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Review article

Human exposure to ozone in school and office indoor environments

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ABSTRACT

Handling Editor: Robert Letcher Keywords: Air pollution Ozone Indoor sources Outdoor sources School environments Office environments *Background:* Although it is recognized that ozone causes acute and chronic health effects and that even trace amounts of ozone are potentially deleterious to human health, information about global and local exposures to ozone in different indoor environments is limited. To synthesize the existing knowledge, this review analyzes the magnitude of and the trends in global and local exposure to ozone in schools and offices and the factors controlling the exposures.

Methods: In conducting the literature review, Web of Science, SCOPUS, Google Scholar, and PubMed were searched using 38 search terms and their combinations to identify manuscripts, reports, and directives published between 1973 and 2018. The search was then extended to the reference lists of relevant articles.

Results: The calculated median concentration of ozone both in school $(8.50 \,\mu\text{g/m}^3)$ and office $(9.04 \,\mu\text{g/m}^3)$ settings was well below the WHO guideline value of $100 \,\mu\text{g/m}^3$ as a maximum 8 h mean concentration. However, a large range of average concentrations of ozone was reported, from 0.8–114 $\mu\text{g/m}^3$ and from 0 to $96.8 \,\mu\text{g/m}^3$ for school and office environments, respectively, indicating situations where the WHO values are exceeded. Outdoor ozone penetrating into the indoor environment is the main source of indoor ozone, with median I/O ratios of 0.21 and 0.29 in school and office environments, respectively. The absence of major indoor ozone sources and ozone sinks, including gas-phase reactions and deposition, are the reasons for lower indoor than outdoor ozone concentrations. However, there are indoor sources of ozone that are of significance in certain indoor environments, including printers, photocopiers, and many other devices and appliances designed for indoor use (e.g., air cleaners), that release ozone either intentionally or unintentionally. Due to significantly elevated outdoor ozone concentrations during summer, summer indoor concentrations are typically elevated. In addition, the age of a building and various housing aspects (carpeting, air conditioning, window fans, and window openings) have been significantly associated with indoor ozone levels.

Conclusions: The existing means for reducing ozone and ozone reaction products in school and office settings are as follows: 1) reduce penetration of outdoor ozone indoors by filtering ozone from the supply air; 2) limit the use of printers, photocopiers, and other devices and appliances that emit ozone indoors; 3) limit gas-phase reactions by limiting the use of materials and products (e.g. cleaning chemicals) the emissions of which react with ozone.

1. Introduction

Ozone is an atmospheric trace gas with high oxidizing potential. Its presence is essential in the stratosphere but is undesirable in the troposphere because it can react easily with many compounds, thus generating oxygenated organic species and particles (Finlayson-Pitts and Pitts Jr., 2000).

Human exposure to ozone is primarily by inhalation, but reactions on skin are also reported (Weschler, 2016). Acute and chronic health effects and the contributions of ozone to morbidity and mortality are summarized in (WHO, 2006). More recent studies have shown that daily exposure to high levels of ozone may cause DNA damage, as previously reported for operators in photocopier centers (Kleinsorge et al., 2011; Manikandan et al., 2010; Mortimer et al., 2002). According to Nazaroff (2013), outdoor ozone is also a pollutant of special concern. Of particular importance is the exposure of children to ozone, as exposure could have lifelong consequences. Moreover, it is widely known that the physiology of children and adults is different. Although

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children have higher air intake per kg of body weight, their airways are narrower, which makes them potentially more vulnerable to air pollutants (Moya et al., 2004). According to the U.S. Environmental Protection Agency, EPA (2017), long-term exposure to higher concentrations of ozone may be linked to permanent lung damage (e.g., abnormal lung development in children). Further, ozone has been associated with school absenteeism due to respiratory illnesses, medication use, respiratory problems associated with asthma, decreased respiratory functions, and increased hospital admissions for asthma (Demirel et al., 2014; Gilliland et al., 2001; Lee et al., 2004; Lin et al., 2008; McConnell et al., 2002; Penard-Morand et al., 2005; Romieu et al., 1992; Sheffield et al., 2015). It has been estimated that a $10 \,\mu g/m^3$ increase in 1 h maximum ozone leads to a grand mean of 0.21% increase in mortality, without controlling for other air pollutants (Levy et al., 2005).

In the urban atmosphere ozone is formed by reactions between nitrogen oxides (NO_x) and volatile organic compounds (VOCs) with short atmospheric lifetimes in the presence of sunlight (Seinfield and Pandis, 2016). Outdoor air is the most common source of ozone in indoor air (EC, 2006), and it has been estimated that, depending on the air exchange rate and ozone removal rate, indoor ozone concentrations are 30%–70% of outdoor levels (Weschler and Shields, 1999) when specific indoor sources (e.g., air purifiers, laser printers, photocopiers) are not present (Nicolas et al., 2007). Fig. 1 provides an overview of the key factors and processes affecting indoor ozone concentrations.

