Title: Indoor and outdoor air quality assessment in daycare centers in Ghent (Belgium) in view of outdoor sleeping in an urban environment

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Abstract

Within Flanders, there is an increasing trend to let children sleep outdoors while in daycare. However, within an urban environment, the densely spread emission sources might affect the air quality and possibly limit the areas where outdoor sleeping is favourable. Nevertheless, there is a lack of data regarding the atmospheric pollution levels in and around daycare centers (DCC). Therefore, the focus of this study is to chemically characterize the air quality indoors, outdoors, and in specifically designed cubicles for outdoor sleeping at 12 DCCs spread over the city of Ghent (Belgium). The measuring of a very broad range of different pollutants, provides unique data for indoor and outdoor air quality at daycare centers in Ghent. The use of axial tube (for volatile organic compounds, VOCs) and Radiello (for NO_2, SO_2, O_3) passive samplers enables multi-component sampling, resulting in time-weighted average concentrations for one week. Forty-seven VOCs are identified and, for the majority (40), indoor to outdoor concentration (I/O) ratios higher than one are found. For the remaining seven compounds (e.g. benzene) outdoor concentrations are a factor 1.3-17.9 (median) higher than indoors. Median indoor TVOC concentrations are 152 µg/m^3 and 142 µg/m^3 for the September and January campaign, respectively. Outdoors, these median TVOC concentrations are much lower (24.5 µg/m^3 and 30.6 µg/m^3). For NO_2, no noticeable differences are observed between average indoor and outdoor concentrations (indoors 12 ± 3 and 13 ± 2 µg/m^3 for the September and January campaign, respectively, and outdoors 11 ± 3 and 14 ± 4 µg/m^3). The highest (outdoor) measured concentration is 21 ± 1 µg/m^3. SO_2 concentrations are below 2.62 µg/m^3 (LOQ). Average indoor O_3 concentrations are 4 ± 3 µg/m^3 and 2 ± 2 µg/m^3 for the September and January campaign, respectively. Much higher values are measured outdoors (46 ± 3 µg/m^3 and 40 ± 11 µg/m^3), but the concentrations stayed well below legal standards.

Keywords*: indoor air quality, outdoor air quality, daycare centers, volatile organic compounds, NO_2, SO_2, O_3, TD-GC-MS, PTR-QiToF-MS*

1. Introduction

There is a growing awareness of the adverse health effects of air pollution (both indoors and outdoors) following both acute and chronic exposure, with rapidly expanding evidence linking air pollution with an increased risk of respiratory and cardiovascular diseases (Clark et al., 2020) (Mannucci et al., 2015). Children are more susceptible to the effects of air pollution than adults due to their higher breathing frequency, immature immune and respiratory system, and higher volume of inhaled air per body weight. For example, asthma and allergies are very common diseases in childhood, and indoor exposure to volatile organic compounds (VOCs) can increase the occurrence (Jia et al., 2019) (Madureira et al., 2015) (Salvi, 2007). Since people spend more than 80% of their time indoors, good indoor air quality (IAQ) in e.g. schools, offices, homes and daycare centres is an important determinant of human health and people's well-being (Cakmak et al., 2014) (Hoge Gezondheidsraad, 2017) (WHO, 2010).

Outdoor EU air quality standards for the protection of human health – e.g. concentrations of particulate matter (PM), ozone (O_3), and nitrogen dioxide (NO_2) – are currently not met in large parts of the EU. This is particularly true for urban areas and is mainly attributed to the high level of emissions from road traffic and residential combustion (European Environment Agency, 2019). Exposure to traffic-related air pollution during infancy has shown adverse, possibly long-lasting effects on lung function (Schultz et al., 2016). Industry (e.g. for 49% of SO_2 emissions), traffic (e.g. 63% of the NO_x emissions) and households (e.g. 18% of NMVOCs emissions) are major contributors to air pollution in Flanders. The production of energy (e.g. 32% of the SO_2 emissions), non-road vehicles, land-use changes, agri- and horticulture (e.g. 95% of the NH_3 emissions), etc. are also sources of outdoor air pollution (VMM, 2018).

This outdoor air pollution should be kept in mind when talking about the habit of letting children sleep outdoors during the daytime, originating from Scandinavian countries (Berti et al., 2020). The initial aim to prevent diseases and reduce the high infant mortality is nowadays replaced by the stimulation of well-being of children and families, and daycare centres (DCCs) in other European countries are starting to take this on as well. There is scarce scientific knowledge about the advantages and disadvantages, but the experiences of parents show that children take longer naps outdoors than indoors and that infants are livelier after sleeping outdoors (Tourula et al., 2008) (Tourula et al., 2010). Sleep is an essential element of health and affects the well-being of the whole family (Nevarez et al., 2010) (Tourula, 2011). However, the impact of the environment (air quality) must also be taken into account, especially in urban areas, where more than 70% of the EU population lives (European Environment Agency, 2019). Therefore, it is important to evaluate whether sleeping outdoors is not exposing the children to higher concentrations of air pollutants when compared to the traditional indoor bedrooms.

The focus of this study is to investigate the group of VOCs in detail, both in indoor as well as in outdoor air, by a systematic approach. In-depth untargeted screening of all samples is performed, followed by the quantification of the 47 identified VOCs. In addition, the exposure of children to the inorganic gaseous pollutants NO_2, SO_2 and O_3 was also investigated. These multi-component measurements took place in two sampling campaigns (September 2019 and January 2020) at twelve DCCs in the city of Ghent, Belgium. VOCs represent an important, large and diverse class of indoor air pollutants as their concentration levels are frequently elevated indoors (Cakmak et al., 2014) (Walgraeve et al., 2011). It is paramount to determine the presence and concentration of individual VOCs, as some are more problematic than others. NO_2 is one of the most common air pollutants in ambient and indoor air and just like the secondary pollutant ozone, it is one of the most important pollutants in urban areas (Kornartit et al., 2010) (Melkonyan & Kuttler, 2012). Ambient NO_2 exposure may increase the risk of respiratory tract infections through the pollutant's interaction with the immune system (Chen et al., 2007). Well-established evidence has been produced for short-term effects, especially on respiratory and cardiovascular systems, associated to ozone exposure (Nuvolone et al., 2018). SO_2 is irritating when inhaled at high concentrations and can cause respiratory problems, especially in individuals suffering from asthma or chronic lung disease (VMM, 2019a).

This unique dataset is being created to answer research objectives such as which of the considered pollutants characteristic are for the indoor and outdoor air respectively (I/O ratios). Concentrations for the sampling campaign in September and January will be compared to each other and it will be investigated if there are notable differences between the locations. Measurements took also place in specially designed cubicles for outdoor sleeping, to investigate if there is an effect of the different bed configurations. These cubicles are partially closed from the environment, which makes it interesting to evaluate whether a difference in air quality between these outdoor beds and the ambient outdoor air exists. The materials of the bed themselves or the mattress/sheets can emit pollutants, and chemicals emitted by the child itself can possibly accumulate. The measurements of gaseous pollutants inside these cubicles are an unique aspect of this study.

2. Materials and methods

2.1 Sampling campaigns and locations

In Ghent (51°03’N 3°43’E), Belgium, two measuring campaigns (19-26 September 2019 and 14-21 January 2020) were set up at twelve DCCs (A-L) spread over the city (Figure A1). Table A1 provides more information about the ventilation systems and environment of the DCCs. Three types of outdoor sleeping can be distinguished: small plastic camp beds, wooden indoor beds installed under a large roof, and outdoor sleeping cubicles (Kids-F, NL) (Figure A2). By using passive sampling, week-average concentrations were obtained. Passive sampling offers advantages as there is no need for electricity and longer sampling periods are possible (Sarigiannis et al., 2011).

NO_2, SO_2, and O_3 were measured with Radiello samplers (Salvatore Maugeri Foundation, Padova, Italy) indoors in the living area, outdoors, and inside the outdoor cubicles where available (three locations (DCC A, B, and H) use specific outdoor sleeping cubicles). Axial samplers were deployed for VOC sampling in parallel to the Radiello samplers. Additionally, one VOC sampler was installed in the sleeping room at each location (Table 1). In the bedroom, no NO_2/SO_2 and O_3 samplers were placed since no (other) sources than in de living area are expected here. This resulted per sampling campaign in 39 VOCs samplers and two times (NO_2/SO_2 and O_3) 27 Radiello samplers in total. The outdoor samplers are positioned in such a way that they are representative for the air that children would breathe while sleeping outside (i.e. at the place where the DCCs offers already outdoor sleeping (6 out of the 12 DCCs at the moment of the sampling campaign), or where they are plan to do so in the future). An overview of the outdoor sampling positions is given in the supplementary part (Table A1).

Table 1: Overview of the sampler locations. x = measured at the twelve DCCs. Outdoor cubicles specifically designed for outdoor sleeping are present at DCC A, B, and H.

	Indoors		**Outdoors**	
	Living area	**Sleeping room**	**Outdoors**	**Outdoor cubicles**
VOCs	x	x	x	DCC A, B, H
NO_2/SO_2 and O_3	x		x	DCC A, B, H

2.2 Axial tube passive sampling and analysis of VOCs

2.2.1 Sample preparation

For the sampling of VOCs, Markes stainless steel sorbent tubes (O.D.: ¼’’; length: 3½’’; I.D.: 5 mm) filled with ±200 mg of Tenax TA (35/60 mesh) (Markes, Llantrisant, United Kingdom) were used. The tubes were first conditioned at 300°C for 1 h with a Helium flow of minimum 50 mL/min flowing through each tube. Clean and dry He gas ([H_2O] <20 ppb$_v$; [O_2] <10 ppb$_v$; [C_xH_y] <100 ppb$_v$; [CO/CO_2] <500 ppb$_v$; [N_2] <1 ppb$_v$) was provided by Air Products (Diegem, Belgium). The tubes were loaded with a gaseous internal standard (IS; perdeuterated toluene (Tol-d8; min. 99.96 atom. %D; Sigma Aldrich)), prepared by a two-phase system according to the procedure described by (Demeestere et al., 2008) (see supplementary information). The tubes were loaded with 10.83 ng Tol-d8.

2.2.2 TD-GC-MS analysis

In total, 39 VOC sampling tubes (3 tubes at each location (outdoor, sleeping room, living area) and 3 locations with an extra sample for the outdoor bed) were passively exposed for one week by using aluminium diffusion caps. After exposure, the samples were immediately closed with ¼’’ brass caps, wrapped into aluminium foil, and stored (for max. 14 days) in a desiccator before analysis using TD-GC-MS. To allow for blank correction, 8 and 11 extra sampling tubes for the sampling campaign in September and January respectively were prepared, loaded with Tol-d8 and kept closed at both ends with ¼” brass caps until analysis.

The desorption of the analytes preconcentrated on the Tenax TA sorbent tubes was performed in a Unity Series 2 Thermal Desorption system (Markes International, Llantrisant, UK) at 260°C under a He flow of 20 mL/min for 10 min. The analytes were refocussed on a microtrap cooled at -10°C and filled with 29.0 mg of Tenax TA 35/60 and 28.3 mg of Carbograph 1TD 40/60 sorbent. Next, the temperature of the microtrap was rapidly increased to 280°C and held for 3 min. The analytes were carried by a He flow and injected onto a 30 m FactorFour VF-1ms low bleed bounded phase capillary GC column (Varian, Sint-Katelijne-Waver, Belgium; 100% polydimethylsiloxane, internal diameter 0.25 mm, film thickness 1 µm), after splitting 5 mL/min of the He-flow. The column head pressure was set at a constant value of 50 kPa. The GC (Focus GC, Thermo Scientific, Milan, Italy) oven temperature was initially set at 35°C (for 10 min) and then gradually increased up to 240°C in four stages. First, a heating rate of 2°C/min was used to increase the temperature from 35°C to 60°C. Next, a temperature of 170°C was obtained by heating the GC oven at 8°C/min. Third, it was increased to 220°C at 15°C/min. Finally, at a heating rate of 15°C/min, a temperature of 240°C was reached and held for 10 min before cooling down to 35°C. The MS-transfer line was heated to 240°C. Masses from m/z 15 to 300 during the first 10 min and afterwards from m/z 29 to 300 were recorded in full scan mode (5 scans per sec) on a DSQ II Quadrupole MS (Thermo Scientific, Austin, TX, USA), hyphenated to the GC. Ionization was performed by electron ionization at 70 eV.

Chromatograms and mass spectra were processed using XCalibur (Thermo Scientific, version 2.2) and AMDIS (Automated Mass Spectral Deconvolution and Identification System, version 2.66) software. AMDIS was used for an untargeted screening of all samples to find and select the compounds present. Identification of VOCs was based on the comparison of the mass spectra with the US National Institute of Standards and Technology (NIST) library (NIST Mass Search Program for the NIST/EPA/National Institutes of Health (NIH) Mass Spectral Library, version 2.0) and the comparison of retention times with standards (see section 2.2.3). The data were quantitatively processed by using a set of characteristic ions, for each selected target compound (Table A2). Targets were only quantified if the signal to noise (S/N) ratio was 10 or higher (limit of quantification, LOQ), and a blank correction was done when the compound could be quantified in the blank (i.e. when S/N ratio ≥10) (Walgraeve et al., 2011).

2.2.3 Calibration and concentration calculation

Liquid calibration mixtures were prepared to calculate relative response factors (RRFs = (peak area analyte/mass analyte loaded on the tube) / (peak area Tol-d8/mass Tol-d8 loaded on the tube)) in accordance with the methodology described by Demeestere et al. (2008) and analysed just before or after the TD-GC-MS analysis of a series of samples. A total set of 84 frequently found VOCs (LC-MS grade), purchased at Acros Organics (Geel, Belgium) or Sigma-Aldrich (Bornem, Belgium), and all having a purity of at least 99.8%, were used as standard compounds. The VOCs were dissolved in two different 100 mL MeOH (Optima® LC-MS grade, Fisher Scientific) solutions (A and B), each containing 42 compounds at a concentration of ± 50 ng/µL of each VOC, including Tol-d8 as internal standard. 1 µL of stock solution A or B was loaded in threefold onto conditioned Tenax TA tubes with a NanoVolume Syringe (SGE Analytical Science), via a homemade injector system (150°C, 3 min) under a 100 mL/min He flow. An overview of the presence of the selected VOCs in calibration mixture A or B is given in Table A2.

The concentration (µg/m^3) of a VOC in the sampled air can be calculated by dividing the mass (ng) of the component in the sample (Eq. 1), by the amount of the sampled air. This volume is obtained by multiplying the exposure time of the corresponding sample (min) and the uptake rate (UR) (mL/min).

Effective Uptake Rates (UR_{eff}) determined by Walgraeve et al. (2011) were used where possible. Otherwise, the UR_{eff} was calculated by the empirical model of Jia et al. (2017), as class-average UR_{eff} or as an overall average UR_{eff}. Theoretical calculated URs (UR_{ideal}) may lead to underestimations (Walgraeve et al., 2011) and were therefore not applied. An overview of the used values for the URs, together with the presence of the selected compounds in the calibration mixtures (A or B) is given in Table A2.

$$m_{component,S} = \left(\frac{PA_{component,S}}{PA_{IS,S}} - \frac{PA_{component,blank}}{PA_{IS,blank}}\right) . \frac{m_{IS,S}}{RRF} \qquad (1)$$

After screening all the samples, 47 VOCs could be identified at the different locations. For 31 of them analytical standards could be obtained and a concentration calculation was done as described above. For the remaining 16 compounds, an average RRF (based on all the RRFS of the other 31 compounds, n = 31.3 = 93) of 0.6 ± 0.4 was used.

