



Normative Problems of the Nitrogen Oxides Concentration Limiting in the Human Residence Environment

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1. Introduction

Internal air is a multiphase mixture of ingredients (gases and vapours and liquid & solid suspended substances) that surrounds the human in enclosed spaces (Miller 1997, Bieniek 2008). It is an extremely dynamic system. Its composition is characterized by high variability in a short period of time, both in terms of chemical compounds and levels of their concentrations (Klepeis et al. 2015). Together with physical factors, the chemical composition forms the quality of the internal air (Hess-Kosa 2011, Daisey & Liroy 1987).

The problem of internal air quality assessment and its impact on human health is now the subject of studies conducted by specialists from different fields of science worldwide. (Capello & Gaddi 2018, Nadadur & Hollingsworth 2015). It is considered that the internal environment is some kind of barrier protecting human beings from the changing and often negative impact of external factors (Woolley 2017, Pluschke & Schleibinger 2018). However, due to the wide variety of presented contaminants, it can cause health problems and even pose a threat to the health and well-being of users of enclosed spaces (Szkarowski & Maliszewska 2018).

The expansion of Poland's gas infrastructure foresees that by 2022, nearly 90% of Polish citizens will be inhabited by municipalities that are gasified (Strategia 2016). Natural gas is a cheap and convenient to use fuel, but you should be aware of the dangers of its use. The combustion of organic fuels is always associated with the emission of harmful substances such as nitrogen oxides (NO_x) and carbon oxides (CO and CO₂).

2. Purpose of work and research methods

Paradoxically, in the legal regulations of many countries, there are no precisely defined rules on the content of harmful substances in rooms with permanent human beings. More often speaking about the working environment, while living residence, where a person spends a similar part of his life is neglected. At the time, even the relatively low levels of harmful substances, with a long exposure period, could pose a serious risk to health.

Risks for carbon monoxide are fairly well identified. Many social campaigns make the public increasingly aware of the risks that CO may pose. Carbon dioxide exhibits poisonous properties only at very high concentrations, in the order of several volume percent. The issue of nitrogen oxides emissions looks much worse.

The only device that emits combustion products directly into the indoor air of living quarters are gas cookers. The purpose of this work is to analyse the normative problems that hinder the control and reduction of the NO_x content emitted during the combustion of natural gas in gas cookers to the internal air of flats.

With such defined work objectives, a combined research method has been used. It is composed in succession with:

- analysis of the normative bases of different countries in terms of reducing the NO_x content in the human environment;
- analysis of the impact of existing ventilation regulations on the NO_x content of the kitchen area;
- experimental verification of the received conclusions.

3. The analysis of the standards concerning NO_x content

3.1. External air

Poland has the same levels of permissible concentrations of pollutants in atmospheric air as other Europe Union countries (Air quality 2018). Table 1 shows the permissible concentration levels for the protection of human health and Table 2 summarised data in terms of plant protection (Rozporządzenie 2012).

Table 1. Permissible levels for nitrogen dioxide in terms of human health

Substance	Averaging period of measurement	Acceptable level, mg/m ³	The permissible frequency of exceeding the acceptable level during the year	Deadline for reaching the acceptable level
Nitrogen dioxide (NO ₂)	One hour	0.20	18 times	2010
	One year	0.04	–	2010

Table 2. Permissible levels for nitrogen oxides in terms of plant protection

Substance	Averaging period of measurement	Acceptable level, mg/m ³	Deadline for reaching the acceptable level
Nitrogen oxides (NO _x) ¹⁾	One year	0.03	2003

¹⁾ Sum of nitrogen dioxide and nitric oxide, expressed as nitrogen dioxide. The levels of the substance in air for gaseous pollutants are determined in the conditions: Temperature 293 K, pressure 101.3 kPa.

3.2. Work area

In Poland, the regulations determining the permissible levels of harmful substances in the working area specify 556 substances, including nitrogen oxides (Rozporządzenie 2018). Information on nitrogen oxides is presented in the Table 3.

