

VARIATION OF OZONE AND AEROSOL PARTICLE NUMERICAL CONCENTRATIONS ON THE WORKING PREMISES UNDER DIFFERENT MICROCLIMATIC PARAMETERS

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Abstract. Investigations of air quality on the working and residential premises as well as its effect on human health and materials have been started many years ago and are still intensively carried out now. This is a result of application of new materials in the field of construction, implementation of new technological processes as well as intensive use of various devices at home and workplaces. It is known that a high concentration of ozone and aerosols could have negative effects on human health as well as on materials. Quite frequently not quite new copying machines which might be a strong source of pollutant emission are used for copying. An experiment was carried out on the working premises with three operating copying machines and a laser printer. The changes and distribution of ozone concentration and numerical concentration of aerosol particles ($d > 0.4 \mu m$) from the source of emission, i. e. a copying machine, were investigated. Measurements were carried out under different conditions of copying work intensity and ventilation. The microclimate parameters (temperature gradients, noise level, relative humidity, light) and spectrum of aerosol particles (0.4-2.0 µm) were measured as well. A copying machine was found to be the main source of ozone and aerosol particles. Intensity of copying work largely determined dynamics of these pollutants on the office premises. The maximum ozone concentration and minimum concentration of aerosol particles were estimated during automatic copying. It was found that ozone concentrations outside could not be the main and significant source of this pollutant in the room. Positive relationship between ozone concentration and temperature gradient was found; the correlation coefficient was 0.85, and the negative one (-0.81) was between aerosol particles and temperature gradient. During experiment it was found that the pollutants under examination were mostly influenced by relative humidity and temperature gradient.

Keywords: the working premises, copying machine, ozone, aerosol particles, copying work intensity, microclimate parameters.

1. Introduction

Plenty of pollutants, chemical and physical factors influence indoor atmospheric environments where people spend more than 90% of their time (Hayes 1991; Sundel 2004). Consequently, exposure to air pollutants is often greater indoors than outdoors, even when outdoor concentrations are higher, as is typically the case with ozone (Zhao *et al.* 2007). Indoor exposures to ozone may represent a significant fraction of total exposure (Weschler *et al.* 1989). Outdoor ozone is transported indoors as a consequence of ventilation and infiltration, and indoor concentrations are often in the range of 20–70% of outdoor concentrations (Weschler and Shields 1999).

Ozone is a common indoor pollutant (Weschler *et al.* 1989). It can be produced by indoor sources including devices designed to generate ozone (Boeniger 1995), certain air cleaners (Niu *et al.* 2001; Britigan *et al.* 2006), and some printers and photocopiers (Lee *et al.* 2001). Also, it is known that the products of indoor chemistry, initiated by ozone, can be more irritating, odorous and damaging to materials than their precursors (Weschler and

Shields 1997). The concentration of indoor ozone depends on a number of factors, including the outdoor concentration, air exchange rates, indoor emission rates, surface removal rates, and reactions between ozone and other chemicals in the air (Weschler 2000).

A range of pollutants is known to be emitted from photocopiers: VOCs, ozone, formaldehyde, nitrogen dioxide and respirable particles (Brown 1999). Ozone forms during copying when photoreceptor and paper are inserted or discharged as well as when UV lamp operates during photocopying (Black and Worthan 1999). Photocopiers work demonstrates that ozone emissions can increase between periods of routine maintenance. For example, ozone emissions from five different photocopiers ranged from 16 to 131 µg/copy before maintenance (Weschler 2000). For five different dry process photocopiers, the ozone emission rate varied from 1.2 to 6.3 mg/h with an average value of 5.2 mg/h (Aoki and Tanabe 2007).

