temperature combustion through reactions between nitrogen and oxygen. Both nitrogen in fuel and nitrogen in the air can participate in the reactions. Formation of NO can be followed by further oxidation into NO₂ in the atmosphere. NO₂ is the most toxic of the nitrogen oxides (grouped generically as NO_x) and it is very important from the health point of view. Emissions of nitrogen oxides have generally increased, a fact which has resulted in a lot of studies dealing with nitrogen oxides exposure, particularly over the last 15 years. Measurements of NO₂ were carried out in residential and industrial premises over 2-day periods both in winter and summer. Personal exposures were found to vary substantially among the 43 respondents, ranging from 10.2 - 87.5 ppb with a mean of 43.7 ± 16 ppb in winter and 23.6 ± 7.8 ppb in summer. The results also showed that the season of the year, house size and outdoor occupation could significantly influence exposure as well as factors such as the type of fuel used and the ambient air quality level. Personal exposure to NO₂ was significantly greater in winter than in summer, and respondents living in smaller houses and/or having outdoor occupations were exposed to higher NO₂ levels (Kulkarni & Patil, 1998.). Another study was focused on measurements of nitrous acid (HONO) and NO₂ concentrations in a total of 119 houses. Indoor samplers were deployed in the living rooms of the houses for 6 days and outdoor samplers were placed near the houses. Average indoor HONO and NO₂ concentrations were higher than outdoor levels. The results suggest that both HONO and NO₂ are generated by indoor sources (Lee et al., 1999). Subsequent surveys focused on characterisation of the air quality in buildings and the investigation of any seasonal changes in the air quality. There was some indication of much higher concentrations of NO and NO₂ on the ground floor of the buildings and there was some evidence of a seasonal gradient when the concentrations of air contaminants obtained during the winter and summer survey were compared (Nayebzadeh et al., 1999). In one study carried out in Hong Kong (Chao & Law, 2000) a group of 60 people was selected for a study of their personal exposure to NO₂. All the participants spent most of their time at home and in an air-conditioned office environment. It was found that the average personal exposure to NO₂ was 46.0 µg/m³, which was quite close to the average level of 47.3 µg/m³ of the 12 participants involved in the measurement exercises in their home environment. The 12 residential premises were studied for NO₂ levels in different locations in both the indoor environment and the surrounding outdoor environment. It was found that the average NO₂ level in the indoor environment (living room, bedroom and kitchen) was 55.2

$\mu\text{g}/\text{m}^3$ while the corresponding outdoor NO_2 level was $71.8 \text{ mg}/\text{m}^3$. The study indicated that cooking activities in the kitchen had a strong impact on its NO_2 level; while cooking was going on, the average indoor NO_2 level was $59.7 \mu\text{g}/\text{m}^3$ but when it wasn't, the NO_2 level was $41.8 \mu\text{g}/\text{m}^3$. The NO_2 level in an air-conditioned office was low and the impact from other outdoor sources was not significant due to the small amount of time spent in those micro-environments. In another study researchers determined that NO_2 concentrations were higher during the heating season. NO_2 was measured in 77 homes, using Palmes tubes placed in the kitchen, living room, and outdoors. The mean outdoor NO_2 concentration for all measurements was 19 ppb, with levels significantly higher in the heating season (21 ppb vs. 17 ppb). Indoor concentrations were higher than outdoor concentrations. The kitchen concentrations were the highest with a maximum concentration of 85 ppb, suggesting the gas stove as the major source of indoor NO_2 . During the heating season, 40% of the measurements made in kitchens exceeded the Environmental Protection Agency's annual average National Ambient Air Quality Standard level (Zota et al., 2005). The effectiveness of an under-floor air distribution system in controlling thermal comfort and indoor air quality in an office building was evaluated. The indoor values of NO_2 concentrations did not exceed the recommended threshold limits in buildings with under-floor air distribution systems (Yusof et al., 2006). Several studies have focused on comparison of indoor and outdoor NO_x concentrations. According to one study (Lawrence et al., 2005) the outdoor concentrations of NO_x were higher than indoor concentrations measured for an 8-h period in a day and carried out in three environments, i.e. rural, urban and roadside. The full day variations of NO_x were in an urban location. Also, for indoor concentrations two peaks dominate, one in the morning and the other during the evening/night, during which time activities such as cooking take place. For outdoor concentrations there are no such dominating peaks but regular small peaks were found which are believed to be due to the use of heavy diesel generators for supplying electricity. Other research has demonstrated that mean levels of NO_2 in houses with gas cookers are appreciably greater than levels in houses with electric cookers. The difference was shown to be greater for the kitchen, although it was also noticeably different for levels in living rooms and bedrooms. The effect of outdoor sources may be more noticeable in homes with electric cookers, as there are no obvious indoor sources of nitrogen dioxide. Furthermore, the mean levels for single and double-glazed homes have been compared to see whether window type affected indoor levels of NO_2 . The results showed that in the kitchen the monitored levels in double-glazed homes were higher, although the difference was not statistically significant (Cotteril & Kingham, 1997). The NO_2 concentration in an office was measured during a 5-month period to study the horizontal flow of pollutants across the building and the indoor-outdoor ratio (IO) with respect to outdoor concentrations. The horizontal distribution pattern of measured NO_2 varied across the building. The IO ratio for NO_2 was <1 across the building demonstrating the influence of outdoor contaminants on indoor air quality (Katiyar & Khare, 2007). The study (Šenitková et al., 2000) suggested that higher indoor nitrogen oxides concentrations occur mainly through energy conservation with its concomitant tendency to decrease the air exchange rate. The highest concentrations of nitrogen oxides were achieved during gas burning in the kitchen and showed dependence on the intensity of gas burning, the amount of gas burned and on the layout of the room(s). In another study (Kajtár & Leitner, 2007) NO_x distribution is described during the use of a

gas cooker in the kitchen by computational fluid dynamics simulation. The aim of the study was to estimate the exhaust ventilation rate necessary to provide the required indoor air quality. Another paper (Kornartit et al., 2010) reports on the measurement and analysis of indoor and outdoor NO₂ concentrations and their comparison with measured personal exposure in various microenvironments during winter and summer. In winter, indoor NO₂ concentrations were found to be strongly correlated with personal exposure levels. The NO₂ concentration in houses where a gas cooker was used was higher during winter in all rooms than in houses with an electric cooker, whereas there was no significant difference noticed in summer. The average NO₂ levels in kitchens with a gas cooker were twice as high as those with an electric cooker, with no significant difference in the summer period. A time-weighted average personal exposure was calculated and compared with measured personal exposures in various indoor microenvironments. Interestingly, NO₂ personal exposure levels were found to be higher during summer than in winter.

In the present work, indoor NO_x concentrations were measured in order to understand their contribution to indoor sources and to develop a mathematical model for predicting the occurrence of indoor nitrogen oxides depending on the intensities of combustion through the use of gas stoves.