Table 3. Permissible levels of nitrogen oxides in the working areas

Item No	Substance (CAS number)	Exposure duration	Permissible concentration	
			ppm	mg/m ³
1	Nitrogen monoxide (10102-43-9)	15 min.	-	-
		8 hr	1.33	2.5
2	Nitrogen dioxide (10102-44-0)	15 min.	0.80	1.5
		8 hr	0.37	0.7

3.3. Living quarters

Indoor air that surrounds man in enclosed spaces creates a peculiar microclimate. It is different from the composition of the outer atmosphere often can be much more polluted. The broadly understood internal air quality control is therefore an indispensable element in ensuring the comfort of users of enclosed spaces. The existing rules and recommendations for the permissible levels of NO₂ in residential areas in the selected countries have been analysed.

In **Poland**, according to the authors, mainly for economic reasons, the problem of internal air quality and its impact on users' health has been and remains marginalised. It is shown, among other things, by the smaller attention to quality of construction and finishing materials in comparison, for example, with the Scandinavian countries. In addition, internal air quality issues have in Poland a much lower societal interest compared to the assessment of pollution of other environmental elements such as atmospheric air or surface water. In Poland the rules for human housing define the permissible concentrations only for 35 harmful substances (Zarządzenie 1996). Unfortunately, these rules do not consider nitrogen oxides.

In the **United States**, there are national air quality standards for living quarters. The permissible concentration ranges for NO₂ are shown in Table 4 (Campagna et al. 2017).

Table 4. US permissible NO₂ concentrations in living quarters

Substance (CAS number)	Exposure duration	Permissible concentration	
		ppm	mg/m ³
Nitrogen dioxide (10102-44-0)	30 min.	0.45	0.85
	24 h	0.21	0.39
	One year	0.05	0.09

In **Germany**, the Dangerous Substances Regulation (GefStoffV) sets out two thresholds in the premises for individual substances. The RWI threshold corresponds to the concentration level of the substance in the internal air, for which there is currently no evidence that it has a negative impact on health. Exceeding the RWI value is undesirable for health reasons and requires appropriate action to minimize side effects.

The RWII threshold corresponds to the proven effects of harmfulness, based on the current toxicological and neurological knowledge. Achieving or exceeding this concentration of a substance requires immediate action as it may

pose a health hazard, especially for sensitive persons who are living in these areas for a long time. Depending on the exposure time, the threshold II may be defined as short-term (RWII K) or long-term (RWII L).

Table 5 shows the threshold values established by the German IRK/AOLG Working Group (German Committee 2005).

Table 5. German threshold values for NO₂ in living quarters

Substance (CAS number)	Threshold RWI		Threshold RWII		Exposure duration	Year of establish- ment
	ppm	mg/m ³	ppm	mg/m ³		
Nitrogen dioxide (10102-44-0)	0.04	0.08	0.13	0.25	1 h	2018

In **Canada**, the reference concentration (RfC) is used to determine the standards for air quality in residential spaces (RIAQG). This concentration means the content of pollutants in the air below which persons (including sensitive subgroups) do not suffer adverse health effects. For NO₂, the RfC values are shown in Table 6 (Government 2015).

Table 6. Canadian NO₂ thresholds for living quarters

Substance (CAS number)	Exposure duration	Threshold RfC		Noticeable effects after RfC exceeded
		ppm	mg/m ³	
Nitrogen dioxide (10102-44-0)	Short-term (1 h)	0.09	0.17	Decreased lung function and increased respiratory response of asthmatics
	Long-term (> 24 h)	0.01	0.02	Higher frequency of days with respiratory symptoms and/or taking medications for children with asthma

4. Health effects of exposures to nitrogen dioxide

The accumulation of nitrogen oxides in poorly ventilated rooms is irritating to the respiratory tract. Much of NO₂ is not absorbed in the upper respiratory tract, but penetrates deep into the lungs, causing toxic effects.