2.3 Radiello passive sampling and analysis of NO_2, SO_2, and O_3

Sampling of NO_2, SO_2 and O_3 was performed by Radiello diffusive sampling devices, using cartridges 166 (NO_2 and SO_2) and 172 (O_3), mounted in blue diffusive bodies (code 120-1). The Radiello cartridge 166 is made of microporous polyethylene coated with triethanolamine (TEA). Nitrogen and sulphur dioxides chemisorb as nitrite and sulphite/sulphate ions respectively. Cartridge 172 consists of a micropore polyethylene tube filled with silica gel coated with 1,2-di(4-pyridyl)ethylene. The latter undergoes an acid-catalysed ozonolysis when exposed to air containing ozone and leads to two molecules of 4-pyridyl aldehyde. Because of the temperature-dependent sampling rate values of NO_2 and O_3, the temperature was logged every 6 min with a Smartbutton ACR datalogger (ACR Systems Inc., Canada).

Sample treatment, calibration and calculations are performed as specified by Radiello (Instituti Clinici Scientifici Maugeri, 2006). Briefly, nitrite and sulphite/sulphate ions were extracted from the Radiello cartridge 166 with 5 mL milli-Q water and measured by ion chromatography using an ICS-90 Ion Chromatography System (Dionex), equipped with a 10 µL sample loop, an IonPac AS14A-5 µm analytical column (3 x 150 mm), a conductivity cell and an ion suppressor unit (AMMS III 2-mm, Dionex). The mobile phase was 8 mM Na_2CO_3 - 1 mM $NaHCO_3$ (Dionex), and 50 mN H_2SO_4 (95.0 - 98.0%, Sigma-Aldrich) was used as regenerant (both in milli-Q water). Every extract, together with two blank extracts (unexposed cartridges), was measured twice. Calibration of the instrument was done by injecting solutions of sodium nitrite ($NaNO_2$) (≥97.0% ACS reagent, Sigma Aldrich) and sodium sulphate (Na_2SO_4) (≥99% ACS reagent, Sigma Aldrich) in Milli Q water, with a concentration of the corresponding ions (NO_2^- and SO_4^{2-}) ranging from about 0.1 to 20 mg/L and from 0.25 to 50 mg/L, respectively. No standards were made for sodium sulphite (Na_2SO_3) because this was never measurable in the samples (direct/further oxidation to SO_4^{2-}). NO_2 and SO_2 concentrations calculation was done by using a sampling rate value Q_{298} of 0.141 ± 0.007 ng/ppb/min and 0.466 ± 0.022 ng/pbb/min, respectively, as indicated in the Radiello user manual (Instituti Clinici Scientifici Maugeri, 2006). The indicated errors corresponds to a 5% uncertainly on the final calculated concentrations.

Ozone concentrations were obtained by quantifying 4-pyridyl aldehyde by UV-VIS analysis, after derivatisation. A 3-methyl-2-benzothiazolinone hydrazone hydrochloride hydrate (MBTH) (97%, Sigma

Aldrich) solution was prepared by dissolving 5 g MBTH and 5 mL of concentrated sulphuric acid per L demineralised water.

Five mL of this solution was added to the samples and – after filtering through a 0.45 µm micropore filter membrane (Whatman syringe filter – 13 mm diameter disposable filter device) – an UV-1650 pc UV-VIS Spectrophotometer (Shimadzu) was used to measure the absorbance at 432 nm. Every sample, together with three blanks, was measured three times. Calibration was done by using 4-pyridyl aldehyde (Sigma Aldrich). Out of the MBTH-solution, 4.5 mL was added to 0.5 mL of different 4-pyridyl aldehyde solutions in a concentration range of 11.2 – 112 µg/mL.

2.4 PTR-QiToF-MS for source identification

Proton Transfer Reaction-Time of Flight Mass Spectrometry (PTR-QiToF-MS) was used for the headspace analysis of building materials obtained from DCC H to identify possible sources of detected VOCs. Samples of Noraplan Stone rubber top layer of the floor, the floor insulation Kingspan Kooltherm K3 and the wall insulation Kingspan Kooltherm K8 were put in a Nalophan bag filled with 15 L of N_2 (Alphagaz2, $N_2 \geq 99.9999\%$, Air Liquide) and, after 5 hours of equilibration, connected to the PTR-QiToF-MS (IONICON Analytik GmbH, Innsbruck, Austria) inlet to analyse the compounds in the headspace.

To analyse the headspace of the pure compound 2-chloropropane (see section 3.1.1.4), a dark penicillin bottle (119.78 mL, Novolab) was prepared with 3 mL of 2-chloropropane (Acros Organics) and after one night of equilibrating, 0.05 mL headspace of this bottle was added to a Nalophan bag filled with 7 L N_2. Blank correction for every measurement was done by subtracting the signal of a blank Nalophan bag filled with only N_2.

The operating parameters in the drift tube of the PTR-QiToF-MS device were set at a temperature of 80°C, pressure of 3.7 mbar, and a drift voltage of 1000 V, resulting in an electric field strength to gas number density ratio (E/N) of 144 Td (Townsend). Volatile compounds with a proton affinity higher than that of water (165.2 kcal/mol or 691 kJ/mol) were ionized via proton transfer reactions with H_3O^+ ions generated in the ion source of the instrument. The resulting product ions were separated in the time-of-flight region, according to their mass-to-charge (m/z) ratio and detected at their protonated mass. 1,3-diiodobenzene ($C_6H_5I^+$ (m/z = 203.943 and $C_6H_5I_2^+$ (m/z = 330.848)), continuously supplied by the built-in PerMaScal unit, was used as calibration gas for the mass scale (Venneman et al., 2020). The parameters of the PTR were set via 'PTR-Manager 3.0.1.208', those of the MS via the 'TPS PTRQ web interface'. PTR-QiToF-MS raw data were recorded by TofDAQ Viewer version 1.2.99 (Tofwerk AG, Thun, Switzerland) and the resulting data files were post-processed using PTR-MS Viewer version 3.2.12 (IONICON Analytik GmbH, Innsbruck, Austria).

3. Results and discussion

3.1 VOC concentrations at DCCs in Ghent

After screening all the samples, 47 VOCs could be identified at the different locations. For each sample, the Total Volatile Organic Compound (TVOC) concentration is calculated (Table A3) as the sum of the 31 VOCs for which a RRF is determined. The minimum, median and maximum concentrations are included in Table 2. The obtained indoor TVOC concentrations range from 61 µg/m^3 (DCC H) to 826 µg/m^3 (DCC G) and from 35 µg/m^3 (DCC C) to 623 µg/m^3 (DCC G) in the September and January sampling campaign, respectively. The Belgian Official Gazette reports an indoor guideline value of 300 µg/m^3 and intervention value of 1000 µg/m^3, valid for a chronic exposure time (>365 days, valid for lifetime exposure) (Federale Overheidsdienst Justitie, 2018). The outdoor TVOC (n=31) concentrations are on average a factor of 7 (between 2.5 and 21) lower than indoors (n=24, average concentration of the living area and the sleeping room, compared to the outdoor concentration, for September and January). The TVOC concentrations outdoors range between 16 µg/m^3 (DCC F) and 30 µg/m^3 (DCC G) (median 25 µg/m^3) in September and between 22 µg/m^3 (DCC K) and 36 µg/m^3 (DCC G) (median 31 µg/m^3) in January. Both indoors and outdoors, the highest TVOC concentrations are measured at DCC G, a newly constructed building (see section 3.1.1.3), located in the Old Docks and where construction works were still going on in the neighbourhood.

Table 2 also shows the overview of the minimum, median and maximum measured concentrations of the individual VOCs both indoors (living area and sleeping room) and outdoors. Standards for VOC-concentrations in non-industrial settings are scarce. Comparing the concentration levels with Flanders indoor guideline values and selected chronic reference values (RV) by the Superior Health Council of Belgium for the VOCs (Table 2), no exceedances were found (Federale Overheidsdienst Justitie, 2018) (Hoge Gezondheidsraad, 2017).
The Belgian Official Gazette reports an indoor intervention value for benzene (for a chronic exposure time (>365 days, valid for lifetime exposure)) of 0.4 µg/m^3 if the outdoor concentration is lower or equal to 0.4 µg/m^3, otherwise the intervention value is equal to the outdoor concentration (Federale Overheidsdienst Justitie, 2018). According to the WHO, no safe level of exposure can be recommended (WHO, 2010). However, the Superior Health Council of Belgium mentions a RV of 10 µg/m^3 for benzene provoking non-genotoxic effects (Hoge Gezondheidsraad, 2017). The indoor measured benzene concentrations, ranging from 0.39 µg/m^3 till 1.29 µg/m^3, often exceed 0.4 µg/m^3 but the corresponding outdoor concentrations (ranging from 0.52 µg/m^3 till 1.94 µg/m^3) were always higher (I/O ratios ranging from 0.53 till 1.0, median 0.74). Therefore, the intervention value is never exceeded. The larger outdoor concentrations indicate a (strong) outdoor source like e.g. traffic. Individual benzene concentrations can be found in Figure A3.
When comparing with other studies, the median indoor benzene concentration of 0.73 µg/m^3 (n=47) is well below the indoor median benzene concentrations found by Madureira et al. (2015) in classrooms in Porto (median 2.5 µg/m^3, n=73) and concentrations reported by Jia et al. (2019) in child care centres in Singapore (median 4.37 µg/m^3, n=32). The median benzene concentrations of five different IAQ studies (SAMI-LUX schools, Clean Air Low Energy Schools, BIBA schools, FLIES day-care centres, CRIPI nurseries) in Belgian schools and daycare centres were between 1.1 µg/m^3 and 5.0 µg/m^3 (Hoge Gezondheidsraad, 2017). Raysoni et al. (2017) report similar concentrations as found in our study (median indoor benzene concentrations between 0.31 µg/m^3 and 0.87 µg/m^3 for four different schools in Texas) (Raysoni et al., 2017). The median benzene concentration indoors in public buildings and schools in the frame of the AIRMEX project – measured during 2003-2008 in 11 cities over Europa – was 2.6 µg/m^3 (Geiss et al., 2011).

Forty of the 47 measured VOCs show I/O ratios higher than one or they are even not quantifiable outdoors (e.g. 2-(2-ethoxyethoxy)-ethanol), indicating the presence of (a) (strong) indoor source(s) for most of the VOCs (Figure A4). Possible indoor VOC sources are the use of cleaning products, air fresheners, fragrances, and many consumer products (e.g. emitting limonene and α-pinene), but also construction materials and furniture/decoration such as coatings, paints, floor materials and wood products (e.g. emitting BTEX) (Arı et al., 2020) (Chin et al., 2014) (Mendell, 2007). The presence of humans can also be a source of VOCs themselves. Breath is a significant source of pollutants emitted indoors (e.g. ethyl acetate, isopropanol, butanone) and a wide range of secondary pollutants can be formed by the reaction of human skin oils with ozone (Kruza & Carslaw, 2019) (Mazzatenta et al., 2021).
For the remaining seven VOCs (benzene, benzaldehyde, benzoic acid, acetophenone, dichloromethane, benzonitrile and phenol) larger concentrations outdoors compared to indoor were found (median I/O ratio <1), during both sampling campaigns (Figure A4).

3.1.1 An in-depth look at the VOCs present in the indoor air of the DCCs

The most abundant VOCs measured indoors are isopropanol, nonanal, and ethyl acetate (median concentration > 4 µg/m^3 for living area and sleeping room in both sampling campaigns), together counting for 20-85% (median 50%, n=47 samples) of the TVOC (n=31) concentration. These VOCs are found in washing and cleaning products as well as in air fresheners, cosmetics, and personal care products. Other possible sources are polishes and waxes, coatings or adhesives, and paints. Isopropanol is also frequently used as a disinfectant (ECHA, 2020) (Wade, 2018).

n-Undecane, benzaldehyde, limonene, 1-butanol and 2-butanone concentrations are generally also present at higher concentrations compared to the other VOCs (median value for at least one indoor data set > 3 µg/m^3), together counting for 7.5-68% (median = 21%, n=47) of the TVOC concentration.

3.1.1.1 Living area versus sleeping room

The TVOC concentrations of the living areas are higher than the corresponding values in the sleeping rooms at 10 of the 12 locations in September, and at 10 of the 11 locations in January (Table A3). The ratio of the concentration measured in the living area, relative to this of the corresponding sleeping room, is for 15 (September) and 24 (January) of the 47 VOCs in at least 75% of the datapoints above one (Figure 1). This can be explained by the diversity of activities that take place in the living areas (e.g. changing diapers, eating, washing hands). For all remaining VOCs, the ratio of ‘one’ falls between the 25th and 75th percentile, indicating an overall similar concentration in both areas.

Table 2: VOC concentrations [µg/m³]: minimum, median and maximum values both indoors (living area and sleeping room) and outdoors for September and January. In total, 47 VOCs could be identified. Only data for the 31 for which RRFs are determined, are shown in this table, together with the TVOC concentration (sum 31 VOCs). Guidelines: (1) Flanders indoor guideline values applicable to a chronic exposure time (>365 days, valid for lifetime exposure) and (2) intervention value reported by the Belgian Official Gazette (Federale Overheidsdienst Justitie, 2018); (3) Selected chronic RVs by the Superior Health Council of Belgium for some substances provoking non-genotoxic effects (Hoge Gezondheidsraad, 2017); (*) C_9-C_{14}-alkanes 250 µg/m³; C_4-C_{11}-aldehydes 650 µg/m³.