2. Formation and sources of nitrogen oxides

Nitrogen compounds form during combustion. In fact, the atmosphere itself is the source of much of the nitrogen leading to the formation of nitrogen oxides (NO_x). Molecular nitrogen (N₂) makes up most of the gases in the earth's atmosphere (79% by volume). Because N₂ is relatively nonreactive under most atmospheric conditions, it seldom enters into chemical reactions, but under pressure and at very high temperatures, it will react with O₂:



Approximately, 90–95% of the nitrogen oxides generated in combustion processes are in the form of nitric oxide (NO), but other nitrogen oxides can form, especially nitrogen dioxide (NO₂), so air pollution experts refer to NO and NO₂ collectively as NO_x. In fact, in the atmosphere the emitted NO is quickly converted photo-chemically into NO₂.



Equation (3) shows the photochemical dissociation of NO₂. Equation (4) shows the formation of ozone from the combination of O and molecular O₂ where M is any third-body molecule (principally N₂ and O₂ in the atmosphere). Equation (5) shows the oxidation of NO by O₃ to form NO₂ and molecular oxygen. These reactions represent a cyclic pathway driven by photons represented by $h\nu$.

Nitric oxide is a colourless, odourless gas and is essentially insoluble in water. Nitrogen dioxide has a pungent acid odour and is somewhat soluble in water. At low temperatures

such as those often present in the ambient atmosphere, NO_2 can form the molecule $\text{NO}_2\text{-O}_2\text{N}$ or simply N_2O_4 that consists of two identical simpler NO_2 molecules. This is known as a dimer. The dimer N_2O_4 is distinctly reddish-brown and contributes to the brown haze that is often associated with photochemical smog.

Both NO and NO_2 are harmful and toxic to humans, although atmospheric concentrations of nitrogen oxides are usually well below the concentrations expected to lead to adverse health effects. The low concentrations are due to the moderately rapid reactions that occur when NO and NO_2 are emitted into the atmosphere. Much of the concern about regulating NO_x emissions stems from the wish to suppress the reactions in the atmosphere that generate the highly reactive molecule ozone (O_3). Nitrogen oxides play key roles in important reactants in O_3 formation. Ozone forms photo-chemically (i.e. the reaction is caused or accelerated by light energy) in the lowest level of the atmosphere, known as the troposphere, where people live. Nitrogen dioxide is the principal gas responsible for absorbing the sunlight needed for these photochemical reactions. So, in the presence of sunlight, the NO_2 that forms from NO incrementally stimulates photochemical smog-forming reactions because nitrogen dioxide is very efficient at absorbing sunlight in the ultraviolet portion of its spectrum. This is why ozone episodes are more common during the summer and in areas with ample sunlight.

High temperature/high pressure conditions exist in internal combustion engines, like those in automobiles (known as "mobile sources"), which is why NO_x is one of the major mobile source air pollutants. These conditions of high temperature and pressure can also exist in boilers such as those in power plants, so NO_x is also commonly found in high concentrations leaving fossil fuel power generating stations. In addition to atmospheric nitrogen, other sources exist, particularly the nitrogen in fossil fuels.

The nitrogen oxides generated from atmospheric nitrogen are known as "thermal NO_x " since they form at high temperatures, such as near burner flames in combustion chambers. Nitrogen oxides that form from fuel or feedstock are called "fuel NO_x ." A significant fraction of fuel nitrogen remains in the bottom ash or in unburned aerosols in the gases leaving the combustion chamber, i.e. the fly ash (Vallero, 2008). In the low-oxygen area of flames, NO can be formed via fuel radicals with nitrogen – so called "prompt NO_x " (Baumbach, 1996).

Indoor nitrogen oxides air pollution can be generated by outdoor or indoor sources. The primary indoor sources of nitrogen oxides are unvented fuel burning appliances (gas ranges, kerosene space heaters etc.), heating appliances and tobacco smoking. Increased levels of nitrogen oxides can occur in homes with gas cookers, paraffin heaters and certain portable bottled gas heaters as well as with badly maintained central heating boilers or gas fires. The rate of production is governed by three main factors: the amount of oxygen, the flame temperature, and the rate of cooling of the combustion products (since NO formation and conversion proceed more rapidly at higher temperatures). The average level in homes without combustion appliances is about half of that outdoor; in homes with kerosene heaters or unvented gas space heaters, indoor levels often exceed outdoor levels. Immediate concentrations of NO_x , occurring during cooking are still more dangerous, however, mainly during winter, when ventilation is insufficient.

3. Measurement of nitrogen oxides

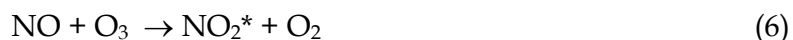
Measurements of nitrogen oxides were carried out in order to determine indoor and outdoor NO_x concentrations during the year, as well as the influence of air exchange and air

tightness of building structures on indoor NO_x occurrence depending on the indoor sources of NO_x . The significant part of measurements of NO_x concentrations focused on the dynamics of NO_x concentrations changes during the combustion process for various rates of gas burning in modelled conditions for the purpose of determining relevant parameters influencing NO_x occurrence. The outcome of the modelled measurements was the development of a mathematical model for predicting the occurrence of indoor nitrogen oxides.

3.1 Method

The investigation included online gas measurement for NO , NO_2 and NO_x by the 'Thermo Environmental Instruments Inc. - model 42 - Chemiluminescence $\text{NO-NO}_2\text{-NO}_x$ Analyzer'. This analyzer was located in the middle of the room, at the level of the breathing zone of a sitting person - 105 cm above the floor. The results showed concentration peaks during cooking, but with large differences resulting from different rates of gas burning.

The principal method used for measuring NO_2 is based on chemiluminescence (6), (7):



where $h\nu$ represents a photon of solar radiation of energy and NO_2^* is the NO_2 molecule in the excited state.

NO_2 concentrations are determined indirectly from the difference between the NO and NO_x ($\text{NO} + \text{NO}_2$) concentrations in the atmosphere. These concentrations are determined by measuring the light emitted from the chemiluminescent reaction of NO with O_3 (similar to the reaction of O_3 with ethylene noted for the measurement of O_3), except that O_3 is supplied at a high constant concentration, and the light output is proportional to the concentration of NO present in the ambient air stream (Vallero, 2008).

3.2 Indoor and outdoor NO_x occurrence during the year

Indoor sciences have been studied over the last twenty years at the Civil Engineering Faculty of the Technical University of Košice in Slovakia. Initially the monitoring of indoor pollutants (radon, particles, VOCs, odours, nitrogen oxides, etc.) was undertaken in buildings located in the east part of Slovakia. Seasonal differences in pollutant levels were confirmed. It was shown in this study that indoor NO_x concentrations were significantly higher than outdoor ones especially in the winter (Fig. 1.). All buildings studied used gas appliances, mainly in the kitchens (Šenitková, 2000).

