



## Outdoor and indoor ozone level – A potential impact on human health

Nivo ozona u otvorenom i zatvorenom prostoru – mogući uticaj na zdravlje ljudi

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### Abstract

**Background/Aim.** Air pollution outside and inside is still one of the most sensitive issues. The aim of this study was to assess the ozone level in ambient air and working premises in terms of its possible influence on human health. **Methods.** The study was based on the results obtained in Lithuanian conditions. Continuous ozone measurement data from the rural monitoring station in Preila over the period 1995–2011 were analyzed. More than 180,000 hourly values were examined according to the requirements in the Directive 2008/50/EC. The World Health Organization (WHO) and European Union indicators the Sum of Ozone Means Over 35 ppb (SOMO 35), the maximum daily 8-hour mean concentration of ozone higher than 100 and 120  $\mu\text{g}/\text{m}^3$  were estimated. Indoor ozone concentrations in copying and welding rooms were evaluated. The ozone concentration was measured with the ozone analyzer O<sub>3</sub>41M. **Results.** The frequency distribution of ozone hourly concentrations at the Preila station showed that less than 1% of the data were higher than 120  $\mu\text{g}/\text{m}^3$  and 6% of them higher than 100  $\mu\text{g}/\text{m}^3$ , that could have the adverse effect on human health, during 1995–2011. The investigations made in working premises showed that near a copying machine the ozone concentration can reach 330  $\mu\text{g}/\text{m}^3$ , however in the room, i.e. 0.5 m from the machine, the average ozone concentration during automatic copying was 165  $\mu\text{g}/\text{m}^3$  and during manual copying it was 50  $\mu\text{g}/\text{m}^3$ . Measurements in a welding room showed that the ozone concentration was in the range of 380–1,850  $\mu\text{g}/\text{m}^3$  at the distance of 25 cm from the electrode and at the distance of 1 m from the source the ozone concentration decreased 2.5 times. **Conclusion.** The danger of the ambient ozone level to human health practically was not observed in Lithuanian conditions. However, almost 6% of the data exceed the new WHO guideline of 100  $\mu\text{g}/\text{m}^3$  during the measurement time. Indoor ozone during welding reached a higher level than during copying that can cause human health problems.

### Key words:

ozone; air pollution; air pollution, indoor; welding; health; risk assessment.

### Apstrakt

**Uvod/Cilj.** Zagađenje vazduha u otvorenom i zatvorenom prostoru i danas je jedno od najosetljivijih pitanja. Cilj ove studije bio je da se proceni nivo ozona u vazduhu i radnim prostorijama u pogledu moguceg uticaja na ljudsko zdravlje. **Metode.** Studija se zasnivala na rezultatima dobijenim u litvanskim uslovima. Analizirani su podaci kontinuiranog merenja ozona iz ruralnog monitoring centra u mestu Preila u periodu 1995–2011. Više od 180 000-časovnih vrednosti ispitane su u skladu sa zahtevima iz Direktive 2008/50/EC. Procenjivani su pokazatelji Svetske zdravstvene organizacije i Evropske unije *the Sum of Ozone Means Over 35 ppb* (SOMO 35), maksimalna dnevna 8-časovna srednja koncentracija ozona viša od 100 i 120  $\mu\text{g}/\text{m}^3$ . Takođe, procenjivane su koncentracije ozona u zatvorenom prostoru gde su vršeni kopiranje i zavarivanje. Koncentracija ozona merena je analizatorom ozona O<sub>3</sub>41M. **Rezultati.** Distribucija učestalosti koncentracije ozona po satima na Preila stanici pokazala je da je manje od 1% izmerenih vrednosti bilo više od 120  $\mu\text{g}/\text{m}^3$  i 6% izmerenih vrednosti 100  $\mu\text{g}/\text{m}^3$ , što bi moglo imati negativan uticaj na ljudsko zdravlje tokom 1995–2011. Istraživanja rađena u radnim prostorijama pokazala su da pored aparata za kopiranje koncentracija ozona može da dostigne 330  $\mu\text{g}/\text{m}^3$ . Međutim, u sobi, na 0,5 m od aparata, prosečna koncentracija ozona tokom automatskog kopiranja bila je 165  $\mu\text{g}/\text{m}^3$ , a tokom ručnog kopiranja 50  $\mu\text{g}/\text{m}^3$ . Merenja u sobi gde je vršeno zavarivanje pokazala su da je koncentracija ozona bila u opsegu 380–1 850  $\mu\text{g}/\text{m}^3$  na rastojanju od 25 cm od elektrode, a na rastojanju od 1 m od izvora koncentracija ozona bila je snižena 2,5 puta. **Zaključak.** Opasnost od ambijentalnog nivoa ozona za ljudsko zdravlje praktično nije uočena u litvanskim uslovima. Međutim, skoro 6% od izmerenih vrednosti prelazi novu smernica Svetske zdravstvene organizacije od 100  $\mu\text{g}/\text{m}^3$  tokom vremena merenja. Nivo ozona u zatvorenom prostoru za vreme zavarivanja dostizao je višu vrednost nego tokom kopiranja, što može biti štetno po zdravlje ljudi.

