RELATIVE HUMIDITY AND TEMPERATURE IMPACT TO OZONE AND NITROGEN OXIDES REMOVAL RATE IN THE EXPERIMENTAL CHAMBER

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Abstract. The indoor ozone and nitrogen oxides concentration depends on their outdoor concentrations, indoor sources and their removal rates on the surfaces. The paper presents experimental data of the of ozone and nitrogen oxide removal in the chamber. The ambient conditions were simulated by four combination of different temperature and relative humidity levels. The impact of temperature ($T = 20\ °C$ and $T = 30\ °C$) and relative humidity ($RH = 24\%$ and $RH = 80\%$) on the deposition velocity was estimated. It was found that the ozone removal in the chamber was influenced more by temperature, and the nitrogen oxides removal was affected more by relative air humidity. The results showed that higher ambient temperature ($T = 30\ °C$) increased the ozone removal rate in the chamber 3 times on an average, and high relative humidity ($RH = 80\%$) increased the NO_x removal rate 4.5 times in comparison with the initial conditions ($T = 20\ °C$; $RH = 24\%$). The obtained deposition velocities varied in the range $(0.0045-0.0137)$ cm s^{-1} for ozone and in the range $(0.002-0.0129)$ cm s^{-1} for nitrogen dioxide.

Keywords: ozone, nitric oxides, relative humidity, temperature, chamber, deposition velocity.

1. Introduction

Indoor air pollution is a major environmental health problem affecting everyone because high levels of air pollutants can harm human health and have damaging effects on materials. Human activities itself can be significant sources of indoor air pollution. If outdoors hourly ozone (O_3) concentration usually varies about $50-60$ ppb and only sometimes the maximum hourly ozone concentrations may exceed 150 ppb in rural areas and 235 ppb in urbanized regions of Europe in summer (EEA 2010), so indoor ozone concentration can often exceed human health standards in the workplaces (Valuntaite, Girgzdiene 2008). However, in the absence of indoor sources, concentrations indoors are lower than outdoors because ozone reacts with indoor surfaces and certain gaseous contaminants (Nicolas *et al*. 2007). Ozone is an irritant that can cause coughs, chest discomfort and irritation of the nose, throat and trachea (Weschler 2006; Selin *et al*. 2009). A powerful oxidant, ozone can react with practically any biological tissue.

Nitrogen dioxide (NO_2) and nitric oxide (NO) is also the oxides of principal health concern since it is known to cause lung damage at high concentrations. Indoor nitrogen oxides can come from outdoor-to-indoor transport and/or indoor combustion processes (e.g. gas appliances, smoking, woodstoves etc.). Over 24 h period indoor nitric oxide concentrations can range from several hundred ppb to less than 1 ppb, while indoor nitrogen

dioxide levels are normally between 20 and 50 ppb, and seldom drop below 10 ppb (Weschler *et al*. 1994; Levy *et al*. 1998). Between ozone and nitrogen oxides exists the relationships. Ozone oxidizes nitric oxide to nitrogen dioxide:

$$O_3 + NO \rightarrow NO_2 + O_2. \qquad (1)$$

This reaction is very fast at 25 °C and its 2nd-order rate constant is $4.43 \cdot 10^{-4}$ ppb^{-1} s^{-1} (Atkinson *et al*. 1992). Indoor, in the absence of sunlight, this reaction is a source of NO_2, and a sink for O_3 and NO (Weschler *et al*. 1994). If ozone is present indoors and there are no indoor sources of NO, the amount of indoor NO is much smaller than that outdoors (Fuchs *et al*. 2010).

Ozone reacts with nitrogen dioxide to generate the nitrate radical:

$$O_3 + NO_2 \rightarrow NO_3 + O_2. \qquad (2)$$
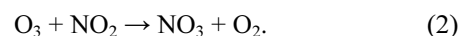
This reaction is also very fast at 25 °C, its 2nd-order rate constant is $7.87 \cdot 10^{-7}$ ppb^{-1} s^{-1} (Atkinson *et al*. 1992). Indoor, given the absence of direct sunlight, reaction (2) may be of comparable importance to outdoors at night (Weschler, Shields 1997).

