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Assessment of indoor air quality in office buildings across Europe – The OFFICAIR study

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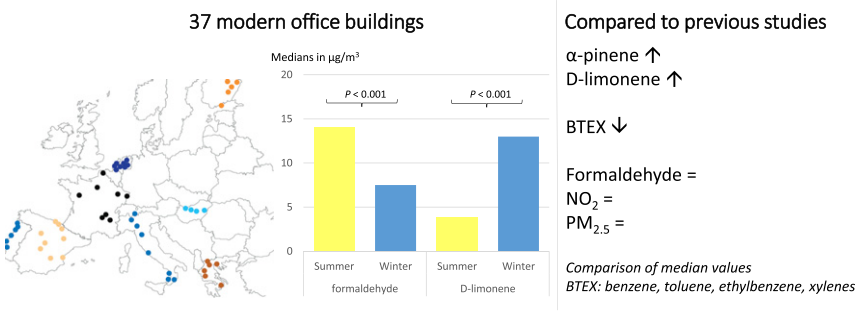
HIGHLIGHTS

- VOCs, aldehydes, O_3, NO_2 and $PM_{2.5}$ were measured in 37 office buildings in 2 seasons.
- The α-pinene and D-limonene concentrations were higher compared to those from past studies.
- The indoor concentrations in summer and winter varied significantly.
- An influence of floor level on indoor concentrations was observed for some pollutants.
- An evaluation of IAQ in terms of respiratory health effects was performed.

GRAPHICAL ABSTRACT



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ABSTRACT

The European project OFFICAIR aimed to broaden the existing knowledge regarding indoor air quality (IAQ) in modern office buildings, i.e., recently built or refurbished buildings. Thirty-seven office buildings participated in the summer campaign (2012), and thirty-five participated in the winter campaign (2012 – 2013). Four rooms were investigated per building. The target pollutants were twelve volatile organic compounds, seven aldehydes, ozone, nitrogen dioxide and particulate matter with aerodynamic diameter <2.5 µm ($PM_{2.5}$). Compared to other studies in office buildings, the benzene, toluene, ethylbenzene, and xylene concentrations were lower in OFFICAIR buildings, while the α-pinene and D-limonene concentrations were higher, and the aldehyde, nitrogen dioxide and $PM_{2.5}$ concentrations were of the same order of magnitude. When comparing summer and winter, significantly higher concentrations were measured in summer for formaldehyde and ozone, and in winter for

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VOC
Terpene
Particulate matter
Seasonal variability
Spatial variability

benzene, α-pinene, D-limonene, and nitrogen dioxide. The terpene and 2-ethylhexanol concentrations showed heterogeneity within buildings regardless of the season. Considering the average of the summer and winter concentrations, the acetaldehyde and hexanal concentrations tended to increase by 4–5% on average with every floor level increase, and the nitrogen dioxide concentration tended to decrease by 3% on average with every floor level increase. A preliminary evaluation of IAQ in terms of potential irritative and respiratory health effects was performed. The 5-day median and maximum indoor air concentrations of formaldehyde and ozone did not exceed their respective WHO air quality guidelines, and those of acrolein, α-pinene, and D-limonene were lower than their estimated thresholds for irritative and respiratory effects. $PM_{2.5}$ indoor concentrations were higher than the 24-h and annual WHO ambient air quality guidelines.

1. Introduction

A growing fraction of the active population is working in office buildings worldwide. Existing studies on indoor air quality (IAQ) in office buildings have focused on specific issues such as emissions from printers and photocopiers (Cacho et al., 2013; Tang et al., 2012), the efficiency of outdoor air filtration (Fisk et al., 2000), or hot and humid climatic zones (Zuraimi et al., 2004). Existing studies have also dealt with some specific parameters such as relative humidity (Wolkoff and Kjærgaard, 2007), man-made vitreous fibers (Salonen et al., 2009a), particulate matter (PM) (Horemans and Van Grieken, 2010) or semi-volatile organic compounds (Fraser et al., 2013; Watkins et al., 2013). The BASE (Building Assessment Survey and Evaluation) study performed in 100 office buildings across the United States over a five-year period (1994–1998) is one of the first large studies regarding volatile organic compounds (VOCs) in indoor air within offices (http://www.epa.gov/iaq/base/). In Europe, Finnish studies provided insights on VOCs, aldehydes, ammonia, and PM in office buildings (Lappalainen et al., 2013; Salonen et al., 2009b). Moreover, several EU-funded projects (IAQ-AUDIT, HOPE and AIRMEX) performed IAQ measurements in office buildings. Within IAQ-AUDIT (European Audit Project to Optimize Indoor Air Quality and Energy Consumption in Office Buildings), fifty-six office buildings in nine European countries were audited during the heating season of 1993–1994 (Bluyssen et al., 1996). The main aim was to develop assessment procedures and guidance on ventilation and source control to assure good IAQ and optimize energy use in office buildings. The HOPE (Health Optimization Protocol for Energy Efficient Buildings) project aimed to determine if energy efficiency, IAQ, comfort and occupant satisfaction can be achieved simultaneously in European buildings; IAQ measurements were performed in a subsample of two offices in the UK (Aizlewood and Dimitroulopoulou, 2006). Lastly, in the AIRMEX (European Indoor Air Monitoring and Exposure Assessment) project, hydrocarbons, aromatic compounds, alcohols, and carbonyls were monitored in the period 2003–2008 in buildings from eleven European cities, including offices (Geiss et al., 2011; Kotzias et al., 2009).

Office buildings have extensively evolved to become controlled environments with sophisticated ventilating and air-conditioning systems. However, little is known about IAQ in these so-called 'modern' office buildings. In recent years, office buildings have not been extensively studied compared to other equally important indoor environments in terms of time spent by the population, such as dwellings and schools. Moreover, in addition to the reported health effects due to indoor air pollutants (World Health Organization [WHO], 2010), the indoor environmental quality in offices may affect cognitive performance and even subclinical disturbances may lead to losses in work productivity (Wargocki and Wyon, 2013).

To increase knowledge about IAQ in modern office buildings, the European OFFICAIR project was focused on office buildings built or refurbished after 2001. The objectives were manifold (Bluyssen et al., 2016) and included three field campaigns with dedicated objectives and methods. One objective was targeted at identifying possible characteristics in terms of indoor air pollutants and/or concentrations to better understand their variabilities over time and space and to assess the associated health effects. One of the three field campaigns, namely the 'detailed study', specifically targeted this objective. It comprised IAQ measurements in office buildings distributed among the eight participating countries: Finland, France, Greece, Hungary, Italy, the Netherlands, Portugal, and Spain. The measurements were repeated in two seasons, with the heating off (called the summer campaign) and with the heating on (called the winter campaign). In addition, a technical description of the investigated rooms was performed, an on-line questionnaire on the perceived comfort and health was filled in by the office workers, and on-line performance and reactivity tests were performed by the same workers.