### Ključne reči:

ozon; vazduh, zagađenje; vazduh, zagađenje u zatvorenom prostoru; zavarivanje; zdravlje; rizik, procena.

## Introduction

Air pollution is a major environmental health problem affecting many peoples in the world. The pollutants can be emitted from various sources into the boundary level of atmosphere. The large problem is also air pollution in the indoor environment where people spend the largest part of time.

Ozone is the one of the main pollutants the level of which is under regulations of many organizations. It is a secondary pollutant and mostly is formed through photochemical reactions appearing in areas with a high traffic density during warm periods of the year. The average ozone concentrations can be higher during spring and summer months, thus increasing the potential toxicity and harmful effect on human health<sup>1</sup>.

The maximum daily 8-hour mean concentration, determined from 8-hour running averages over the day, is the parameter on which the European Union (EU) ozone long-term and target value for the protection of human health is based. EU in the Directive 2008/50/EC set the long-term objective for the protection of human health (daily maximum 8-hour average concentrations of  $120 \mu\text{g}/\text{m}^3$ ) that should not exceed  $120 \mu\text{g}/\text{m}^3$  on more than 25 days *per* calendar year averaged over 3 years. The target value was exceeded in approximately 22% of the area of 27 EU member states territory and affected approximately 16% of the total population in this territory in 2010<sup>2</sup>. As it was pointed in the report the situation in 2010 was not fundamentally different but in contrast to the last three summers there were some exceedances in northern Europe and the average number of threshold exceedances increased slightly in northwestern, central and eastern Europe.

The World Health Organization (WHO) declares stricter requirements, reduced guideline for ozone from the existing level of  $120 \mu\text{g}/\text{m}^3$  to  $100 \mu\text{g}/\text{m}^3$ , since the latest studies have shown health effects at ozone concentrations below  $120 \mu\text{g}/\text{m}^3$  but without clear evidence of a threshold<sup>3</sup>. This concentration should provide adequate protection of public health, though some health effects in some sensitive individuals may occur below this level.

The WHO recommended another indicator for human health impact – the Sum of Ozone Means Over 35 ppb (SOMO 35). SOMO 35 is an indicator of the accumulated ozone concentration in excess of 35 ppb ( $70 \mu\text{g}/\text{m}^3$ ) during the whole year. The decision to select this value was based on some arguments and one of them told that a statistically significant increase in mortality risk estimates was observed at ozone concentrations above  $50\text{--}70 \mu\text{g}/\text{m}^3$ . The largest contributions to SOMO 35 are made during the summer months, although some contributions can also be during colder months as March or October.