3.3 Influence of air exchange on NO_x concentrations

The effect of air exchange by ventilation on indoor nitrogen dioxide occurrence was studied in buildings with natural and forced air replacement during and after the combustion process. Monitoring of NO_2 concentrations was carried out during the use of a gas stove with gas consumption from 0.2 to 0.4 m^3 for 30 minutes. The increase and decrease of NO_2 concentrations were monitored depending on the air exchange in the room. The measurements were performed with exhaust ventilation turned both off and on and at different adjustments of air exhaust. The maximum concentration of NO_2 with natural air exchange was 321.1 $\mu\text{g}/\text{m}^3$, and 185.9 $\mu\text{g}/\text{m}^3$ with an air exhaust rate of 130 m^3/h , i.e. the air

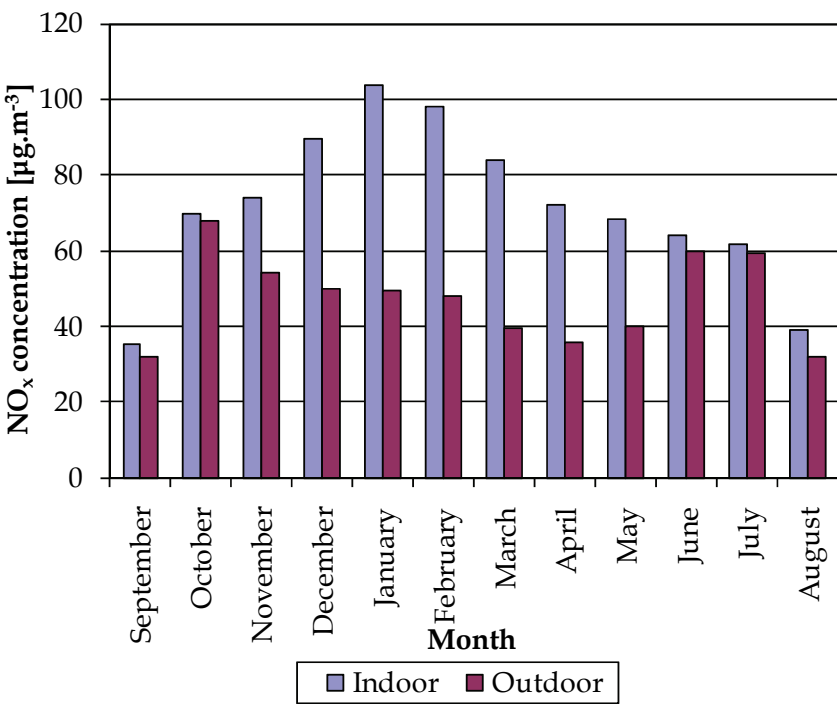


Fig. 1. Mean indoor and outdoor NO_x concentrations throughout the year

exchange rate was about 4 times per hour. Before the start of the combustion process, background concentrations of NO₂ ranging from 26.6 to 38.3 µg/m³ were determined, as were concentrations of NO₂ at the natural ventilation of 87.8 µg/m³ for 35 minutes after burning. In contrast, the value of NO₂ concentration reached only 20.3 µg/m³ with forced air replacement. The dynamics of the increase and decrease of NO₂ concentrations monitored in the room with natural and forced ventilation is evident from Fig. 2.

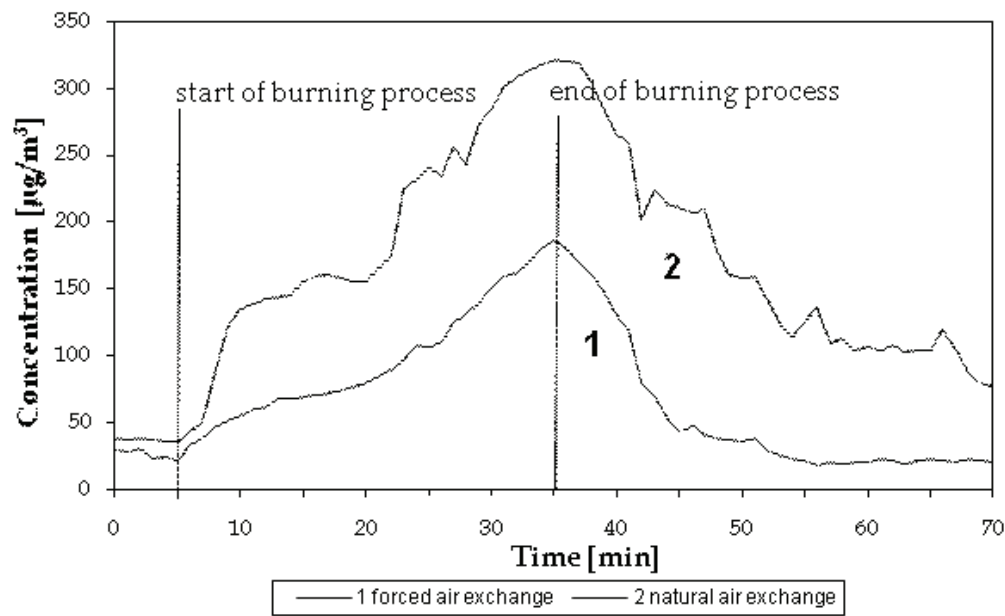


Fig. 2. NO₂ concentrations depending on air exchange

In rooms with a similar layout and volume, higher values of NO_2 concentrations were reached, especially when the combustion process did not provide sufficient air exchange (insufficient natural ventilation by infiltration). NO_2 concentrations exceeded the Slovak safety limit value of $200 \mu\text{g}/\text{m}^3$ during and after the combustion process. The measurements of NO_2 concentrations have confirmed that the level of nitrogen oxides indoors depends not only on the rate of combustion and on the volume of the room but also on the manner and rate of air exchange, and in particular on the specific distribution conditions.

3.4 Influence of building structures on NO_x concentrations

In order to verify the influence of air tightness of openings, NO_2 concentrations were measured in kitchens with both wooden and plastic windows. The gap permeability coefficient for wooden windows was $i_{lv} = 1.4 \times 10^{-4} \text{ m}^2/(\text{s} \cdot \text{Pa}^{0.67})$, the determined air exchange, $n = 1.8$. The gap permeability coefficient for plastic windows as determined by the manufacturer was $i_{lv} = 0.02 \times 10^{-4} - 0.4 \times 10^{-4} \text{ m}^2/(\text{s} \cdot \text{Pa}^{0.67})$, the determined air exchange, $n = 0.2$. Measurements of NO_2 concentrations in buildings with wooden and plastic windows were made with other parameters (rates of gas burning, time and burning conditions etc.) equal. Outdoor NO_2 concentrations, depending on the time of measurement and diffusion conditions, varied from $17.3 \mu\text{g}/\text{m}^3$ to $36.1 \mu\text{g}/\text{m}^3$. The increase of NO_2 concentrations was monitored, depending on the intensity of gas burning, for sixty minutes and the decrease of NO_2 concentrations for ninety minutes. The influence of tightness of windows on the dynamics of NO_2 concentration changes for the intensity of gas combustion from 0.1 to $0.2 \text{ m}^3/\text{h}$ is shown in Fig. 3.