This paper presents the results of the IAQ measurements, the IAQ seasonal and spatial variabilities, and the evaluation of IAQ in terms of potential adverse health effects based on WHO air quality guidelines or estimated thresholds for irritative and respiratory effects.

2. Materials and methods

2.1. Building recruitment

Each country independently selected the buildings from the approximately 20 investigated in the first phase of the project (Bluyssen et al., 2016). The selection was carried out based on several common criteria, especially the willingness of the building manager to take part in this further step of the project and at least 40 workers to achieve enough participants for the questionnaires and performance tests. Moreover, the 'detailed study' included questionnaires on perceived IAQ and comfort as well as performance tests. Therefore, buildings with symptoms that are associated with environmental parameters other than air quality were excluded. These symptoms include stress and/or work overload and were identified during the first phase of the project (Bluyssen et al., 2016).

Thirty-seven office buildings took part in the 'detailed study' summer campaign, among which two office buildings withdrew for the winter campaign. The buildings were located in Finland (3 buildings), France (9), Greece (5), Hungary (5), Italy (4), the Netherlands (4), Portugal (4), and Spain (3). The locations of the buildings are shown in Fig. S1 in the Supplementary material. The summer campaign occurred between June 18, 2012 and October 19, 2012. Four buildings were investigated between May 13, 2013 and June 14, 2013. The winter campaign occurred between November 5, 2012 and April 19, 2013. Two buildings were investigated between November 15 and November 22, 2013.

2.2. Target pollutants

The target pollutants were twelve VOCs, seven aldehydes, ozone (O_3), nitrogen dioxide (NO_2) and particulate matter with aerodynamic diameter <2.5 μm ($PM_{2.5}$). The VOCs and aldehydes are listed in Table 1. These pollutants were chosen based on a literature review of indoor air pollutants in office environments published in the framework of the OFFICAIR project (Wolkoff, 2013) and based on their potential

Table 1
Target pollutants monitored in the frame of the OFFICAIR project, and sampling and analysis specifications.

Pollutants	Methods
VOCs (**n = 12**) benzene, toluene, xylenes, ethylbenzene, n-hexane, trichloroethylene, tetrachloroethylene, α-pinene, D-limonene, 2-butoxyethanol, 2-ethylhexanol, styrene	Sampling: Radiello® passive sampler: diffusive body code 120–2 and cartridge code 145 Analysis: thermal desorption, gas chromatography mass spectrometry, ISO 16017–2 standard, analyzed at CNR-IIA, Italy
Aldehydes (**n = 7**) formaldehyde, acetaldehyde, acrolein, propionaldehyde, benzaldehyde, hexanal, glutaraldehyde	Sampling: Radiello® DNPH passive sampler: diffusive body code 120–1 and cartridge code 165 Analysis: high performance liquid chromatography coupled with UV detection, ISO 16000–4 standard, analyzed at CNR-IIA, Italy
Ozone (O_3)	Sampling: Radiello® passive sampler: diffusive body code 120–1 and cartridge code 172 Analysis: UV–Vis spectrophotometry, analyzed at ELTE, Hungary
Nitrogen dioxide (NO_2)	Sampling: Gradko® circular diffusive sampler Analysis: UV–Vis spectrophotometry, analyzed at Gradko International Ltd., UK
$PM_{2.5}$	Sampling: low-volume aerosol sampler, quartz fiber filters Analysis: gravimetric (BS EN 12341: 2014), weighed at ELTE, Hungary

association with irritation symptoms, cardiovascular and pulmonary effects, or degraded work performance.

2.3. Sampling

Sampling was carried out over five days, from Monday morning (approximately 9 am) to Friday afternoon (approximately 5 pm) at four locations in each building, except for $PM_{2.5}$ (one location per building). The locations were chosen to be as diverse as possible. The sampling devices were placed preferentially on four different floors in the building and at different orientations: North, South, East, and West. Moreover, the diversity of rooms was considered, namely open spaces and cellular offices if the building had both types of working spaces. The duration of the sampling was a compromise between the willingness to characterize average concentrations in office buildings to assess long-term worker exposure and technical feasibility. Similarly, the number of sampling locations per building was a compromise between extensive IAQ characterization for the whole building and budget and feasibility issues.

The samplers were placed in the center of each room, not closer than 1 m to the wall, at the height of the breathing zone of seated occupants, i.e., approximately 110 cm; ventilation channels and heating sources, including the sun, were avoided. In practice, the samplers were fixed to a wire attached to the ceiling or placed in a metallic rack. Any adhesive or material that emits VOCs was strictly avoided when placing the samplers.

The specifications of the passive and active sampling techniques are provided in Table 1. Before sampling, the passive samplers were stored in a refrigerator (<4 °C). After sampling, the VOC, aldehyde and O_3 samplers were sent in refrigerated packages to the respective central laboratories. NO_2 samplers were returned at ambient temperature to the central laboratory.

$PM_{2.5}$ sampling was performed at one location per building (one out of the four with gaseous pollutant sampling) using low-volume aerosol samplers equipped with $PM_{2.5}$ heads (Mihucz et al., 2015; Szigeti et al., 2016). The flow rate was checked before and after each sampling and adjusted if needed. $PM_{2.5}$ was collected onto quartz fiber filters (Ø 37 or 47 mm, 450 μm thickness, 3-mm pore size; Whatman QM-A grade) supplied by GE Healthcare (Little Chalfont, Buckinghamshire, UK). Before sampling, the filters were conditioned at 20 ± 1 °C and 50 ± 5% relative humidity for 48 h and weighed on a Mettler Toledo XP26DR balance with a precision of 2 μg. After sampling, the filters were sent back to the central laboratory in refrigerated packages.

2.4. Analysis and quality control

The specifications of the analytical methods are provided in Table 1.