	Concentration [µg/m³] September 2019									**Concentration [µg/m³] January 2020**									**Guidelines [µg/m³]**	
	Living area (n=12)			Sleeping room (n=12)			Outdoors (n=12)			Living area (n=12)			Sleeping room (n=11)			Outdoors (n=12)				
Compound	Min	Med	Max	Min	Med	Max	Min	Med	Max	Min	Med	Max	Min	Med	Max	Min	Med	Max	(1)	(3)
Alkanes																				
n-hexane	0.11	1.15	27.7	0.62	1.03	2.59	<LOQ	0.62	0.96	1.02	1.35	4.44	0.94	1.33	5.73	0.94	1.38	2.79		700
methylcyclopentane	0.26	0.30	7.43	0.20	0.30	21.2	0.16	0.27	0.71	0.42	0.51	24.1	0.34	0.49	16.8	0.39	0.50	2.01		
cyclohexane	0.29	0.52	43.8	0.31	0.50	123	0.18	0.28	0.76	0.69	0.98	153	0.39	0.66	100	037	0.49	1.19		6000
n-heptane	0.53	0.71	5.85	0.51	0.63	16.2	0.18	0.32	0.46	0.38	0.50	13.3	0.36	0.47	9.29	0.27	0.31	0.68		21000
n-octane	0.95	1.89	2.78	0.99	1.62	4.14	0.11	0.22	0.47	0.51	1.07	2.08	0.39	1.02	2.12	0.14	0.17	0.32		
n-nonane	0.24	0.63	1.10	0.36	0.57	1.30	0.07	0.14	0.24	0.22	0.40	0.59	0.23	0.33	0.77	0.10	0.11	0.25	250* (C_9-C_{14})	
n-decane	0.71	1.47	44.9	0.53	1.31	18.1	0.17	0.23	0.45	0.71	1.49	30.7	0.45	0.72	15.1	0.19	0.23	0.37		
n-undecane	0.42	2.39	18.4	0.35	1.58	18.4	0.08	0.18	0.99	0.55	3.64	12.0	0.34	1.62	10.7	0.09	0.15	0.28		
n-dodecane	0.78	1.71	13.2	0.43	1.32	10.6	0.03	0.15	0.57	0.65	1.23	3.68	0.24	0.86	2.47	0.04	0.06	0.36		
n-tridecane	0.13	0.84	4.47	0.13	0.84	8.51	<LOQ	0.02	0.23	0.38	2.84	8.24	0.29	2.11	7.39	<LOQ	0.03	0.30		
Aromatic hydrocarbons																				
benzene	0.45	0.59	0.73	0.39	0.59	0.67	0.52	0.71	0.79	0.84	1.04	1.29	0.89	1.03	1.20	1.20	1.54	1.94		10
toluene	1.34	2.61	6.49	1.41	2.83	3.97	1.08	1.73	4.74	1.34	2.42	6.18	1.37	2.05	6.12	1.05	2.06	9.01	5000	260
ethylbenzene	0.33	0.46	14.4	0.25	0.45	25.3	0.15	0.22	0.33	0.28	0.42	2.60	0.22	0.36	1.98	0.21	0.23	0.52		260
m/p-xylene	0.95	1.26	49.5	0.73	1.13	85.3	0.44	0.66	1.19	0.71	1.00	8.73	0.71	0.87	6.41	0.59	0.68	1.68		200
o-xylene	0.35	0.45	19.8	0.26	0.42	39.3	0.17	0.22	0.43	0.29	0.43	3.42	0.29	0.34	2.62	0.23	0.26	0.67		
styrene	0.19	0.41	1.65	0.17	0.39	1.98	0.03	0.05	0.18	0.21	0.53	1.31	0.15	0.40	1.21	0.08	0.11	0.21	260	850
benzaldehyde	3.14	4.28	29.1	2.08	3.49	25.4	3.21	5.27	10.3	<LOQ	4.53	38.4	2.52	5.21	38.1	6.63	9.19	10.5		
phenol	0.79	1.48	7.60	0.80	1.17	5.31	1.15	1.68	2.12	<LOQ	0.93	1.33	0.26	0.90	1.26	0.90	1.23	1.67		
Terpenes																				
α-pinene	0.46	1.81	34.0	0.41	1.84	40.0	0.05	0.09	0.14	0.07	2.19	22.1	0.32	1.21	14.4	0.11	0.16	0.44		450
3-carene	0.06	0.45	8.82	<LOQ	0.41	6.18	<LOQ	<LOQ	<LOQ	<LOQ	0.27	7.89	<LOQ	0.16	4.02	<LOQ	<LOQ	<LOQ		1500
limonene	0.20	5.21	41.4	0.22	2.65	22.4	0.02	0.04	0.08	2.41	11.2	128	0.69	3.37	46.3	0.06	0.11	0.15		450
Alcohols																				
isopropanol	10.1	49.2	147	9.70	19.2	80.0	0.21	0.38	6.38	14.3	69.1	248	2.36	34.3	183	<LOQ	0.62	2.75		
1-butanol	0.99	3.83	15.1	1.18	3.85	25.0	<LOQ	<LOQ	<LOQ	<LOQ	1.92	8.64	<LOQ	2.57	9.51	<LOQ	<LOQ	<LOQ		
1-pentanol	0.33	1.37	4.55	0.25	1.64	27.0	<LOQ	<LOQ	0.07	<LOQ	0.54	2.86	<LOQ	0.69	8.79	<LOQ	<LOQ	<LOQ		
2-ethyl-1-hexanol	1.32	2.66	5.94	1.27	2.37	5.85	<LOQ	0.19	0.70	0.66	1.51	9.77	0.43	1.94	6.92	<LOQ	0.12	0.21		
Aldehydes																				
nonanal	12.9	22.8	60.1	5.95	22.3	68.0	<LOQ	0.30	2.19	<LOQ	10.8	30.4	2.45	8.59	31.3	<LOQ	0.63	1.57	*	
Ketones																				
2-butanone	2.01	3.40	62.1	1.88	2.59	144	0.76	1.04	1.35	0.30	3.07	32.6	0.66	1.86	16.0	0.92	1.05	1.42		
acetophenone	0.90	1.89	3.54	0.48	1.33	3.79	2.72	4.34	5.42	<LOQ	1.18	3.94	0.55	1.11	2.49	3.14	4.09	4.91		
Chlorinated hydrocarbons																				
dichloromethane	0.04	0.07	1.67	0.03	0.07	5.85	0.04	0.07	0.13	0.19	0.24	5.57	0.19	0.25	3.89	0.34	0.48	0.61		
Esters																				
ethyl acetate	2.79	16.5	49.1	1.58	9.55	171	1.07	1.88	10.4	1.14	4.44	97.7	1.00	9.55	47.6	0.69	1.14	3.81		
Nitriles																				
benzonitrile	0.15	0.20	0.28	0.15	0.16	0.32	0.22	0.29	0.50	<LOQ	0.18	0.30	0.14	0.19	0.75	0.45	0.61	0.84		
TVOC	92.6	173	422	60.7	84.4	826	15.5	24.5	29.7	78.7	194	623	35.2	124	390	22.4	30.6	35.6	300 (1)	1000 (2)

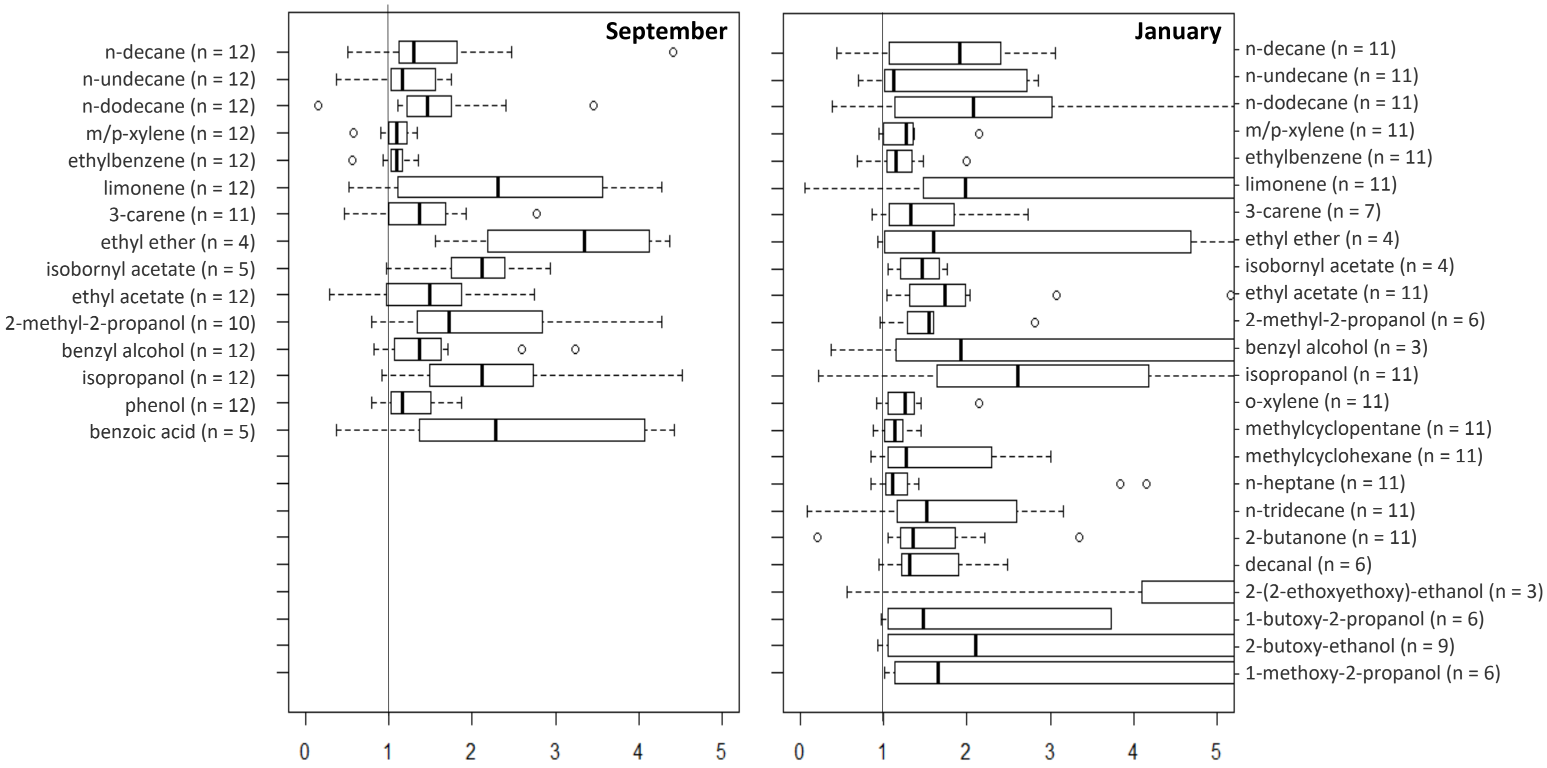

Figure 1: Concentration ratios living area/sleeping room (concentration measured in the living area relative to the concentration in the sleeping room). The boxes shown represent the 25, 50 (median) and 75 percentiles. This figure shows the VOCs of which the 25th percentile is higher than one. Left: September, Right: January. Both graphs are enlargements of a ratio from zero to five.

3.1.1.2 Indoor variation between September 2019 and January 2020 sampling campaigns

The ratio of the measured concentrations in January, relative to those in September, is calculated for the living area (for all VOCs with a concentration > LOQ at minimum 5 locations) (Figure A5). DCC G is left out of consideration here because of the notable decreases through time in indoor VOC concentrations at this location (new building, see section 3.1.1.3). 18 VOCs show similar concentrations in January as in September (ratio of one between the first and third quartile). 17 VOCs are found at lower concentrations in January (more than 75% of the data with a ratio below one), and 7 VOCs at lower concentrations in September (more than 75% of the data with a ratio above one). Literature shows different results about indoor seasonal VOC changes. Chin et al. (2014) measured overall the highest VOC levels in spring and fall, and the lowest in summer and winter in homes of children with asthma in Detroit, Michigan, USA (Chin et al., 2014). Paciência et al. (2016) mention that in the four indoor environments considered in their review (schools, housing, offices, and other indoor), the mean concentration of most VOCs was higher in the cold season (Paciência et al., 2016). Concentrations up to three times higher in winter than in summer were measured in apartments by Rehwagen et al. (2003) in Leipzig, Germany (Rehwagen et al., 2003). Seasonal changes can be caused by many factors including changes in indoor emissions due to seasonal variations in indoor activities (Al-Khulaifi et al., 2014). The air exchange rate (AER) is also an important factor, which depends on the indoor-outdoor temperature difference, the presence and use of air conditioners and forced ventilation systems, window opening, wind speed, age and condition of the house (building airtightness) (Chin et al., 2014).

3.1.1.3 Specific observations in DCCs regarding VOCs

At DCC A in the sleeping room, elevated concentrations compared to the other locations in the September campaign were measured for n-undecane, n-dodecane, n-tridecane, phenol, 2-butanone, isopropanol, α-pinene, limonene, decanal and acetic acid. Concentrations for these compounds are 2.7 (acetic acid) to 21.7 (α-pinene) times higher than the median measured concentrations in the sleeping rooms of the 12 DCCs in the September campaign. In the same sleeping room in the January campaign, only n-undecane, n-tridecane and α-pinene were still identified as an outlier (i.e. outside 1.5 times the interquartile range above the upper quartile and below the lower quartile). The concentration of 2-butanone, for example, decreased from 29.1 µg/m^3 in September to 2.24 µg/m^3 in January, and the limonene concentration from 22.4 µg/m^3 to 3.83 µg/m^3. Also, some ethers (1-butoxy-2-propanol, 2-butoxy-ethanol, 1-methoxy-2-propanol and 2-(2-ethoxyethoxy)-ethanol)), for which a tentative quantification was done, are considered as outliers at this location in September (e.g. more than 30 times larger values of 1-butoxy-2-propanol compared to the median measured concentrations in the sleeping rooms of the 12 DCCs in September). The use of cleaning products or freshly washed textile (VOCs in washing products) in September is a possible explanation since the majority of the above mentioned VOCs are found in household-, cleaning and personal care products and the concentrations in the second sampling are significantly lower, indicating a temporary source (ECHA, 2020).

Other outliers indoors are mainly found at DCC G, situated in a newly constructed building with new furniture, and DCC L, where a new building section was added in 2018. The brand new DCC G was only opened two weeks before the first sampling campaign.
The highest measured TVOC concentration was measured at DCC G in September (826 µg/m^3). The main contributor to this value is ethyl acetate (171 µg/m^3), followed by 2-butanone (144 µg/m^3), cyclohexane (123 µg/m^3), and m/p-xylene (85.3 µg/m^3). All these compounds can be emitted by building materials such as varnishes, sealing agents and flooring (Federal Public Service, 2016) (Suzuki et al., 2020). The TVOC concentration at DCC G was a factor of 2 (living area September) to 6 (sleeping room September) times higher than the average TVOC concentration of the other 11 DCCs. Brown (2002) – in his study about buildings with new materials introduced during renovation, extension or construction in Melbourne, Australia – mentions a VOC pollution of one to two orders of magnitude higher in new and renovated buildings than in established dwellings and it did not decay to the latter for several weeks to months. The baseline TVOC concentration (~200 µg/m^3) was reached after ca. 35 weeks (Brown, 2002).

29 VOCs were identified as outliers at DCC G compared to the other 11 locations in the living area and/or sleeping room in September and/or January. A comparison between the concentrations for these 29 compounds measured at older buildings (average concentrations of three daycare centres (DCC C, D and K) that have not been renovated for at least the past eight years), with the new building section of 2018 in DCC L and the newly constructed building (DCC G of 2019), is given in Table 3. On average, 2 to 50 times higher concentrations for these 29 VOCs are measured at DCC G in comparison to the mean of the three older buildings. VOCs emitted from furniture and building materials have been recognized as major indoor pollutants (Pei et al., 2016). It has been pointed that in renovated or new buildings, VOC concentration levels are often several orders of magnitude higher. Interior coatings for example can increase indoor air pollution due to VOC emissions, and sources of aldehydes in homes

include building materials, hardwood, plywood, laminate floorings, adhesives, paints and varnishes (Missia et al., 2010).

For 17 and 24 VOCs at DCC G in the living area and sleeping room, respectively, concentrations are drastically reduced in the second sampling campaign, i.e. four months later (Table 3). O-xylene, m/p-xylene and ethylbenzene for example have decreased with an average factor of 5.7 and 13.7 for the living area and sleeping room, respectively. Noguchi et al. (2016) also measured VOC concentrations (26 target compounds) in a newly built DCC just after completion of the building and three months later in Tokyo, Japan. The TVOC concentrations three months later were approximately one-tenth of the values measured just after completion (Noguchi et al., 2016). In our study, the TVOC concentration at DCC G decreased from 826 µg/m³ to 390 µg/m³ in the sleeping room in 4 months. On the other hand, in the living area, the TVOC concentration increased from 442 µg/m³ to 623 µg/m³. The main contributor in the living area in January is isopropanol (221 µg/m³), a compound found in paints and coating products, but also in consumer products like wash liquids/detergents, disinfectant sprays, and air fresheners (ECHA, 2020). Shin and Jo (2013) did a 24-month longitudinal survey of VOC concentrations in 25 new houses in high-rise apartment buildings in Daegu, Korea. The indoor TVOC concentrations exhibited a decreasing tendency over the whole follow-up period (mean TVOC concentration decreased from 881 µg/m^3 for the first month to 432 µg/m³ for the last month). Many individual VOCs associated with building materials and furniture emissions (e.g. toluene) showed this decreasing trend over 24 months as well, whereas certain VOCs associated with household product emissions (e.g. naphthalene, assumed to be emitted mainly from mothballs) did not (Shin & Jo, 2013).