There have been number of chamber studies that have investigated the mechanism and kinetics of the heterogeneous reaction between NO_2 and H_2O (Goodmann *et al*. 1999; Brink, Spolestra 1998; Kleffmann *et al*. 1998). These studies have shown that under the conditions investigated, the reaction is first order with

respect to both NO_2 and H_2O. Using a mass balance approach to study this reaction, Febo and Perrino (1991) have suggested the following stoichiometry:

$$2NO_2 + H_2O \rightarrow HONO + HNO_3. \qquad (3)$$

Under the experimental conditions of Febo and Perrino (1991) 98% of the HONO was released from the surfaces into the gas phase while all of the HNO_3 remained adsorbed on surfaces. Most reactions that occur indoors proceed faster as the temperature increases.

The deposition velocities of ozone and nitrogen oxides vary for material surfaces (Grøntoft, Raychaudhuri 2004). Grøntoft (2004) found that the deposition velocity of O_3 on a gypsum board at 30% *RH* was 0.12 cm s^{-1}, at 90% *RH* − 0.15 cm s^{-1}, while the deposition velocity of NO_2 on a gypsum board was equal to 0.025 cm s^{-1} and 0.084 cm s^{-1}, respectively. For the experimental conditions tested, the major determining factor in the NO_2 surface removal rate was the presence of the synthetic carpet. Without carpeting, the NO_2 surface removal rate ranged from − 0.07 to 0.15 h^{-1}, corresponding to deposition velocities of − 0.0004 to 0.0009 cm s^{-1}. When the carpet was introduced into the chamber, the NO_2 surface removal rate increased approximately an order of magnitude, ranging from 0.89 to 1.3 h^{-1} (deposition velocity from 0.0056 to 0.0081 cm s^{-1}) with a mean of 1.12 ± 0.13 h^{-1} (deposition velocity 0.007 ± 0.0008 cm s^{-1}). Other investigations (Wainman *et al.* 2001) founded that in the chamber NO_x deposition velocity on Teflon with carpet at 50% *RH* was 0.033 cm s^{-1}, and at 70% *RH* increase up to 0.039 cm s^{-1}; NO_x deposition velocity on wallpaper with carpet at 50% *RH* was 0.032 cm s^{-1}, and at 70% *RH* decrease up to 0.029 cm s^{-1}. Nazaroff *et al.* (1993) presented data of converse results obtained by some investigators studying relationship between ozone and NO_2 deposition velocities on the different surfaces and relative humidity.

The deposition velocities of ozone on particular material surfaces, at different temperatures and humidity, have been measured experimentally in living rooms and offices (Avol *et al.* 1998; Weschler *et al.* 1989). However in such conditions ozone is removed by reactions in the gas phase and by deposition and reaction on the different room surfaces. The influence of temperature and relative humidity on ozone deposition velocities on the numerous building surfaces in this occurrence is difficult to evaluate. The influence of the temperature on ozone and nitrogen oxides deposition velocity in the chamber practically is not investigated.

The aim of this study was to exam the influence of relative humidity and temperature to ozone and nitrogen dioxide deposition velocity in the experimental chamber made from the homogeneous material.

2. Investigation methodology

The purpose of the present study is to examine the effect of temperature and relative humidity on the decay rate of ozone and nitrogen dioxide concentration. The temperature and relative humidity ranges were taken during the experiment considering mostly observed their marginal indoor values. The different combinations of these variables were created in the experimental chamber:

   1 combination − *T* = 20 °C; *RH* = 24%;
   2 combination − *T* = 30 °C; *RH* = 24%;
   3 combination − *T* = 20 °C; *RH* = 80%;
   4 combination − *T* = 30 °C; *RH* = 80%.

The chamber size was 1.5 m lengthy, by 1.5 m width and by 1.8 m height with a volume of 4.05 m^3, a surface area of 15.3 m^2, and a surface to volume ratio (S/V) of 3.8 m^{-1}. The chamber was constructed from the polyethylene which was choosen considering that in general saturated polymers such as polyethylene are relatively inert to ozone's effects. The chamber was initially "conditioned" prior to the experiments by repeatedly exposing them to ozone in excess of 1000 ppb for several hours. This conditioning should have removed any organics on the surfaces that may have potentially reacted with NO_2. After each experiment chamber surface was rinsed with distilled water and dried.

Ozone was generated by the mercury lamp DRT 400. Maximum concentration in the empty chamber was achieved approximately after 10 minutes. Ozone concentration later varied within the limits of 5 percent. When the maximum concentration was achieved, ozone generation was stopped and its variation was measured. Ozone concentration was measured using an ozone analyzer ML9811 (Monitor Labs), which has a detection limit of 1 ppb.