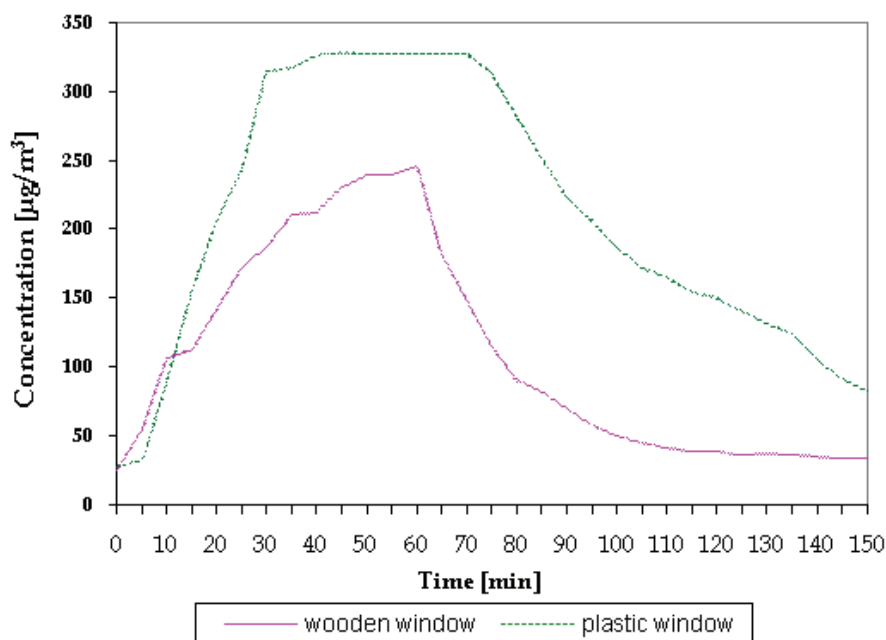


Fig. 3. Dynamics of NO_2 concentration changes

In kitchens with wooden windows a maximum NO_2 concentration of $246.1 \mu\text{g}/\text{m}^3$ was reached after 60 minutes of gas burning at a consumption rate of 0.1 to $0.2 \text{ m}^3/\text{h}$. In kitchens with plastic windows, a significantly higher concentration of $315.2 \mu\text{g}/\text{m}^3$ was reached after 30 minutes of gas burning at a consumption rate of 0.1 to $0.2 \text{ m}^3/\text{h}$. During the combustion process, the NO_2 concentrations increased linearly in both kitchens with wooden and plastic

windows. After completion of the combustion process, the concentrations decreased and after 90 minutes a value of $33.7 \mu\text{g}/\text{m}^3$ was reached in kitchens with wooden windows. In kitchens with plastic windows the concentrations increased linearly for only the first 30 minutes, at which point they reached a value of $315.2 \mu\text{g}/\text{m}^3$. Then, for 45 minutes the concentrations steadied and values ranged from 318.7 to $328.4 \mu\text{g}/\text{m}^3$. Concentrations decreased 15 minutes after the end of the combustion process and a value of $82.4 \mu\text{g}/\text{m}^3$ was reached after 90 minutes. The dynamics of the changes in concentrations showed a need for a targeted air exchange during gas stove combustion.

3.5 Indoor NO_x concentrations in modelled conditions

Measurements were made to detail NO_x production and determine the resultant indoor concentrations while gas appliances were being used. The intensity of gas combustion varied from $0.1 \text{ m}^3\cdot\text{h}^{-1}$ to $0.7 \text{ m}^3\cdot\text{h}^{-1}$. As kitchens had been identified as the most polluted rooms, measurements were taken there. The levels of background concentrations of NO , NO_2 and NO_x were monitored. The increase and decrease of NO_x concentrations was found at the same time.

Changes in NO_x concentrations for the range of gas consumption intensities were studied. The changes while allowing the combustion process to continue at a maximum rate for 60 min, and for decreasing rates of gas usage, were then followed for 90 min after combustion had stopped. The mean NO_x concentrations for the intensity of gas consumption from $0.1 \text{ m}^3\cdot\text{h}^{-1}$ to $0.7 \text{ m}^3\cdot\text{h}^{-1}$ are presented in Table 1.

Before the start of the combustion process the average outdoor i.e. indoor background concentrations of NO_x were determined and mean values from 21.0 to $27.9 \mu\text{g}/\text{m}^3$ were recorded. The combustion process took 60 minutes and a linear increase was recorded for 5 - 60 minutes depending on the intensity of gas burning. After the completion of combustion, mean NO_x concentrations achieved high levels from 1785.9 to $2889.2 \mu\text{g}/\text{m}^3$. 90 minutes after the end of the combustion process, the mean values of NO_x were from 279.1 to $1293.6 \mu\text{g}/\text{m}^3$. The concentrations of NO_x after 1 h of gas combustion were shown to depend on the level of gas consumption ranging from $0.1 \text{ m}^3\cdot\text{h}^{-1}$ to $0.7 \text{ m}^3\cdot\text{h}^{-1}$ (Fig. 4).