The threshold for perceptibility of smell and irritating effects is similar and ranges from 0.23-0.41 mg/m³.

Repeated, short-term exposure to NO₂ worsens the existing respiratory tract disease and may induce the development of chronic obstructive disorders, including asthma (Kroczyńska-Bednarek 2008).

Dutch researchers have shown that hourly exposure to NO₂ already at a concentration of 0.21 mg/m³ causes an increase in bronchial hyperreactivity in patients with asthma.

Exposure to high concentrations (0.56 mg/m³) results in impaired lung ventilation and a significant increase in airway resistance. On average, an increase in NO₂ concentration of 0,10 mg/m³ causes a decrease in FVC (Forced Vital Capacity) by 15% (Ackermann-Liebrich et al. 1997).

Sensitive to NO₂ are especially children in whom exposure to this gas for several days, even in low concentrations, can increase the frequency of morning asthma symptoms. Chronic exposure can promote the development of chronic bronchitis and increase susceptibility to respiratory infections (Mortimer et al. 2002).

5. Analysis of standards related to ventilation

The probability of high NO_x concentrations in a residential area (in case of heavier load of gas equipment or malfunctioning ventilation) is quite high (WHO Guidelines 2010).

Nitrogen dioxide in the internal environment is the result of both infiltration of NO₂ from the outside air and produced by combustion sources at home. The impact of NO_x emissions from gas equipment can be minimized when the rooms are well ventilated and exhaust gases are effectively evacuated outside. However, this impact can become significant if the rooms are poorly ventilated.

The requirements concerning residential ventilation differ in particular European countries. Some of them are presented in Table 7 (Indoor air 2015).

In Poland, ventilation of apartments is provided mainly by natural systems. The exhaust is carried by gravity ventilation ducts, and the supply through disordered infiltration and airing. In this case, it is very difficult to achieve the air exchange at the recommended level.

In "Technical conditions to which buildings should correspond" (Regulation 2002), we find contradictions that additionally heighten the scale of the problem.

Table 7. Ventilation requirements for living quarters in selected EU countries

Country, standard and status	Ventilation level for the entire building	Kitchens	Bathroom + WC	WC only
Belgium (NBN D 50-001) Required	3.6 m ³ /h per m ² of area	Open kitchens, min. 75 m ³ /h (exhaust)	Min. 50 m ³ /h (upper limit 75 m ³ /h)	Min. 25 m ³ /h
Poland (PN-B- 03430:1983/AZ3:2000) Recommended to use	Min. 20 m ³ /h per person	30-70 m ³ /h (gas cookers)	50 m ³ /h	30 m ³ /h
UK (Approved Document F) Recommended to use	46.8-104.4 m ³ /h (depending on the number of rooms)	46.8-216 m ³ /h (exhaust)	28.8-54 m ³ /h (exhaust)	21.6 m ³ /h (exhaust)
EN 15251 European Standard	1.26-1.76 m ³ /h per m ² of floor surface	50.4-100.8 m ³ /h	36 -72 m ³ /h	25.2 -50.4 m ³ /h

According to §150.9 of the Regulation “*In a room with solid fuel furnaces, liquid or with gas appliances that collect air for combustion from a room and with a gravity exhaust gas duct, the use of mechanical exhaust ventilation is forbidden*”.

But according to §176.2 of the same document “*Boilers for gaseous fuels with a total thermal power of up to 30 kW can be installed in rooms not intended for permanent human stay*”.

As a result of these regulations, it is possible to install low-power gas boilers both in the bathrooms and in the kitchens. This is space where residents spend a lot of time. If we install gas devices there, then mechanical ventilation in these rooms is forbidden. In this case, the concentration of harmful substances in the room is largely dependent on the varying conditions, such as the correctness of channel selection, outside air temperature, wind power and others. Using a ventilation hood over a gas stove is contrary to the provision of § 150.9 of the Regulation.