The nitrogen oxides were obtained by means of the reaction between sulphuric acid and sodium nitrite. During the chemical reaction between sulphuric acid and sodium nitrite there are released nitrogen oxides into the air. 0.1 N sulphuric acid was dripped from burette into the glass with 20% aqueous solution of sodium nitrite for about 4 minutes in average speed 0.1 ml min^{-1}. Nitrogen oxides concentrations were measured using a chemiluminescent (Environnement S. A.) nitrogen oxides analyzer AC32M. Its detectable limit is 0.4 ppb. Ozone and nitrogen oxides analyzers were calibrated before experiments.

To accelerate mixing of air in the chamber, the fan was used that enabled also to maintain a stable level of temperature and relative humidity. Their sensors were placed in the chamber centrum at the 1 m height. Temperature and relative humidity were measured with sensors DD100 and DD101, respectively, of data logger DrDAQ. The resolution of temperature sensor was 0.1 °C and of relative humidity 0.2%.

Ozone and NO_x concentration and other parameters in the chamber were measured continuously by averaging data of 2 minute. Analogue signals were converted into digital by the converter ADC−16.

3. Results of experiment

In order to estimate ozone and nitrogen oxides removal in the chamber, the experiments were carried out under the different conditions of additive generation: only ozone was generated, only sodium nitrite was dripped into sulf acid, sodium nitrite was dripped into sulf acid and ozone was generated simultaneously.

Ozone removal. Ozone concentration was generated in the chamber under different conditions. Each examination of the ozone removal under different 4 combinations of temperature and relative humidity and presence or not presence of NO_x was repeated in the chamber for several times. The ozone removal profiles for a given material showed good repeatability during replicate experiments.

The first attempt was to estimate ozone removal rate under different conditions without the presence of NO_x. The combinations of conditions resulted in ozone peak concentration ranging from 200 to 270 ppb. The data were normalized to the maximum concentration, i.e, all measured concentration values where divided by maximum concentration, so all values fell within the range (0–1) and that enabled to compare obtained ozone decline curve shapes. The empirical first-order decay model was used to interpret the monitored data. The ozone removal rates were estimated and they were converted to deposition velocities v_d by dividing them by the nominal surface area to volume ratio (S/V) of 3.8 m^{-1}.

Ozone removal in the chamber after generation can be expressed as

$$dC/dt = -kC , \qquad (4)$$

where C ozone concentration (ppb), t time (s), k ozone removal rate in the chamber. The curves show the different tendencies of ozone concentration removal (s^{-1}) in the chamber. The solution of Eq. 4 is

$$C = C_0 e^{-k(t-t_0)} . \qquad (5)$$

The representative examples of the change of ozone concentration are presented in Fig. 1.

The obtained results (Fig. 1, combination 1) showed that the ozone deposition velocity at $T = 20$ °C and at $RH = 24\%$ was 0.0045 cm s^{-1}; increase of temperature up to 30 °C resulted in the higher ozone removal rate, i.e. the measured deposition velocity was 0.0134 cm s^{-1} (Fig. 1, combination 2).

Meanwhile, at $T = 20$ °C; at $RH = 80\%$, i.e. at higher relative humidity (Fig. 1, combination 3), the deposition velocity increased up to 0.0093 cm/s, and at higher temperature and at higher relative humidity ($T = 30$ °C; $RH = 80\%$) v_d was 0.0094 cm s^{-1} (Fig. 1, combination 4).
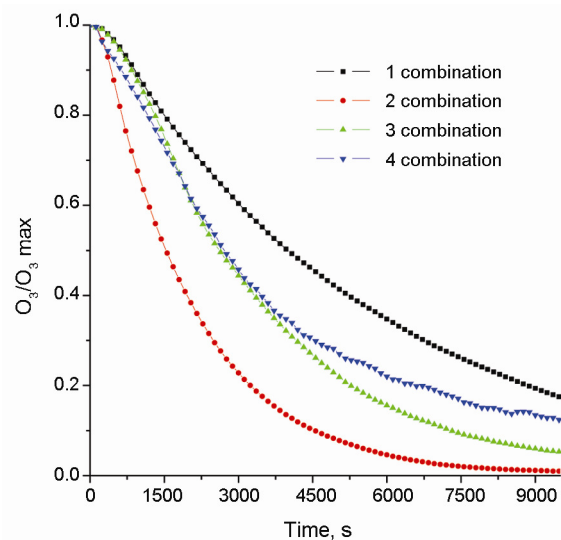

Fig. 1. The change of ozone concentration by different conditions (without the source of NO_x)

It is evident from the obtained data that the increase of temperature by 10 °C increased the ozone removal rate in the chamber almost 3 times, while the increase of relative humidity from 24% up to 80% increased the ozone removal rate 2 times. The magnitude of these effects depends on the nature of the surface. The similar tendencies were reported by other researchers (Grøntoft, Raychaudhuri 2004; Klenø *et al.* 2001).