The VOC samplers were analyzed within one month after their arrival at the central laboratory. Before analysis, the samplers were stored at 4 °C in dark conditions. The analyses of VOCs were performed by thermal desorption coupled with capillary gas chromatography (GC) - mass spectrometry (MS). The thermal desorption was achieved by sequential tube desorber model 1000 (DANI Instruments, Pavia, Italy), while the GC–MS consisted of a TRACE ULTRA chromatograph coupled with a single quadrupole detector both supplied by THERMO (Bremen, Germany). In the first step, the tubes were heated at 300 °C for 10 min with a helium gas flow rate of 50 mL min^{-1} (purity: 99.999%; Rivoira, Milan, Italy). During this phase, the eluted VOCs were transferred from the tube into a cold trap (Tenax TA) thermostated at −30 °C. After the primary desorption, the cold trap was rapidly heated to 250 °C (secondary desorption) and then maintained at this temperature for 3 min. The analytes were injected onto a capillary column (GsTEK-624, length: 60 m, inner diameter: 0.32 mm, film thickness: 1.8 μm) via a heated transfer line at 200 °C. The column oven temperature was initially at 40 °C for 5 min, increased to 230 °C at a rate of 6.0 °C min^{-1} and then maintained at 230 °C for 5 min. Helium was used as the carrier gas with a constant flow rate of 1.0 mL min^{-1}. The GC interface temperature was set to 250 °C, while the source temperature and emission current were 230 °C and 100 μA, respectively. The chromatograms were acquired in full scan mode (scanned mass range was from 35 to 300 amu, in electron ionization mode at 70 eV). The quantitative identification of target VOCs was based on retention times and the ion ratios of the qualifier and quantification ions. Six-point calibration (20, 50, 100, 200, 500, and 1000 μg analyte per tube) was applied.

The aldehyde samplers were analyzed within two weeks after their arrival at the central laboratory. The analysis was performed according to the LC ISO 16000–4 standard (2011). The aldehydes were extracted from the DNPH-cartridges by introducing 2 mL of acetonitrile (Ultra Gradient, ROMIL) into the cartridge and shaking for 30 min. The solution was then filtered and analyzed via high-performance liquid chromatography (Shimadzu, Kyoto, Japan) coupled with UV detection at a wavelength of 360 nm. The system consisted of a pump, a UV–Vis detector (SPD-M20A), an injection valve with a volume of 20 μL and a reversed-phase column (Restek Ultra C-18, 100 Å, 150 mm × 4 mm, 5 μm) maintained at 25 °C (CTO-10AS VP). The mobile phase was an acetonitrile-water gradient. The analyses were conducted with a flow rate of 1 mL min^{-1} for 3.5 min, followed by 1.7 mL min^{-1} for 35 min before returning to 1 mL min^{-1}. The aldehyde concentrations were calculated using a four-point calibration curve (ranging between 0.05 and 1 mg mL^{-1}) and standard solutions of the corresponding DNPH-aldehyde derivatives in acetonitrile. The peak areas were matched using the known hydrazone concentrations, and the concentrations of the corresponding aldehydes were calculated using conversion factors from the manufacturer.

The O_3 samplers were analyzed within one week after their arrival at the laboratory. During transport and until analysis, the samplers were stored at 4 °C in dark conditions. The adsorbing material was mixed with 5 mL of a 3-methyl-2-benzothiazolinone hydrazone hydrochloride (MBTH) solution (Sigma Aldrich, St Louis, USA) to obtain the corresponding MBTH-azide. After 1 h of reaction, the samples were filtered through 0.45 μm membrane filters. Finally, the absorbance of the solutions was measured at 430 nm with a UV–Vis spectrophotometer

(Lambda 15, Perkin-Elmer). Standard solutions of 4-pyridylaldehyde were used for calibration.

The NO_2 analysis was completed by Gradko International Ltd., UK, within three weeks after exposure. The concentrations of nitrite ions and hence chemically adsorbed NO_2 were quantitatively determined via UV–Vis spectrophotometry with reference to a calibration curve derived from the analysis of standard nitrite solutions (U.K.A.S. Accredited Methods).

For the chromatographic methods (VOCs and aldehydes), the limits of detection (LODs) were defined as 3 times the signal-to-noise ratio (3:1). The limits of quantification (LOQs) were defined as 10 times the signal-to-noise ratio (10:1). In the case of the spectrophotometric methods (O_3, NO_2), the LOD and LOQ values were calculated as 3 and 10 times the standard deviation of the lot blank values, respectively. The LODs and LOQs are reported in Table S1.

One field blank per chemical parameter (VOCs, aldehydes, O_3, and NO_2) was placed in one room of each building. Moreover, for all the parameters except VOCs, one non-exposed and non-transported on-site sampler per purchased package and per shipment (i.e., a lot blank) was sent to the central laboratory and analyzed. Some duplicates were sampled in a limited number of buildings for method validation purposes. The VOC, aldehyde, O_3, and NO_2 concentrations were all expressed in μg m^{-3} after adjustment of the sampling rates on temperature and subtraction of the corresponding field blank value. The sampling rates were adjusted based on the average temperature measured during the sampling period at each sampling site and using the equations provided by the manufacturers.

The $PM_{2.5}$ sampling and gravimetric analysis were performed following the EN 12341: 2014 standard. This procedure has already been extensively described elsewhere (Mihucz et al., 2015; Szigeti et al., 2016).

2.5. Description of the investigated office rooms

All of the investigated rooms were described with respect to general information (surface, ceiling height, surface of windows, number of occupants, etc.); building materials (floor, walls and ceiling); furniture; equipment (computers, printers, humidifiers, etc.); heating, ventilating and air-conditioning (HVAC) systems; operability of windows; and activities within the room (use of products such as glues, white boards, air fresheners, cleaning products, etc.). This information was collected by the project staff members accompanied by a facility manager during the sampling week using a checklist.

2.6. Data analysis

The concentrations below the LODs and LOQs were set at LOD/2 and LOQ/2, respectively, to calculate the indoor concentration distributions. For VOCs and aldehydes, when available, raw values given by the laboratory were kept for the statistical analyses.

The seasonal variabilities were studied with the Wilcoxon matched pairs test (non-parametric test; repeated measurements). This analysis was conducted using the XLSTAT 2014 software (Addinsoft, Paris, France).

The spatial variability of air pollutants in office buildings was determined through the calculations of intra-class correlation (ICC) coefficients with a two-stage model: office level and building level. The model is given by

$$y_{ij} = \mu_Y + u_{oj} + \varepsilon_{ij} \quad (1)$$

where $i = 1, \ldots, r$ and $j = 1, \ldots, b$ with r the number of rooms and b the number of buildings, and y_{ij} is the measured concentration of compound Y in room i within building j, μ_Y is the true mean concentration, u_{oj} is the deviation from μ_Y due to the effect of building j, and ε_{ij} is the deviation from building j due to the effect of room i. The variance components are $var\,(\varepsilon_{ij}) = \sigma_\varepsilon^2$ and $var\,(u_{oj}) = \sigma_u^2$. σ_ε^2 represents the variance between rooms, and σ_u^2 represents the variance between buildings. The ICC coefficients were calculated as follows:

$$ICC = \frac{\sigma_u^2}{\sigma_u^2 + \sigma_\varepsilon^2} \quad (2)$$

Furthermore, to study the influence of the floor level, this variable was included in the 'empty model', after having been centered. The model is given by

$$y_{ij} = \beta_{0j} + \beta_1 \times \text{floor level}_{ij} + \varepsilon_{ij} \quad (3)$$

where $i = 1, \ldots, r$ and $j = 1, \ldots, b$ with r the number of rooms and b the number of buildings, β_{0j} is the mean concentration in building j, and β_1 is the deviation due to the fixed effect of the floor level. The normality of the log-transformed concentrations was checked for each pollutant in each season using the Shapiro-Wilk test. These analyses were performed with SAS version 9.4 (SAS Institute, NC, Cary, USA) using the 'MIXED' procedure. The statistical analyses relative to the investigation of the spatial and seasonal variations were carried out only for pollutants with >50% of the indoor concentrations above the respective LOQs.