In general, it is more challenging to control outdoor ozone than other outdoor air pollutants because it is a secondary pollutant and its formation processes depend not only on the availability of precursors but also on other factors, such as region, season, and time of day (Finlayson-Pitts and Pitts Jr., 2000). In order to protect human health the World Health Organization (WHO) has provided a guideline value of $100 \,\mu\text{g/m}^3$ as the maximum 8 h mean ozone concentration (WHO, 2006). Governmental organizations have issued various of recommendations or standards for outdoor air ozone. In Europe, 120 µg/ m³ (8 h average, allowing 25 exceedances per year) is the reference value, and $240 \,\mu g/m^3$ (1 h average) is the alert threshold (E.C. Ozone Directive, 2016). In the United States, the National Ambient Air Quality Standard (NAAQS) for ozone is 0.070 ppm (8 h average) (US EPA, 2018). In Australia, the National Air Quality Standard for ozone is 0.08 ppm (4 h average) (Australian Government, 2018). For residential indoor air, Health Canada set an 8h average of $40 \,\mu g/m^3$ (Health Canada, 2010). Unlike other air pollutants the concentrations of which have been decreasing, ozone concentrations worldwide are either decreasing much slower, are remaining unchanged, or are even increasing, despite the efforts to control them (Abeleira and Farmer, 2017; Karlsson et al., 2017; Stowell et al., 2017; Wang et al., 2017). The increase could be a baseline or seasonal increase or an increase in the frequency and magnitude of high ozone episodes. This is due to changes in the types and concentrations of precursor compounds that influence the pathways and kinetics of atmospheric chemistry. Fisk (2015) projected that climate change-related increases in ozone—which are due to changes in air movement, cloud cover, humidity, and the emission rates of reactive VOCs and NO_x —will result in substantial adverse health effects. Based on the available evidence, this is to a great extent a consequence of indoor exposures. Fann et al. (2015) analyzed scenarios for the United States and concluded that climate change will support an increase in ambient ozone levels until 2030. Melkonyan and Wagner (2013) provide a similar projection for Germany.

While ozone is a concern in any type of indoor environment, it is of particular significance in schools and offices. The similarity between these environments is firstly in that children spend a good part of the day at school, and a large fraction of the adult population in urbanized countries spend many hours a day in an office environment. Therefore, it is of great importance to ensure that classrooms and offices are safe, healthy environments (Salthammer et al., 2016). Secondly there is a similarity in when these environments are occupied, which is during daytime when the outdoor ozone concentrations are the highest and hence their impact on indoor concentrations, particularly in schools and offices. There are also differences within these two types of environments and between them. For example, office tend to be more often mechanically ventilated than schools, however, there schools, which are mechanically ventilated and offices, which are naturally ventilated. However, to date there has been little focus on indoor ozone in schools and offices, with most published studies concentrating on carbon dioxide, organic compounds, or particles. Therefore, the aim of this work was to assess the magnitude and trends of global and local exposure to ozone in schools and offices and the factors controlling that exposure, based on published literature. Our specific objectives were to: (i) assess the concentrations and exposures occurring indoors; (ii) conclude on the apportionment between outdoor air as a source and indoor source contribution; (iii) review the new ozone generating devices available on the market and the patterns of their usage (e.g., in different countries by different population groups); and (iv) make recommendations about mitigating indoor ozone.

2. Material and methods

A Web of Science, SCOPUS, Google Scholar and PubMed search of



Fig. 1. The key factors affecting indoor ozone concentrations, including ozone infiltration from outdoors, emissions from indoor sources, deposition (e.g., surface removal), and chemical reaction (e.g., with unsaturated hydrocarbons).

the literature published between 1973 and 2018 (until July 2018) was performed. Altogether, 38 search terms (see Table S1 in the supplementary material (SM)) and different combinations of those terms were used. Searches included combinations of at least four terms simultaneously, and each combination included at least two of the following terms were used each time: ozone, school, environment, exposure, concentration, printers, and photocopy machines. The search included original peer-reviewed scientific journal articles, literature reviews, and conference articles (full papers). The search was then extended to the reference lists of relevant articles (based on their abstract and/or full text). The decision to look at certain articles in more detail was based on article titles. Some of the articles were downloaded free from the internet, and we also used the electronic databases of Aalto University. Texas University, and Queensland University of Technology. From over 200 publications identified in the initial search, 141 publications were selected for inclusion in the review analysis. If necessary, ozone concentration was converted from ppm or ppb to $\mu g/m^3$ (1.0 ppb = 2.0 $\mu g/m^3$) m^3 at T = 293 K and P = 1013 mbar).

3. Results and discussion

3.1. Ozone in ambient air

In 1839, the chemist Christian F. Schoenbein reported on a new compound formed during the electrolysis of water and named it "ozone". Fortunately, Schoenbein also discovered that in an aqueous solution potassium iodide (KI) reacts with ozone in the formation of I_2 and that this reaction can be used to determine ozone in air. Therefore, data on tropospheric ozone concentrations have been available since the mid-19th century (Rubin, 2001). Anfossi et al. (1991) reanalyzed data from the time period between 1868 and 1893 for Northern Italy and concluded that the tropospheric ozone levels in Europe have increased by more than two times within 100 years.

In addition to the analysis conducted to reveal the trends occurring in ozone concentrations, a lot of research have been devoted over the past decades to uncover the mechanisms of ozone formation. In particular it has been demonstrated that the chemical atmospheric reactions that lead to the formation and depletion of ozone occur quickly, and therefore considerable changes in concentration take place within less than one hour. Consequently, ozone is now continuously monitored using ultraviolet absorption spectrometry at 254 nm (Daumont et al., 1992). Due to the rapidly changing conditions, ozone concentrations are usually provided as 1 h, 8 h, 1 month, and annual averages. This is exemplified in Fig. 2 for Bad Cannstatt in Stuttgart, Germany (urban background), a polluted urban area, showing the diurnal, seasonal, and annual fluctuations of ozone. Today, there is a dense network of ozone monitoring stations worldwide.

In 2015, the European Environment Agency (EEA) (2017) collected ozone data from 1813 stations in the 28 EU member states and eight other European countries. Based on this data, it can be concluded (as presented in Fig. 3) that the reference level of $120 \,\mu\text{g/m}^3$ is frequently exceeded in Central and Southern Europe. In the annual report by EEA (2017), the observed ozone concentrations are published as 93.2 percentiles, which represents the 26th highest value in a complete series (340/365 = 0.0932). In 2015, 18 EU member states and 4 other reporting countries registered concentrations above the O₃ target value > 25 times. Conformity with the WHO value (8 h mean of $100 \,\mu\text{g/m}^3$) was observed at 4% of all stations and at 8 of 495 rural background stations. Ozone episodes with peak concentrations of 200 µg/m³ sometimes occur in Europe (Saavedra et al., 2012), but extreme values $> 250 \,\mu\text{g/m}^3$ have primarily been reported for other parts of the world, such as the United States (Weschler, 2000; Zhang and Lioy, 1994), China (Duan et al., 2008), India (Verma et al., 2017), Mexico (Velasco and Retama, 2017) and Brazil (Andrade et al., 2017). Lefohn et al. (2017) have demonstrated the local and global variety of ozone concentration distributions by analyzing and comparing data from 481 monitoring sites in the European Union, United States and China.