The kitchen hood connected to the ventilation duct is undoubtedly the installation of mechanical exhaust ventilation and its simultaneous use with gravity ventilation is forbidden. However, cooker hoods are often assembled by users as specific type of the technical equipment of the building. Only the flat users

decide about the purchase and the designers have no influence on its parameters, such as flow, noise and connecting place.

Installing the hood, it is necessary to provide the air supply during its operation. Only under this condition the operation of the main ventilation system will not be disrupted. Unfortunately, for the most part, housing users do not realize the importance of this issue.

A growing problem is also the issue of the proper selection of gravity channels at the stage of the construction design. Ventilation ducts for each flat should be individually calculated by the right selection of their cross sections and the numbers, for the reference conditions, such as outdoor temperature + 12°C, internal temperature + 20°C, and wind speed 0 m/s (Polska Norma 2000).

Most often calculations are not made. Then they are usually built in brick technology with a cross section of 140×140 mm or made of ceramic hollow block with a diameter of 150 mm, without considering their height and location in the apartment. As a consequence, the ventilation air flow recommended in Polish standards is practically impossible (Opaliński & Rabczak 2003).

A separate issue is the amount of air delivered to the rooms. In the case of properly operating gravity ventilation, the volume flow rates of the extract air and infiltrating air should be equal. This is usually done through leaks in the window and door openings and airing, rarely through window air grates (Szkolenie 2014).

In heating season the dangerous concentration of hazardous substances in the residential space increases significantly. In this time the amount of airing rooms is limited due to avoid heat loss. The outside air can only get to the rooms through leaks in the windows and door joinery. However, currently used technology of tight joinery has radically changed the issue of air exchange, making leakage infiltration unworkable or dysfunctional. It should be considered that new windows without installed inlets are unable to provide the required amount of inflow air.

Please note that gas cookers are most likely to charge the air necessary for the combustion process directly from the room in which they are installed. In the case when a gas stove is installed in the room, the inflow of fresh air takes on special significance. The necessary amount of fresh air must be supplied for the fuel combustion process.

This amount of air must be additionally supplied, regardless of the air necessary for hygienic and sanitary purposes. If during combustion the natural gas, the amount of air supplied is lower than required, then harmful substances will appear in the combustion products next to carbon dioxide. These substances will be that more, than the imbalance in delivery of air for burning process will be higher.

6. Experimental research

The above analysis shows that gas cookers can be a very important source of NO_x emissions. If the kitchen hoods are absent and ventilation is functioning improperly, nitrogen oxides generated during the work of the cookers, are accumulated in the kitchen first. Then they extend to space of other rooms.

Properly assess the impact of indoor air on human health and well-being can only be based on experimental studies. In the world literature, only a limited number of such studies can be found. An example is research published by Slovak scientists (Šenitkova & Vilčeková 2009). Figure 1 presents the seasonally results of research on the concentration of nitrogen dioxide in the internal environment, depending on the type of room.

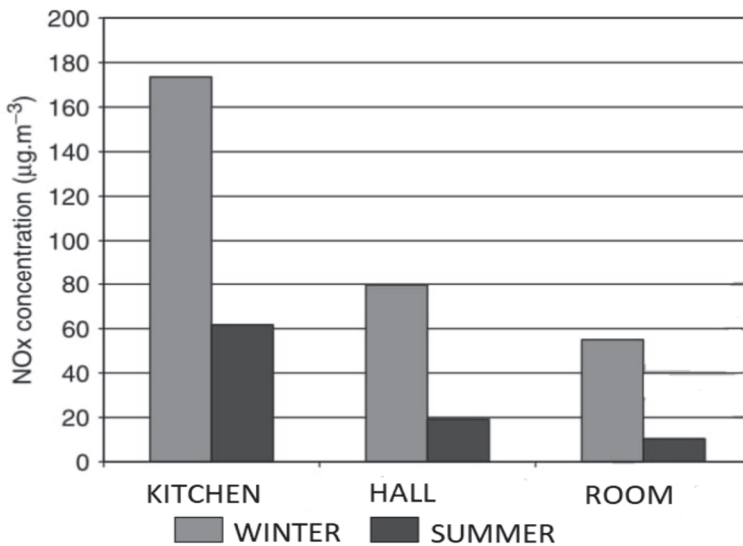


Fig. 1. NO_x concentration in the internal area, depending on the type of room and the season

As can be seen from these data, kitchens are particularly dangerous for human health. In extreme cases, differences recorded between concentrations in the kitchen and in other rooms were up to 300%.