3. Results and discussion

3.1. Building and room characteristics

A total of 148 rooms in 37 office buildings were investigated. All rooms were occupied during the measurements, with a median number of 7 occupants. The median floor area of the rooms was 64 m^2 (standard deviation, SD: 129 m^2), with the smallest being 8.5 m^2 and the largest being an open space of 600 m^2. The median surface available for each worker was 11 m^2. The majority of the rooms were open spaces (n = 101, 68%) and to a lesser extent, cellular offices (n = 47; 32%). About one-third of the rooms were close, i.e., within 50 m, to a road with heavy traffic (n = 57; 39%). The majority of the rooms had carpet on the floor (n = 76; 51%) or synthetic smooth floor coverings (n = 44; 30%), painted walls (n = 112; 76%), and mineral fiber tiles (n = 77; 52%) or exposed concrete/plaster (n = 28; 19%) on the ceiling. None of the rooms were naturally ventilated. Nearly all rooms were equipped with a balanced mechanical ventilation system (n = 137; 92%); some had only exhaust (n = 7; 5%) or only supply (n = 4; 3%). In addition, the windows were not openable in 33 rooms (22%). Nearly all rooms were equipped with a cooling system (n = 141; 95%). One or more printers or copiers were present in 96 rooms (65%). The use of air fresheners was reported in one room.

3.2. Indoor air quality

The percentages of concentration values above the LODs and LOQs as well as the VOC, aldehyde, O_3 and NO_2 concentration distributions are reported in Table 2a (summer) and Table 2b (winter) for the whole set of buildings. The mean concentrations at the country level and statistical differences between countries are provided in Tables S2a and S2b. Results for indoor temperature and relative humidity are reported in Table S3 for the whole set of buildings and in Tables S4a and S4b at the country level per season.

With the exception of trichloroethylene and tetrachloroethylene, all the target pollutants were detected in both seasons in >75% of the offices. Among VOCs, the highest median concentration was measured for toluene in summer (4.7 μg m^{-3}) and for D-limonene in winter (13 μg m^{-3}). In both seasons, formaldehyde represented the highest indoor concentration among the investigated aldehydes, with median values equal to 14 and 7.5 μg m^{-3} in summer and winter, respectively. A relatively high indoor concentration (290 μg m^{-3}) was measured for tetrachloroethylene in winter in one of the buildings. A dry-cleaning

Table 2a
Indoor concentrations (μg m^{-3}) of air pollutants monitored in office buildings in the frame of the OFFICAIR project during the summer campaign.

Pollutant	n (rooms)	% > LOD	% > LOQ	Min.	P5	Median	Mean	SD	P95	Max.
Benzene	146	99	72	<LOD	<LOQ	1.0	1.4	1.3	3.6	10
Toluene	146	99	92	<LOD	<LOQ	4.7	8.1	8.5	20	63
Xylenes	145	97	90	<LOD	<LOQ	2.5	3.8	5.1	8.3	40
Ethylbenzene	146	95	88	<LOD	<LOQ	1.1	1.8	2.0	4.1	14
n-Hexane	146	95	90	<LOD	<LOQ	1.4	1.9	1.6	4.5	9.1
Trichloroethylene	145	8	6	<LOD	<LOD	<LOD	<LOD	–	0.1	0.8
Tetrachloroethylene	145	20	13	<LOD	<LOD	<LOD	<LOQ	–	0.6	1.0
α-Pinene	146	96	96	<LOD	0.8	3.0	4.2	6.3	8.7	66
D-Limonene	146	99	95	<LOD	0.4	3.9	4.7	4.0	9.4	34
2-Butoxyethanol	146	98	93	<LOD	<LOQ	2.5	5.7	10	21	80
2-Ethylhexanol	145	99	93	<LOD	<LOQ	3.8	4.7	4.5	15	31
Styrene	146	99	92	<LOD	<LOQ	0.9	1.0	1.1	2.0	12
Formaldehyde	143	100	100	4.7	7.3	14	16	7.6	29	49
Acetaldehyde	143	100	99	<LOQ	3.5	6.1	6.4	2.2	10	16
Acrolein	143	98	95	<LOD	0.8	2.4	2.5	1.1	4.3	5.5
Propionaldehyde	143	99	98	<LOD	1.3	2.4	2.8	1.5	5.5	11
Benzaldehyde	143	100	88	<LOQ	<LOQ	0.9	1.0	0.5	1.9	4.9
Glutaraldehyde	143	100	59	<LOQ	<LOQ	1.1	1.3	0.8	3.0	3.9
Hexanal	143	100	100	4.6	5.7	10	11	5.0	18	35
Ozone	146	86	64	<LOD	<LOD	5.6	9.0	9.7	32	42
Nitrogen dioxide	112	100	100	2.7	6.8	16	16	5.1	24	29

P5: 5th percentile; P95: 95th percentile; Min.: minimum concentration; Max.: maximum concentration; LOD: limit of detection; LOQ: limit of quantification; SD: standard deviation.

shop was located on the ground floor, which is likely responsible for the high concentration. Indoor concentrations of the same order of magnitude were measured in the four offices investigated in this building located on the 4th and 5th floors.

$PM_{2.5}$ was sampled in a limited number of buildings: 16 in summer and 22 in winter. In summer, the indoor $PM_{2.5}$ concentrations ranged between 2.7 μg m^{-3} in Finland and 17 μg m^{-3} in Hungary (n = 16; median = 9.2 μg m^{-3}; mean and SD: 9.7 ± 4.6 μg m^{-3}). In winter, the indoor $PM_{2.5}$ concentrations ranged between 3.4 μg m^{-3} in Finland and 32 μg m^{-3} in Hungary (n = 22; median = 16 μg m^{-3}; mean and SD: 15 ± 8.4 μg m^{-3}).

A comparison of these observations with those of studies performed in offices worldwide is presented in Table 3. The comparison included studies with at least 10 buildings to avoid any specific situation and with measurements performed after 2000. In most of the studies, the measurements were performed in all seasons. The reported medians and means were compared with those observed in the OFFICAIR project.