Homogeneous reactions between ozone and terpene can be a significant source of secondary organic aerosol

indoors (Weschler 2000; Sarwar et al. 2003). Some VOC emitted by different wood-based materials products (e. g. d-limonene, α -pinene) could react with ozone fast enough to generate sub-micron particles (Aoki and Tanabe 2007). The office with a limonene source had significantly higher levels of sub-micron particles when indoor ozone levels were elevated. Increase of $2 \mu g/m^3$ was observed in the mass concentration of particles with diameters less than 0.7 µm when a small amount of perfume was exposed to just 15 ppb of ozone in a ventilated chamber of 11 m³ (Sarwar et al. 2003). Some investigations of aerosol concentration variations in residential houses are presented in (Morawska et al. 2003). The average numerical and mass concentrations of particles of 2.5 µm in diameter during indoor activities were $(18.2\pm3.9)\cdot10^3$ particles/cm³ and $(15.5\pm7.9) \,\mu\text{g/m}^3$, respectively, and under non-activity conditions - $(12.4\pm2.7)\cdot10^3$ particles/cm³ $(11.1\pm2.6) \mu g/m^3$, respectively (Morawska et al. 2003).

According to (Wilson *et al.* 2000), for efficient control of health effects, however, exposures to particles from different sources, having potentially different health effects or exposure-response relationships, should be estimated separately. In the United States the National Ambient Air Quality Standard for particles is being revised to include a concentration limit for particles less than 2.5 μ m in diameter: 65 μ g/m³ in 24 h or 15 μ g/m³ as an annual average (Weschler and Shields 1999).

Indoor concentrations of ozone are reduced by deposition and decomposition/reaction on indoor surfaces. The rate of this deposition is material specific. Ozone half-life in the standard office room mostly varies between 7 and 10 minutes (Weschler 2000). According to the Lithuanian Hygiene Standard HN 35:2002 the threshold limit value of ozone concentration in workplace air is 200 μ g/m³, given the average of eight hours (HN 35:2002).

The ambient parameters as temperature, gradient of temperature, relative humidity, indoor-air velocity, and air-ventilation rate are physical factors that influence not only indoor thermal comfort but also the indoor pollutant level. The investigations of the influence of temperature on formaldehyde emission parameters from building materials and the influence of humidity on the emissions of phthalate esters from vinyl flooring are presented (Weschler and Little 2007).

An interest in the question whether ozone-induced reaction products (mainly aldehydes and sub-micron particles) may have a more important adverse effect on human health than their precursors has been recently raised (Mølhave *et al.* 2005; Weschler 2004), (Fiedler *et al.* 2005). A long-term study of the variability of ultra fine-and coarse-particle matter (PM) has been performed in a townhouse in Reston, Virginia (Wallace and How-ard-Reed 2002). The study concluded that indoor sources were responsible for 50–90% of indoor PM in non-smoking households. More important there is an indoor source if some equipment that can produce ozone is in operation. Therefore, a high concentration of ozone

and aerosol particles inside the premises is a matter of great concern.

The aim of this work was to evaluate the level and variations of ozone and aerosol particle concentration in workplaces at different ambient conditions and intensity of the copying process.

2. Investigation methodology

Investigation of ozone and aerosol particle numerical concentration variation was carried out in a room of 16 m^2 and 42.3 m^3 , where copying machines were the source of pollutant emission. A detailed description of the premises is provided in (Valuntaité and Gigždiené 2007). The maximum intensity of a copying machine was 120 copies per minute.

Ozone concentration was measured by the commercial ozone analyser O₃ 41M (O₃ 41M 1990). Operation of this analyser is based on the principle of ultraviolet absorption. The interval of ozone measuring is 0-2000 μ g/m³, and sensitivity is 1 μ g/m³. The aerosol particle measurer A3-5 was used for measurement of numerical concentration of 0.4 µm aerosol particles as well as for estimation of spectrum of the particles on the premises. The numerical concentration was measured with this measurer of particles with $d > 0.4 \mu m$. Spectrum of aerosol particles was measured every hour, and the particles with d > 0.5; 0.6; 0.7; 0.8; 0.9; 1.0; 1.1; 1.5 and 2.0 µm were recorded for 2-minute periods. Concentration of ozone and aerosol particles was measured continuously, and data were presented as 1-minute average. Data were recorded automatically in the computer.