People spend about 90% time indoors where they receive most of their exposure to pollutants<sup>4</sup>. Investigation results relating high outdoor concentrations of pollutants with human health effects can be inaccurate due to the impact of indoor exposure. Typical indoor ozone concentrations vary between  $2\text{--}20 \mu\text{g}/\text{m}^3$ . Exposure to air pollutants is strongly

influenced by microenvironmental phenomena. For example, at an air exchange rate of 2/h, the indoor ozone concentration is about 1/3 of the outdoor ozone concentration due to indoor reactions<sup>5</sup>. These reactions increase indoor concentrations of products including formaldehyde, other carbonyl species, aerosols, and carboxylic acids. Therefore, personal exposure to ozone decreases while exposure to these byproducts increases<sup>6</sup>.

Not only many materials but also some technological processes can be a source of pollutant emission as well. The old copying machines, which produce higher pollutant concentrations compared to the new design copying machines, are often used in the offices of developing or poor countries. However, the increase of indoor pollutant emissions due to the abundance of new sources (e.g., ozone generators, electrostatic air filters, laser printers and some technological processes) is also evident<sup>7,8</sup>. The threshold value of pollutants in workplace air can vary depending on the regulations and guidelines in different countries; e.g., the ozone threshold value of 100 or  $200 \mu\text{g}/\text{m}^3$  is typically set. According to the Lithuanian hygiene standard HN 23 : 2007 the threshold limit value of ozone concentration in workplace air is  $200 \mu\text{g}/\text{m}^3$ , given the average of eight hours, and the maximum permissible ozone concentration, given the average of 5–10 minutes, is  $600 \mu\text{g}/\text{m}^3$ . Considering the fact that exposure occurs both indoors and outdoors for all the pollutants it is reasonable to propose using the same air quality guidelines for both indoor and outdoor exposures<sup>2</sup>.

Unceasing interest in atmospheric ozone is great mainly because of its effect on human health and the role in climatic change. Inhaled ozone can aggravate chronic diseases such as emphysema, bronchitis and asthma. Studies indicate that various pulmonary function parameters can be reduced by exposures to ozone concentrations in the range of  $160\text{--}360 \mu\text{g}/\text{m}^3$  for a period of 1–8 hours<sup>2</sup>. Ozone concentration level in Lithuania is lower in comparison to southern and western part of Europe<sup>9–13</sup>. The hourly mean of ozone concentration higher than  $120 \mu\text{g}/\text{m}^3$  was observed rarely. An increase in the 8-hour average ozone concentration by  $100 \mu\text{g}/\text{m}^3$  is expected to induce the 25% increase in symptom exacerbation among adults and asthmatics involved in normal activities as well as the 10% increase in hospital admissions for respiratory conditions<sup>9</sup>.

Weschler<sup>13</sup> argues that mortality and morbidity associated with ambient ozone may be due, in part, to exposure to indoor ozone and its byproducts. Based on data from US urban communities during a 14-year period<sup>2</sup>, it is estimated a 0.52% increase in daily mortality for a  $20 \mu\text{g}/\text{m}^3$  increase in the previous week's local ambient ozone level.

The aim of this study was to evaluate the level and variations of ozone concentration in Lithuanian ambient air and different workplaces from the viewpoint of the potential influence on human health.

## Methods

Ozone concentrations were monitored by using a commercial UV ozone analyzer O341M (Environment s.a.). The

lower detection level of the instrument is  $2 \mu\text{g}/\text{m}^3$  and the concentration measurement range is  $0\text{--}2000 \mu\text{g}/\text{m}^3$ . Concentration of ozone was measured continuously and obtained data were saved according to experiment requirements as one minute or one hour averages. The air was sucked through the Teflon lines and the sample flow rate was  $1.5 \text{ L}/\text{min}$ .