The authors also began their own series of experimental studies. The purpose of these measurements was:

- estimation of the possible concentration of harmful substances arising during the normal use of gas equipment,
- assessment of extent of influence on a human body of harmful substances presented in the internal air;

- the development of technical and organizational measures to protect human health effectively;
- the development of recommendations enabling improvement and optimization of the standards and regulations in the examined area.

The researches of concentrations of nitrogen oxides have normalized standards. However, for the analysis of chemical contaminants generated in the tested room, for the needs of the experiment, the gas analyzer giving the measurement result in real time was used. The measurements were carried out using the certified flue gas analyser Sigma from MRU, meeting the requirements of the PN-EN 482: 2002 standard.

Measuring points were located in three-dimensional grid nodes with a spacing of 10.0 cm, in the height range from 1.50 to 1.80 m, so that the measurements included the breathing zone of an adult person. The measurements were made with closed windows and doors, under stabilized conditions. The stove was working with all the burners switched on except the oven burner. The measurement results averaged in the above-mentioned height range are depicted in Figure 2.

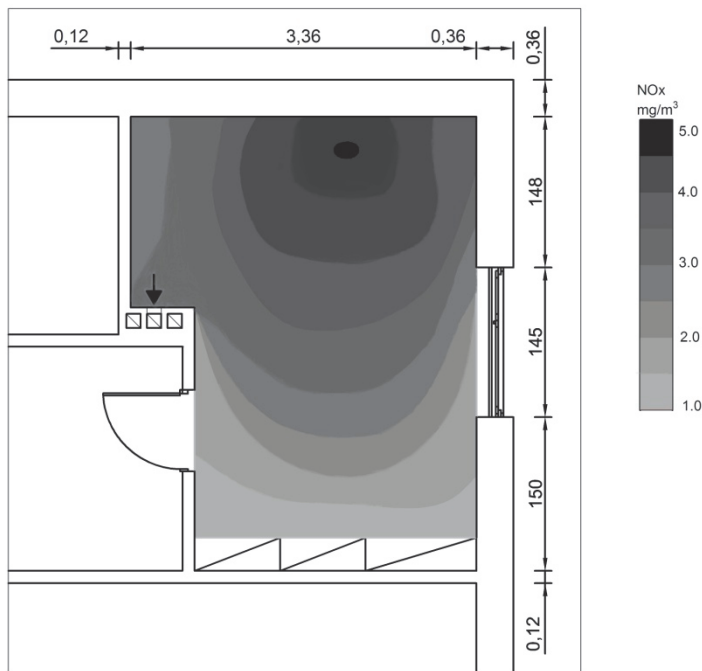


Fig. 2. The maximum concentration of NO_x in the tested room during operation of the gas equipment

Some results of this series have already been published (Szkarowski & Maliszewska 2018). The researches presented in this work were carried out under different conditions in a flat with other gas equipment installed. Room and equipment characteristics are listed in Table 8.