NO_x removal. The similar investigations were made for analysis of NO_x removal in the chamber (Fig. 2). The NO_x was generated by dripping acid into nitrite. NO, NO_2 and NO_x concentration change after stopping to generate NO_x are presented in Fig. 2.

The highest exeeded NO_x concentration was about 45 ppb, NO concentration was only about 3 ppb. The removal rates and deposition velocities of NO, NO_2 and NO_x on the chamber surface were found to be 0.0031 cm s^{-1}, 0.0021 cm s^{-1}, 0.0022 cm s^{-1}, respectively. The temperature was +20 °C and RH was 24%, the NO concentration in the total quantity of NO_x was less than 10%, therefore only the total NO_x concentration was estimated during further investigations.
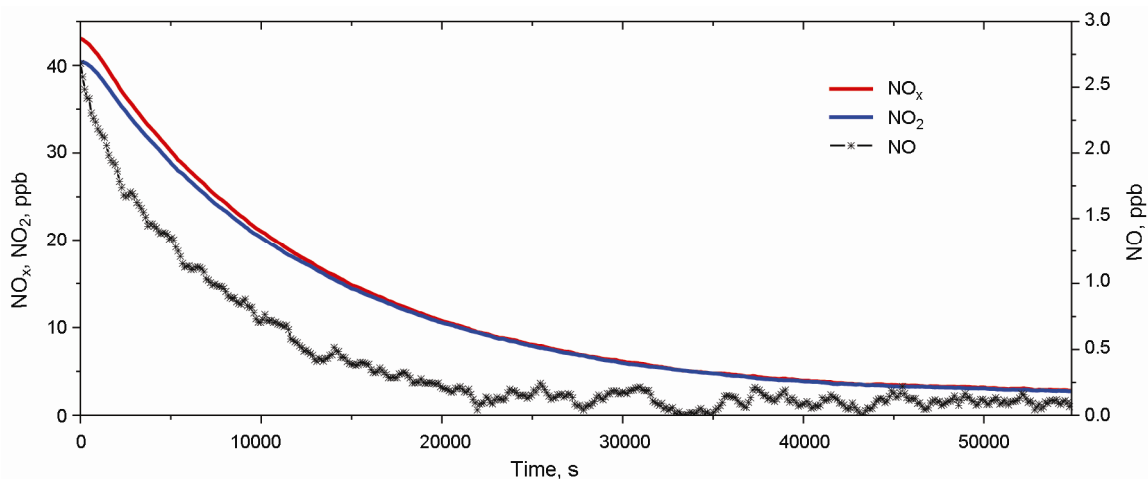

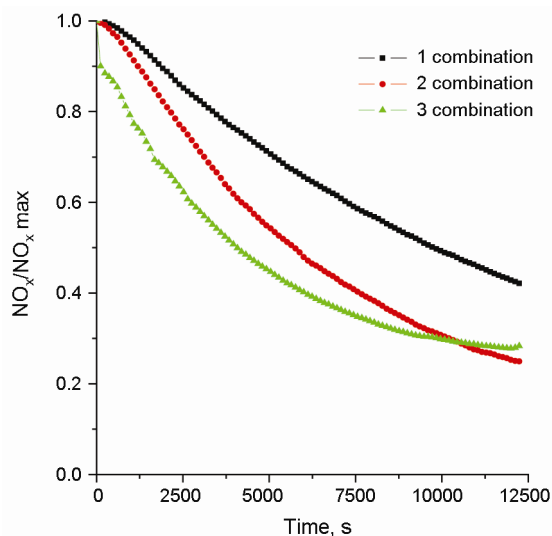**Fig. 2.** Variation of NO, NO_2 and NO_x concentrations in time ($T = 20$ °C; $RH = 24\%$)

Fig. 3. The change of NO_x concentration by different conditions: ($T = 20$ °C; $RH = 24$%), ($T = 30$ °C; $RH = 24$%) and ($T = 20$ °C; $RH = 80$%)

Experiment was continued by changing the values of temperatures and RH as in the ozone removal rate investigation cases. The obtained results are given in Fig. 3.