For Asian metropoles, Verstraeten et al. (2015) found an increasing ozone concentration trends since 2000 and that the tropospheric ozone concentrations over China have increased by about 7% between 2005 and 2010. Gao et al. (2017) reported that in Shanghai, China, has witnessed a substantial increase in ambient ozone concentration, from annual average value of $32.7 \,\mu\text{g/m}^3$ in 2006 to $55.4 \,\mu\text{g/m}^3$ in 2015. As high as $343.5 \,\mu\text{g/m}^3$ a daily maximum ozone concentration was observed in summer 2013 (Pu et al., 2017).

In the Middle East, measured ozone concentrations are comparatively low (Alghamdi et al., 2014; Porter et al., 2014), but model calculations predict a strongly increasing trend within the next three decades (Lelieveld et al., 2014). In Mexico City, the 1 h threshold of 185 ppb (370 μ g/m³) was exceeded 2016 by the first time in 13 years, which is attributed to an increase of 20% in the vehicular fleet and unfavorable weather conditions (Velasco and Retama, 2017). Andrade et al. (2017) analyzed the air quality in the Metropolitan Area of Sao Paulo and did not find a decreasing trend for ozone. Since 2006, an upward trend of 1 h maximum concentrations with peak values > 250 μ g/m³ was observed. Carvalho et al. (2015), who evaluated ozone concentrations in Sao Paulo for the period between 1996 and 2009,



Fig. 2. Outdoor ozone concentrations (urban background) in Bad Cannstatt in Stuttgart, Germany: annual (1 year) averages (1995–2016) and 1 h averages (20.06.2017). The data were obtained from the website of the Landesanstalt für Mesungen Umwelt und Naturschutz Baden-Wuerttemberg (https://udo.lubw.baden-wuerttemberg.de). The diurnal changes (1 h averages) are typical for a polluted summer day in German cities.



Fig. 3. Observed concentrations of ozone in 2015. The figure shows the 93.2 percentile of the ozone maximum daily 8 h average, representing the 26th highest value in a complete series of 365 days. It is related to the ozone target value, allowing 25 exceedances over the $120 \,\mu\text{g/m}^3$ threshold. At the sites marked with red and dark red dots, the 26th highest daily ozone concentrations were above the $120 \,\mu\text{g/m}^3$ threshold. The figure is taken from EEA Report No. 13/2017 (EEA, 2017). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

found decreasing trends for all major pollutants except ozone. Few studies are available for the continent of Africa. However, the trend is similar to that observed for other regions: moderate to high seasonally and annually averaged concentrations (Laakso et al., 2013; Nzotungicimpaye et al., 2014) but extreme peak concentrations in the metropoles (Wheida et al., 2018).

Parrish et al. (2009) reported an increasing trend in tropospheric ozone in the United States and Europe between 1981 and 2007. For Stuttgart, Germany, there is a clearly increasing trend of ozone concentrations between 1995 and 2003. However, a statistical analysis revealed no significant trend for the years between 2004 and 2016. The EEA has analyzed available ozone metrics for the years 2000-2014. In terms of annual averaged concentrations, they report a declining trend at rural sites and an upward trend at traffic stations. The EEA also found that the trends in mean concentration are small and frequently not significant (ECA, 2016). Kleanthous et al. (2014) obtained similar results for the island of Cyprus. They found a non-significant upward trend for the period between 1997 and 2013. Karlsson et al. (2017) reported that in Northern Europe, precursors of ozone including NOx and non-methane-VOCs have decreased between 1990 and 2015, which caused a decrease in the yearly maximum 8 h mean concentrations. However, summer nighttime and winter day and nighttime concentrations have increased. Based on representative concentration pathways (RCP)-based climate model simulations, Stowell et al. (2017) came to a similar conclusion and predicted an increase in ozone health risks in the United States in the 2050s. Both publications highlight the increasing importance of the potential formation of tropospheric ozone.

Ambient ozone is a contributor to global disease burden. Cohen et al. (2017) estimate that exposure to ozone caused an additional 254,000 deaths and a loss of 4.1 million DALYs (disability-adjusted life years) from chronic obstructive pulmonary disease in 2015. Mullins (2018) found that higher ozone levels are associated with poorer human performance in events that heavily tax the respiratory system. Personal exposure to ambient ozone was studied by Niu et al. (2018). The authors highlight that meteorological conditions and activity patters should be considered in individual exposure assessments.

3.2. Ozone in indoor air

We found 13 studies that examined 91 school buildings ($n_{indoor samples} = 525$) and 14 studies ($n_{indoor samples} > 581$) that examined > 136 office buildings (the number of studied offices was not reported in all studies) that reported the indoor concentrations of ozone. A summary of these studies is presented in Tables 1 and 2 and in Tables S2 and S4 in the SM. Detailed information of the studies is available in Tables S3 and S5 in the SM.