The hourly ozone data from the Preila rural monitoring station ( $55^\circ 55'$  North and  $21^\circ 00'$  East) over the period 1995–2011 were used in the analysis. More than 180,000 hourly values were examined. The criteria, according to the requirements in the Directive 2008/50/EC, were used for checking validity when aggregation of data and calculation of statistical parameters were performed. Continuous outdoor ozone measurements were hourly averaged when valid data were collected over less than 75% of the time (i.e. 45 minutes). The applied frequency distribution for hourly ozone data showed that the distribution has a Gaussian distribution shape during the analysed period. Trends in annual ozone concentration means were tested statistically using Mann-Kendall analysis. The Mann-Kendall test is a non-parametric test that has the advantage of robustness against outliers and can be applied to non-normally distributed data with missing values. Sen's non-parametric method was used to estimate the slope of an existing trend.

The indicator SOMO 35 was calculated according to recommendations by using the equation (1) <sup>12</sup>.

$$\text{SOMO } 35_{\text{measured}} = \sum_i \max(0, (C_i - 70)) \quad (1)$$

A correction to full time coverage has been applied:

$$\text{SOMO } 35_{\text{estimate}} = (\text{SOMO } 35_{\text{measured}} \cdot N_{\text{period}}) / N_{\text{valid}} \quad (2)$$

where  $N_{\text{valid}}$  is the number of valid daily values and  $N_{\text{period}}$  the number of days *per* year. The corrected values of SOMO 35 are calculated and used when sufficient valid daily measurements are available. For practical reasons, data coverage of at least 75% is required (i.e.  $N_{\text{valid}} > 273$ ) and the days with missing data should not concentrate in one season.

The indoor ozone investigation was carried out in the working room without sources of ozone as well as in the premises where ozone is emitted during the technological processes (i.e. copying, welding). The standard office room was selected for measurement of inflow of air with ozone from outdoors to indoors. Electric devices emitting ozone during their operation (i.e. laser printer, copying machine, etc.) were not in operation during the experiment. The area of a window was  $1.44 \text{ m}^2$ . Ozone concentration was measured at the points located at different distances from the window (0, 1, 2, 3, 4, 5 m) and at a different height (0, 0.9, 2.8 m) one minute after opening the window.

The ozone concentration was measured in a room of  $16 \text{ m}^2$ , where the source of emission was a copying machine. The maximum intensity of the copying machine was 120 copies per min. The indoor ozone concentration was investigated at a different height (0.1; 1.0; 2.5 m) and at a particular distance from the copying machine (0.1; 0.5; and 1.5 m).

Horizontal and vertical distribution of ozone concentration was investigated in a welding room of  $95 \text{ m}^2$ . Ozone emission from the welding machine was measured at a different distance from the electrode (0.25, 0.5 and 1 m) and at a different height of the room (0, 0.25, 1.5 m). Welding was carried out with the electrode of 4 mm diameter.

## Results and Discussion

Ground-level ozone monitoring in Lithuania has been performed since 1982 <sup>10</sup> and the upward trend of annual mean concentration was established, however the ozone level danger for human health was practically not observed. The frequency distribution of the ozone hourly concentrations at the Preila station showed that less than 1% of the data were higher than  $120 \mu\text{g}/\text{m}^3$  and 6% of data higher than  $100 \mu\text{g}/\text{m}^3$  during 1995–2011 (Figure 1).

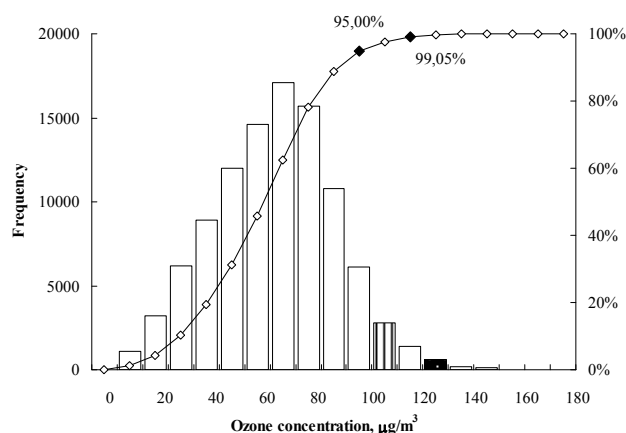


Fig. 1 – The frequency distribution of hourly ozone concentrations, Preila, 1995–2011.