The obtained NO_x deposition velocity ($T = 20$ °C and $RH = 24$%) was equal to 0.0020 cm s^{-1}; at higher air temperature, but at the same RH ($T = 30$ °C and $RH = 24$%), it was 0.0037 cm s^{-1}; and at higher relative humidity ($T = 20$ °C; $RH = 80$%), it was 0.0064 cm s^{-1}. This shows that the increase of relative humidity results in the increased NO_x removal rate in comparison with the initial conditions. In the (Mainman *et al.* 2001) is mentioned that by the heterogeneous reaction of NO_2 with water vapor (H_2O) can be generated nitrous acid (HONO) and nitric acid (HNO_3). The chamber studies in which NO_2 was introduced into humidified air have shown that HONO concentrations increase in a fashion that matches the decrease in NO_2 concentrations. The heterogeneous reaction between NO_2 and H_2O can be expressed as
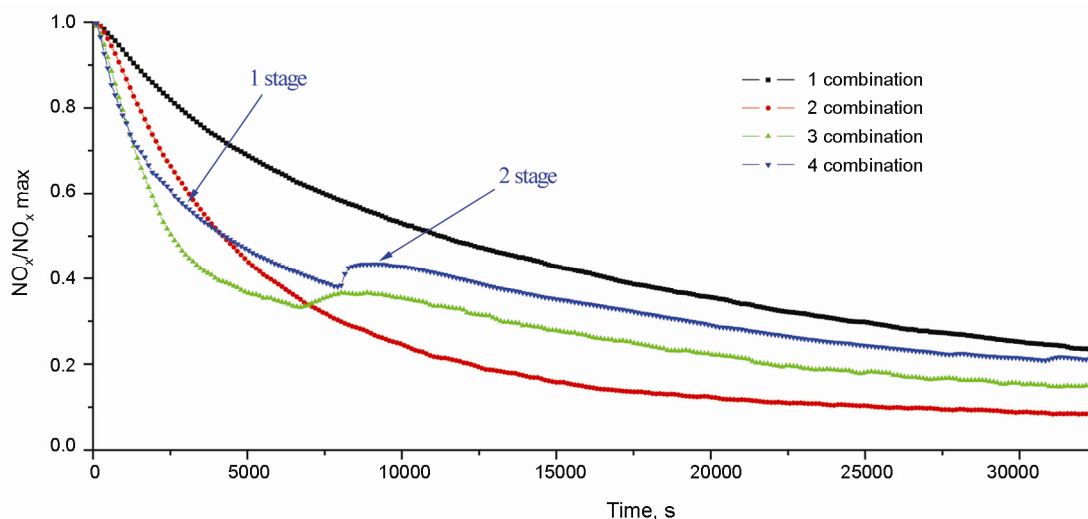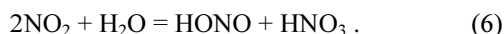
$$2NO_2 + H_2O = HONO + HNO_3 . \qquad (6)$$

Under the experimental conditions presented in (Febo, Perrino 1991), 98% of the HONO was released from the surfaces into the gas phase while all of the HNO_3 remained adsorbed on surfaces. The similar results were obtained during other investigations (Finlayson-Pitts *et al.* 2003). However, on the contrary than in the ozone case, the increase of temperature has less impact on NO_x removal rate than increased relative humidity. During our experiment neither HONO nor HNO_3 have been measured.

The ambient air almost always contains some photooxidants as ozone and nitrogen oxides. The reaction between these pollutants is the essential factor which determines air quality; therefore the experiments were carried out to estimate the ozone and NO_x removal in the chamber containing both compounds. It should be noted that in this case, in addition to deposition on surface, the NO_x and O_3 removal process also includes heterogeneous reactions between these gases in the air and other substance which result from these reactions. It was found that stopping of both ozone and NO_x generation, the NO concentration dramatically decreased within several minutes up to the level which almost did not vary during the remaining part of the experiment, i.e. up to 0.10 ± 0.05 ppb.