Based on these studies, it can be concluded that in school settings the average concentration of ozone (Fig. 4) varied between $0.8 \,\mu g/m^3$ (in Belgium during winter) and $114 \,\mu g/m^3$ (in Mexico during winter) (Blondeau et al., 2005; Bozkurt et al., 2015; Demirel et al., 2014; Dimakopoulou et al., 2017; Gold et al., 1996; HESE, 2006; Mi et al., 2006; Poupard et al., 2005; Stranger et al., 2008; Verriele et al., 2016). In a study comparing school environments in different European cities (HESE, 2006), the highest indoor concentration of ozone (25.9 $\mu g/m^3$) was measured in Uppsala, Sweden, and the lowest was measured in Reims, France (5.3 $\mu g/m^3$). The calculated median concentration (based on the reported average concentrations) in school settings was 8.50 $\mu g/m^3$ (Fig. 5), which was under 10% of the WHO ozone guideline (maximum 8 h mean) value of 100 $\mu g/m^3$ (WHO, 2006).

The average indoor levels of ozone in school settings were much lower than the average outdoor levels $(9.5 \,\mu g/m^3 - 154 \,\mu g/m^3$, Tables S4 and S5). A significant correlation between indoor and outdoor O₃

Table 1

Indoor concentrations (μ g/m³) and indoor/outdoor-ratios of ozone in school buildings in different countries.

Study case (additional information)	Reference	Country/area	Min	Mean	Max	I/O
1	(Gold et al., 1996)	Mexico (Mexico City)	0	114.0	522	NR
2	(Jakobi and Fabian, 1997)	Germany (Freising)	NR	NR	NR	0.66*
3	(Romieu et al., 1998)	Mexico (metropolitan area of Mexico City (MAMC))	NR	29.6*	NR	0.35*
4a (summer)	(Blondeau et al., 2005; Poupard et al., 2005)	France (La Rochelle and its suburbs)	NR	8.5	NR	0.07*
4b (winter)	(Blondeau et al., 2005; Poupard et al., 2005)	France (La Rochelle and its suburbs)	NR	9.6	NR	0.27*
5	(Mi et al., 2006)	China (Shanghai)	1.1	5.3	7.0	0.25
6	(HESE, 2006)	Italy (Sienna, Udine), Norway (Oslo), Sweden (Uppsala), Denmark	3.0	14.3	48.5	0.29
		(Aarhus), France (Reims)				
7a (urban winter)	(Stranger et al., 2008)	Belgium (Antwerp)	< DL	1.5	4.0	0.1
7b (suburban winter)	(Stranger et al., 2008)	Belgium (Antwerp)	< DL	0.8	1.8	0.3
7c (urban summer)	(Stranger et al., 2008)	Belgium (Antwerp)	< DL	3.0	9.9	0.1
7d (suburban summer)	(Stranger et al., 2008)	Belgium (Antwerp)	< DL	2.1	6.4	0.04
8	(Fischer et al., 2013)	Sweden (Göteborg)	0	NR	78.2	NR
9	(Demirel et al., 2014)	Turkey (Eskişehir)	6.23	18.57	55.13	0.21
10	(Jovanović et al., 2014)	Serbia (Zajecar)	8.8	15.5	15.9	0.07
11a (summer)	(Bozkurt et al., 2015)	Turkey (Kocaeli)	NR	12.0	NR	0.2
11b (winter)	(Bozkurt et al., 2015)	Turkey (Kocaeli)	NR	2.3	NR	0.3
12a (non-heating period)	(Kalimeri et al., 2016)	Greece (Kozani)	~ 2.0	NR	~ 20	0.06*
12b (heating period)	(Kalimeri et al., 2016)	Greece (Kozani)	< 0.1	NR	~8	0.06*
13	(Verriele et al., 2016)	France (northern and eastern France)	NR	3.1	NR	NR

NR = not reported; ~ value estimated from the figure in the original article. * calculated mean of the reported average values.

concentrations for all schools has also been reported (Kalimeri et al., 2016). The same authors found a strong correlation between the non-heating period indoor and outdoor ozone levels (R = 0.76, p < 0.05) and a positive correlation (R = 0.63, p < 0.05) between the respective heating period levels.

In office settings, the reported average concentrations of ozone in indoor air varied between 0 and $96.8 \,\mu\text{g/m}^3$ (Fig. 5) (Bako-Biro et al., 2004; Bozkurt et al., 2015; Glas et al., 2015; Hales et al., 1974; Jakobi and Fabian, 1997; Kalimeri et al., 2017; Lee and Hsu, 2007; Mandin et al., 2017; Sabersky et al., 1973; Singh et al., 2014; Wargocki et al.,

1999; Wargocki et al., 2000; Weschler and Shields, 1999; Weschler et al., 1989; Xiang et al., 2016). The lowest mean ozone concentration was measured in Denmark, which was within the designed ventilation rate of $3 L/(s \cdot person)$ (Wargocki et al., 2000), and the highest mean ozone concentration rate was found in an Indian photocopy center during photocopying (Singh et al., 2014). The calculated median concentrations (based on the reported average concentrations) in office settings was $9.04 \mu g/m^3$ (Fig. 5), which was slightly higher than the median concentration in school settings and was less than one tenth of the WHO guideline value (maximum 8-h mean) of $100 \mu g/m^3$ (WHO,

Table 2

Indoor concentrations ($\mu g/m^3$) and indoor/outdoor ratios of ozone in office buildings in different countries.