In DCC L, where a new building section was added in 2018, higher concentrations were measured compared to the other DCCs for some VOCs (TVOC concentrations a factor of 1.8 – 4.0 higher than the average TVOC concentrations of the 3 reference buildings) (Table 3). Several compounds (e.g. o-xylene, m/p-xylene, ethylbenzene, n-dodecane, styrene, 3-carene, 2-butanone, phenol, n-decane, acetophenone and cyclohexane) show in the September campaign (both in the living area and sleeping room) concentrations between the average value in the older reference buildings and those in the new construction building of 2019. Their concentrations decreased four months later (Table 3, ratio concentration January 2020 / September 2019 < 1). It concerns VOCs with typically sources from building materials/furniture (e.g. cyclohexane is found in paints, varnishes and glues; n-decane in adhesives for floors, wood varnishes, carpets and rugs; m/p- and o-xylene in paints, varnishes and adhesives; styrene in insulation materials and plastics) (Federal Public Service, 2016).

3.1.1.4 VOCs released from insulation materials

2-chloropropane is discussed in more detail as it is only found indoors at one location (DCC H) in both the living area and sleeping room, and in both sampling campaigns (September and January). Tentative concentrations (based on an average RRF) between 1.8 and 17.3 µg/m^3 were calculated.

As little information was available about this compound and it was not measured at the other 11 locations, the source of 2-chloropropane at DCC H was unclear at first. It was suspected that the specific floor material could be a source of this compound, or the insulation material since 2-chloropropane can be used in insulation panels as an alternative foam-blowing agent (Oars Weel, 2014). Therefore, samples of the rubber top layer of the floor, the Kingspan Kooltherm floor insulation, and the wall insulation, were subjected to headspace analysis by PTR-QiToF-MS. To compare the spectrum of the pure compound 2-chloropropane with the spectrum of the building materials, headspace of the pure compound was analysed as well.

Table 3: Comparison between the VOC concentrations in older and new buildings. Older = average concentration of three daycare centres (DCC C, D and K) that have not been renovated for at least the past eight years; n=3 except values indicated with an asterisk (*) n=2 and (**) n=1 (concentration <LOQ at one or two locations). N.F. = not quantifiable at none of the three locations. DCC L = new building section added in 2018. DCC G = new construction building of 2019. • Tentative concentrations: Values can be compared relative to each other based on an average RRF.

	Concentration [µg/m^3]												Ratio concentration January 2020 / September 2019			
	September 2019						January 2020									
	Living area			Sleeping room			Living area			Sleeping room			Living area		Sleeping room	
	Older (n=3)	DCC L 2018	New DCC G 2019	Older (n=3)	DCC L 2018	New DCC G 2019	Older (n=3)	DCC L 2018	New DCC G 2019	Older (n=3)	DCC L 2018	New DCC G 2019	DCC L 2018	New DCC G 2019	DCC L 2018	New DCC G 2019
o-xylene	0.39 ± 0.03	2.7	19.8	0.37 ± 0.09	2.0	39.3	0.5 ± 0.3	2.2	3.4	0.30 ± 0.01	1.0	2.6	0.8	0.2	0.5	0.1
m/p-xylene	1.0 ± 0.1	5.6	49.5	1.0 ± 0.3	4.7	85.3	1 ± 1	4.5	8.7	0.75 ± 0.03	2.1	6.4	0.8	0.2	0.4	0.1
ethylbenzene	0.38 ± 0.07	1.5	14.4	0.4 ± 0.1	1.3	25.3	0.4 ± 0.3	1.2	2.6	0.25 ± 0.04	0.6	2.0	0.8	0.2	0.5	0.1
styrene	0.32 ± 0.09	0.8	1.7	0.3 ± 0.1	0.4	2.0	0.4 ± 0.2	0.6	0.9	0.2 ± 0.1	0.4	0.2	0.8	0.5	1.0	0.1
toluene	2.4 ± 0.4	3.6	2.6	2.1 ± 0.4	3.7	3.9	1.8 ± 0.4	2.6	6.2	2 ± 1	1.9	6.1	0.7	2.4	0.5	1.6
benzaldehyde	5 ± 1	4.4	18.2	3.2 ± 0.5	6.35	18.5	3.7 ± 0.7	4.6	18.0	4 ± 1	5.7	27.1	1.0	1.0	0.9	1.5
phenol	1.6 ± 0.5	1.7	7.1	1.1 ± 0.1	1.3	4.3	0.8 ± 0.2	1.3	1.2	0.9 ± 0.3	1.1	1.2	0.8	0.2	0.8	0.3
3-carene	0.3 ± 0.2	5.9	8.8	0.3 ± 0.2*	4.9	6.2	0.19 ± 0.07	2.6	7.9	N.F.	1.0	4.0	0.4	0.9	0.2	0.6
α-pinene	1.6 ± 0.2	17.8	20.5	1 ± 1	17.4	14.4	2.0 ± 0.6	10.1	22.1	0.36 ± 0.06	4.8	13.7	0.6	1.1	0.3	1.0
n-hexane	10 ± 15	1.1	1.2	1.2 ± 0.3	1.0	2.6	1.5 ± 0.6	1.5	4.4	1.2 ± 0.2	1.2	5.7	1.4	3.7	1.2	2.2
n-heptane	0.7 ± 0.3	2.3	5.9	0.7 ± 0.2	3.1	16.2	0.9 ± 0.7	9.0	13.3	0.38 ± 0.03	2.2	9.3	4.0	2.3	0.7	0.6
n-nonane	0.5 ± 0.1	0.7	0.8	0.61 ± 0.06	0.6	1.3	0.37 ± 0.09	0.5	0.4	0.3 ± 0.1	0.4	0.8	0.8	0.5	0.6	0.6
n-decane	2 ± 1	2.8	4.8	0.7 ± 0.2	1.8	8.4	1.5 ± 0.1	2.6	2.2	0.50 ± 0.04	1.2	4.9	0.9	0.5	0.7	0.6
n-undecane	4 ± 5	2.6	18.4	1.2 ± 0.8	1.6	18.4	4 ± 1	8.4	1.2	0.7 ± 0.2	3.2	1.7	3.2	0.1	2.1	0.1
n-dodecane	1.8 ± 0.6	4.3	13.2	0.9 ± 0.6	2.4	10.6	1.2 ± 0.2	3.7	1.3	0.26 ± 0.02	1.8	1.5	0.9	0.1	0.8	0.1
n-tridecane	2 ± 1	0.7	4.5	0.7 ± 0.6	0.7	4.6	3 ± 1	6.5	1.0	0.6 ± 0.1	3.2	1.0	8.7	0.2	4.9	0.2
methylcyclopentane	0.31 ±0.02	0.3	7.4	0.26 ± 0.05	0.2	21.2	1.0 ± 0.8	0.5	24.1	0.45 ± 0.03	0.3	16.8	1.8	3.2	1.7	0.8
2-methyl-butane •	0.6 ± 0.4	7.3	4.3	0.7 ± 0.6	6.5	5.9	0.8 ± 0.3	5.7	9.2	0.9 ± 0.9	3.1	27.5	0.8	2.1	0.5	4.7
cyclohexane	3 ± 4	1.8	43.8	4 ± 7	12.6	123	3 ± 2	1.5	153	1 ± 1	3.7	100	0.8	3.5	0.3	0.8
methylcyclohexane •	3 ± 3	1.6	16.3	2 ± 2	1.0	33.7	0.6 ± 0.5	79.3	27.2	0.3 ± 0.1	9.0	17.2	50	1.7	9.5	0.5
dichloromethane	0.10 ± 0.06	0.2	1.7	0.10 ± 0.04	0.1	5.9	0.24 ± 0.02	0.3	5.6	0.23 ± 0.04	0.3	3.9	1.6	3.3	2.0	0.7
ethyl acetate	10 ± 6	34.9	49.1	6 ± 4	20.3	171	3 ± 2	97.7	83.1	2 ± 1	18.9	47.6	2.8	1.7	0.9	0.3
isopropanol	57 ± 33	50.6	24.4	12 ± 3	22.1	19.9	103 ± 52	64.3	221	10 ± 10	35.9	83.4	1.3	9.1	1.6	4.2
1-pentanol	0.9 ± 0.4	4.5	3.0	1.0 ± 0.6	27.0	15.6	0.5 ± 0.2	2.9	1.2	0.3**	8.8	3.7	0.6	0.4	0.3	0.2
acetophenone	2.5 ± 0.9	2.3	3.1	1.0 ± 0.4	1.7	3.8	0.86 ± 0.05	1.3	3.9	1.9 ± 0.6	1.0	2.0	0.6	1.3	0.6	0.5
2-butanone	2.3 ± 0.4	5.1	62.1	1.98 ± 0.09	10.3	144	13 ± 17	4.1	21.0	1.2 ± 0.5	3.9	16.0	0.8	0.3	0.4	0.1
benzyl alcohol •	1 ± 1	0.9	32.7	0.5 ± 0.3	1.0	39.6	0.3**	0.5	3.1	N.F.	0.3	8.5	0.5	0.1	0.2	0.2
hexanal •	3 ± 2	9.2	7.7	2 ± 1	60.8	31.8	0.44 ± 0.09	3.1	1.5	0.077 ± 0.002	8.3	3.3	0.3	0.2	0.1	0.1
hexanoic acid •	2.5 ± 0.3	9.5	3.2	1.5 ± 0.7	28.5	26.4	1.5 ± 0.5*	6.1	2.0	0.7 ± 0.4	19.8	6.9	0.6	0.6	0.7	0.3

The result of the PTR-QiToF-MS measurements of 2-chloropropane is given in Figure A6 (at the top) . The protonated product ions of 2-chloropropane (m/z = 39.023 ((C_3H_2)H^+), 41.039 ((C_3H_4)H^+), 43.055 ((C_3H_6)H^+)) were clearly found in the sample of the floor insulation (Figure A6 bottom). 2-chloropropane was not detected in the sample of the rubber floor or the wall insulation. These results were confirmed by sampling 300 mL of headspace on Tenax TA sorbent tubes and analysis with TD-GC-MS (split 40 mL/min) (Figure A7). It is remarkable that this DCC was founded in 2012 and that 2-chloropropane, emitted from the insulation material, is still measurable 8 years later. Kingspan insulation consists of phenolic foam which is made by foaming a phenolic resin. According to the description of the patent, the foam-blowing agent includes a chlorinated hydrocarbon containing two to five carbon atoms, possibly 2-chloropropane (Coppock et al., 2014).

Moreover, both in the sample of the Kingspan Kooltherm floor insulation and in that of the wall insulation, there was also a prominent peak of 2-methylbutane measured by TD-GC-MS (Figure A7). The highest tentative concentrations of this compound (no RRF) were found in DCC G, H and L (> 7 µg/m^3). The tentative concentrations measured in the sleeping room and living area at the other 9 locations over the two sampling campaigns (n=35) ranged between 0.2 and 5.4 µg/m^3, with a median and mean of 0.9 and 1.3 µg/m^3 respectively. The largest concentration of 2-methylbutane was measured in January at DCC H in the sleeping room (73.1 µg/m^3), followed by the sleeping room of DCC G (27.5 µg/m^3). This compound is used in fuels, perfumes, personal care products and cosmetics, as well as in paints and coatings (ECHA, 2021). The higher concentrations measured at DCC G and L are possibly explained by the new building/renovations at these locations, and in DCC H by the insulation (Kingspan Kooltherm). The used type of isolation in DCC H and L is polyisocyanurate (PIR) from Isorex and IKO Enertherm, respectively.

3.1.2 An in-depth look at VOCs in the outdoor air

Outdoors, benzaldehyde and acetophenone were measured at the highest concentrations (median values > 4 µg/m^3) compared to the other VOCs in this study and are together counting for 28-53% (median 43%, n=24) of the TVOC concentration.

The average (over the two sampling campaigns) measured median concentrations were compared with the median reported values in the AIRMEX study, for all VOCs which were measured outdoors in both studies. The median concentrations of the BTEX compounds, hexane, heptane, decane, undecane, dodecane and limonene higher in the AIRMEX study compared to our study (ratio ranging from 1.1 for hexane to 54.3 for dodecane). However, the median outdoor concentration for α-pinene and styrene was zero in the AIRMEX study versus 0.13 and 0.08 µg/m^3, respectively, in our measurements (Geiss et al., 2011).

3.1.2.1 Variation between September 2019 and January 2020 sampling campaigns

The ratio of the concentrations in January, relative to those in September, for the VOCs which were measured (>LOQ) outdoors (Figure A8), shows similar concentrations (ratio of one between the first and third quartile) for 14 VOCs in both sampling campaigns. Phenol, n-dodecane, ethyl acetate and benzoic acid are in general found at lower concentrations in January compared to September (more than 75% of the data with a ratio < 1). However, a larger list of VOCs (13) is measured at a general higher concentration in January (more than 75% of the data with a ratio > 1). Outside benzene concentrations range from 0.52 µg/m^3 to 0.79 µg/m^3 in September, with on average 2.3 times higher concentrations in January, ranging from 1.20 µg/m^3 to 1.94 µg/m^3. Outdoor TVOC concentrations were also larger in January (22.4 – 33.8 µg/m^3; median 30.6 µg/m^3) compared to September (15.5 - 29.7 µg/m^3; median 24.5 µg/m^3). The measurements of Bozkurt et al. (2018) in Turkey show in general also lower summer concentrations compared to other seasons and the highest concentration levels were observed in winter. This is the case for the mean concentrations of e.g. n-hexane, n-heptane, n-nonane, cyclohexane, methyl cyclopentane, o-xylene, benzene, ethylbenzene and toluene, which were VOCs that were also measured at higher concentrations in January compared to September in this study, with a median ratio between 1.04 and 3.77. Fuselli et al. (2010) measured significantly higher concentrations in winter than in summer for the monoaromatic hydrocarbons (e.g. winter/summer ratio benzene was 2.3-2.8). Possible explanations are the lower dispersion of air in the winter due to thermal inversion or in general unfavourable meteorological conditions (e.g. lower mixing height), the longer persistence of the compounds due to reduced photochemical degradation, and an increase in pollutant emissions during winter (heating of buildings, traffic (cold starts)) (Bozkurt et al., 2018) (Fuselli et al., 2010) (Paciência et al., 2016).

3.1.2.2 VOC concentrations in the outdoor cubicles designed for sleeping

It was investigated if the specifically designed outdoor cubicles could have an impact on the VOC concentrations. The ratio of the concentration in the outdoor cubicles (DCC A, B and H) relative to that in the open air was calculated, but the effect of the outdoor cubicles on the VOC concentrations in this study is minimal; i.e. for 22 of the 33 compounds for which it was possible to calculate a ratio, the value of one is situated between the 25th and 75th percentile (Figure 2A).