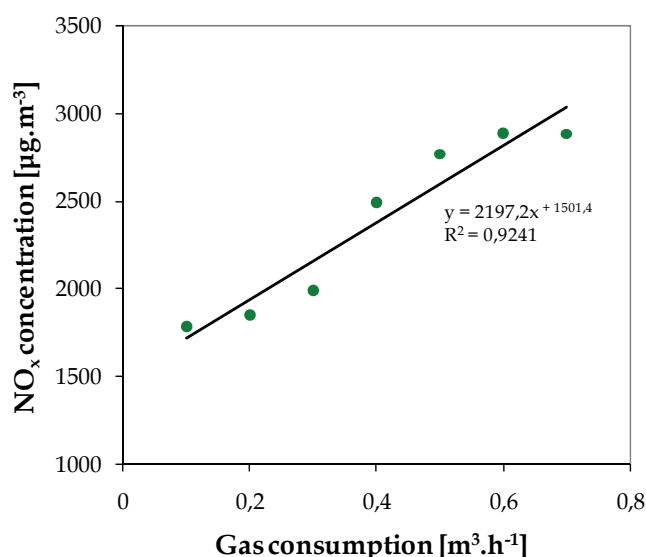


Fig. 4. NO_x concentrations for the gas consumption range studied

Time [min]	Intensity of gas consumption [m³.h ⁻¹]						
	0.1	0.2	0.3	0.4	0.5	0.6	0.7
0	21.0	22.3	27.9	26.3	22.6	24.8	23.3
5	204.7	429.2	756.8	1043.5	1819.2	3058.7	3071.2
10	489.5	829.8	1521.1	2269.8	2875.0	2886.1	2882.5
15	671.5	1140.4	2076.6	2590.6	2758.0	2887.7	2882.1
20	854.5	1366.1	2272.2	2475.9	2771.1	2900.5	2885.8
25	1013.0	1669.2	2060.8	2496.2	2773.1	2892.8	2885.8
30	1131.8	1910.1	1996.4	2497.1	2771.8	2899.5	2884.0
35	1320.8	2089.8	1994.3	2496.2	2771.8	2894.1	2882.6
40	1477.9	2092.7	1990.9	2494.9	2769.6	2892.6	2883.0
45	1565.1	1899.7	1988.2	2495.3	2770.8	2893.2	2886.2
50	1660.9	1857.3	1991.1	2493.0	2769.9	2888.2	2884.7
55	1711.7	1851.3	1990.6	2492.2	2769.2	2889.6	2883.8
60	1785.9	1852.4	1989.0	2490.4	2769.9	2889.2	2885.1
65	1644.4	1857.9	1990.2	2491.6	2769.4	2890.4	2883.9
70	1432.5	1851.0	1989.7	2492.7	2769.8	2890.0	2881.7
75	1282.8	1864.2	1989.0	2496.3	2769.9	2889.6	2882.1
80	1172.5	1937.9	1989.9	2493.2	2772.8	2890.3	2884.7
85	1041.1	1891.0	1987.8	2492.8	2773.6	2890.8	2885.7
90	929.6	1629.7	2028.6	2488.8	2775.4	2890.9	2885.8
95	810.7	1395.7	2142.3	2535.9	2773.1	2890.4	2886.2
100	715.3	1256.4	2003.4	2558.7	2794.8	2882.0	2873.1
105	634.9	1107.5	1759.1	2283.3	2790.3	2940.0	2944.0
110	566.5	994.4	1657.5	2034.3	2622.0	2912.1	2900.9
115	512.8	881.4	1526.9	1807.7	2383.4	2610.6	2627.4
120	468.8	800.7	1425.0	1672.8	2207.8	2404.9	2522.1
125	431.8	705.1	1315.7	1541.4	2009.2	2267.6	2359.9
130	393.5	656.8	1214.2	1420.2	1860.4	2080.5	2159.5
135	362.7	587.5	1126.9	1323.6	1712.4	1859.8	1929.1
140	340.1	538.9	1017.3	1248.1	1538.7	1650.4	1687.8
145	310.3	473.9	920.4	1128.7	1366.9	1469.2	1519.3
150	279.1	441.6	848.8	1021.9	1202.3	1284.7	1293.6

Table 1. Mean NO_x concentrations for various gas usage rates

The oxides formation at a gas consumption rate of 0.1 m³.h⁻¹ is given in Fig. 5. It is evident that an essentially linear increase of NO, NO₂ and NO_x concentrations occurred during the whole combustion period to a final maximum value after 1 h, when gas usage was stopped (NO 1357.4 µg.m⁻³, NO₂ 428.5 µg.m⁻³ and NO_x 1785.9 µg.m⁻³). A decrease in concentrations was then observed for a period of 90 min, after which the concentrations were NO 209.8 µg.m⁻³, NO₂ 69.4 µg.m⁻³ and NO_x 279.1 µg.m⁻³. Importantly, it should be noted that the concentration of NO₂ after 1 h of gas combustion (428.5 µg.m⁻³) was more than double the Slovak safety limit (200 µg.m⁻³). The limit was exceeded 10 min after the start of combustion (270.9 µg.m⁻³) and concentrations remained above the limit for the next 100 min (228.6 µg.m⁻³).

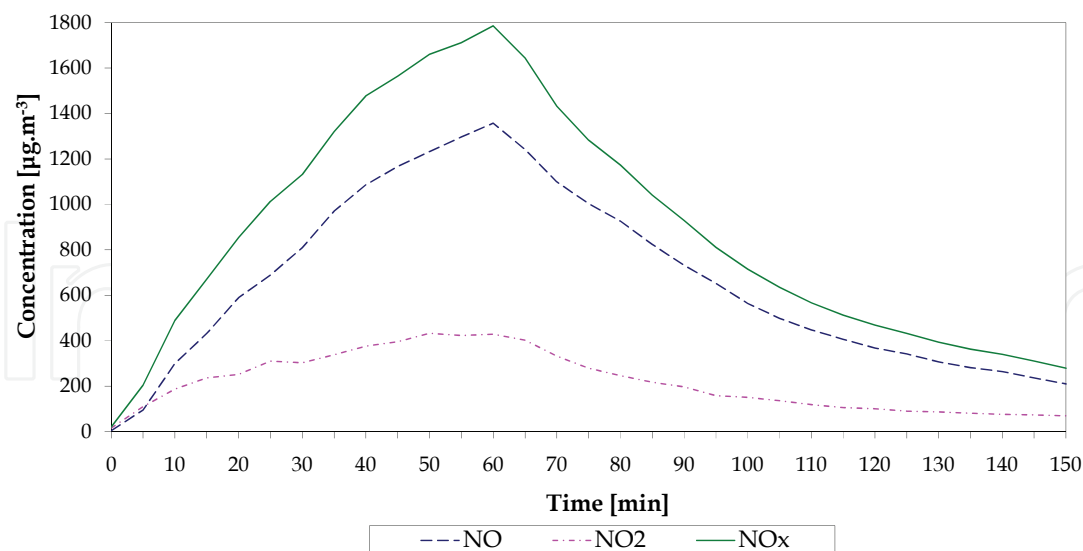


Fig. 5. Nitrogen oxides concentrations at a gas usage rate of $0.1 \text{ m}^3.\text{h}^{-1}$
Oxides formation at a gas consumption rate of $0.2 \text{ m}^3.\text{h}^{-1}$ is given in Fig. 6.