Study case	Reference	Country/area	Min	Mean	Max	I/O
1	(Sabersky et al., 1973)	USA (Los Angeles)	0	NR	476	0.8
2	(Hales et al., 1974)	USA (Pasadena, California)	NR	NR	NR	0.73*
3	(Weschler et al., 1989)	USA (New Jersey)	0	NR	~159	0.47*
4	(Jakobi and Fabian, 1997)	Germany (Freising)	NR	NR	NR	0,65*
5	(Weschler and Shields, 1999)	USA (New Jersey)	6.34	NR	90.9	NR
6a (designed ventilation rate 3 L/(s·p))	(Wargocki et al., 2000)	Denmark	NR	0	NR	0
6b (designed ventilation rate $10 L/(s \cdot p)$)	(Wargocki et al., 2000)	Denmark	NR	8.5	NR	0.22
6c (designed ventilation rate 30 L/(s · p))	(Wargocki et al., 2000)	Denmark	NR	33.8	NR	0.59
7a (pollution source absent)	(Bako-Biro et al., 2004; Wargocki et al., 1999)	Denmark	NR	16.9	NR	NR
7b (pollution source present)	(Bako-Biro et al., 2004; Wargocki et al., 1999)	Denmark	NR	21.1	NR	NR
8a (before photocopying)	(Lee and Hsu, 2007)	Taiwan (Tainan area)	NR	8.45	NR	NR
8b (during photocopying)	(Lee and Hsu, 2007)	Taiwan (Tainan area)	NR	84.5	148	NR
9a (office 1, before photocopying)	(Singh et al., 2014)	India (New Delhi)	NR	5.18*	NR	NR
9b (office 1, highest mean during photocopying)	(Singh et al., 2014)	India (New Delhi)	NR	20.1	NR	NR
9c (office 2, before photocopying)	(Singh et al., 2014)	India (New Delhi)	NR	20,0*	NR	NR
9d (office 2, highest mean during photocopying)	(Singh et al., 2014)	India (New Delhi)	NR	82	NR	NR
10a (summer)	(Bozkurt et al., 2015)	Turkey (Kocaeli)	NR	4.5	NR	0.1
10b (winter)	(Bozkurt et al., 2015)	Turkey (Kocaeli)	NR	2.7	NR	0.36
11	(Xiang et al., 2016)	China. Changsha	NR	9.08	NR	0.25
12	(Kalimeri et al., 2017)	Greece (Athens)	1.15	11.2	28.9	0.21*
13a (summer)	(Mandin et al., 2017)	Europe (Finland, France, Greece, Hungary, Italy, Netherlands, Portugal, Spain)	< LOD	9.0	42	NR
13b (winter)	(Mandin et al., 2017)	Europe (Finland, France, Greece, Hungary, Italy, Netherlands, Portugal, Spain)	< LOD	3.9	39	NR

*Calculated mean of the reported average values.



Fig. 4. Statistical values (min, mean, med, max) of ozone in school settings. See Table 1 for details.

2006).

3.3. Indoor/outdoor ratios

A number of studies have investigated the penetration of outdoor ozone into the indoor environment and indoor to outdoor (I/O) ozone ratios. The reported I/O ratios in school environments varied between 0 and 0.77 (Fig. 6a) (Blondeau et al., 2005; Bozkurt et al., 2015; Demirel et al., 2014; HESE, 2006; Jakobi and Fabian, 1997; Jovanović et al., 2014; Kalimeri et al., 2016; Mi et al., 2006; Poupard et al., 2005; Romieu et al., 1998; Stranger et al., 2008). The highest I/O ratio was measured in Freising, Germany (Jakobi and Fabian, 1997), and the lowest I/O ratio was measured in La Rochelle and its suburbs in France (Blondeau et al., 2005; Poupard et al., 2005). The calculated median concentration (based on the reported or calculated I/O ratios) in school settings was 0.21 (Fig. 6a).

Reported I/O ratios in office environments varied between 0.02 and 0.90 (Fig. 6b) (Bako-Biro et al., 2004; Bozkurt et al., 2015; Hales et al., 1974; Jakobi and Fabian, 1997; Kalimeri et al., 2016; Sabersky et al., 1973; Wargocki et al., 1999; Wargocki et al., 2000; Weschler et al., 1989; Xiang et al., 2016). The highest I/O ratio was measured in Freising, Germany (Jakobi and Fabian, 1997), and the lowest was measured in Athens, Greece (Kalimeri et al., 2017). In the study by Jakobi and Fabian (1997), the measured I/O ratios ranged between 0.02 and 1.00, with an average of 0.5. The calculated median concentration (based on the reported or calculated I/O ratios) in office settings was 0.29 (Fig. 6a).

Romieu et al. (1998) studied ozone concentrations in homes and school buildings in Mexico City and reported that the major predictors of I/O ratios were open windows in the monitoring room, the presence of carpeting, and the use of air filters. They suggested that in rooms where windows were never open between 10 am and 4 pm the I/O ratio would decrease by 36% compared to rooms where windows were usually open during the day. Other conclusions were that the I/O ratio would decrease by 43% with the presence of carpeting in the rooms and that it would decrease by 21% with the use of air filters for 8 h per day. Recently, Dimakopoulou et al. (2017) reported the at-school outdoor level of ozone was the major predictor of personal exposure to ozone and that the length of time windows were open was significantly associated with personal exposure to ozone.

Weschler (2000) evaluated I/O ratios measured by various investigators for buildings with negligible indoor sources of ozone, concluding a) that outdoor to indoor transport of ozone is significant and that b) indoor ozone levels are frequently 30%–70% the corresponding outdoor levels.

For 8 school buildings Blondeau et al. (2005) found that for 8 school buildings the I/O ratios of ozone varied from 0 to 0.45 and were strongly influenced by how airtight the buildings were, that is, the more airtight the building envelope, the lower the ratio. The authors concluded that the I/O concentration ranges could be reasonably used as a basis for assessing the lower and upper limits of children's total exposure to outdoor ozone. All I/O ratios < 1 indicate that the ozone concentration indoors derives from the ozone transport from outdoors to indoors rather than internal sources (Poupard et al., 2005). Analyses



Fig. 5. Statistical values (min, mean, med, max) of ozone in office settings. See Table 2 for details.



Fig. 6. a) The reported I/O ratios (and calculated median value) in school environments; b) the reported I/O ratios (and calculated median value) in office environments.

of the temporal profiles of indoor and outdoor concentrations have revealed that opening windows significantly increases the I/O ozone ratio (Blondeau et al., 2005).