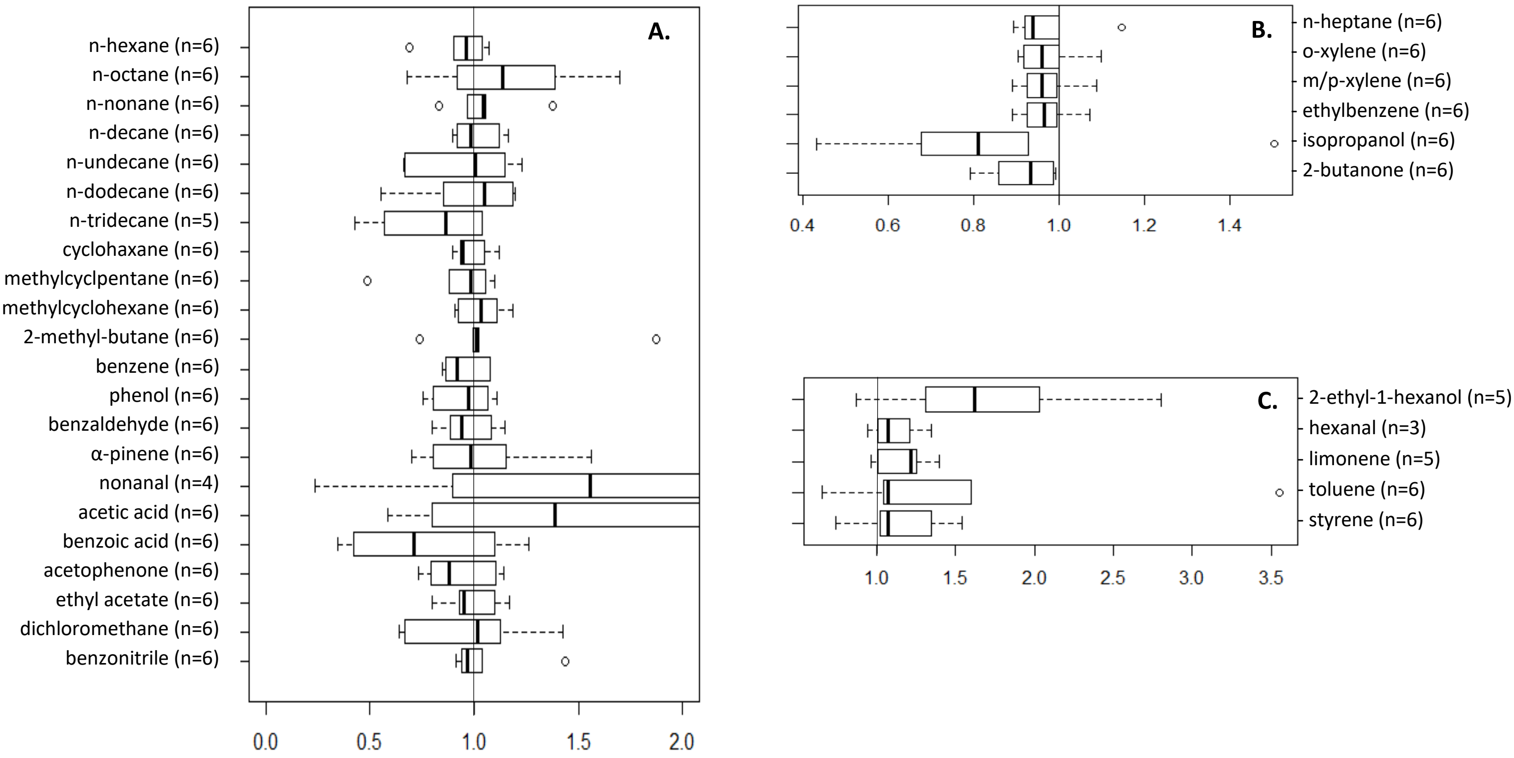

Figure 2: Concentration ratios outdoor bed/outdoors (concentration measured in the outdoor bed relative to the concentration measured in the environmental air). (A) Compounds with a ratio of one between the first and third quartile (enlargements of a ratio from zero to two); (B) Compounds with values of the third quartile lower than one; (C) Compounds with values of the first quartile higher than one. The boxes shown represent the 25, 50 (median), and 75 percentiles.
Ratios were calculated for the three locations (DCC A, B and H) having the cubicles specifically designed for outdoor sleeping, both in September and January (total n=6). However, not all VOCs were (always) detected outdoors. Compounds that were only measurable twice or less ($n \leq 2$), are not plotted. Out of the 33 plotted VOCs, the majority (22) has a ratio of about one (A).

For six compounds (n-heptane, o-xylene, m/p-xylene, ethylbenzene, isopropanol and 2-butanone) the concentration in the outdoor cubicles was slightly lower than in the ambient outdoor air (>75% of the data with a ratio of the concentration outdoor cubicle/outdoors below one, with median ratios between 0.81 and 0.96 for these 6 VOCs) (Figure 2B). For five compounds (2-ethyl-1-hexanol, hexanal, limonene, toluene, and styrene) a slightly higher concentration in the outdoor cubicles was measured (>75% of the data with a ratio of the concentration outdoor cubicle/outdoors larger than one, with median ratios between 1.07 and 1.62 for these 5 VOCs) (Figure 2C). These compounds could be released from the beds (e.g. hexanal and limonene from wood, 2-ethyl-1-hexanol from paint or coating) (Adamová et al., 2020) (ECHA, 2020). However, the differences are quite small with the lowest and highest median ratios of the 11 above-mentioned VOCs being 0.81 for isopropanol (n=6) and 1.62 for 2-ethyl-1-hexanol (n=5). Only a statistically significant difference ($p < 0.05$) was found for 2-butanone when applying a t-test to compare the mean (n=6) ratio with a ratio of one. Moreover, it concerns very small absolute concentrations with median outdoor and outdoor cubicle concentrations between 0.07 and 2.32 µg/m^3 for the above-mentioned VOCs at DCC A, B and H. It can be concluded that the specifically designed cubicles for outdoor sleeping will not significantly enlarge the VOC levels in the cubicles compared to the ambient outdoor environment. The TVOC concentrations in the outdoor beds (ranging from 20.2 to 24.1 µg/m^3 in September and from 28.5 to 33.8 µg/m^3 in January) are on average a factor 5 lower than the concentrations measured in the sleeping rooms of the corresponding DCC.

3.2 NO_2, SO_2, and O_3 concentrations

3.2.1 Nitrogen dioxide

Measured indoor and outdoor NO_2 data are summarized in Figure 3. The average indoor concentration, as well as the values of all the individual indoor samples, were below the guide (20 µg/m^3) and the intervention (40 µg/m^3) value published by the Belgian Official Gazette (Federale Overheidsdienst Justitie, 2018). The guideline of the WHO (40 µg/m^3 as annual average concentration) was always respected outside (WHO, 2010). The highest measured concentration was 21 µg/m^3, outdoors at DCC I during the January sampling campaign.

When comparing the indoor and outdoor NO_2 concentrations, there are some locations (e.g. DCC G in September) where the indoor and outdoor concentrations seems to differ from each other. However, there are no significant differences ($p \geq 0.05$) when comparing the averages of the concentrations indoors and outdoors), indicating in general a homogeneous distribution of NO_2 between the indoor and outdoor air. I/O ratios range from 0.7 to 2.8 (median 1.2, n = 12) in September and from 0.6 to 1.4 (median 1.1, n = 12) in January. It can be concluded that sleeping outdoors will not provide extra exposure to NO_2, and both indoors and outside, guideline values were respected.

As no outliers (i.e. outside 1.5 times the interquartile range above the upper quartile and below the lower quartile) were identified in any of the datasets (indoor September; outdoor September; indoor January; outdoor January), there are no significant differences between the locations, neither indoors nor outdoors.

Despite differences between the two sampling campaigns are not significant ($p \geq 0.05$), measured values in winter (January) are slightly higher (on average a factor 1.4 and 1.2 outdoors and indoors, respectively) than in summer/autumn (September) (Figure 3). Possible explanations are better atmospheric dilution conditions in the summer months, an increased use of fossil fuels for heating in the winter as well as an increase in traffic density (and cold starts) (Bozkurt et al., 2018) (VMM, 2019a). Moreover, winter is characterized by shorter days and reduced solar radiation, causing a decrease in the role of photolysis and shifting the balance of daytime oxidation and nighttime chemistry (Kenagy et al., 2018).

The differences between the concentrations in the outdoor cubicles and the ambient outdoor air of the corresponding locations are negligible (ratio concentrations outdoor cubicle/outdoors ranging from 0.93 to 1.2), so the outdoor cubicles do not influence the NO_2 concentration to which children are exposed when sleeping.

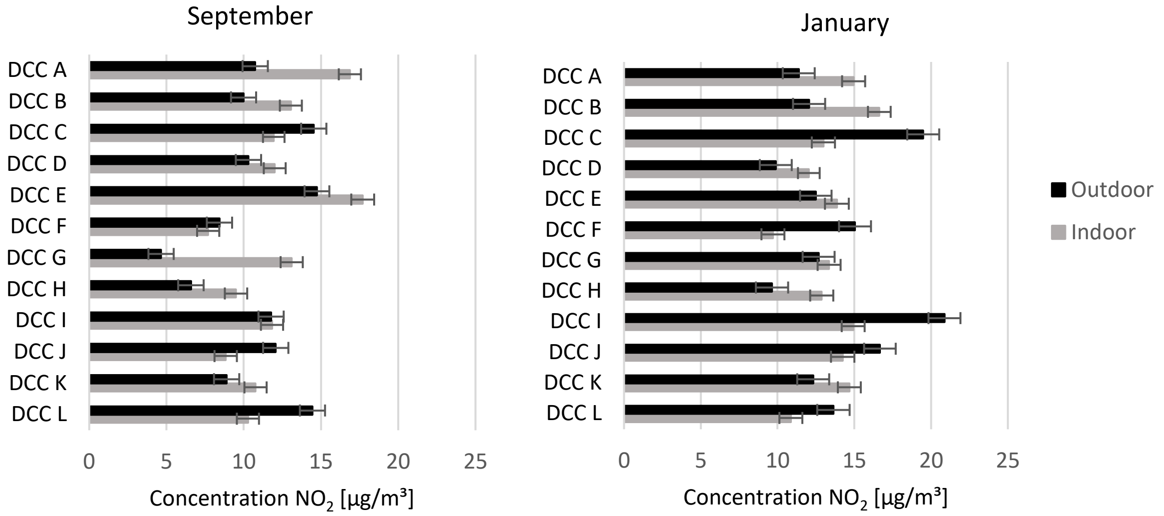

Figure 3: NO_2 concentrations [µg/m^3] measured in September (left) and January (right) at the 12 DCCs, indoors (living area) and outdoors.

3.2.2 Sulphur dioxide

SO_2 concentrations were all below the quantification limit of 1 ppb (2.62 µg/m^3). In Flanders, SO_2 emissions in 2017 were 78% lower than those in 2000, mainly due to the lower sulphur content in fuels. This decrease in the emission is reflected in a decrease of SO_2 concentrations in the ambient air. Where SO_2 concentrations in urban environments in Flanders in 1981 were about 60 µg/m^3, today they are approximately 2 µg/m^3, which corresponds to a value lower than the LOQ of the used method (VMM, 2019a). Also when looking broader, a trend assessment study in Europe (over 22 countries) for the period 2000-2017 shows that the average SO_2 annual mean concentration has decreased by more than 70%. In 2018, this mean concentration for stations in (sub)urban areas was below 4 µg/m^3 (European Environmental Agency, 2020). The WHO reports an air quality guideline (24-hour) of 40 µg/m^3 (World Health Organization, 2021).

3.2.3 Ozone

Measured indoor and outdoor ozone concentrations are summarized in Figure 4. The indoor concentrations (maximum measured concentration 11.2 µg/m^3) are below the indoor guide (40 µg/m^3) and intervention (78 µg/m^3) values published in the Belgian Official Gazette (Federale Overheidsdienst Justitie, 2018). The outdoor concentrations (max. measured concentration 53.7 µg/m^3 (DCC C, January)) are well below the legally binding long-term objective target value for health protection (120 µg/m^3 – maximum 25 exceedances in 3 years) of the European Directive 2008/50/EG. Also, the stricter recommended exposure limit of the WHO (100 µg/m^3) was always respected (Salonen et al., 2018) (VMM, 2019b) (WHO, 2006) (European Union, 2008). However, it should be mentioned that these guidelines are all applicable to an exposure time of 8 hours and the measured values are 7-day averages. The median and average O_3 concentrations measured in Flanders (1990-2018) are in general between 40 and 50 µg/m^3 (VMM, 2018) (VMM, 2019a). These values agree with the measured concentrations in this study.

The outdoor concentrations are significantly ($p < 0.05$) higher than indoors, with average I/O ratios of 0.11 ± 0.07 (n=10) and 0.05 ± 0.04 (n=11) for the September and January sampling campaign respectively. Ozone is a secondary pollutant formed during a photochemical reaction with VOCs and NO_x species. Since indoor sources are often absent, ozone in indoor environments mostly comes from outdoor sources (e.g. intake by ventilation), and the short atmospheric lifetime (half-life of ozone indoors is 7 - 10 min) causes a lower indoor concentration (de Gennaro et al., 2014) (Huang et al., 2019) (Quarcoo et al., 2019) (Salonen et al., 2018) (Weschler, 2000). Moreover, humans are recognized as a sink for ozone in the indoor environment. The chemicals that constitute human skin oils can be classified as wax esters, glycerol, fatty acids, squalene, esters, and sterols and contain unsaturated carbon bonds which readily react with ozone (Kruza & Carslaw, 2019).

It can be concluded that outdoor sleeping will expose the children to a higher ozone concentration than indoors, but the concentrations stayed below the legally binding long-term objective target value for health protection. However, mainly on warm summer days, these values can increase. Regarding outdoor sleeping, it should be followed up if no 'ozone days' (i.e. a day on which the European information threshold of 180 µg/m^3 is exceeded at least at one measurement location) are announced. In 2018 e.g. in Flanders, this information threshold was exceeded on 9 days (at least at one out of the 17 measurement sites) and the highest measured hourly average was 238 µg/m^3 (Sint-Pieters-Leeuw, 3 August 2018) (VMM, 2018) (VMM, 2019a). On these days, sleeping outside is not recommended.

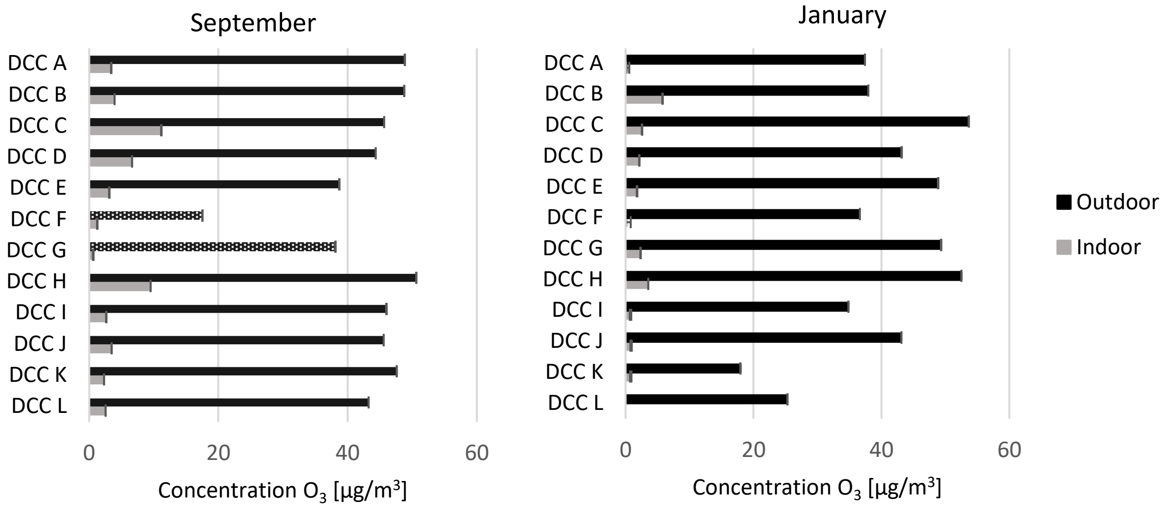

Figure 4: Ozone concentrations [µg/m^3] measured in September (left) and January (right) at the 12 DCCs, indoors (living area) and outdoors. The concentrations of DCC F and G in September are an underestimation (part of the samples were lost because these samples were wet which made it impossible to transfer the silica gel quantitatively).