The results of SOMO 35 calculated and corrected according to equation (2) and the number of days when 8-hour mean was more than 100 and  $120 \mu\text{g}/\text{m}^3$  are presented in Figure 2. Data showed that values of these indicators can differ significantly from year to year.

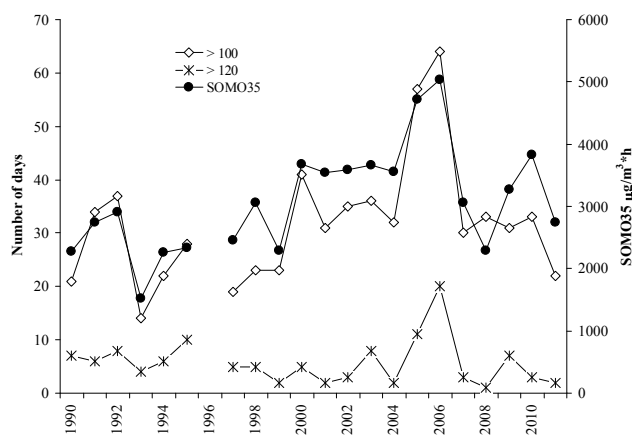


Fig. 2 – The variation of the sum of ozone means over 35 ppb (SOMO 35) and the number of days with ozone concentration  $> 120 \mu\text{g}/\text{m}^3$  and  $> 100 \mu\text{g}/\text{m}^3$ .

The ozone inflow through the open window into the room was measured in summer when outside concentration reached  $120 \mu\text{g}/\text{m}^3$ . Ozone concentration in the office room was equal to zero before opening the window. The ozone concentrations were measured at different distances from the window. The outdoor temperature was  $25^\circ\text{C}$ , and the indoor temperature was lower, i.e.  $22^\circ\text{C}$ .

The obtained results showed that the ozone concentration varied significantly at a distance up to 2 m from the window. Further distribution of ozone in the room was almost uniform, and equal to  $54 \pm 4 \mu\text{g}/\text{m}^3$ , i.e., ozone concentration was almost two times lower than the ozone concentration measured at the initial moment (i.e. outdoors).

Ozone that enters the office room from outdoors completely decays within 1.5 h. During the experiment, half of the total quantity of ozone in the office room decayed within 7 minutes. The obtained results coincide with the results of other authors<sup>13</sup>. It was established that the ratio of indoor ozone concentration ( $75 \pm 5 \mu\text{g}/\text{m}^3$ ) and outdoor ozone concentration ( $120 \pm 11 \mu\text{g}/\text{m}^3$ ) was equal to  $0.63 \pm 0.04$ .

The contribution of ozone emitted during technological processes (copying, welding) to the ozone concentration level in the working premises was estimated. The measurements of vertical dispersion of ozone emitted from the copying machine were performed near the floor, at the height of the copying machine (1 m) and near the ceiling (2.5 m). A variation of horizontal ozone concentration was established at the following distances from the source: 0, 0.5, 1 and 1.5 m (Figure 3).

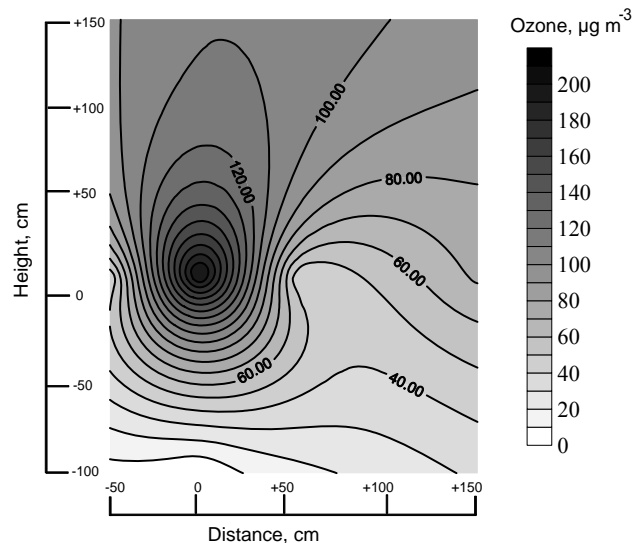


Fig. 3 – Spatial dispersion of ozone concentration in a copying room.