Demirel et al. (2014) found that I/O ozone ratios varied between 0.46 and 1.08 and that the ratios were < 1 for almost all the samples (only 0.08% were > 1), concluding that the absence of major sources in indoor environments, such as gas-phase reactions and deposition, might result in lower indoor ozone concentrations. Other studies came to a similar conclusion, for example, those by Chao (2001), Blondeau et al. (2005), and Weschler (2006).

Jakobi and Fabian (1997) experimentally assessed indoor half-live times of ozone and found them to range from 3 to 15 min, while Weschler (2000) estimated that range to be between 7 and 10 min. In the absence of indoor sources and under the assumption that the indoor ozone concentration is primarily determined by air exchange λ (1/h) and the surface removal rate constant k_m (1/h), I/O can be estimated from Eq. (1) (see Weschler (2006)):

$$I/O = \frac{\lambda}{\lambda + k_m} \tag{1}$$

The rate constants for the removal of ozone by indoor surfaces were reviewed by (Weschler, 2000). The range of reported k_m values was 0.8–7.6 1/h, but most of the values were between 2.5 1/h and 3.5 1/h. We used Eq. (1) to consider two different situations, as presented in

tween 1.0 1/h and 7.0 1/h. When considering typical indoor k_m values around 3 1/h, I/O ratios of 0.1–0.2 and 0.5–0.6 are calculated for low and high air exchange rates, respectively. Even under unfavorable conditions (high air exchange, low k_m) the I/O ratio is below 0.83. At an average air exchange rate of 0.5 1/h, which is typical for living conditions in private dwellings (Salthammer, 2018), the I/O ratio is 0.3 or lower. In school classrooms, the ventilation rates are often below required minimum standards (Fisk, 2017). This is of advantage for low ozone I/O ratios but causes many other problems. By using a simple steady-state model with 80% accuracy and ac-

Fig. 7. The left part of the figure presents a scenario with low air ex-

change rates (0.1–1.0 1/h), while the right part presents a scenario with high rates (1.0–5.0 1/h). In both cases, the removal rates k_m were be-

cording to the ventilation rate and surface removal rate data collected from the literature, Lai et al. (2015) found that the most common indoor-to-outdoor ozone ratios were 0.09, 0.19, and 0.47 for infiltration, mechanical ventilation, and natural ventilation, respectively.

3.4. Factors affecting the indoor ozone concentrations and exposure

3.4.1. Seasonality, temperature, and relative humidity

Several studies (Bozkurt et al., 2015; Mandin et al., 2017) have reported significant seasonal differences in ozone levels and significantly higher outdoor and indoor ozone concentrations during



Fig. 7. Contour plots of calculated ozone I/O ratios as a function of air exchange rates and surface removal rates k_m (see Eq. (1)). Low air exchange rates (0.1–1.0 1/h) were assumed in the left part, while high air exchange rates (1.0–5.0 1/h) were assumed in the right part. In both cases, the ozone surface removal rates k_m ranged from 1.0 1/h–7.0 1/h.

summer. However, Mandin et al. (2017) found in their OFFICEAIR study that the general trends found at the European level were not always observed at the country level. Other studies did not identify any significant seasonal differences regarding indoor ozone levels (Kalimeri et al., 2016).

Higher outdoor ozone concentrations during summer (due to the photochemical production from increased ultraviolet radiation) means that higher amounts of ozone penetrate buildings through window openings, leading to the ozone-initiated formation of formaldehyde involving terpenes (e.g., p-limonene) (Nørgaard et al., 2014; Weschler, 2006). During winter, terpenes are less involved in ozone-initiated reactions and are observed at higher concentrations (Mandin et al., 2017). All this indicates that in the course of a year, a spot measurement during one work week is inadequate to characterize the long-term exposure of occupants to ozone within a building.

Jakobi and Fabian (1997) reported no significant correlation between outdoor temperature and outdoor NO2 or O3, or between outdoor NO₂ and outdoor O₃, and finally, indoor O₃ was increased at higher indoor NO2. Their results suggest that the decay rate of ozone is independent of temperature, at least within the investigated range of room temperatures between 22 °C and 27 °C. Ozone decay was substantially influenced by the surface-to-volume ratio, which is in good agreement with previous studies indicating that deposition onto surfaces is one of the main removal processes of ozone levels indoors. The reaction between ozone and NO (a fast ozone decay reaction) can be excluded in all cases. In their linear regression analysis, Gold et al. (1996) found that both the relative humidity and temperature alone were predictors of indoor ozone concentration and that relative humidity was a stronger predictor of indoor ozone than temperature. Avol et al. (1998) reported that indoor ozone levels were higher when outdoor temperatures were higher; this effect was magnified when windows were open. However, Bernard et al. (1999) reported that temperature has no, or very little effect on the ozone production cycle. According to their study, temperature was a covariable of sunlight (r = 0.85, p = 0.001), and the causal relationship existed only between sunlight and ozone concentration.

3.4.2. Type of ventilation and other housing characteristics

Ventilation has an essential role of controlling the indoor air exposure of different pollutants. It has been recognized that e.g. the type and quantity of chemical compounds (both organic and inorganic) in public buildings such as in schools and offices are highly influenced from ventilation (Lyng et al., 2015; Salonen et al., 2009; Vornanen-Winqvist et al., 2018).

In naturally ventilated school buildings the ventilation is often supplemented by airing rooms (opening doors and windows), and thus concentrations of pollutants, apart from emissions from materials and equipment, largely depend on the concentrations of compounds in the atmospheric air (Stabile et al., 2016). In naturally ventilated buildings, means to reduce indoor ozone level can restrict the ventilation for the portion of day when ozone is elevated and increase ventilation when ozone levels are lower (Weschler, 2006). However, it should be kept in mind that this would have an impact on pollutants generated indoors, particularly CO₂, potentially elevating it's to unactable levels, especially in classrooms with high occupancy density. The study by Gold et al. (1996) concluded that as outdoor ozone concentrations increased, indoor ozone concentrations increased more rapidly with windows and doors open than with windows and doors closed.