Overall, the benzene, toluene, ethylbenzene, and xylene concentrations were lower in the OFFICAIR buildings; the α-pinene and D-limonene concentrations were higher; and the aldehyde, NO_2 and $PM_{2.5}$ concentrations were of the same order of magnitude. Due to the absence of data, no comparisons could be proposed for acrolein, benzaldehyde, glutaraldehyde, and O_3.

3.3. Seasonal variabilities

Significant seasonal differences ($p < 0.05$) were observed for all the target pollutants except for the xylenes ($p = 0.24$). For most of the pollutants, the indoor concentrations were significantly higher in summer than winter. Significantly lower concentrations were measured in summer compared to winter for benzene, α-pinene, D-limonene, and NO_2. These general trends at the European level were not always observed at the country level, as shown in Fig. 1 for formaldehyde, benzene, D-limonene, α-pinene, O_3 and NO_2.

Table 2b
Indoor concentrations (μg m^{-3}) of air pollutants monitored in office buildings in the frame of the OFFICAIR project during the winter campaign.

Pollutant	n (rooms)	% > LOD	% > LOQ	Min.	P5	Median	Mean	SD	P95	Max.
Benzene	137	100	93	<LOQ	<LOQ	1.7	2.1	1.7	5.3	8.9
Toluene	139	96	84	<LOD	<LOQ	3.1	6.1	8.8	25	62
Xylenes	138	92	81	<LOD	<LOD	2.2	3.3	3.7	14	20
Ethylbenzene	138	93	85	<LOD	<LOD	1.0	1.3	1.2	4.5	6.7
n-Hexane	139	96	93	<LOD	<LOQ	1.2	1.5	1.4	4.9	8.6
Trichloroethylene	130	13	12	<LOD	<LOD	<LOD	<LOQ	–	1.1	1.8
Tetrachloroethylene	135	64	38	<LOD	<LOD	<LOQ	8.2	45	4.8	290
α-Pinene	136	90	90	<LOD	<LOD	4.0	6.3	8.3	18	68
D-Limonene	139	100	98	<LOQ	1.2	13	19	18	57	81
2-Butoxyethanol	139	99	86	<LOD	<LOQ	0.4	2.7	6.8	15	43
2-Ethylhexanol	139	94	83	<LOD	<LOD	2.3	3.9	4.4	12	34
Styrene	138	100	75	<LOQ	<LOQ	0.5	0.8	0.7	1.9	5.6
Formaldehyde	140	100	100	1.7	3.3	7.5	8.1	4.1	16	23
Acetaldehyde	140	99	94	<LOD	<LOQ	4.5	4.9	2.0	8.3	12
Acrolein	140	99	64	<LOD	<LOQ	1.0	1.3	1.0	2.9	7.7
Propionaldehyde	140	100	92	<LOQ	<LOQ	1.2	1.4	0.9	2.8	5.7
Benzaldehyde	140	94	23	<LOD	<LOD	<LOQ	<LOQ	–	1.1	4.9
Glutaraldehyde	140	76	19	<LOD	<LOD	<LOQ	<LOQ	–	1.8	4.3
Hexanal	140	100	100	1.5	2.0	4.4	5.0	2.4	9.0	14
Ozone	140	96	46	<LOD	<LOQ	<LOQ	3.9	5.0	11	39
Nitrogen dioxide	128	100	100	4.8	8.7	18	18	6.7	30	39

P5: 5th percentile; P95: 95th percentile; Min.: minimum concentration; Max.: maximum concentration; LOD: limit of detection; LOQ: limit of quantification; SD: standard deviation.

Table 3
Indoor concentrations (μg m^{-3}) measured in office buildings in the frame of the OFFICAIR project compared to those measured in office buildings over the past decade.

Pollutant	Reference	#Buildings	n	Sampling period	Country	% > LOD	P5	Median	P95	AM (SD)	OFFICAIR
Benzene	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	100	n.a.	n.a.	n.a.	8.1	↓
	Geiss et al., 2011	52[b]	188	2003–2008[a]	European Union	n.a.	0.8	2.6	11.9	4.4	↓
Toluene	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	100	n.a.	n.a.	n.a.	110	↓
	Geiss et al., 2011	52[b]	188	2003–2008[a]	European Union	n.a.	1.7	7.1	48	13	↓
m-/p-Xylenes	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	100	n.a.	n.a.	n.a.	12	↓
	Geiss et al., 2011	52[b]	188	2003–2008[a]	European Union	n.a.	1.1	2.9	22	6.2	↓
o-Xylene	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	100	n.a.	n.a.	n.a.	9.6	↓
	Geiss et al., 2011	52[b]	188	2003–2008[a]	European Union	n.a.	0.6	1.2	7.1	2.2	↓
Ethylbenzene	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	100	n.a.	n.a.	n.a.	12	↓
	Geiss et al., 2011	52[b]	178	2003–2008[a]	European Union	n.a.	0.5	1.3	7.4	2.4	↓
Hexane	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	100	n.a.	n.a.	n.a.	7.5	↓
	Geiss et al., 2011	52[b]	134	2003–2008[a]	European Union	n.a.	0.5	1.7	7.6	3.0	↓
Trichloroethylene	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	79	n.a.	n.a.	n.a.	0.12	=
Tetrachloroethylene	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	85	n.a.	n.a.	n.a.	0.92	=
α-Pinene	Salonen et al., 2009b	176	520	2001–2006[a]	Finland	n.a.	n.a.	n.a.	n.a.	2.0	↑
	Geiss et al., 2011	52[b]	160	2003–2008[a]	European Union	n.a.	<LOD	1.5	12	3.2	↑
D-Limonene	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	82	n.a.	n.a.	n.a.	61	↑
	Salonen et al., 2009b	176	520	2001–2006[a]	Finland	n.a.	n.a.	n.a.	n.a.	4.0	↑
	Geiss et al., 2011	52[b]	179	2003–2008[a]	European Union	n.a.	0.3	2.6	33	9.4	↑
2-Butoxyethanol	Geiss et al., 2011	52[b]	74	2003–2008[a]	European Union	n.a.	<LOD	0.25	8.3	4.5	=
2-Ethylhexanol	Salonen et al., 2009b	176	520	2001–2006[a]	Finland	n.a.	n.a.	n.a.	n.a.	0.9	↑
Styrene	Ongwandee et al., 2011	17	68	2009[a]	Thailand (Bangkok)	100	n.a.	n.a.	n.a.	3.2	=
	Geiss et al., 2011	52[b]	128	2003–2008[a]	European Union	n.a.	<LOD	< LOD	2.4	0.2	=
Formaldehyde	Salonen et al., 2009b	176	520	2001–2006[a]	Finland	n.a.	n.a.	n.a.	n.a.	11	=
	Geiss et al., 2011	52[b]	185	2003–2008[a]	European Union	n.a.	4.5	14	32	17	=
Acetaldehyde	Geiss et al., 2011	52[b]	186	2003–2008[a]	European Union	n.a.	2.5	7.2	19	8.5	=
Propionaldehyde	Geiss et al., 2011	52[b]	185	2003–2008[a]	European Union	n.a.	0.8	2.3	9.1	3.0	=
Benzaldehyde	Salonen et al., 2009b	176	520	2001–2006[a]	Finland	n.a.	n.a.	n.a.	n.a.	1.5	=
Hexanal	Salonen et al., 2009b	176	520	2001–2006[a]	Finland	n.a.	n.a.	n.a.	n.a.	2.1	=
	Geiss et al., 2011	52[b]	134	2003–2008[a]	European Union	n.a.	3.0	11	40	16	=
Nitrogen dioxide	Mosqueron et al., 2002	62	62	Dec. 1999 to September 2000	France (Paris)	n.a.	n.a.	44	n.a.	45 (16)	=
$PM_{2.5}$	Horemans and Van Grieken, 2010	10	25	March to May 2008	Belgium	100%	n.a.	16	n.a.	15 (0.9)	=
	Mosqueron et al., 2002	55	55	Dec. 1999 to September 2000	France (Paris)	n.a.	n.a.	26	n.a.	35 (39)	=