3.2.3.1 Variation between September 2019 and January 2020 sampling campaigns

Despite the differences are not significant ($p \geq 0.05$ when comparing the averages of the concentrations of January and September, both indoors and outdoors), the concentrations are in general somewhat higher during the September measurements (median indoor and outdoor concentration of 3.3 µg/m^3 ($n = 12$) and 45.8 µg/m^3 ($n = 10$, two samples lost) respectively) in comparison to January (median indoor and outdoor concentration 1.8 µg/m^3 ($n=12$) and 40.5 µg/m^3 ($n=12$) respectively). Since O_3 is a secondary pollutant formed by photochemical reactions, more sunlight in summer/autumn compared to winter can explain this (Bozkurt et al., 2018). The calm weather and temperature during the first sampling campaign (weekly average measured temperature 16.6 ± 4.2°C) encourage more ventilation by open windows, which can result in higher AERs and can explain the higher concentrations in the summer compared to winter indoors (weekly average measured temperature of the second sampling campaign 6.7 ± 3.5°C).

3.2.3.2 Outdoor cubicles

There are no significant differences between the averages of the datasets (both for September and January) when comparing the outdoor cubicles and the ambient outdoor air, but looking at the separate values, the outdoor concentration is almost always higher than in the outdoor cubicle. The ratio of the concentration in the outdoor cubicle versus outdoors (for DCC A, B and H) is on average 0.8 ± 0.1 ($n=5$), indicating a slightly lower concentration in the cubicles compared to the ambient outdoor air. In the cubicles themselves, there is no ozone source and less irradiation from the sun which is necessary to form photochemical O_3. The presence of children themselves in the cubicles can also decrease the ozone concentration (Kruza & Carslaw, 2019).

4. Conclusions

In general, from this multi-component study (47 VOCs, NO_x, SO_2, and O_3) in the context of DCCs in Ghent, the following conclusions can be made. First, median indoor TVOC concentrations (152 µg/m^3 (September) and 142 µg/m^3 (January)) are much higher than outdoors (24.5 µg/m^3 and 30.6 µg/m^3). Also regarding individual VOCs, concentrations are in general lower outdoors than indoors (40 of the 47 VOCs were found at higher concentrations indoors). Remarkable higher VOC concentrations for specific VOCs are measured in new construction buildings compared to older daycare centres. Some VOCs (e.g. isopropanol and benzyl alcohol) are more abundant in the living area compared to the sleeping room, probably due to the broader range of activities that take place there. 2-chloropropane was only measured at one location indoors, in both sampling campaigns. Headspace analysis with PTR-QiToF-MS showed to be a fast and powerful tool to demonstrate that the floor insulation is the source of this compound, even after more than 8 years.

Second, sleeping outdoors does not cause extra exposure to NO_2 as similar indoor and outdoor levels were measured. Third, there can be concluded that ozone exposure outdoors is larger (median indoor concentrations (n=12) of 3.3 and 1.8 µg/m^3 for September and January respectively versus 45.8 µg/m^3 (n=10) and 40.5 µg/m^3 (n=12) outdoors), but the measured concentrations stayed well below legal standards. Finally, regarding the specifically designed outdoor cubicles, there are no observable differences found in comparison to the ambient outdoor air for NO_2 and VOCs. Ozone concentrations are slightly lower in the cubicles (average ratio outdoor cubicle/outdoors 0.8 ± 0.1 (n=5)). Of course there are also other factors for which the trade-off between inside and outside has to be made (e.g. noise and temperature), but based on our air quality assessment at DCC in Ghent, we can conclude that sleeping outside should not be hampered by the presence of air contaminants (on the contrary), except for ozone days where exceedance of the WHO limit of 100 µg/m^3 can be expected.

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References

Adamová, T., Hradecký, J., & Pánek, M. (2020). Volatile organic compounds (VOCs) from wood and wood-based panels: Methods for evaluation, potential health risks, and mitigation. *Polymers*, *12*(10), 1–21. https://doi.org/10.3390/polym12102289

Al-Khulaifi, N. M., Al-Mudhaf, H. F., Alenezi, R., Abu-Shady, A.-S. I., & Selim, M. I. (2014). Seasonal and Temporal Variations in Volatile Organic Compounds in Indoor and Outdoor Air in Al-Jahra City, Kuwait. *Journal of Environmental Protection*, *05*(04), 310–326. https://doi.org/10.4236/jep.2014.54034

Arı, A., Ertürk Arı, P., Yenisoy-Karakaş, S., & Gaga, E. O. (2020). Source characterization and risk assessment of occupational exposure to volatile organic compounds (VOCs) in a barbecue restaurant. *Building and Environment*, *174*, 106791. https://doi.org/10.1016/j.buildenv.2020.106791

Berti, S., Cigala, A., Sharmahd, N., Boudry, N., & Sabaliauskiene, R. (2020). *The meaning of spaces in ECEC centres. A literature review.* Educas Project.

Bozkurt, Z., Üzmez, Ö. Ö., Döğeroğlu, T., Artun, G., & Gaga, E. O. (2018). Atmospheric concentrations of SO2, NO2, ozone and VOCs in Düzce, Turkey using passive air samplers: Sources, spatial and seasonal variations and health risk estimation. *Atmospheric Pollution Research*, *9*(6), 1146–1156. https://doi.org/10.1016/j.apr.2018.05.001

Brown, S. K. (2002). Volatile organic pollutants in new and established buildings in Melbourne, Australia. *Indoor Air*, *12*(1), 55–63. https://doi.org/10.1034/j.1600-0668.2002.120107.x

Cakmak, S., Dales, R. E., Liu, L., Kauri, L. M., Lemieux, C. L., Hebbern, C., & Zhu, J. (2014). Residential exposure to volatile organic compounds and lung function: Results from a population-based cross-sectional survey. *Environmental Pollution*, *194*, 145–151. https://doi.org/10.1016/j.envpol.2014.07.020

Chen, T.-M., Kuschner, W. G., Gokhale, J., & Shofer, S. (2007). Outdoor Air Pollution: Nitrogen Dioxide, Sulfur Dioxide, and Carbon Monoxide Health Effects. *The American Journal of the Medical Sciences*, *333*(4), 249–256. https://doi.org/10.1097/MAJ.0b013e31803b900f

Chin, J.-Y., Godwin, C., Parker, E., Robins, T., Lewis, T., Harbin, P., & Batterman, S. (2014). Levels and sources of volatile organic compounds in homes of children with asthma. *Indoor Air*, *24*(4), 403–415. https://doi.org/10.1111/ina.12086

Clark, H., Coll-Seck, A. M., Banerjee, A., Peterson, S., Dalglish, S. L., Ameratunga, S., Balabanova, D., Bhan, M. K., Bhutta, Z. A., Borrazzo, J., Claeson, M., Doherty, T., El-Jardali, F., George, A. S., Gichaga, A., Gram, L., Hipgrave, D. B., Kwamie, A., Meng, Q., … Costello, A. (2020). A future for the world's children? A WHO–UNICEF–Lancet Commission. In *The Lancet* (Vol. 395, Issue 10224, pp. 605–658). Lancet Publishing Group. https://doi.org/10.1016/S0140-6736(19)32540-1

Coppock, V., Zeggelaar, R., Takahashi, H., & Kato, T. (2014). *Phenolic foam*. Patent No. 8772366. https://www.freepatentsonline.com/8772366.html

de Gennaro, G., Dambruoso, P. R., Loiotile, A. D., Di Gilio, A., Giungato, P., Tutino, M., Marzocca, A., Mazzone, A., Palmisani, J., & Porcelli, F. (2014). Indoor air quality in schools. *Environmental Chemistry Letters*, *12*(4), 467–482. https://doi.org/10.1007/s10311-014-0470-6

Demeestere, K., Dewulf, J., de Roo, K., de Wispelaere, P., & van Langenhove, H. (2008). Quality control in quantification of volatile organic compounds analysed by thermal desorption-gas chromatography-mass spectrometry. *Journal of Chromatography A*, *1186*(1–2), 348–357. https://doi.org/10.1016/j.chroma.2007.11.036

ECHA. (2020). *Substance infocard*. European Chemicals Agency. https://echa.europa.eu/nl/substance-information/-/substanceinfo/

ECHA. (2021). *Substance Infocard 2-methylbutane*. https://echa.europa.eu/substance-information/-/substanceinfo/100.001.039

European Environment Agency. (2019). *Outdoor air quality in urban areas*. https://www.eea.europa.eu/airs/2018/environment-and-health/outdoor-air-quality-urban-areas#tab-based-on-data

European Environmental Agency. (2020). Air quality in Europe - 2020 report. In *EEA Report* (Issue No 09/2020).

European Union. (2008). Directive 2008/50/EC of the European Parliament and of the Council. In *Official Journal of the European Union*.

Federale Overheidsdienst Justitie. (2018). *Belgisch Staatsblad - Moniteur Belge (Belgian Official Gazette)*.

Fuselli, S., de Felice, M., Morlino, R., & Turrio-Baldassarri, L. (2010). A three year study on 14 VOCs at one site in Rome: Levels, seasonal variations, indoor/outdoor ratio and temporal trends. *International Journal of Environmental Research and Public Health*, *7*(10), 3792–3803. https://doi.org/10.3390/ijerph7103792

Geiss, O., Giannopoulos, G., Tirendi, S., Barrero-Moreno, J., Larsen, B. R., & Kotzias, D. (2011). The AIRMEX study - VOC measurements in public buildings and schools/kindergartens in eleven European cities: Statistical analysis of the data. *Atmospheric Environment*, *45*(22), 3676–3684. https://doi.org/10.1016/j.atmosenv.2011.04.037

Hoge Gezondheidsraad. (2017). *Gezondheidsraad Indoor Air Quality in Belgium September 2017. 8794*.

Huang, Y., Yang, Z., & Gao, Z. (2019). Contributions of indoor and outdoor sources to ozone in residential buildings in nanjing. *International Journal of Environmental Research and Public Health*, *16*(14). https://doi.org/10.3390/ijerph16142587

Instituti Clinici Scientifici Maugeri. (2006). *Radiello*.

Jia, S., Sankaran, G., Wang, B., Shang, H., Tan, S. T., Yap, H. M., Shen, J., Gutiérrez, R. A., Fang, W., Liu, M., Chang, V. W. C., Ng, L. C., & Fang, M. (2019). Exposure and risk assessment of volatile organic compounds and airborne phthalates in Singapore's Child Care Centers. *Chemosphere*, *224*, 85–92. https://doi.org/10.1016/j.chemosphere.2019.02.120

Kenagy, H. S., Sparks, T. L., Ebben, C. J., Wooldrige, P. J., Lopez-Hilfiker, F. D., Lee, B. H., Thornton, J. A., McDuffie, E. E., Fibiger, D. L., Brown, S. S., Montzka, D. D., Weinheimer, A. J., Schroder, J. C., Campuzano-Jost, P., Day, D. A., Jimenez, J. L., Dibb, J. E., Campos, T., Shah, V., … Cohen, R. C. (2018). NOx Lifetime and NOy Partitioning During WINTER. *Journal of Geophysical Research: Atmospheres*, *123*(17), 9813–9827. https://doi.org/10.1029/2018JD028736

Kornartit, C., Sokhi, R. S., Burton, M. A., & Ravindra, K. (2010). Activity pattern and personal exposure to nitrogen dioxide in indoor and outdoor microenvironments. *Environment International*, *36*(1), 36–45. https://doi.org/10.1016/j.envint.2009.09.004

Kruza, M., & Carslaw, N. (2019). How do breath and skin emissions impact indoor air chemistry? *Indoor Air*, *29*(3), 369–379. https://doi.org/10.1111/ina.12539

Madureira, J., Paciência, I., Rufo, J., Ramos, E., Barros, H., Teixeira, J. P., & de Oliveira Fernandes, E. (2015). Indoor air quality in schools and its relationship with children's respiratory symptoms. *Atmospheric Environment*, *118*, 145–156. https://doi.org/10.1016/j.atmosenv.2015.07.028

Mannucci, P. M., Harari, S., Martinelli, I., & Franchini, M. (2015). Effects on health of air pollution: a narrative review. *Internal and Emergency Medicine*, *10*(6), 657–662. https://doi.org/10.1007/s11739-015-1276-7

Mazzatenta, A., Pokorski, M., & di Giulio, C. (2021). Volatile organic compounds (VOCs) in exhaled breath as a marker of hypoxia in multiple chemical sensitivity. *Physiological Reports*, *9*(18). https://doi.org/10.14814/phy2.15034

Melkonyan, A., & Kuttler, W. (2012). Long-term analysis of NO, NO 2 and O 3 concentrations in North Rhine-Westphalia, Germany. *Atmospheric Environment*, *60*(December), 316–326. https://doi.org/10.1016/j.atmosenv.2012.06.048

Mendell, M. J. (2007). Indoor residential chemical emissions as risk factors for respiratory and allergic effects in children: A review. *Indoor Air*, *17*(4), 259–277. https://doi.org/10.1111/j.1600-0668.2007.00478.x

Missia, D. A., Demetriou, E., Michael, N., Tolis, E. I., & Bartzis, J. G. (2010). Indoor exposure from building materials: A field study. *Atmospheric Environment*, *44*(35), 4388–4395. https://doi.org/10.1016/j.atmosenv.2010.07.049

Nevarez, M. D., Rifas-Shiman, S. L., Kleinman, K. P., Gillman, M. W., & Taveras, E. M. (2010). Associations of early life risk factors with infant sleep duration. *Academic Pediatrics*, *10*(3), 187–193. https://doi.org/10.1016/j.acap.2010.01.007

Noguchi, M., Mizukoshi, A., Yanagisawa, Y., & Yamasaki, A. (2016). Measurements of volatile organic compounds in a newly built daycare center. *International Journal of Environmental Research and Public Health*, *13*(7), 1–14. https://doi.org/10.3390/ijerph13070736

Nuvolone, D., Petri, D., & Voller, F. (2018). The effects of ozone on human health. *Environmental Science and Pollution Research*, *25*(9), 8074–8088. https://doi.org/10.1007/s11356-017-9239-3

Oars Weel. (2014). 2-chloropropane. *Workplace Environmental Exposure Level*, *3*, 1–5.