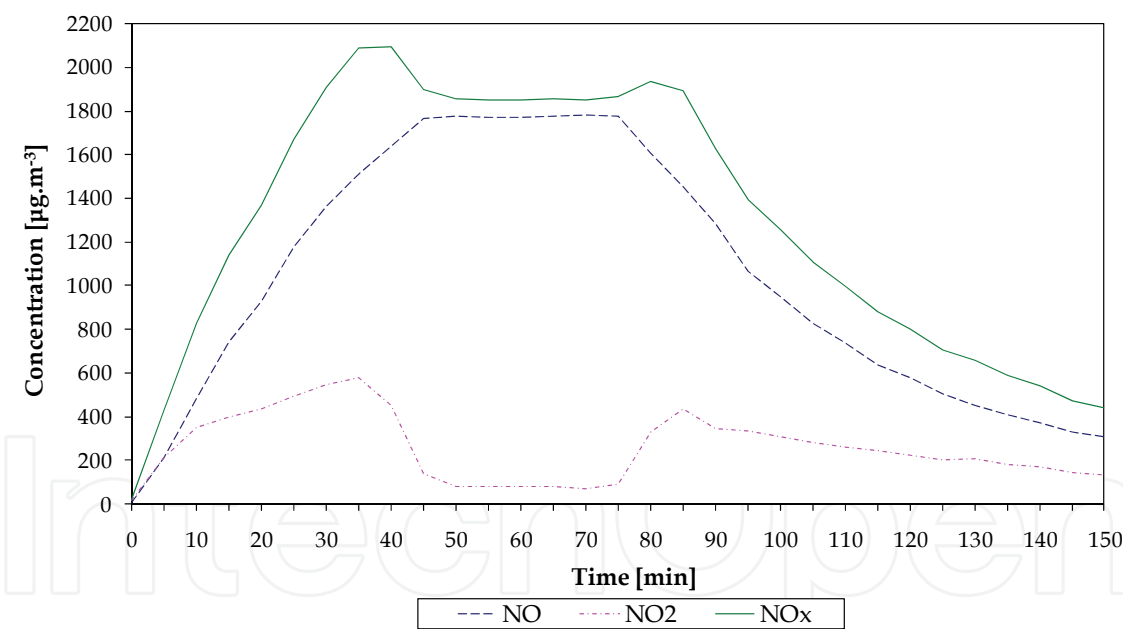


Fig. 6. Nitrogen oxides concentrations at a gas usage rate of $0.2 \text{ m}^3.\text{h}^{-1}$
The figure shows that a linear increase in concentrations of NO, NO₂ and NO_x was not sustained throughout the combustion process. Concentrations of NO₂ and NO_x rose linearly for 35 minutes, reaching an NO₂ value of $577.2 \mu\text{g.m}^{-3}$ and an NO_x value of $2089.8 \mu\text{g.m}^{-3}$. Subsequently the concentrations decreased and for 25 minutes were almost constant, with NO₂ values ranging from 68.8 to $88.2 \mu\text{g.m}^{-3}$ and NO_x from 1851.0 to $1864.2 \mu\text{g.m}^{-3}$. As a consequence of natural air exchange, an increase of NO₂ concentrations and decrease of NO concentrations occurred 15 minutes after the completion of combustion. 90 minutes after the

end of combustion, the concentrations were NO 306.8 $\mu\text{g.m}^{-3}$, NO₂ 134.8 $\mu\text{g.m}^{-3}$ and NO_x 441.6 $\mu\text{g.m}^{-3}$.

4. Mathematical modelling of indoor nitrogen oxides

Modelling is a powerful tool that is being used extensively by scientists to investigate air movement and airflow patterns and to evaluate the performance of HVAC systems. Numerous models have been developed to study indoor air quality (IAQ). Although designers, architects, code enforcement officials, and other building scientists have been using modelling as a powerful tool to address indoor IAQ issues, more development is needed to increase the accuracy and the validity of the IAQ models. These are being used to predict the dispersion of air contaminants within the building environment. Air pollution concentration can be estimated both spatially and as a function of time. The aim of IAQ models is to provide a way to link information about sources, distribution, dynamic changes of pollutant concentrations and also building factors to estimate indoor pollutant concentrations. Many of the latest indoor air models include exposure modelling and risk modelling. The type of model used depends on the planned uses of the model. The most common uses of IAQ models are the following: (i) estimating population exposure to various indoor pollutants, (ii) estimating the impact of individual sources on pollutant concentration and (iii) estimating the impact of individual sources and IAQ control options on personal exposure (Spengler et al., 2001).

Designing a model and modelling can substantially contribute to problem solving and to obtaining new understanding of system behaviour with respect to air component dynamic changes. A mathematical description of the processes can generalise experimental results so that they may be used to interpret other processes physically similar to those introduced in the model. Although modelling doesn't achieve all the rules of similarity, it does provide overall information about a system's behaviour, which is only a little different from a real situation. Also, although it is only an approximation, the model does simplify description of the real physical measurements.

Most research institutes and universities focus on modelling and predicting outdoor and indoor pollutants. Most of the models developed to predict human exposures use empirical data on indoor/outdoor ratios (I/O) to determine indoor microenvironment concentrations as a function of time. These models are described in various studies. For example, one study (Sexton et al., 1983) describes a simple deterministic model, relating exposure to background ambient levels, indoor values, and human activities. Results suggest that indoor nitrogen dioxide concentrations in private dwellings vary primarily because of outdoor levels and the type of cooking fuel used, but are also affected by factors such as air-exchange rates and strength of indoor sources. Estimates of population exposures are obtained by combining observed distributions of nitrogen dioxide concentrations from outdoor and indoor settings with information about the number of people and amount of time spent in each microenvironment. One group of researchers (Lee et al., 1998) developed regression models for indoor NO₂ exposure. Using analytical methods to choose descriptive proxies and evaluate the tradeoffs in their implementation can help epidemiological studies improve their designs. Other researchers (Axley et al., 1994) developed and applied mathematical models to predict indoor air concentrations resulting from the transport of outdoor gas-phase nitrogen oxides, ozone and nitric acid indoors; and integral expressions that may be used to estimate model parameters from dynamic measurements of contaminant

concentration. Another study (Dimitroulopoulou et al., 2001, Ashmore et al., 1999) deals with a modelling of indoor exposure to nitrogen dioxide in the UK. A dynamic multi-compartment computer model was developed to describe the physical processes determining indoor pollutant concentrations as a function of outdoor concentrations, indoor emission rates and building characteristics. The model has been parameterised for typical UK homes and workplaces and linked to a time-activity model to calculate exposures for a representative homemaker, schoolchild and office worker, with respect to NO_2 . This model can be used to distinguish three types of exposures: outdoor exposure, indoor exposure resulting from penetration of outdoor air and indoor exposure resulting from indoor sources. The model predicts the above types of personal exposures for a representative homemaker, schoolchild and office worker, based on indicative activity patterns. The deterministic modelling approach applied here combines two types of models: (i) A physical model, used to calculate hourly indoor air pollutant concentrations for different microenvironments as a function of outdoor concentrations, building characteristics and indoor source emissions. This physical model, entitled INTAIR, is a simple dynamic compartment model which solves the resulting set of differential equations using a fourth-order Runge-Kutta scheme. (ii) An exposure model, used to calculate personal exposures by combining the movement of typical individuals through a series of microenvironments with the modelled ME concentrations. Another study (Baxter et al., 2007) seeks to utilize publicly available data and questionnaire responses to predict residential indoor concentrations of traffic-related air pollutants for lower socioeconomic status (SES) urban households. Indoor and outdoor 3–4 day samples of NO_2 were collected in 43 low SES residences across multiple seasons from 2003 to 2005. Home characteristics and occupant behaviours were collected via a standardized questionnaire. Additional housing information was collected through property tax records, and ambient concentrations were collected from a centrally located ambient monitor. The contributions of ambient concentrations, local traffic and indoor sources to indoor concentrations were quantified using regression analyses. Comparing models based on covariate selection using p-values or a Bayesian approach yielded similar results, with traffic density within a 50 m buffer of a home and distance from a truck route as important contributors to indoor levels of NO_2 . The Bayesian approach also highlighted the uncertainty in the models.

The difficulty of predicting NO_x emissions during combustion of fossil fuels is caused mostly by the complexity of the mechanisms of NO_x formation, mechanisms which depend on many factors. Some partial mechanisms have not yet been completely clarified so it is not possible to describe them exactly. Consequently, empirical correlations between fuel properties, combustion regime and NO_x emissions cannot be properly calculated. This affects the degree to which calculations can be informative in a mathematical model of indoor NO_x production. The final result has to be assembled from the results obtained from statistically suitable collections of measurements as well as more controlled laboratory measurements.