Table 8. Characteristics of the experimental room and equipment

Room	Kitchen			
Location	Three-storey building, first floor			
Area	13.26 m ²			
Height	2.53 m			
Volume	34.24 m ³			
Type of ventilation	Gravity ventilation duct 14x14 cm			
Indoor gas equipment				
Type of device	Four-burner gas stove with oven			
Burner type	Power, kW	Maximum gas flow, dm ³ /h	Quantity, PCs.	Maximum gas flow, dm ³ /h
Big	3.0	238.9	1	238,9
Normal	1.9	151.3	2	302,6
Auxiliary	1.0	79.6	1	79.6
Gas oven	3.0	238.9	1	238.9
Total	Maximum power, kW		Maximum gas flow, dm ³ /h	
	10.8		860.0	

The figure shows the results with the measured maximum NO_x concentration in the tested room. A total of 30 experimental studies have been carried out so far, from February to April, in average operating conditions of gas appliances. The obtained results indicate that in 70% the norms of permissible levels of nitrogen oxides in relation to work rooms were exceeded. Almost every study (98%) has exceeded the NO₂ concentration above the level of 1,0 mg/m³, which may affect the increase of bronchial hyperreactivity in patients with asthma, additionally promote the development of chronic bronchitis and increase susceptibility to respiratory infections in children.

7. Conclusions

1. Nitrogen oxides are a very dangerous for human health. The gas cookers are the main source of air pollution with nitrogen oxides in flats. Exhaust gases from cookers go directly to the living quarters, and in consequences the average exposure time of nitrogen oxides to inhabitants is several hours.
2. Polish standards, in contrast to other countries, do not specify the permission limits for NO_x in living quarters. The most similar, comparing the time of staying in a polluted atmosphere, are standards for working area.
3. The inconsistency of regulations regarding the ventilation of gas supplied rooms and the high tightness of the currently used window joinery, practically prevent effective ventilation of the rooms and removal of the generated NO_x.
4. The performed part of experimental research confirms the conclusions of the analysis. In a large part of the kitchen room, at the level of breathing, there are concentrations exceeding the standards for the working space and acceptable standards for the external environment.
5. The results of the work prove the need to introduce standards for NO_x content in the living quarters in Poland. In addition, the clarification of building regulations on ventilation would allow the use of solutions for human-friendly air quality at the design stage.

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Abstract

The gas supply of apartments, especially with the use of natural gas, is welcomed by the majority of residents. Jest to komfortowe, łatwe w użyciu i stosunkowo tanie źródło energii. The Polish Gas Development Program foresees that in 2022 to 90% of residents of the country will be provided with gas supply. However, this inevitably involves the emission of exhaust gases from gas equipments into air in apartments. Nitrogen oxides (NO_x) are considered to be the most dangerous components of combustion products for human health. They are created even during complete combustion of fuel and efficiently operating burners.

Air in apartments is a separate type of people's environment compared to the outside atmosphere and the working area. The man spends in this environment a considerable part of his life comparable with the working time. However, in standards of many countries it is not considered. This complicates the analysis and effective solution of human health protection against the effects of NO_x . The sanitary standards of several countries were analysed from this point of view (Tab. 1-6). The Polish norms define the requirements in relation to the air of the apartments, but paradoxically lack nitrogen oxides in them. Therefore, it is currently only possible to orientate on the permissible NO_x concentration in the working environment.

The second important issue is the effectiveness of ventilation, which should ensure the discharge of gases generated during the combustion of natural gas. The standards and building regulations of several countries were analysed from this point of view (Tab. 7). It was found that Polish standards for kitchen ventilation are less stringent in terms of global trends. In addition, national building regulations have contradictions that hinder the use of effective solutions at the design stage.

Additional difficulties arise from the very high tightness of the currently used window joiners. This practically prevents the supply of adequate air to the premises through infiltration, which is assumed in the standards.

In order to verify the obtained conclusions, the literature experimental data in this field were analysed (Fig. 1). The own experimental series was then carried out (Tab. 8, Fig. 2). The average operating conditions of the standard equipment have been tested. It has been proven that the concentration of NO_x in the kitchen air can significantly exceed the Polish permissible compartments for the working environment and global requirements for living quarters.

In order to ensure the protection of human beings against NO_x emitted into the air of gas-supplied housing is requested to introduce urgent changes to national sanitary standards.