Measurements were carried out at 40 cm distance and 50 cm height from the copying machine. Such distances were chosen in order to assess the working conditions of an operator as precisely as possible. A person working with a copying machine normally is at such distances and breathes such an air. Air on the premises was sucked through Teflon straw. An analogue signal was converted into a digital one by the exchanger ADC– 16; this helped to monitor data directly with the programs PicoLog and Microsoft Excel.

The measurement was performed at a different intensity of copying. Three copying machines and one laser printer operated in the room. Ozone emmission from the laser printer was insufficient and was mostly switched off during our investigation. Intensity of the copying process was grouped into four clusters:

- 1 -copying isn't in progress;
- 2 between copying;
- 3 from 1 to 20 copies/min.;
- 4 from 21 to 60 copies/min.;
- 5 from 61 to 120 copies/min.

Copying machines did not operate during the 1st variant, therefore, only the background of the contaminants under study was recorded. The time periods between separate copying-work stages were chosen for the second variant. At that time the ozone concentration source was turned off, and concentration declined. If the copying process had not started, the concentration of ozone reached the background level in solitary instances. Dry deposition rate of ozone on the premises was estimated as 0.035 cm/s.

The parameters of microclimate were also measured on the premises during experiment: temperature gradient, relative humidity, lighting, level of noise. There were two goals of the noise measurement: to establish what noise level is at the place where operators work and to find the main source of the noise.

DrDAQ datalogger with installed light, temperature and noise detectors was applied in order to establish microclimate parameters. Temperature was measured at two different heights on the working premises: at the ceiling and at the floor, and the temperature gradient was estimated. Measurements of relative humidity, lighting and noise were carried out at the height of a copying machine and at a distance of 1 m from a copying machine.

The investigation was carried out, and all the changes were recorded during working hours, i. e. at the same copying work intensity, copying mode (automatic or manual), changes of the premises ventilation regime, etc.

The statistical characteristics of the variation of both pollutant concentrations were evaluated. Box and Whisker charts were used to display the statistical analyses.

3. Measurement results

The measurements were carried out continuously on 8-12 January 2007. Variation of ozone concentration and aerosol numerical concentration on the working premises is provided in Fig. 1. Intensity of copying work varied during working hours. Work started at 8:00 a. m., and no activity was carried out on the premises after 7:00 p. m.

Ozone concentration varied from 1 to 330 µg/m^3 , and aerosol particles were in the interval of $(10-480) \cdot 10^6$ particles/m³ during working hours on the office premises. At night-time the concentration of ozone on the office premises varied about 3 µg/m³, i. e. ozone on the premises was decomposed, but about $30 \cdot 10^6$ particles/m³ of aerosol was found after working hours. These concentrations characterize the background of the premises. Statistical analysis of variation of these contaminants is provided in Fig. 2.

Variation of relative humidity was low during experiment: it was $26\pm2\%$ at night-time and $28\pm3\%$ during working hours; only on January 10 it was a little higher – $34\pm6\%$, as the window was opened and air from outside got into the premises. According to the data from Environmental Protection Agency, the relative humidity in Vilnius during the experiment varied from 66 to 97%, and on January 10 it was at least 87%. Temperature outside varied from 2 to 11 °C, and the wind speed was 0.7–3.0 m/s. Ozone concentrations were low and typical of winter-time. The highest ozone concentration outside was detected on January 11 when the inside ozone value reached 35 µg/m³. These data show that concentrations outside could not be the main and significant source of ozone on the premises.

Sound level during working hours did not exceed 75 dBA, and light intensity -63 lx. The detected close sound level and ozone concentration variation courses (the correlation coefficient of 0.76) indicate that the main noise source was a copying machine.

The obtained results show (Fig. 3) that increasing intensity of copying work on the premises also increases the concentration of ozone. The highest average ozone concentration of 166 μ g/m³ was detected during the work of the fifth operation mode of a copying machine. With increase of copying work intensity, higher concentrations of aerosol particles (d > 0.4 μ m) were also detected. Only at the fifth mode of operation, when the mode is automatic and the sheets are not reversed by an operator, the average numerical concentration of aerosol particles decreased on the premises. Such a dependence allows to state that the source of ozone is a copying machine, and the main source of aerosol particles is the process of copying, for example, dust of paper; mechanical formation of aerosol particles.