Paciência, I., Madureira, J., Rufo, J., Moreira, A., & Fernandes, E. de O. (2016). A systematic review of evidence and implications of spatial and seasonal variations of volatile organic compounds (VOC) in indoor human environments. *Journal of Toxicology and Environmental Health - Part B: Critical Reviews*, *19*(2), 47–64. https://doi.org/10.1080/10937404.2015.1134371

Pei, J., Yin, Y., & Liu, J. (2016). Long-term indoor gas pollutant monitor of new dormitories with natural ventilation. *Energy and Buildings*, *129*, 514–523. https://doi.org/10.1016/j.enbuild.2016.08.033

Quarcoo, M. A., Strickland, P. O., & Shendell, D. G. (2019). Indoor ozone estimation from outdoor ozone and LBNL relocatable classroom study data. *Atmospheric Environment*, *213*(May), 491–498. https://doi.org/10.1016/j.atmosenv.2019.06.036

Raysoni, A. U., Stock, T. H., Sarnat, J. A., Chavez, M. C., Sarnat, S. E., Montoya, T., Holguin, F., & Li, W. W. (2017). Evaluation of VOC concentrations in indoor and outdoor microenvironments at near-road schools. *Environmental Pollution*, *231*, 681–693. https://doi.org/10.1016/j.envpol.2017.08.065

Rehwagen, M., Schlink, U., & Herbarth, O. (2003). Seasonal cycle of VOCs in apartments. *Indoor Air*, *13*(3), 283–291. https://doi.org/10.1034/j.1600-0668.2003.00206.x

Salonen, H., Salthammer, T., & Morawska, L. (2018). Human exposure to ozone in school and office indoor environments. *Environment International*, *119*(May), 503–514. https://doi.org/10.1016/j.envint.2018.07.012

Salvi, S. (2007). Health effects of ambient air pollution in children. In *Paediatric Respiratory Reviews* (Vol. 8, Issue 4, pp. 275–280). https://doi.org/10.1016/j.prrv.2007.08.008

Sarigiannis, D. A., Karakitsios, S. P., Gotti, A., Liakos, I. L., & Katsoyiannis, A. (2011). Exposure to major volatile organic compounds and carbonyls in European indoor environments and associated health risk. *Environment International*, *37*(4), 743–765. https://doi.org/10.1016/j.envint.2011.01.005

Schultz, E. S., Hallberg, J., Bellander, T., Bergström, A., Bottai, M., Chiesa, F., Gustafsson, P. M., Gruzieva, O., Thunqvist, P., Pershagen, G., & Melén, E. (2016). Early-life exposure to traffic-related air pollution and lung function in adolescence. *American Journal of Respiratory and Critical Care Medicine*, *193*(2), 171–177. https://doi.org/10.1164/rccm.201505-0928OC

Shin, S. H., & Jo, W. K. (2013). Longitudinal variations in indoor VOC concentrations after moving into new apartments and indoor source characterization. *Environmental Science and Pollution Research*, *20*(6), 3696–3707. https://doi.org/10.1007/s11356-012-1296-z

Suzuki, N., Nakaoka, H., Nakayama, Y., Takaya, K., Tsumura, K., Hanazato, M., Tanaka, S., Matsushita, K., Iwayama, R., & Mori, C. (2020). Changes in the concentration of volatile organic compounds and aldehydes in newly constructed houses over time. *International Journal of Environmental Science and Technology*, *17*(1), 333–342. https://doi.org/10.1007/s13762-019-02503-3

Tourula, M. (2011). *The childcare practice of children's daytime sleeping outdoors in the context of northern Finnish winter*.

Tourula, M., Isola, A., & Hassi, J. (2008). Children sleeping outdoors in winter: Parents' experiences of a culturally bound childcare practice. *International Journal of Circumpolar Health*, *67*(2–3), 269–278. https://doi.org/10.3402/ijch.v67i2-3.18284

Tourula, M., Isola, A., Hassi, J., Bloigu, R., & Rintamäki, H. (2010). Infants sleeping outdoors in a northern winter climate: Skin temperature and duration of sleep. *Acta Paediatrica, International Journal of Paediatrics*, *99*(9), 1411–1417. https://doi.org/10.1111/j.1651-2227.2010.01814.x

Venneman, J., Vandermeersch, L., Walgraeve, C., Audenaert, K., Ameye, M., Verwaeren, J., Steppe, K., Van Langenhove, H., Haesaert, G., & Vereecke, D. (2020). Respiratory CO2 Combined With a Blend of Volatiles Emitted by Endophytic Serendipita Strains Strongly Stimulate Growth of

Arabidopsis Implicating Auxin and Cytokinin Signaling. *Frontiers in Plant Science*, *11*(September), 1–25. https://doi.org/10.3389/fpls.2020.544435

VMM. (2018). *Uitstoot 2000-2017 en luchtkwaliteit 2018 in vlaanderen*.

VMM. (2019a). *Jaarrapport Lucht Emissies en concentraties van luchtverontreinigende stoffen*.

VMM. (2019b). *Streef- en advieswaarden ozon*. https://www.vmm.be/lucht/ozon/streef-en-advieswaarden-ozon

Wade, G. L. (2018). *Isopropyl alcohol*. Britannica. https://www.britannica.com/science/isopropyl-alcohol

Walgraeve, C., Demeestere, K., Dewulf, J., van Huffel, K., & van Langenhove, H. (2011). Diffusive sampling of 25 volatile organic compounds in indoor air: Uptake rate determination and application in Flemish homes for the elderly. *Atmospheric Environment*, *45*(32), 5828–5836. https://doi.org/10.1016/j.atmosenv.2011.07.007

Weschler, C. J. (2000). Ozone in Indoor Environments : Concentration and Chemistry. *Indoor Air*, 269–288.

WHO. (2006). *WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide Summary of risk assessment*.

WHO. (2010). WHO guidelines for indoor air quality: selected pollutants. *World Health Organization*.

World Health Organization. (2021). WHO global air quality guidelines. *Coastal And Estuarine Processes*, 1–360.

Supplementary Information

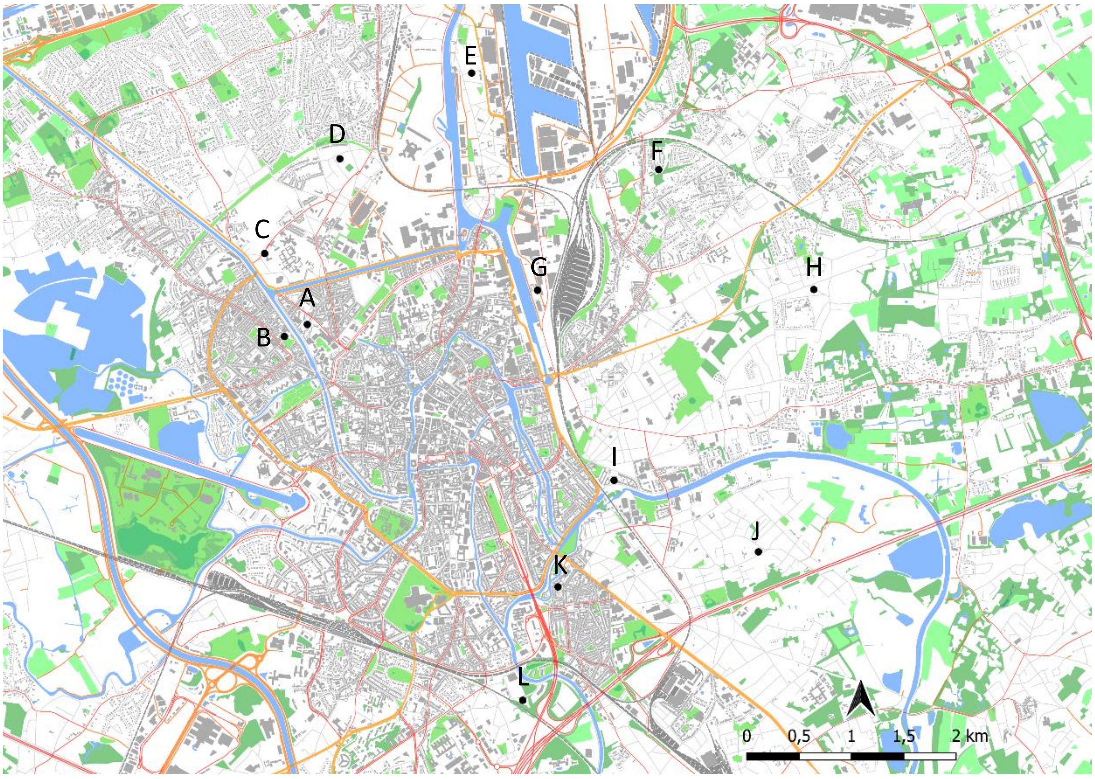

Figure A1: Sampling locations (12 DCCs) in Ghent (51°03'N 3°43'E), Belgium.

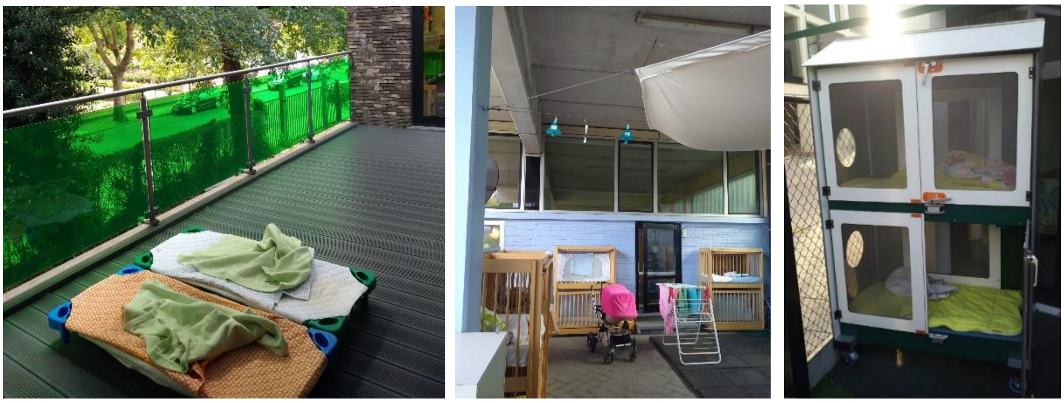

Figure A2: Different types of outdoor sleeping (left: plastic camps beds, middle: wooden indoor beds installed under a large roof, right: outdoor sleeping cubicles (Kids-F, NL)).

Table A1: Extra information about the ventilation system and environment of the DCCs, and the position of the outdoor samplers. When no structural protection from precipitation was available, Radiello Outdoor Shelter devices were used for the outdoor measurements.

Location (DCC)	**Extra information ventilation system and environment**	**Position outdoor samples**
A	Window ventilation grill	± 2.5 m from the building, under a shelter Backside of the building, at the playground
B	Mechanical ventilation supply New building since 2016 Park + busy street in vicinity	± 2 m from the building, under a party tent (open at all sides) Backside of the building, at the playground
C	Ventilation through windows Busy neighbourhood	± 2.5 m from the building, under a roof At the terrace on the first floor
D	No busy streets in the environment	± 10 m from the DCC building, under a shelter close to a wall (< 0.5 m) Backside of the building
E	Ventilation through windows Busy street, near the port	± 5 m from the building, under a roof Backside of the building, at the playground/terrace
F	Mechanical ventilation supply Renovation in 2016	± 10 m from the building, under a shelter Backside of the building, at the playground
G	New building, open since September 2019 Near old docks, R40 and railway. Construction works were still going on in neighbourhood.	± 3 m from the building, under a roof Backside of the building, at a terrace
H	Mechanical ventilation supply: ventilation system permanently on between 6 a.m. and 6 p.m	± 1.5 m from the building, under a roof At the back of the building, at the playground
I	Mechanical ventilation supply + window ventilation grill Near the Schelde and R40	± 3 m from the building, under a shelter Backside of the building, at the playground
J	Central ventilation system (mechanical ventilation supply) Near E17	± 1 m from the building, under a roof At the terrace on the first floor Roadside in front of the building
K	Window ventilation grill Near R40 with the Schelde in between	± 1.5 m from the building, under a roof Backside of the building, at the playground
L	New extra building in 2018 Near E17 and railway	± 1 m from the building, under a shelter At the playground/garden side (right ride of the DCC)

Sample preparation - Loading tubes with internal standard Toluene-d8

The conditioned tubes were loaded with a gaseous internal standard (IS; perdeuterated toluene (Tol-d8; min.99.96 atom.%D; Sigma Aldrich)), prepared by a two-phase system according to the procedure described by (Demeestere et al., 2008). In short, 20 µL of a stock solution containing Tol-d8 (226.3 µg/mL) in methanol (Optima® LC-MS grade, Fisher Scientific) was added to 20 mL of demineralized water in a dark penicillin bottle (119.78 mL) (Novolab). The bottles were immediately sealed with a Mininert valve (Supelco, Sigma-Aldrich Belgium) and PTFE tape, and placed in a water bath at a constant temperature of 25°C for at least 12 h. From the headspace of these two-phase systems, 500 µL (pressure-lok series A syringe 500 µL, Supelco) corresponding to 10.83 ng Tol-d8, was loaded through a home-made heated (150°C) injector onto the sorbent tubes under a 100 mL/min He flow. After injection, the He stream was kept on for 3 min before the tubes were closed again with ¼'' brass caps and PTFE ferrules, wrapped in aluminium foil and stored in a desiccator.

Table A2: Selected VOCs, divided into classes, with their retention time (RT), presence in calibration mixture A or B and the used uptake rate (UR). The 5th column refers to the source of the UR: 1 = effective UR determined in indoor environment by Walgraeve et al. (2011) (with stdev if the UR could be determined in more than one sampling campaign); 2 = effective UR calculated based on the formula of Jia et al. (2017) ($U_{eff} = \alpha \, . \, UR_{ideal}$ with the 7-day passive sampling efficiency 7d-α = 0.3626 . ln(boiling point) - 1.2324); 3 = average class-based UR (for 2-methyl-butane n = 11, for o-xylene n = 7, for 3-carene n = 2); 4 = average calculated UR (n = 25) based on all the URs values in this table with source 1 or 2. The right column shows the selected characteristic ions (not overlapping with ions of neighbouring compounds).

Compound	RT	Calibration mixture	UR (mL/min)	Source UR	Selected ions
Alkanes					
2-methyl-butane	3.39	-	0.26 ± 0.05	3	41; 57; 72
n-hexane	7.14	A	0.16	1	41; 71; 86
methylcyclopentane	8.81	B	0.20	1	55; 56; 69; 84
cyclohexane	11.28	B	0.25 ± 0.20	1	69; 84
n-heptane	14.77	B	0.26 ± 0.05	1	70; 71; 100
methylcyclohexane	16.78	-	0.24	2	55; 83; 98
n-octane	24.32	A	0.27 ± 0.10	1	71; 85; 114
n-nonane	29.39	B	0.34 ± 0.07	1	43; 85; 128
n-decane	32.69	B	0.28 ± 0.02	1	57; 85; 142
n-undecane	35.33	A	0.28	1	43; 57; 85; 156
n-dodecane	37.47	A	0.26	1	43; 57; 71; 85; 170
n-tridecane	39.17	B	0.30	2	57; 71; 85
Aromatic hydrocarbons					
benzene	10.65	A	0.27 ± 0.03	1	51; 52; 63; 77; 78
toluene	20.27	A	0.32 ± 0.04	1	65; 91; 92
ethylbenzene	27.32	A	0.35 ± 0.04	1	91; 106
m & p-xylene	27.77	A/B	0.36 ± 0.04	1	91; 105; 106
styrene	28.60	A	0.36 ± 0.05	1	78; 104
o-xylene	28.79	A	0.36 ± 0.05	3	91; 105; 106
benzaldehyde	30.89	B	0.41 ± 0.06	1	51; 77; 105; 106
benzoic acid	36.59	-	0.29 ± 0.07	4	77; 105; 122
phenol	31.91	B	0.43 ± 0.03	1	66; 94
Terpenes					
α-pinene	30.78	A	0.20 ±0.03	1	92; 93; 121; 136
3-carene	33.16	A	0.24 ± 0.05	3	77; 91; 93; 121; 136
limonene	33.61	B	0.27 ± 0.04	1	67; 68; 93; 107; 121; 136
Alcohols					
isopropanol	3.60	B	0.29 ± 0.07	4	31; 45; 59
2-methyl-2-propanol	4.19	-	0.29 ± 0.07	4	93; 95; 121; 136
1-butanol	11.02	B	0.29 ± 0.07	4	31; 43
1-pentanol	20.58	B	0.29 ± 0.07	4	29; 31; 55; 57; 70
2-ethyl-1-hexanol	33.27	A	0.29 ± 0.07	4	31; 41; 56; 57; 70; 83; 112
benzyl alcohol	33.32	-	0.29 ± 0.07	4	77; 79; 108
Aldehydes					
hexanal	22.69	-	0.17	1	44; 56; 72; 82
nonanal	35.08	A	0.29 ± 0.07	4	29; 41; 57; 70; 82; 98
decanal	37.36	-	0.29 ± 0.07	4	41; 43; 57
Ketones					
2-butanone	6.19	B	0.29	2	43; 71; 72; 73
acetophenone	34.16	A	0.39	2	51; 77; 105; 120

Continuation Table A2

Chlorinated hydrocarbons					
2-chloro-propane	3.75	-	0.29 ± 0.07	4	78; 80; 81
dichloromethane	4.29	B	0.29 ± 0.07	4	49; 84; 86
Ethers					
ethyl ether	3.85	-	0.29 ± 0.07	4	31; 45; 59; 74
1-methoxy-2-propanol	11.55	-	0.29 ± 0.07	4	31; 45; 47
2-butoxy-ethanol	29.13	-	0.29 ± 0.07	4	29; 41; 45; 57; 87
1-butoxy-2-propanol	30.48	-	0.29 ± 0.07	4	45; 57; 87
2-(2-ethoxyethoxy)-ethanol	32.18	-	0.29 ± 0.07	4	45; 59; 72
Acids					
acetic acid	7.32	-	0.29 ± 0.07	4	60; 40
hexanoic acid	31.97	-	0.29 ± 0.07	4	60; 73
Esters					
ethyl acetate	7.3	B	0.23 ± 0.03	1	70; 88
isobornyl acetate	39.19	-	0.31	2	93; 95; 121; 136
Nitriles					
benzonitrile	31.5	B	0.29 ± 0.07	4	76; 103

Table A3: TVOC concentrations [µg/m³] calculated as the sum of the 31 VOCs for which RRFs were determined. Indoor sample of DCC E in the sleeping room was lost. Values in bold are higher than the Belgian guideline value (300 µg/m³).