In air-tight buildings with a mechanical ventilation system, the balancing and controlling of the ventilation system and pressure differences are necessary in order to keep pollution exposure (especially impurities infiltrating from the structures) as low as possible (Vornanen-Winqvist et al., 2018). It has been reported that in buildings with mechanical ventilation systems, some types of filters (charcoal or chemically impregnated filters) can remove a large fraction of ozone (Kelly and Kinkead, 1993; Shair, 1981; Weschler, 2006; Zhong et al.,

2017). Aldred et al. (2016) concluded in their study, that carbon filtration during the ozone season was beneficial and economically viable for indoor ozone removal in commercial office buildings, long-term health-care facilities, and K–12 schools. Chatzidiakou et al. (2015) found that exposure to indoor ozone levels in the non-heating season increased the hazard ratio of nasal and general symptoms; however, in the heating season exposure was not significantly related to any SBS (sick building syndrome) symptoms.

Apart from ventilation, the air quality in schools and offices is mostly influenced by the presence and types of decoration and finishing materials including building and structural materials (Śmiełowska et al., 2017). Placing ozone-scavenging materials or material coatings indoors can be employed as passive removal methods of ozone reduction (Darling et al., 2016). Several studies have published findings about building materials or decorative material coatings (e.g. paint, plaster) for passive reduction of ozone (Cros et al., 2012; Darling et al., 2016; Darling et al., 2012; Kunkel et al., 2010).

Location of the building may also affect the ozone levels and exposure to indoor ozone. Gül et al. (2011) noticed in their school study that nitrogen dioxide and ozone levels were higher in industrial areas than in non-industrial areas, and pupils in industrial areas suffered more respiratory health problems.

3.4.3. Indoor chemistry

In the indoor environment, many chemical reactions occur, which might either consume or produce ozone. These reactions are fast enough to quickly influence the ozone indoor concentration and the kind and concentration of indoor chemicals (Weschler, 2000). As known from classical atmospheric chemistry, many other oxidants like the hydroxy radical OH (Gligorovski et al., 2014; Weschler and Shields, 1996), nitrous acid (HONO) (Gligorovski, 2016), peroxyacyl nitrate (PAN) (Jakobi and Fabian, 1997), nitric oxides (NO_x) and hydroperoxyl (HO₂) (Waring and Wells, 2015) are involved and thousands of complex and often multifunctional products can be produced from the reaction with unsaturated VOCs (Weschler and Carslaw, 2018). The main route to OH formation indoors is through reaction of O_3 with alkenes and monoterpenes (Weschler and Carslaw, 2018). Another pathway for the removal of indoor ozone is the reaction with nitric oxide (NO).

Indoor homogenous and heterogenous chemistry, particularly terpene/ozone reactions, contribute to the depletion of indoor ozone (Morrison, 2010). However, the bimolecular gas-phase reaction strongly depends on the kinetic reaction constant, which is between $10\cdot10^{18}$ and $2000\cdot10^{18}$ cm²·molecule⁻¹·s⁻¹ (Toftum et al., 2008). Porous materials, such as clay, can be used to passively remove ozone (Darling et al., 2016). Humans are also known as major ozone sinks due to reactions with skin (Fischer et al., 2013; Wisthaler and Weschler, 2010), clothing (Rai et al., 2013), and hair (Pandrangi and Morrison, 2008). Fadeyi et al. (2013) calculated v_d (deposition velocity) values between 14.4 m/h and 22.3 m/h per person under the assumption of a 1.7 m^2 body surface. Fischer et al. (2013) found that in a classroom the ozone removal caused by 24 children and one teacher was approximately 2.6 times greater than that of the available surfaces of the classroom and its furniture. Fadeyi (2015) reviewed ozone surface deposition velocities attributed to humans and estimated values between 5.4 m/h and 22.3 m/h per person. The contribution of humans to ozone removal can be calculated from Eq. (2) (Fadeyi et al., 2013):

$$k_m = N \cdot v_{d_person} \cdot \frac{A_{person}}{V}$$
⁽²⁾

where N is the number of people in the room, v_{d_person} is the deposition velocity per person, A_{person} is the body surface per person, and V is the room volume. For a typical classroom scenario (N = 20-30, $V = 240 \text{ m}^3$, $A_{person} = 1.7 \text{ m}^2$ and $v_{d_person} = 5-22 \text{ m/h}$), removal rates k_m range between 0.7 1/h and 4.7 1/h. Under these conditions, occupants may act as sinks for > 50% of indoor ozone. Fadeyi et al. (2013) measured a 33% reduction of ozone when a 240 m³ chamber was

occupied by 18–20 workers. It should also be considered that the total inhalation rate of occupants ($\approx 8 \text{ l/min}$) accounts for approximately 5% of the room volume. Xiang et al. (2016) found that ozone deposition velocities to human surfaces reported in the literature range from 0.1–0.6 cm/s (3.6–21.6 m/h).

In another study, extreme ozone concentrations > 1500 ppm occurred during the disinfection of buildings. However, no one was present during and immediately after the treatment of the building with ozone (Poppendieck et al., 2007a; Poppendieck et al., 2007b). Classical tropospheric photochemistry is unlikely in indoor environment. Under laboratory conditions, Salthammer and Fuhrmann (2007) observed a 10% decrease of the NO₂ concentration and an increase of the NO concentration in a glass chamber when the air was illuminated from outside the chamber with a UV/VIS sunlight simulator, but ozone was not measured in this experiment. However, the bond of molecular oxygen breaks under irradiation at wavelengths < 240 nm (UV-C). This might lead to the formation of high ozone levels in ultraviolet photocatalytic oxidation (PCO) reactors, which are used in the decomposition of indoor VOCs (Farhanian and Haghighat, 2014).