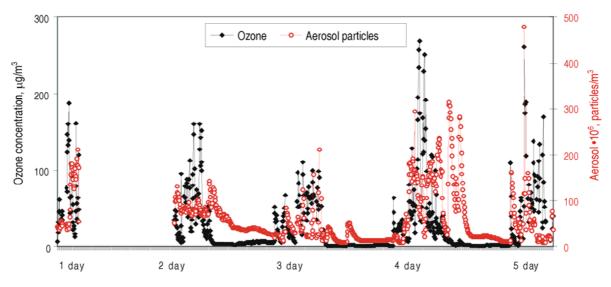


Fig. 1. Dynamics of ozone and aerosol particle concentrations in the workroom, January 8-12, 2007

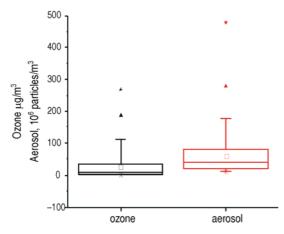


Fig. 2. Statistical analysis of ozone concentration and aerosol particle numerical concentration during experiment. The horizontal lines in the boxes denote the 25^{th} , 50^{th} , and 75^{th} percentile values. The error bars denote the 5^{th} and 95^{th} percentile values. The triangles indicate the 99^{th} percentile values and the stars denote the maximum values. The squares in the boxes denote the average of the ozone or aerosol concentrations

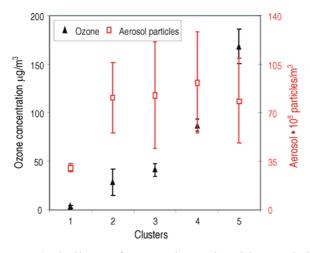


Fig. 3. Change of ozone and aerosol particle numerical concentration with increase of copying intensity

It is known (Aoki and Tanabe 2007) that aerosol particles can form when ozone affects some building materials, as the materials can emit some VOCs, for example, α -pinene and α -limonene. These chemicals react with ozone rapidly and generate sub-micron particles as secondary products.

In order to establish what size particles varied most on the working premises, a study of variation of aerosol particles spectrum was carried out on the office premises during working hours. The obtained results are provided in Fig. 4.

As it could be expected, concentration of aerosol particles of 0.4–0.5 μ m varied most during copying work, and variation of concentration of particles exceeding 1.5 μ m was low in the working room. Quantity of 0.4 μ m aerosol particles varied from 20 \cdot 10⁶ to 60 \cdot 10⁶ particles/m³, and concentration of 2.0 μ m aerosol particles distributed in the interval of (5–8) \cdot 10⁵ particles/m³ on the office premises. Concentration of aerosol particles with the size of 0.8–1.1 μ m varied by about 1 \cdot 10⁶ particles/m³.

Unhomogeneous relationship is found between ozone and aerosol particle ($d > 0.4 \mu m$) numerical concentration. Aerosol particles can be generated both by ozone leak due to heterogeneous reactions on their surface and as a result of ozone reaction with other materials under certain environmental conditions, as it was mentioned above. In the first case the relationship between ozone and particles is usually negative, and in the second case the relationship can also be positive. For example, on January 11 (Fig. 5), when the window was closed, a negative weak relationship between these two contaminants was detected (the correlation coefficient was -0.26). But a positive relationship was also detected between these contaminants (the correlation coefficient was +0.48), for example, on January 10 (Fig. 6).

The day was distinguished for a higher humidity and lower temperature in the room. It shows that there are more than one particles source in the room, and one of them dominates depending on the microclimate conditions. It should be noticed that concentration of ozone decreased to the background level very quickly after work was stopped, and higher concentrations of aerosol particles were monitored for longer periods.