4.1 Input parameters

The research task to describe NO_x production in the combustion process relies on the measurement of NO_x as influenced by those parameters which directly characterize the combustion process, i.e. intensity of gas combustion I ($\text{m}^3 \cdot \text{s}^{-1}$), exposure period t (s), amount of indoor air m (kg) and air change n (s^{-1}).

All the given variables are defined using basic dimensions, allowing their direct use for dimensional analysis to create a mathematical model of NO_x production. All the variables can be simply measured during combustion allowing us to then compare NO_x production under the particular working conditions of the chosen combustion device on the basis of direct measurements and by means of the created mathematical model described in the following section.

4.2 Dimensional analysis application

Dimensional analysis offers a method for reducing complex physical problems to the simplest form prior to obtaining a quantitative answer. At the heart of dimensional analysis is the concept of similarity. In physical terms, similarity refers to some equivalence between two things or phenomena that are actually different. Mathematically, similarity refers to a transformation of variables that leads to a reduction in the number of independent variables that specify a problem (Sonin, 2001). The behaviour of a physical system is defined by the complete set of dimensionless variables formed by the relevant physical variables. This fact suggests that if two systems have the same numerical values for all the defining dimensionless variables, then these two systems are dimensionally similar. Further, if there are two dimensionally similar systems, then their behaviour can be closely correlated and hence the results of measurements on either one can be “projected” to the other. This is the basis for dimensional modelling. Therefore the first step in a model experiment is to construct a complete set of dimensionless variables relevant to the system. These dimensionless variables are then made to be equal for the model and the prototype (Szirtes & Rozsa, 2006).

The mathematical model describing NO_{xi} formation is based on the formation of dimensionless arguments π_i , from the stated variables that influence formation of the oxides of nitrogen. The valuable property of these arguments is that in all existing systems of units they have the same numerical size and they have no dimension. The formation of a mathematical model rests on derivation of functional dependence from the expressed dimensionless variables, which in general always has an exponential character. Transformation of this function into logarithmic co-ordinates corresponds to a linear character that makes the work with the model easier and enables one to determine simply the parameters of linear function (Čarnogurská, 1998, 2000).

The model given here has universal validity for all combustion devices that have at least approximately similar geometric characteristics. For every combustion device the parameters of linear function, i.e. regression coefficients, have to be determined separately.

The general relationship between the selected variables which affect NO_{xi} production can be expressed in the form:

$$\varphi(I, t, m, \text{NO}_{xi}, n) = 0 \quad (8)$$

The dimensional matrix-relationship created (9) has the rank of a matrix where $r = 3$ and its lines are dimensionally independent of each other. From $n = 5$ independent variables a matrix r can be set up with $(n-r)$ dimensionless arguments.

$$\begin{array}{c} t \quad I \quad m \quad \text{NO}_{xi} \quad n \\ \begin{array}{l} s \\ m \\ kg \end{array} \left| \begin{array}{ccccc} 1 & -1 & 0 & 0 & -1 \\ 0 & 3 & 0 & -3 & 0 \\ 0 & 0 & 1 & 1 & 0 \end{array} \right| \end{array} \quad (9)$$

For the general form of the argument Π is valid:

$$\Pi = t^{x_1} \cdot I^{x_2} \cdot m^{x_3} \cdot NO_{xi}^{x_4} \cdot n^{x_5} \quad (10)$$

From the condition that the left side of the identity equals one (ordinary number Π), the resulting exponent of every basic dimension is equal to zero. By applying this condition for every element from r basic dimensions, we obtain a system of three linear equations with five unknowns. To solve the system, we have to select two unknowns (and this always twice) and then calculate the rest.

For the selected $x_4 = 1, x_5 = 0$ the exponents will be calculated from the linear equations system, thus $x_1 = 1, x_2 = 1, x_3 = -1$.

For the selected $x_4 = 0, x_5 = 1$ the exponents will be calculated from the linear equations system, thus $x_1 = 1, x_2 = 0, x_3 = 0$.

For these the dimensionless argument is then valid:

$$\begin{array}{ccccc} & x_1 & x_2 & x_3 & x_4 & x_5 \\ \Pi_1 & \left| \begin{array}{ccccc} 1 & 1 & -1 & 1 & 0 \end{array} \right| \\ \Pi_2 & \left| \begin{array}{ccccc} 1 & 0 & 0 & 0 & 1 \end{array} \right| \end{array} \quad (11)$$

The required dimensional homogeneous function in dimensionless form is

$$\Psi(\Pi_1, \Pi_2) = 0 \quad (12)$$

or after adjustment and backward transformation of dimensions for the particular variables, the function will have the form:

$$\Psi\left(\frac{t \cdot I \cdot NO_{xi}}{m}, t \cdot n\right) = 0 \quad (13)$$

The real course of the dependence of dimensionless arguments Π_1 to Π_2 calculated according to the relationship (13) from the measured values.

$$\Pi_1 = A \cdot \Pi_2^B \quad (14)$$

From this a regression straight line for the calculation of the regression coefficients can be calculated by the method of least squares.

After completing relationship (14) using relationship (13) and its modification, we obtain the relationship characterising NO_x production in the form:

$$NO_{xi} = \frac{10^9 \cdot A \cdot t^{B-1} \cdot n^B \cdot m}{I} \quad (15)$$

where A and B are regression coefficients calculated by the method of least squares.

The mathematical formula (15) presents a mathematical model of NO_{xi} production through using of gas appliances in the kitchen. The indoor NO_x is given by the formula:

$$\text{NO}_x = \text{NO}_{xi} + \text{NO}_{xo}$$

(16)

NO_{xi} – indoor nitrogen oxides concentration [$\mu\text{g}.\text{m}^{-3}$]
 NO_{xo} – outdoor nitrogen oxides concentration [$\mu\text{g}.\text{m}^{-3}$]

5. Verification of the mathematical model

The mathematical model was verified for various amounts of gas consumption in the range of 0.1 – 0.7 $\text{m}^3.\text{h}^{-1}$. As an illustration, the verification for gas consumptions of 0.1 $\text{m}^3.\text{h}^{-1}$ and of 0.2 $\text{m}^3.\text{h}^{-1}$ using a model experimental room is given below. The amount of air was 28.28 kg and the air change rate $3.56 \times 10^{-4} \text{ s}^{-1}$. The air change rate was determined on the basis of decreasing NO_x concentration per hour.
On the basis of measured input parameters, NO_{xi} concentrations were determined through the located constants and regression coefficients obtained by the method of least squares. These values are presented in Table 2.