The ozone concentration in the copying room was about  $4 \mu\text{g}/\text{m}^3$  at night, whereas during operation of copying machines it varied from 10 to  $330 \mu\text{g}/\text{m}^3$ . The copying process mostly was not intensive (from 1 to 20 copies/min) during the experiment, thus the measured ozone concentrations ranged from 30 to  $50 \mu\text{g}/\text{m}^3$ . With the increase in intensity of copying process, the indoor ozone concentration increased.

The maximum ozone concentration was measured during automatic copying (the average concentration was  $165 \mu\text{g}/\text{m}^3$ ) while during manual copying the measured ozone concentration was lower (the average concentration was  $50 \mu\text{g}/\text{m}^3$ ). Therefore, it may be stated that the intensity of copying process has a major impact on the ozone concentration level in office premises.

The maximum ozone concentration ( $330 \mu\text{g}/\text{m}^3$ ) was measured at the height of the copying machine. The ozone emitted by the copying machine is rising towards the ceiling. The surface ozone concentration near the floor varied from 7 to  $20 \mu\text{g}/\text{m}^3$ , and near the ceiling it varied from 10 to  $110 \mu\text{g}/\text{m}^3$ . The results of the ozone concentration distribution at different distances from the copying machine are presented in Figure 3.

Welding is another technological process when high ozone concentration can occur. Ozone can be produced by ultraviolet light from the welding arc, and also in greater quantities by gas metal arc welding (GMAW) or short-arc, gas tungsten arc welding (GTAW) or heli-arc, and plasma arc cutting. The exposure to ozone generated in GMAW and plasma arc welding may produce excessive mucus secretion, headache, lethargy, eye irritation and irritation and inflammation of the respiratory tract. In extreme cases, excess fluid and even hemorrhage may occur in the lungs. The irritant effects of gas on the upper respiratory tract and the lungs can be detected with some delay.

The investigations of ozone distribution during welding showed a very high ozone level (Figure 4) in the room. Due

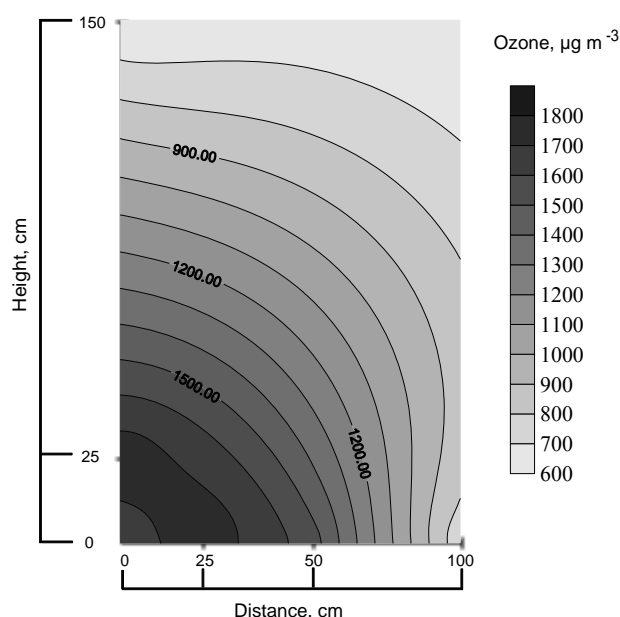


Fig. 4 – Spatial dispersion of ozone concentration in a welding room.

to intensive turbulent flow, the ozone concentration near the welding instrument just after its formation is heterogeneous, therefore large variations of concentrations were observed. The obtained results showed that the ozone concentration decreases with increase of the distance from the electrode. The

maximum ozone concentration was found at the distance of 25 cm from the electrode where the ozone concentration varied in the range of 380–1,850  $\mu\text{g}/\text{m}^3$ , and at the distance of 1 m from the source, the ozone concentration decreased 2.5 times.