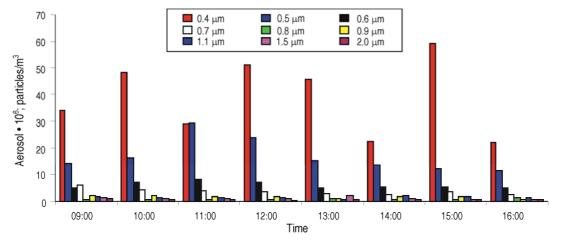


Fig. 4. Variation of the numerical concentration of aerosol particles during working hours

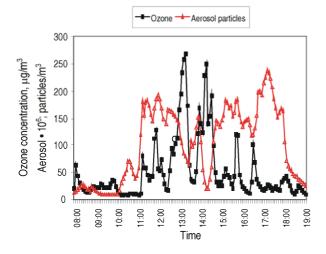


Fig. 5. Variation of ozone and aerosol numerical concentrations (without additional room ventilation)

The indoor pollutant concentration depends on the air movement in the room. The rate at which outdoor air replaces indoor air is described as the air exchange rate. When there is little infiltration, natural ventilation, or mechanical ventilation, the air exchange rate is low and pollutant levels can increase. The intesity of air movement on the premises can be correlated with temperature gradient. Therefore, the relationship between pollutant concentration and temperature gradient in a working room was investigated. The obtained data show that in the case of a higher temperature gradient the concentration of ozone in the room increased (Fig. 6). Temperature gradient on the office premises varied at an interval of 0.7-1.9 K/m, and ozone concentration varied at an interval of $2-110 \mu g/m^3$.

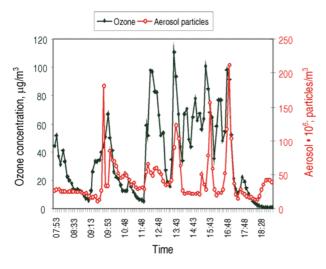


Fig. 6. Variation of ozone and aerosol numeracal concentrations (with additional room ventilation)

A positive relationship (Fig. 7) was established; the correlation coefficient was 0.85. This dependence is not linear. Partially it can be explained as follows: increase of gradient enhanced the air mixing in the room as well as transport of ozone from the source of emission to the point of measuring.

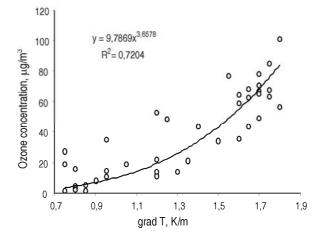


Fig. 7. Relationship between ozone concentration and temperature gradient, January 10

The investigation of relationship between concentration of aerosol particles (d > 0.4 μ m) and temperature gradient showed that there was a contrary relationship between these parameters; a negative correlation coefficient was obtained (-0.81) (Fig. 8). Air is ionized during copying. Some studies (Lee *et al.* 2004). Grabarczyk (2001) showed that the emission of air ions that charge aerosol particles could reduce the concentration of airborne dust in indoor environments. In Lee *et al.* (2004) it was found that particle mobility at high ion emission rates could enhance the deposition of particles on walls and other indoor surfaces. These observations can explain the established relationship between aerosol concentration and temperature gradient.

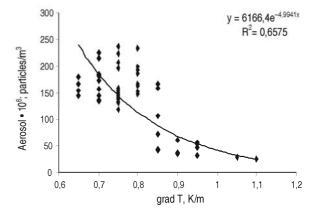


Fig. 8. Relationship between aerosol particle concentration and temperature gradient

The obtained results of the spectrum measurement showed that the numerical distribution of emitted particles during copying appeared to be unimodal and dominated the numerical concentrations of particles with sizes smaller than 0.5 μ m. The relationship between ozone concentration and different-size particles was analysed. The data showed that the relationship between ozone concentration and aerosol particles with the diameter of 0.5 μ m was negative; however, it was not linear. The best definition of this correlation was logarithmic approxima-

tion. The correlation coefficient between these variables was minus 0.75. It will be observed that in the case of low ozone concentrations ($<60 \ \mu g/m^3$), this relationship is not elastic, and only when ozone concentration is higher than 60 $\ \mu g/m^3$, this relationship becomes stronger. This shows that in the case of high ozone concentrations (the case of intensive copying work), particle deposition on the surface was prevailing comparing with their emission. A positive relationship between the particles of 2.0 $\ \mu m$ and ozone concentration was detected, the correlation coefficient was 0.49. It was not strong, though this dependence might be defined by logarithmic approximation.