n: number of sampling locations; P5: 5th percentile; P95: 95th percentile; LOD: limit of detection; AM: arithmetic mean; SD: standard deviation; n.a.: not available; ↓/↑ indicates that a lower/higher indoor concentration was measured within OFFICAIR and the '=' sign indicates that the indoor concentration measured within OFFICAIR was of the same order of magnitude. The buildings studied by Ongwandee et al. (2011) were all mechanically ventilated. Horemans and Van Grieken (2010) instrumented only naturally ventilated buildings. The type of ventilation system was not reported in the other studies.

[a] Sampling all over the year(s).

[b] The results reported from AIRMEX (Geiss et al., 2011) do not distinguish office buildings and schools.

These seasonal trends had never been shown for 'modern' office buildings. They remain consistent with the observation of Konopinski (1985) for formaldehyde in one office building in the US. They are also consistent with what was already observed in schools for benzene and formaldehyde (Canha et al., 2016), and terpenes (Chatzidiakou et al., 2014), as well as in dwellings for benzene (Langer et al., 2016), formaldehyde (Salthammer et al., 2010), terpenes (Schlink et al., 2010), O_3 (Cattaneo et al., 2011), and NO_2 (Cattaneo et al., 2011; WHO, 2010). Even if office buildings are expected to be more controlled environments due to the presence of HVAC systems compared to schools and dwellings, the season nevertheless shows an effect. In summer, the higher temperature leads to increased material emissions indoors, e.g., formaldehyde emissions. In addition, higher outdoor O_3 concentrations (due to the photochemical production from increased ultraviolet radiation) penetrate inside the building through window openings and lead to the ozone-initiated formation of formaldehyde involving terpenes, e.g., D-limonene (Nørgaard et al., 2014; Weschler, 2006). In winter, terpenes are less involved in ozone-initiated reactions and are observed at higher concentrations. Still, in winter, higher benzene and NO_2 emission rates from combustion sources such as heating systems combined with a higher atmospheric stability (low mixing layer height and low wind speed) occur outdoors and impact IAQ. These results are important because they indicate that, on a year time frame, a spot measurement during one working week may be inadequate to characterize the long-term exposure of office workers within a building.

3.4. Spatial variabilities

The homogeneity of the IAQ within a given building in comparison with the overall variability of IAQ between office buildings was assessed using the ICC calculations for both seasons, see Table 4. For a given indoor air pollutant, the concentrations in office rooms are considered to be correlated (i.e., the variance between rooms is lower than the variance between buildings) when the ICC is above 60%; conversely, the concentrations are considered heterogeneous (i.e., the variance between rooms is higher than the variance between buildings) when the ICC is below 40%.

The ICCs show different ranges depending on the pollutant and the season. For benzene, O_3 and NO_2, whose main sources are outdoor ones (no combustion sources in offices in contrast to domestic environments), the concentrations in the different locations in a building appear to be homogeneous, showing a similar impact of outdoor air quality. Regardless of the location of the rooms, they are all impacted by the outdoor environment (ICCs >60% at both seasons). For VOCs associated with the use of products, furniture or building materials, such as α-pinene, D-limonene, 2-ethylhexanol (Missia et al., 2010; Nazaroff and Weschler, 2004) that may vary from one room to another depending on the decoration and occupant habits and activities, the indoor concentrations differ more within a given building than between the office buildings (ICCs <40% at both seasons).

Table 5 reports the five pollutants for which the influence of the floor level on the indoor concentration could be studied. For the other