3.4.4. Printers, photocopiers and other devices and appliances

Printers and photocopiers have been found to be significant sources of ozone and other pollutants in office environments (Destaillats et al., 2008; Lee and Hsu, 2007; Singh et al., 2014), and even low levels of ozone emitted by printers and photocopiers can react with other indoor pollutants, resulting in secondary pollutants and the generation of ultrafine aerosol particles (Destaillats et al., 2006a; Destaillats et al., 2006b; Norgaard et al., 2014; Singer et al., 2006). The effect of most other devices and appliances (such as desktops and notebook computers) on indoor ozone levels are generally smaller or even negligible (Destaillats et al., 2008). However, several studies have found that things such as air cleaning devices may increase indoor ozone concentrations to harmful levels (Poppendieck et al., 2014).

Destaillats et al. (2008) evaluated the available literature up to 2007 and reported ozone emission rates of up to $7900 \,\mu$ g/h and unit. However, the situation significantly improved after the introduction of ecolabels such as, for example, RAL-UZ-205 (2017). Modern printers and photocopiers usually release only small amounts of ozone (Morawska et al., 2009). Nevertheless, high ozone concentrations can still be measured in photocopy centers, as reported by Singh et al. (2014) for eight locations in Delhi, India.

Today, there are many devices and appliances designed for indoor use that can release ozone either intentionally or unintentionally. Siegel (2016) pointed out that a device intended as an air cleaner that intentionally emits any compound into indoor air should not be considered a true air cleaner because the contamination can outweigh any air cleaning benefits. A study by Hubbard et al. (2005) focused on ozone generators as air purifiers. The authors measured peak concentrations of ozone of between $55 \,\mu g/m^3$ and $102 \,\mu g/m^3$ in different types of dwellings during the operation of such devices. Waring et al. (2008) studied portable air cleaners in a $14.75 \,m^3$ stainless steel chamber and measured ozone emission rates of between $3.3 \,mg/h$ and $4.3 \,mg/h$ per unit. To estimate ozone indoor concentrations based on emission rates, Hubbard et al. (2005) use Eq. (3), which assumes steady-state conditions in a well-mixed environment in the absence of outdoor ozone and significant indoor chemistry.

$$C_{03} = \left[\frac{ER/V}{\lambda + v_d \cdot (A/V)}\right] \tag{3}$$

 C_{O3} is the concentration of ozone (µg/m³), ER is the emission rate (µg/h), V is the volume of the room (m³), λ is the air exchange rate (1/h), v_d is the deposition velocity (m/h), and A is the surface of the room (m²). For typical building products, deposition velocities differ widely (Grøntoft and Raychaudhuri, 2004; Lamble et al., 2011; Lin and Hsu, 2015; Schripp et al., 2012). In his comprehensive review, Fadeyi (2015)

reported ozone deposition velocities between 0.1 m/h (paint on polyester film) and 28.8 m/h (gypsum board), with the exception of glass (v_d < 0.01 m/h).

Poppendieck et al. (2014) investigated the generation of ozone by two types of in-duct electrostatic precipitators (ESP) in a test house. During continuous operation of the ESP under different conditions, the ozone concentration was between $40 \,\mu g/m^3$ and $151 \,\mu g/m^3$, and emission rates were between 21.8 mg/h and 60.4 mg/h. Zhang and Jenkins (2017) provided a comprehensive compilation of ozone emissions from consumer products and home appliances. The ozone concentration was strongly dependent on the distance to the operating device. The highest room concentration was measured during 60 min operation of the laundry water treatment device connected to a washing machine, with $485 \,\mu g/m^3$ (max) and $209 \,\mu g/m^3$ (mean).

4. Conclusions

The findings of our literature review reveal a) the increasing importance of the potential formation of tropospheric ozone; b) the calculated median concentration of ozone was $8.50 \,\mu\text{g/m}^3$ (average $0.8 \,\mu g/m^3 - 114 \,\mu g/m^3$) and $9.04 \,\mu g/m^3$ (average $0 - 96.8 \,\mu g/m^3$) in school and office settings, respectively, which is significantly lower than the WHO 8-h guideline value $(100 \,\mu g/m^3)$ for ozone. However, there are situations both in school and office settings where the WHO value is exceeded. The median I/O ratio was 0.21 (average I/O ratios 0–0.77) and $0.29 \,\mu\text{g/m}^3$ (average I/O ratios 0.02–0.90) in school and office settings, respectively; c) there are significant correlations between indoor and outdoor O_3 concentrations for school and office buildings; d) the absence of major sources in indoor environments, gasphase reactions and deposition, might result in lower indoor ozone concentrations; e) there are revealing seasonal differences for the ozone levels and higher indoor and outdoor concentrations of ozone during the summer season, but the general trends at the European level were not always observed at the country level; f) the type of ventilation, type of filters, open windows, and the presence and types of decoration and finishing materials were significantly associated with indoor O₃ levels; g) indoor chemistry, particularly terpene/ozone reactions, might contribute to the depletion of indoor ozone; and h) printers and photocopiers and many devices/appliances designed for indoor use (e.g., air cleaners) can release ozone either intentionally or unintentionally. Although it is very costly and difficult to reduce the concentration of ozone outdoors, there are some means to reduce the concentration of indoor ozone and its oxidation products to below the WHO guideline value. These include, for example, the use of suitable filters, keeping windows and doors closed when concentration of ambient ozone is high, the use of suitable building or decorative material coatings (e.g. paint, plaster) for passive reduction of ozone, and reducing the use of ozone releasing devices/appliances and products with ozone-reacting constituents. We should be also noted that ambient ozone concentration alone is not a suitable surrogate to evaluate individual exposure, and factors affecting personal ozone exposure, especially activity patterns (e.g. windows opening and using the ozone releasing devices) and outdoor activities should be taken into account.

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Appendix A. Supplementary data

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