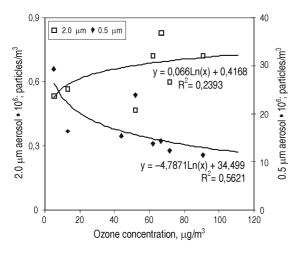


Fig. 9. Relationship between aerosol particles of different size and ozone concentration

4. Conclusions

1. The maximum ozone concentration and the minimum of aerosol particle numerical concentration were determined in the case of automatic copying. The background ozone concentration on the office premises was approximately 3 μ g/m³, and that of aerosol was approximately 30 \cdot 10⁶ particles/m³. After switch-on of a copying machine, the ozone concentration on the premises varied from 330 μ g/m³, and the aerosol – (10–480) \cdot 10⁶ particles/m³.

3. It was found that the ozone concentration outside could not be the main or significant source of ozone on the premises during the experiment.

4. Among microclimatic parameters the most significant influence on the change of ozone and aerosol particle concentrations on the premises is exerted by relative humidity and temperature gradient.

5. The numerical concentration of aerosol of 0.4–0.5 μ m varied more significantly, i. e. (11–59) \cdot 10⁶ particles/m³ to compare with aerosol larger than 1.5 μ m, i. e. (0.5–1.8) \cdot 10⁶ particles/m³.

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OZONO KONCENTRACIJOS IR AEROZOLIO DALELIŲ SKAITINĖS KONCENTRACIJOS DARBO PATALPOSE ESANT SKIRTINGIEMS APLINKOS RODIKLIAMS

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Santrauka

Darbo ir gyvenamųjų patalpų oro kokybės, jo poveikio žmogaus sveikatai ir medžiagoms tyrimai, pradėti pasaulyje prieš daugelį metų, yra intensyviai tęsiami ir dabar. Tai yra susiję su naujų medžiagų naudojimu statybose, naujų technologinių procesų diegimu ir intensyviu įvairių prietaisų naudojimu buityje ir darbo vietose. Žinoma, kad didelės ozono ir aerozolių koncentracijos gali pakenkti žmonių sveikatai, turėti neigiamos įtakos medžiagoms. Gana dažnai yra naudojami ne visai nauji kopijavimo aparatai, kurie gali būti stiprūs teršalų emisijos šaltiniai.

Eksperimentas buvo atliekamas darbo patalpoje, kurioje veikė trys kopijavimo aparatai ir lazerinis spausdintuvas. Buvo tiriama ozono koncentracijos ir aerozolio dalelių ($d > 0,4 \mu m$) skaitinės koncentracijos pokyčiai, jų sklaida nuo emisijos šaltinio – kopijavimo aparato. Matavimai atlikti esant skirtingam kopijavimo intensyvumui ir skirtingoms ventiliacijos sąlygoms. Lygiagrečiai matuoti aplinkos rodikliai (temperatūros gradientas, triukšmo lygis, santykinė drėgmė, apšvietimas) ir aerozolio dalelių ($0,4-2,0 \mu m$) spektras. Nustatyta, kad pagrindinis ozono ir aerozolio dalelių šaltinis buvo kopijavimo aparatas. Didelės įtakos šių teršalų dinamikai biuro patalpose turėjo kopijavimo intensyvumas. Didžiausia ozono ir mažiausia aerozolio dalelių koncentracija nustatyta esant automatiniam kopijavimui. Kopijavimo aparatų darbo metu patalpoje nustatyta vidutinė ozono koncentracija 60 µg/m³, o aerozolio dalelių – 78 · 10⁶ vnt/m³. Nustatyta, kad ozono koncentracijos lauke negalėjo būti pagrindinis arba svarbus šio teršalo patalpoje šaltinis. Automatinio kopijavimo metu nustatyta maksimali vidutinė ozono koncentracija 166 µg/m³. Nustatytas teigiamas ryšys tarp ozono koncentracijos ir temperatūros gradiento, koreliacijos koeficientas – 0,85, tačiau tarp aerozolio dalelių ir temperatūros gradiento gautas neigiamas koreliacijos koeficientas (-0,81). Darbo patalpoje labiausiai kito 0,4 $-0,5 \mu$ m aerozolio dalelė, o didesnių kaip 1,5 µm dalelių kiekis buvo mažiausias. Tiriant aplinkos rodiklių poveikį ozono ir aerozolio dalelių sklaidai patalpoje gauta, kad tiriamiems teršalams didžiausios įtakos turėjo santykinė drėgmė ir temperatūros gradientas.