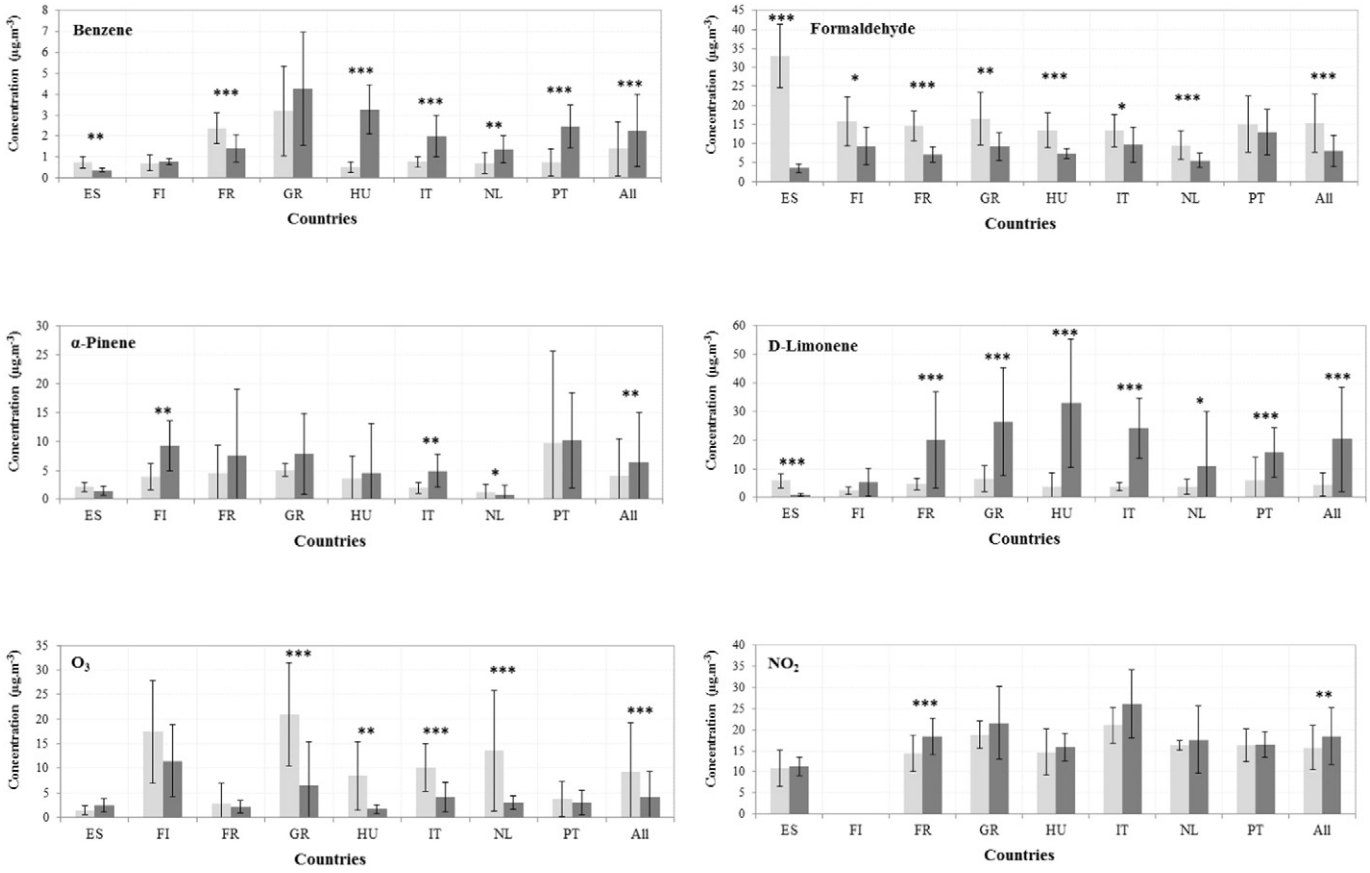

Fig. 1. Summer and winter mean concentrations and standard deviations (μg m^{-3}) of selected indoor air pollutants monitored in office buildings in the frame of the OFFICAIR project. Light grey: summer campaign, dark grey: winter campaign; ES: Spain, FI: Finland, FR: France; GR: Greece, HU: Hungary, IT: Italy, NL: the Netherlands and PT: Portugal; *: $p < 0.05$, **: $p < 0.01$ and *** $p < 0.001$ indicate statistical differences between seasons in the Wilcoxon matched pair test.

pollutants, either <50% of the indoor concentrations were above the respective LOQ or the log-transformed concentrations were not normal.

Table 5 shows the significant association ($p < 0.05$) between the floor level and acetaldehyde, hexanal, and NO_2 indoor concentrations averaged between summer and winter. No influence of the floor level was observed for a given season. The aldehyde average concentrations tend to increase by 4–5% on average with every floor level increase, and the average NO_2 concentration tends to decrease by 3% on average with every floor level increase. For NO_2, considering that most of the investigated rooms had openable windows, the influence of floor level can be explained by the distance to the outdoor traffic.

3.5. Evaluation of IAQ in relation to potential adverse health effects

An evaluation of IAQ in terms of potential adverse health effects can be performed based on a comparison of the results of this study with WHO air quality guidelines or estimated thresholds for irritative and respiratory effects. In this context, indoor concentrations derived from 5-day measurements were evaluated for formaldehyde, acrolein, α-pinene, D-limonene, O_3 and $PM_{2.5}$. This evaluation can only be considered preliminary due to the difficulties in assessing both the acute irritative effects from the mean values obtained for samples collected for 5 days and the long-term respiratory effects from 5 days of continuous monitoring without interruption outside of office hours.

3.5.1. Formaldehyde

The median formaldehyde concentrations during the summer (14 μg m^{-3}) and winter (8 μg m^{-3}) were below the WHO Indoor Air Quality Guideline (IAQG) of 100 μg m^{-3} derived in 2010 based on sensory irritation as the critical effect (WHO, 2010). The same conclusion can be reached for the maximum concentrations reported, 49 and 23 μg m^{-3} for summer and winter, respectively. The WHO guideline value is a health-based limit of exposure considered protective against both acute and long-term effects of formaldehyde exposure (sensory irritation, effects on lung function, nasopharyngeal cancer and myeloid leukemia). Nevertheless, it refers to 30-min average concentrations, which means that it should not be exceeded during any 30-min interval during the day. The WHO (2010) IAQG is supported by the assessment of Golden (2011), in which a formaldehyde indoor air limit of 0.1 ppm (0.125 μg m^{-3}) was considered to protect even particularly susceptible individuals from both irritation effects and any potential cancer hazard. Further, the value of 0.1 mg m^{-3} has been strengthened by Nielsen et al. (2013) following an evaluation of the scientific literature regarding formaldehyde toxicity since the WHO (2010) IAQG was published. However, the objective threshold for sensory irritation is above 800 μg m^{-3} for constant exposure (Mueller et al., 2013).

3.5.2. Acrolein

During the summer and winter campaigns, the median and maximum acrolein concentrations were below the Critical Exposure Limit (CEL) of 21 μg m^{-3} for short-term effects, derived within the EPHECT (emissions, exposure patterns and health effects of consumer products in the EU) project based on sensory irritation as the critical effect, and the CEL of 10 μg m^{-3} for long-term effects based on lesions in the nasal respiratory epithelium (Trantallidi et al., 2015). Nevertheless, elevated maximum concentrations were reported, reaching 26% CEL (short-term) and 55% CEL (long-term) during summer and 37% CEL (short-term) and 77% CEL (long-term) during winter. However, the

Table 4
Intra-class correlation coefficients (ICCs) for the air pollutants measured in office buildings in the frame of the OFFICAIR project (two-stage model).

Pollutant	Summer campaign	Winter campaign	Both seasons
Benzene	**66**	**90**	**88**
Toluene	53	46	55
Xylenes	*27*	**81**	56
Ethylbenzene	*27*	**74**	49
n–Hexane	49	58	60
α–Pinene	*18*	*23*	*32*
D–Limonene	*9*	43	44
2–Butoxyethanol	50	**88**	**82**
2–Ethylhexanol	*38*	*36*	*35*
Styrene	*20*	55	41
Formaldehyde	**72**	**82**	**73**
Acetaldehyde	56	**72**	58
Acrolein	**79**	*38*	**66**
Propionaldehyde	36	**68**	43
Benzaldehyde	46	*18*	*35*
Glutaraldehyde	58	55	51
Hexanal	54	52	60
Ozone	**87**	**70**	**89**
Nitrogen dioxide	**77**	**83**	**82**

Bold ICC: >60%, concentrations are considered to be homogeneous within a building; Italic and underlined ICC: <40%; concentrations are considered to be heterogeneous within a building; Grey cells: 40% < ICC < 60%, no conclusion can be drawn. ICCs were not calculated for $PM_{2.5}$ due to the small data set.

well-known difficulty of analyzing acrolein must be kept in mind (Herrington and Hays, 2012).