The total NO_x removal rate and, at the same time, the deposition velocity increased in comparison with the case without the ozone source under the certain conditions ($T = 20$ °C and $RH = 24$%) – 0.0022 cm s^{-1}. When temperature was increased up to 30°C, the deposition velocity increased up to 0.0049 cm s^{-1} (Fig. 4, combination 2). It should be noted that NO_x removal in the chamber at the increased relative humidity (in our case RH was 80%, Fig. 4 combination 3 and 4) and at certain ozone concentration is significantly more complicated. The data analysis allows to assume that in the first stage, approximately within 150 minutes from the beginning of the experiment, at $T = 20$ °C; $RH = 80$%, ozone was used (Fig. 4, combination 3) both in reactions with NO_x, and by the process of deposition on the surface. If temperature is increased up to +30 °C, this stage lasts up to 164 minutes. Further decrease of NO_x concentration dramatically slows down and deposition velocity becomes 0.0016 cm s^{-1} both at 24 °C and at 30 °C.

Fig. 4. Variation of NO_x concentration in time (with source of ozone emission)

Presence of nitrogen oxides also increases ozone removal rate in the chamber. The ozone removal process in the chamber by different T and RH combinations are presented in the Fig. 5. NO_x generation was stopped when its concentration achieved 30 ppb.

The obtained results (Fig. 5, combination 1) showed that in the chamber containing the source of both ozone and nitrogen oxides, the ozone deposition velocity at $T = 20$ °C and at $RH = 24$% was 0.0054 cm s^{-1}, and the increase of temperature by 10 °C resulted in the increased ozone removal rate, i.e. the measured deposition velocity was 0.0137 cm s^{-1} (Fig. 1, curve 2), and at higher relative humidity, i.e. at $T = 20$ °C; $RH = 80$% (Fig. 1, curve 3), the deposition velocity was 0.0117 cm s^{-1}. It is evident from the obtained results that the increase of temperature by 10 °C, and the increase of relative humidity by 56% increased the ozone removal rate in the chamber 2.5 and 2 times, respectively.

The summarized results of ozone and nitrogen oxides deposition velocities under different conditions are presented in the Table 1.
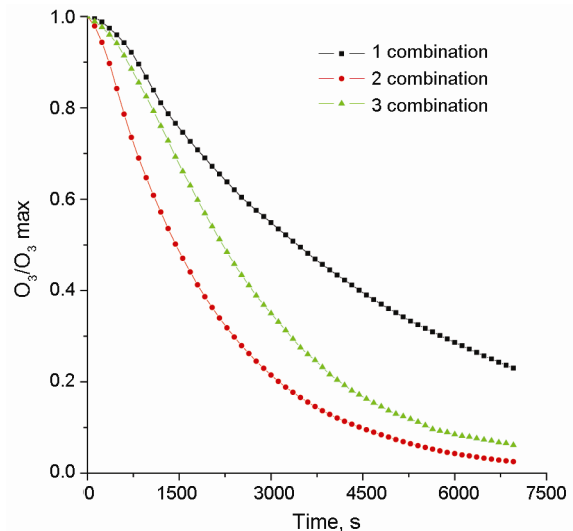

Fig. 5. The change of ozone concentration by different conditions: (1 combination − $T = 20$ °C; $RH = 24$%), (2 combination − $T = 30$ °C; $RH = 24$%) and (3 combination − $T = 20$ °C; $RH = 80$%) (with NO_x source)

Table 1. Ozone and nitrogen oxides deposition velocities

	O_3 deposition velocity, cm s^{-1}			
	Experiment condition			
	$T = 20$ °C; $RH = 24$%	$T = 30$ °C; $RH = 24$%	$T = 20$ °C; $RH = 80$%	$T = 30$ °C; $RH = 80$%
With the source of NO_x concentration	0.0054	0.0137	0.0117	−
Without the source of NO_x concentration	0.0045	0.0134	0.0093	0.0094
	NO_x deposition velocity, cm s^{-1}			
	Experiment condition			
	$T = 20$ °C; $RH = 24$%	$T = 30$ °C; $RH = 24$%	$T = 20$ °C; $RH = 80$%	$T = 30$ °C; $RH = 80$%
With the source of O_3 concentration	0.0022	0.0049	0.0129* 0.0016**	0.0095* 0.0016**
Without the source of O_3 concentration	0.0020	0.0037	0.0064	−

* first stage; ** second stage (see, Fig. 4)

The obtained data show that the removal process of the ozone and nitrogen dioxide is dependent on the ambient conditions. By change only of the temperature and relative humidity it is possible to reduce human dangerous ozone and nitrogen dioxides levels in the working and leaving rooms.