Gas consumption [$\text{m}^3.\text{h}^{-1}$]	A	B
0.1	4.0000E	1.8420
0.2	2.0000E	1.7765
0.3	2.0000E	1.8179
0.4	3.0000E	1.8593
0.5	4.0000E	1.6603

Table 2. Regression coefficient values

As an illustration, the relationship for NO_{xi} production at a gas consumption rate of 0.1 $\text{m}^3.\text{h}^{-1}$ is obtained from:

$$\text{NO}_{xi} = \frac{4.0\text{E} - 09 . t^{1.8420-1} . n^{1.8420} . m}{I}$$

(17)

The values of measured and calculated NO_{xi} concentrations are compared in Table 3. The differences between measured and calculated NO_{xi} concentrations are considered acceptable. The deviations reflect two things: the selection of relevant parameters is a simplification; their degree of involvement and the number of factors on which NO_{xi} production depends, which they represent, are both unknown.
The relationship for NO_{xi} production at a gas consumption rate of 0.2 $\text{m}^3.\text{h}^{-1}$ is obtained from:

$$\text{NO}_{xi} = \frac{2.0\text{E} - 09 . t^{1.7765-1} . n^{1.7765} . m}{I}$$

(18)

The values of NO_{xi} concentrations measured and calculated for consumption of 0.2 $\text{m}^3.\text{h}^{-1}$ are compared in Table 4. A linear increase of NO_{xi} concentrations continued for forty minutes from the start of combustion, i.e. only this amount of time was required for complete combustion of gas burning.

t [min]	NO _{xi,measured} [µg.m ⁻³]			NO _{xi,calculated} [µg.m ⁻³]	Deviation [%]
	min	max	mean		
5	195.7	219.5	204.7	243.9	20.67
10	426.7	534.5	489.5	420.6	16.34
15	593.4	740.3	671.5	583.2	16.97
20	712.7	992.5	854.5	737.3	19.54
25	884.5	1161.7	1013.0	885.4	18.43
30	999.8	1262.0	1131.8	1028.9	17.1
35	1213.4	1478.5	1320.8	1168.6	13.89
40	1410.5	1566.1	1477.9	1305.1	12.15
45	1499.4	1645.3	1565.1	1439.0	10.77
50	1571.1	1747.4	1660.9	1570.5	10.02
55	1639.6	1776.3	1711.7	1700.0	11.53
60	1762.1	1803.3	1785.9	1827.7	11.25

Table 3. Measured and calculated NO_{xi} concentrations for gas consumption of 0.1 m³.h⁻¹

t [min]	NO _{xi,measured} [µg.m ⁻³]			NO _{xi,calculated} [µg.m ⁻³]	Deviation [%]
	min	max	mean		
5	316.2	556.0	429.2	421.2	33.89
10	632.6	1016.2	829.8	705.6	24.98
15	1003.8	1290.2	1140.4	958.5	15.16
20	1230.8	1499.1	1366.1	1192.8	13.26
25	1527.1	1779.5	1669.2	1414.3	14.80
30	1785.4	2031.0	1910.1	1625.9	14.54
35	1960.2	2201.3	2089.8	1829.9	12.00
40	2048.6	2151.4	2092.7	2027.3	4.48

Table 4. Measured and calculated NO_{xi} concentrations for gas consumption of 0.2 m³.h⁻¹

6. Interpretation of mathematical model

Current knowledge as well as the results of monitoring in a selected group of residential buildings in situ confirmed the significant occurrence of indoor nitrogen oxides, especially with the burning of gas in kitchens, and lead to the construction of a mathematical model for predicting the occurrence of nitrogen oxides. Subsequently, a mathematical model based on the dimensional analysis method was verified for each intensity of gas burning. The calculated values of NO_{xi} concentrations were compared with average measured concentrations of NO_{xi}. The variance between the average measured and calculated values of NO_{xi} is shown in Fig. 7. This difference was established as an absolute error which ranged from values of 4.48% - 34.05%. The average absolute error is 17.86%. This value is acceptable, and confirms that the proposed mathematical model is useful for predicting the occurrence of indoor nitrogen oxides.

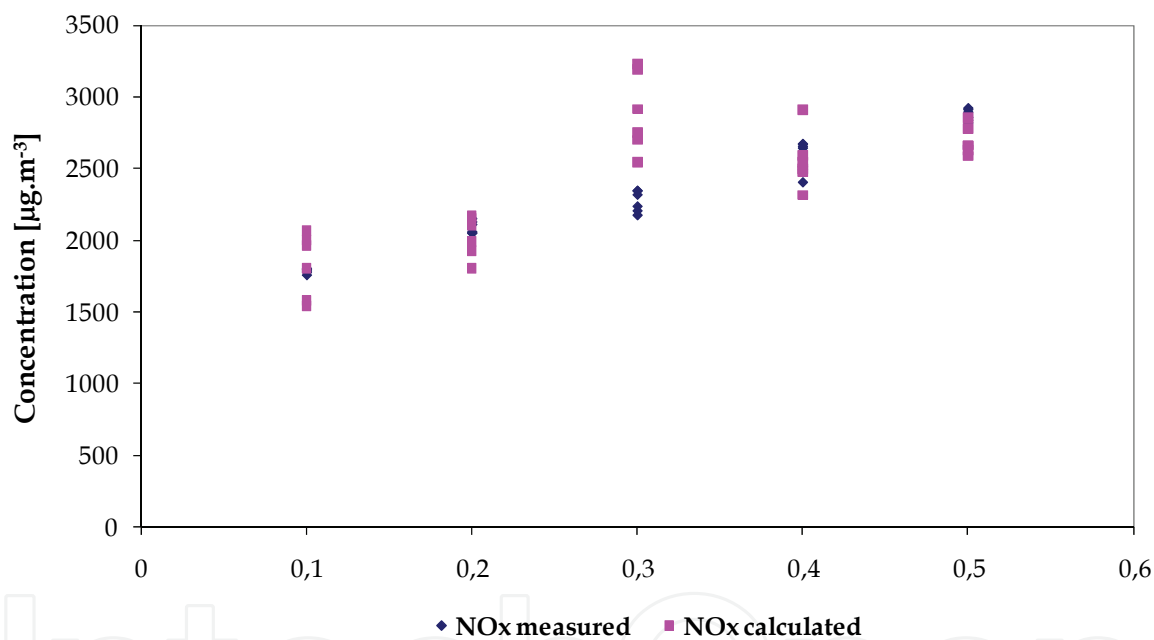


Fig. 7. Variance of measured and calculated NO_{xi} concentrations for various gas intensities

7. Conclusion

The study of indoor pollutants affecting indoor air quality is a modern problem. One solution is the use of mathematical methods to predict the level of pollutant concentrations. This chapter proposes a possible mathematical approach to indoor NO_x modelling. The correct selection and clear determination of all relevant variables at the beginning of construction of the mathematical model was important. The experimental measurements confirmed that NO_x production is mainly affected by intensity of gas combustion, exposure period, and rate of air exchange. A mathematical description of the physical processes influencing NO_x production has been derived and used to predict the occurrence of these

gases. The model was verified with satisfactory results for gas consumption in the range of $0.1 - 0.7 \text{ m}^3 \cdot \text{h}^{-1}$. Differences between measured and calculated indoor NO_{xi} concentrations are probably because the selected relevant variables for construction of the mathematical model do not give a complete and fully accurate picture of the combustion processes leading to NO_{xi} production. Results from the mathematical modelling approach can be improved through better understanding of input parameters and precise verification. The dimensional analysis method can be used for modelling and prediction of indoor pollutants. The presented mathematical model can be applicable for indoor nitrogen oxides prediction within the building design process and building operation management.

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