The upward trend of the annual ozone mean concentration was established in Lithuania during the last 25 years. Mann-Kendall trend analysis showed that the annual mean ozone concentration increased (0.73  $\mu\text{g}/\text{m}^3$ ,  $p < 0.001$ ). The increasing trend (0.69  $\mu\text{g}/\text{m}^3$ ,  $p < 0.001$ ) was also found during a warm (April–September) period. Nevertheless, the ozone level ( $> 180 \mu\text{g}/\text{m}^3$ ) danger to human health practically was not observed in Lithuania. A total of 6% of the data exceed the new WHO guideline of 100  $\mu\text{g}/\text{m}^3$ , i.e. they were observed over more than 10,000 hours. The number of days when high ozone concentration was observed varied no significantly year by year (Figure 2) except in 2005 and 2006. The warm seasons (April – September) of this period were hot, especially in 2006 and the favorable conditions for formation of high ozone concentration were established. It should be noted that such concentrations are observed mostly during spring and summer months when people spend more time outside. Another situation is observed by analyzing the variation of the indicator SOMO 35. The similar ground-level ozone concentrations are observed in Latvia and higher ones in Poland. Ambient ground-level ozone is created from ozone precursors, the largest source of which are cars and other vehicles. Ozone pollution is usually a problem in southern cities of Europe and not in northern cities. In northern countries, ozone causes a larger problem in rural areas. However, the obtained results of ozone level analysis allow us to stay that the ambient ozone level in Lithuania is not dangerous for people to date. Another situation was determined with indoor ozone concentrations. The obtained results suggest that the outdoor ozone concentration under Lithuanian conditions does not cause risks to human health

in door. Only when an additional source of indoor ozone exists, the total level of ozone in separate cases may exceed the permissible concentration level.

As was revealed from measurement results in workplaces, the ozone concentration sometimes can exceed the level dangerous for workers. Our proposition is somewhat contradictory to the results published by other authors, for instance, Kowalska and Zajusz-Zubek<sup>14</sup> and coincides with that of Zhou et al.<sup>15</sup>. The ozone concentration is strongly dependent on the distance of measurement places from sources (Figure 3). Measurements made in the subject's breathing zone are often considered to be the most accurate estimate of a person's "true" exposure. Therefore, the operator can be in a different situation than other people in the same room, who can be affected by the lower ozone concentration. Today, newer models use a different system to reduce the amount of ozone produced by the copying machine. Our investigations were performed with an old machine. As early as 1978, Allen et al.<sup>16</sup> investigated photocopying machines and found that ozone concentrations up to 490  $\mu\text{g}/\text{m}^3$  could be generated in poorly ventilated rooms.

### Conclusion

The danger of the outdoor ozone level to human health practically was not observed in Lithuania during 1995–2011. However, 6% and 1% of all data exceed the World Health Organization guidelines of 100  $\mu\text{g}/\text{m}^3$  and 120  $\mu\text{g}/\text{m}^3$ , respectively.

The indicator of the human health impact, the sum of means over 35 ppb (SOMO 35) varied year from year and reached a maximum of 6,000  $\mu\text{g}/\text{m}^3 \cdot \text{h}$  value in 2006.

The maximum ozone concentration during welding varied from 380 to 1,850  $\mu\text{g}/\text{m}^3$ , and in the copying room it was in the range of 50–330  $\mu\text{g}/\text{m}^3$  and can be dangerous for human health.

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