The second necessary action is to eliminate inconsistencies in national building regulations in terms of kitchen ventilation. This would allow the use of effective technical solutions already at the design stage.

Keywords:

gas supply, gas equipment, health protection, nitrogen oxides, emissions, sanitary standards, building regulations, experimental research

Normatywne problemy ograniczenia stężenia tlenków azotu w środowisku przebywania ludzi

Streszczenie

Gazyfikacja mieszkań, szczególnie z użyciem gazu ziemnego, jest mile widziana przez większość mieszkańców. Jest to komfortowe, łatwe w użyciu i stosunkowo tanie źródło energii. Program rozwoju gazownictwa Polski przewiduje, że w roku 2022 do 90% ludności kraju zostanie ogarnięto zaopatrzeniem w gaz. Jednak to nieuchronnie wiąże się z emisją spalin od urządzeń gazowych do powietrza w mieszkaniach. Za najgroźniejsze dla zdrowia ludzi składniki spalin uznaje się tlenki azotu (NO_x). Powstają one nawet przy pełnym spalaniu paliwa i sprawnie działających palników.

Powietrze w mieszkaniach stanowi odrębny rodzaj otoczenia ludzi w porównaniu z atmosferą zewnętrzną i środowiskiem pracy. Człowiek spędza w tym środowisku sporą część swego życia porównywalną z czasem pracy. Jednak w normach wielu krajów tego się nie uwzględnia. Utrudnia to analizę i skuteczne rozwiązanie zagadnienia ochrony zdrowia ludzi przed działaniem NO_x . Przeanalizowano sanitarne normy kilku krajów z tego punktu widzenia (Tab. 1-6). Normy polskie definiują wymagania w stosunku do powietrza mieszkań, jednak paradoksalnie brakuje w nich tlenków azotu. Dlatego orientować się w tej sprawie można obecnie tylko na dopuszczalne stężenie NO_x w środowisku pracy.

Drugą istotną kwestią jest skuteczność wentylacji, która powinna zapewnić odprowadzanie spalin powstających w trakcie spalania gazu ziemnego. Z tego punktu widzenia przeanalizowano normy i przepisy budowlane kilku wybranych krajów (Tab. 7). Ustalono, że normy polskie w zakresie wentylacji kuchni są mniej rygorystyczne względem trendów światowych. Ponad to w krajowych przepisach budowlanych występują sprzeczności utrudniające zastosowanie efektywnych rozwiązań na etapie projektowania.

Dodatkowe utrudnienia sprawia bardzo wysoką szczelność obecnie stosowanej stolarki okiennej. Praktycznie uniemożliwia to doprowadzanie odpowiedniej ilości powietrza do pomieszczeń na drodze infiltracji zakładanej w normach.

W celu weryfikacji uzyskanych wniosków przeanalizowano literaturowe dane doświadczalne (Rys. 1) w tym zakresie. Następnie przeprowadzono własną serię eksperymentalną (Tab. 8, Rys. 2). Przetestowano przeciętne warunki eksploatacji standardowego wyposażenia. Udowodniono, że stężenie NO_x w powietrzu kuchni może znacznie przekraczać polskie dopuszczalne przedziały dla środowiska pracy i światowe wymagania w stosunku do pomieszczeń mieszkalnych.

Żeby zapewnić wymagania ochrony ludzi przed działaniem NO_x emitowanych do powietrza mieszkań zaopatrywanych w gaz wysunięto wniosek o wprowadzeniu pilnych zmian w krajowych normach sanitarnych.

Drugim koniecznym działaniem uznano wyeliminowanie niespójności w krajowych przepisach budowlanych w zakresie wentylacji kuchni. To umożliwiłoby zastosowanie efektywnych rozwiązań technicznych już na etapie projektowania.

Słowa kluczowe:

zaopatrzenie w gaz, kuchenki gazowe, ochrona zdrowia, tlenki azotu, emisja, normy sanitarne, przepisy budowlane, badania doświadczalne.