3.5.3. α-Pinene

The 5-day median and maximum concentrations reported during the summer and winter campaign were considerably below the short-term CEL of 45 mg m^{-3} developed within the EPHECT project based on sensory irritation as the critical effect (Trantallidi et al., 2015). For the evaluation of long-term effects of α-pinene exposure, a comparison with the Guide Value II of 2 mg m^{-3} set by the German Indoor Air Hygiene Commission for respiratory tract inflammation (Sagunski and Heinzow, 2003) can be performed but only in terms of order of magnitude. Specifically, this health-based limit of exposure was derived by incorporating an Assessment Factor (AF) of 2 for children, which is not applicable in the case of this study because it focuses on the health of office workers.

3.5.4. D-Limonene

Median D-limonene concentrations during the summer (4 μg m^{-3}) and winter (13 μg m^{-3}) were well below the short-term CEL of 90 mg m^{-3} derived in the framework of the EPHECT project based on sensory irritation (Trantallidi et al., 2015). Similarly, this conclusion can be stated for the maximum concentrations of 34 and 81 μg m^{-3} reported for the summer and winter campaign, respectively. For the evaluation of long-term effects of D-limonene exposure, the previously mentioned 5-day median and maximum concentrations were lower than the Derived No Effect Level (DNEL) of 3.6 mg m^{-3}, chosen by Petry et al. (2014) for the purposes of a health risk evaluation of selected VOCs emitted from scented candles. This long-term limit value was derived on the basis of a subacute inhalation toxicity study of aerosolized D-limonene in rats and default assessment factors according to European Chemicals Agency REACH Guidance documents (ECHA, 2012).

3.5.5. Ozone

Median 5-day O_3 concentrations both for the summer and the winter were below the WHO Air Quality Guideline (AQG) of 100 μg m^{-3} for lung effects (daily maximum 8-h mean) (WHO, 2005). The maximum concentrations reported were also below this limit, reaching 42% and 39% of the AQG for the summer and winter, respectively. This AQG is considered to provide adequate protection for public health, although some health effects may occur below 100 μg m^{-3}. No guideline value can be specified for O_3 that fully protects human health (WHO, 2005). However, this AQG is an ambient guideline value; epidemiological studies have also been taken into account for its derivation. According to animal studies and in accordance with human exposure studies, sensory irritation would be expected at >0.1 ppm (200 μg m^{-3}) O_3 at high D-limonene concentrations (Wolkoff et al., 2012).

3.5.6. $PM_{2.5}$

As a preliminary approach in the evaluation of $PM_{2.5}$ concentrations, the ambient WHO AQG of 25 μg m^{-3} (24-h mean) and 10 μg m^{-3} (annual mean) for respiratory and cardiovascular effects can be used (WHO, 2005). The WHO notes that complete protection for every individual against all possible adverse health effects of PM is unlikely to be guaranteed by any limit value due to the substantial inter-individual variability in exposure and in the response to a given exposure; therefore, the lowest concentrations possible in the context of local constraints, capabilities and public health priorities should be achieved (WHO, 2005). Additionally, the EUROPART European interdisciplinary group of researchers concluded for indoor particles that "there is inadequate scientific evidence for establishing limit values or guidelines for particulate mass or number concentrations" (Schneider et al., 2003). Furthermore, indoor generated particles (e.g., secondary organic aerosols) have different composition and morphology than ambient (combustion/traffic) particles (WHO, 2005). Therefore, comparison with the WHO AQG should be cautious. In this context, the median 5-day $PM_{2.5}$ concentrations, 9 and 16 μg m^{-3} for the summer and winter, respectively, were below the 24-h limit but exceeded the annual one. The maximum concentrations, 17 and 32 μg m^{-3} in summer and winter, respectively, appeared high compared to both AQGs.

Table 5
Influence of the floor level on indoor air concentrations (two-stage model; *p*-values in brackets).

Pollutant	Summer campaign		Winter campaign		Both campaigns	
	Intercept	Floor level	Intercept	Floor level	Intercept	Floor level
Acetaldehyde	/		/		**1.6 (< 0.0001)**	**0.04 (0.0017)**
Acrolein	/		−0.01 (0.9395)	0.06 (0.1556)	/	
Propionaldehyde	/		0.17 (0.0343)	0.04 (0.0918)	/	
Hexanal	/		0.05 (0.6917)	0.03 (0.3755)	**2.0 (<0.0001)**	**0.05 (0.0377)**
Nitrogen dioxide	/		/		**2.8 (<0.0001)**	**−0.03 (0.0412)**

"/" means that the normality of the log-transformed concentration residues was not verified. Statistical differences between floor levels are bold.

3.6. Strengths and limitations

The strength of the study was the large set of data collected from a large number of European office buildings. These buildings were selected on a voluntary basis; the degree of representativeness of the studied sample regarding the stock of 'modern' office buildings in Europe cannot be assessed.

The IAQ evaluation can only be considered preliminary. Limitations are evident due to the following aspects: (1) short-term, i.e., hourly, exposure data are needed to assess acute effects; (2) the sampling of indoor air pollutants included time outside of the office hours; (3) 5-day sampling may not reflect a 'long-term' situation; and (4) in the case of O_3 and $PM_{2.5,}$ comparison of the exposure data in offices was performed with ambient AQGs in the absence of IAQGs or other adequate thresholds for respiratory effects. Furthermore, regarding PM, the number of sampling sites was limited. Moreover, the health significance of increased exposure is difficult to assess, as it depends on the toxicity of the particles.

4. Conclusions

The OFFICAIR project provides a reference dataset for indoor concentrations obtained from 5-day sampling (approximately 100 h) in 37 'modern' mechanically-ventilated office buildings in 8 European countries. The indoor concentrations in summer and winter varied significantly for all the pollutants except xylenes, with some concentrations being higher in summer (e.g., aldehydes and O_3) or in winter (benzene, terpenes, and NO_2). The terpene and 2-ethylhexanol indoor concentrations also varied markedly between rooms within a building. These results are useful for the development of sampling strategies in the context of building certification or of checking compliance with IAQ guideline values. Sampling during at least two different seasons and on the ground floor and an upper floor are recommended.

Regarding potential adverse health effects, the 5-day median and maximum indoor air concentrations of formaldehyde and ozone were lower than their respective WHO indoor and ambient air quality guidelines; those of acrolein, α-pinene, and D-limonene were lower than their estimated thresholds for irritative and respiratory effects; and the indoor concentrations of $PM_{2.5}$ appeared high when compared to the 24-h and annual WHO AQGs.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.scitotenv.2016.10.238.

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