Reikšminiai žodžiai: darbo patalpos, kopijavimo aparatas, ozonas, aerozolio dalelės, kopijavimo intensyvumas, aplinkos rodikliai.

ИЗМЕНЕНИЕ КОНЦЕНТРАЦИИ ОЗОНА И СЧЕТНОЙ КОНЦЕНТРАЦИИ ЧАСТИЦ АЭРОЗОЛЯ В РАБОЧИХ ПОМЕЩЕНИЯХ ПРИ РАЗЛИЧНЫХ МИКРОКЛИМАТИЧЕСКИХ ПАРАМЕТРАХ

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Резюме

Исследования качества воздуха рабочих и жилых помещений, а также влияния воздуха на здоровье человека и материалы, издавна проводившиеся в различных странах мира, интенсивно проводятся и в настоящее время. Это связано с использованием новых материалов в строительстве, внедрением новых технологических процессов и интенсивным использованием различных приборов в быту и на рабочих местах. Известно, что большая концентрация озона и аэрозолей может наносить вред здоровью людей и оказывать отрицательное воздействие на материалы. Зачастую для копирования используются не новые копировальные аппараты, которые могут являться сильными источниками эмиссии загрязнений.

Эксперимент проводился в рабочем помещении, в котором работали три копировальных аппарата и лазерный принтер. Исследовалось изменение концентрации озона и счетной концентрации аэрозольных частиц (d > 0,4 мкм), а также его рассеяния от источника эмиссии – копировального аппарата. Измерения проводились при разной интенсивности копирования и разных условиях вентиляции. Параллельно проводились измерения параметров окружающей среды (градиент температуры, уровень шума, относительная влажность, освещение) и спектра аэрозольных частиц (0,4-2,0 мкм). Установлено, что основным источником озона и аэрозольных частиц являлся копировальный аппарат. Интенсивность копирования в большей части обусловила динамику этих загрязнений в офисном помещении. Наибольшая концентрация озона и наименьшая концентрация аэрозольных частиц установлена при автоматическом копировании. Во время работы копировальных аппаратов в рабочем помещении установлена средняя концентрация озона – 60 мкг/м³, а аэрозольных частиц – $78 \cdot 10^6$ ед./м³. Установлено, что концентрация озона вне помещения не могла быть основным либо важным источником загрязнения в помещении. Во время автоматического копирования установлена максимальная средняя концентрация озона – 166 мкг/м³. Установлена положительная связь между концентрацией озона и градиентом температуры, коэффициент корреляции составил 0,85, однако между аэрозольными частицами и градиентом температуры получен отрицательный коэффициент корреляции (-0,81). В рабочем помещении наибольшим изменениям подверглись аэрозольные частицы размером 0,4–0,5 мкм, а количество частиц размером свыше 0,5 мкм было наименьшим. При исследовании воздействия параметров среды на рассеяние озона и аэрозольных частиц установлено, что наибольшее влияние на исследуемое загрязнение оказывали относительная влажность и градиент температуры.

Ключевые слова: рабочее помещение, копировальный аппарат, озон, аэрозольные частицы, интенсивность копирования, параметры среды.

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