4. Conclusions

This study provides additional evidence of the influence of temperature and relative humidity on ozone and nitrogen oxides removal rate.

1. The most significant influence of microclimatic parameters on the change of ozone and nitrogen oxides concentrations in the chamber have relative humidity and temperature.

2. It was found that higher ambient temperature ($T = 30$ °C) increases the rate of ozone and NO_x removal in the chamber 3 times on an average in comparison with the initial conditions ($T = 20$ °C; $RH = 24$%).

3. The obtained results showed that high relative humidity ($RH = 80$%) increased the ozone removal rate in the chamber 2 times on an average, and the NO_x removal rate – 4.5 times in comparison with the initial conditions ($T = 20$ °C; $RH = 24$%).

4. It was noticed that the ozone removal is influenced more by temperature, and the NO_x removal is affected more by relative air humidity.

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SANTYKINIO ORO DRĖGNIO IR TEMPERATŪROS ĮTAKA OZONO IR AZOTO OKSIDŲ SUIRIMO GREIČIUI EKSPERIMENTINĖJE KAMEROJE

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S a n t r a u k a

Ozono ir azoto oksidų koncentracija patalpose priklauso nuo šių teršalų koncentracijos lauke, jų šaltinių patalpose ir šių teršalų suirimo greičio ant skirtingų paviršių. Darbe nagrinėjamas ozono ir azoto oksidų suirimas eksperimentinėje kameroje. Skirtingos mikroklimatinės sąlygos buvo sukuriamos naudojant keturių skirtingų temperatūrų ir santykinių oro drėgnių lygių derinius. Vertinta temperatūros (T = 20 °C ir T = 30 °C) ir santykinio drėgnio (RH = 24 % ir RH = 80 %) įtaka tiriamų teršalų suirimui. Nustatyta, kad ozono suirimui daugiau įtakos turėjo temperatūra, o NO_x irimui – santykinis oro drėgnis. Nustatyta, kad aukštesnė aplinkos temperatūra (T = 30 °C) paspartina ozono suirimą kameroje vidutiniškai tris kartus, o didelis santykinis drėgnis (RH = 80 %) paspartina NO_x suirimą 4,5 karto, lyginant su pradinėmis sąlygomis (T = 20 °C; RH = 24 %). Eksperimento metu ozono suirimo greitis kito nuo 0,0045 iki 0,0137 cm s^{-1}, o azoto oksidų – nuo 0,002 iki 0,0129 cm s^{-1}.

Reikšminiai žodžiai: ozonas, azoto oksidai, santykinis oro drėgnis, temperatūra, kamera, suirimo greitis.

ВЛИЯНИЕ ОТНОСИТЕЛЬНОЙ ВЛАЖНОСТИ И ТЕМПЕРАТУРЫ НА СКОРОСТЬ РАСПАДА ОЗОНА И ОКСИДОВ АЗОТА В ЭКСПЕРИМЕНТАЛЬНОЙ КАМЕРЕ

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Р е з ю м е

Концентрации озона и оксидов азота в помещениях зависят от их концентрации в открытом воздухе, источниках внутри помещений и скорости их распада на поверхностях. В статье представлены экспериментальные данные по распаду озона и оксидов азота в камере при разных микроклиматических условиях, созданных с применением четырех комбинаций температур и относительной влажности. Оценивалось воздействие температуры (T = 20 °C и T = 30 °C) и относительной влажности воздуха (RH = 24% и RH = 80%) на скорость осаждения. Было обнаружено, что на распад озона в камере большее влияние оказывает температура, а на распад оксидов азота – относительная влажность воздуха. Результаты показали, что при повышении температуры окружающей среды (T = 30 °C) увеличилась скорость распада озона в камере в 3 раза по сравнению со средней, а при высокой относительной влажности (RH = 80%) увеличилась скорость распада NO_x в 4,5 раза по сравнению с начальными условиями (T = 20 °C; RH = 24%). Полученные для озона скорости осаждения варьировали в пределах (0,0045−0,0137) см с$^{-1}$, а для оксида азота – в пределах (0,002−0,0129) см с$^{-1}$.

Ключевые слова: озон, оксиды азота, относительная влажность, температура, камера, скорости осаждения.

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