	TVOC concentration [µg/m³] (sum of 31 VOCs)					
	September			**January**		
DCC	**Living area**	**Sleeping room**	**Outdoors**	**Living area**	**Sleeping room**	**Outdoors**
A	229	**326**	23.4	134	129	31.9
B	160	77.8	21.7	142	77.2	27.3
C	152	64.7	29.7	106	35.2	27.4
D	102	85.1	25.9	204	35.8	32.4
E	110	61.8	24.1	250	-	33.8
F	152	83.8	15.5	94.9	227	30.8
G	**422**	**826**	29.7	**623**	**390**	35.6
H	92.6	60.7	25.7	78.7	52.6	25.9
I	246	146	18.4	**354**	240	31.9
J	220	155	24.9	219	124	28.2
K	187	68.0	22.6	184	53.5	22.4
L	266	264	27.5	**329**	165	30.4

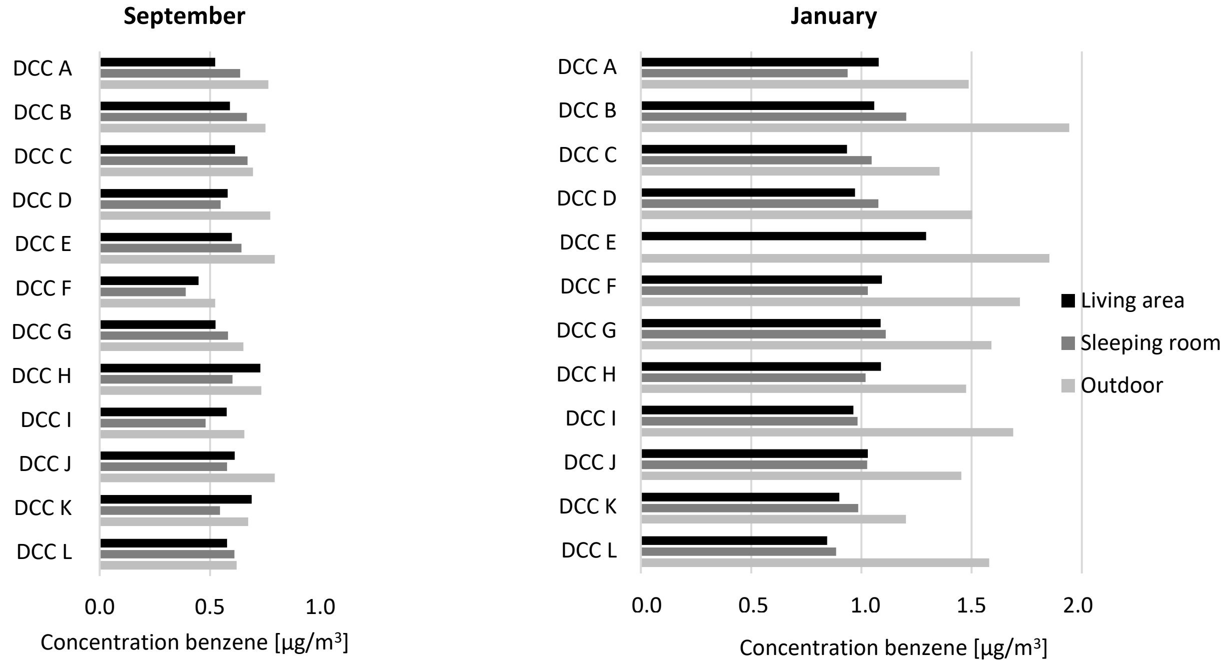

Figure A3: Benzene concentrations [µg/m³] in September (left) and January (right) measured indoors (living area and sleeping room) and outdoors. Sample of DCC E in the sleeping room in January was lost. Indoor concentrations are always lower than outdoors (except for DCC K in September: outdoors 0.67 µg/m³ and living area 0.69 µg/m³, however, this very small difference is negligible).

The larger outdoor concentrations indicate a (strong) outdoor source like e.g. traffic. Both indoor and outdoor concentrations are higher in the January campaign compared to the September measurements. Possible explanations are the lower dispersion of air in the winter due to thermal inversion or in general unfavourable meteorological conditions (e.g. lower mixing height), the longer persistence of the compounds due to reduced photochemical degradation, and an increase in pollutant emissions during winter (heating of buildings, traffic (cold starts)) (Bozkurt et al., 2018) (Fuselli et al., 2010) (Paciência et al., 2016). The difference between indoor and outdoor concentrations in the September campaign is smaller than in the January campaign. The calm weather and temperature during the first sampling campaign encourage more ventilation by open windows, which can result in higher AERs and consequently higher infiltration of benzene, resulting in similar concentrations indoors and outdoors.

Indoor/outdoor ratios

Six compounds are not quantifiable (<LOQ) at any location in the outdoor air (1-butanol, 3-carene, isobornyl acetate, 2-chloropropane, 2-(2-ethoxyethoxy)-ethanol and 2-butoxy-ethanol), neither in September nor in January. Six other VOCs (1-pentanol, 2-methyl-2-propanol, benzyl alcohol, ethyl ether, 1-methoxy-2-propanol and 1-butoxy-2-propanol) are measured outdoors at maximum 3 locations in September and January at very low concentrations (≤ 0.4 µg/m^3). For the remaining 35 VOCs, indoor/outdoor ratios are calculated where possible (hexanoic acid, decanal and hexanal are not or only at 1 location measurable outdoors in January). For the indoor concentration, an average of the living area and sleeping room is used.

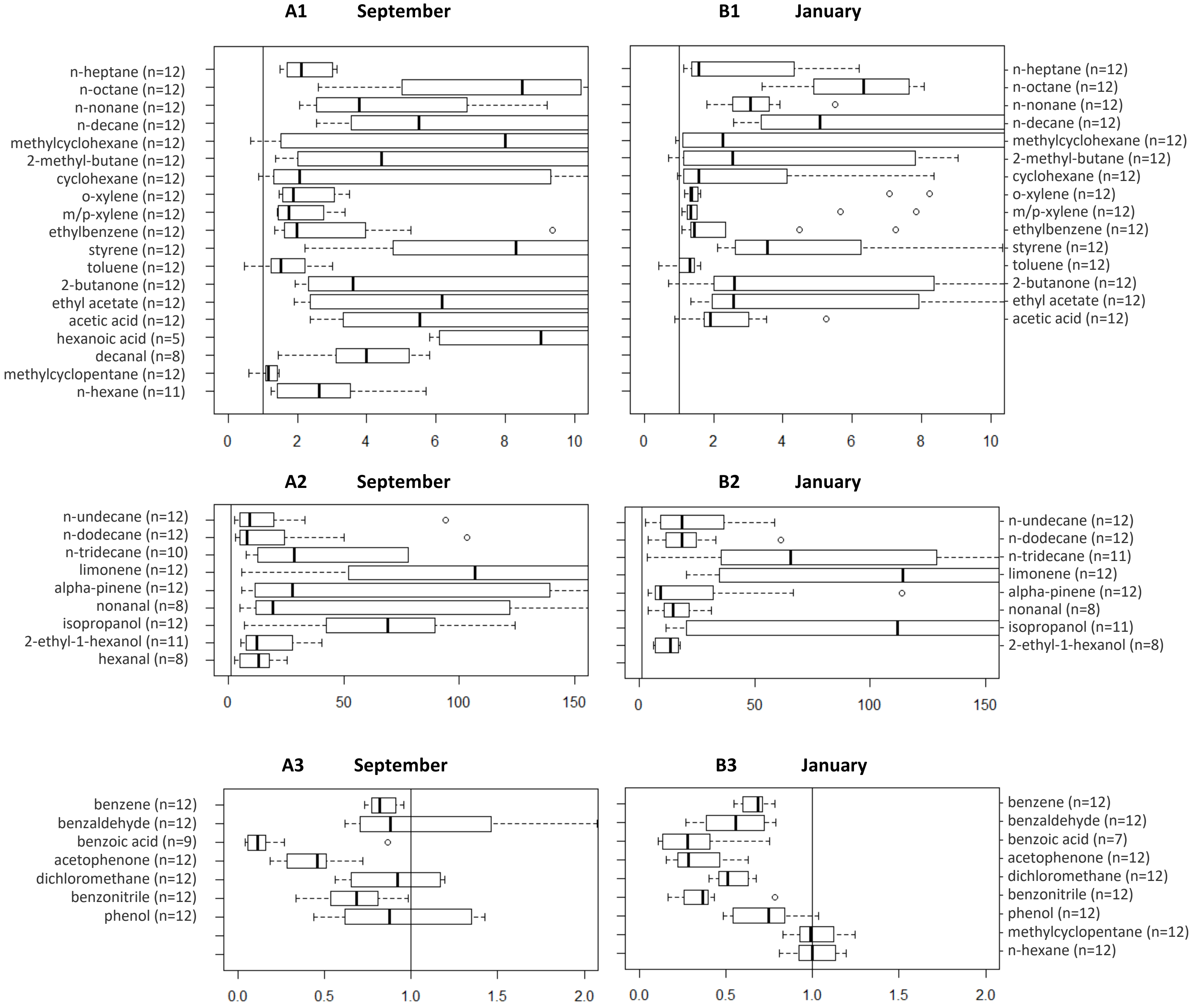

Figure A4: Indoor/outdoor ratio of measured VOC concentrations. Left: September, Right: January. A1, B1, A2, B2: Compounds with a ratio of the first quartile higher than one. A3, B3: Compounds with a median ratio lower than one. A1, B1: enlargements of a ratio from zero to ten. A2, B2: enlargements of a ratio from zero to 150. A3, B3: enlargements of a ratio from zero to two. Methylcyclopentane and n-hexane show in September slightly higher ratios than one; in January ratios around one.
The boxes shown represent the 25, 50 (median) and 75 percentiles. Indoor values are the average of the concentrations measured in the living area and sleeping room.

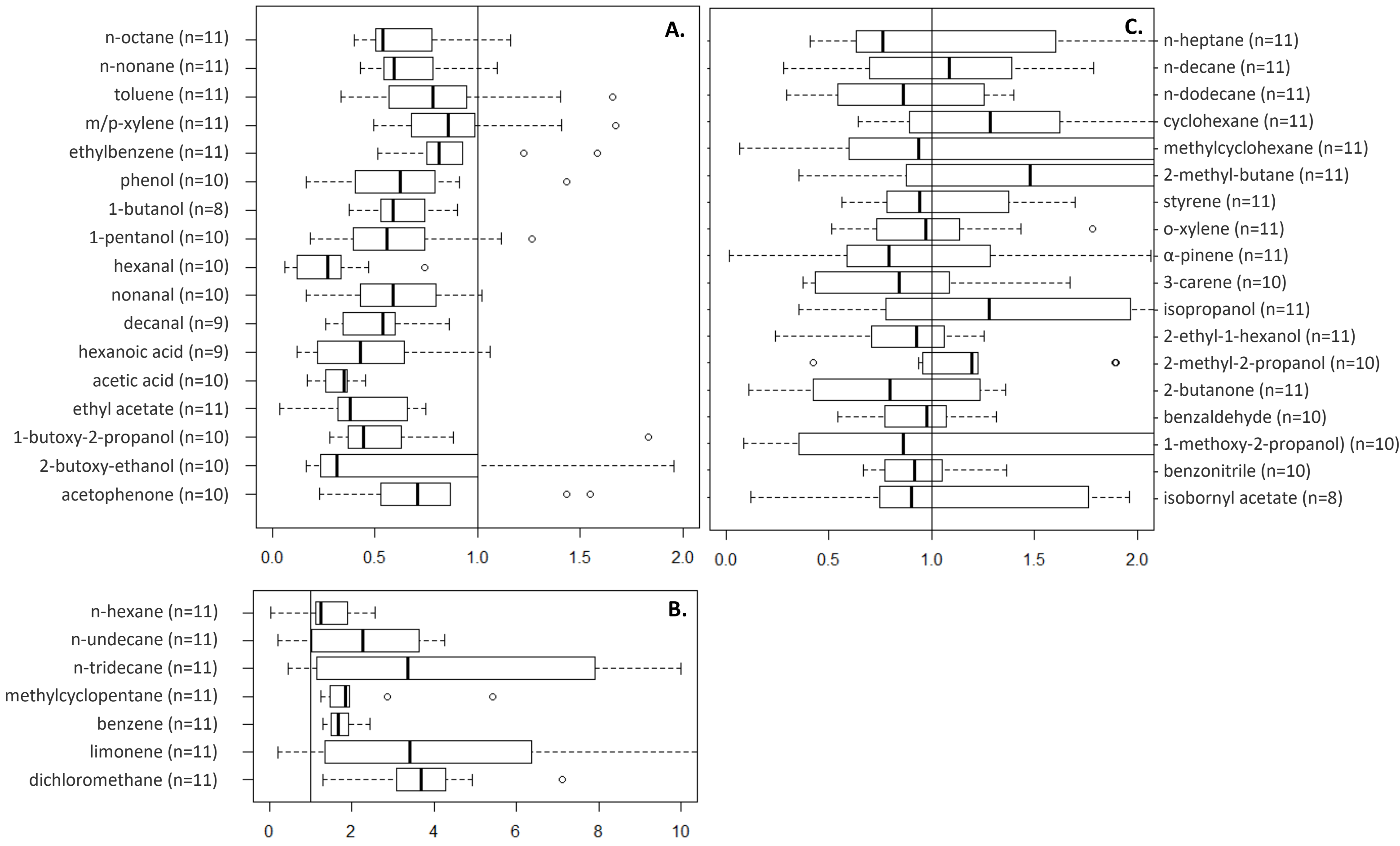

Figure A5: Indoor (living area) concentration ratios January/September. (A.) VOCs with the 75th percentile below one; (B.) VOCs with 25th percentile higher than one; (C.) 'one' between 25th and 75th percentile. A and C are enlargements of a ratio from zero to two; B is an enlargement of a ratio from zero to ten. DCC G (new building) is left out of consideration here because of the notable decrease in indoor VOCs at this location. The boxes shown represent the 25, 50 (median) and 75 percentiles.

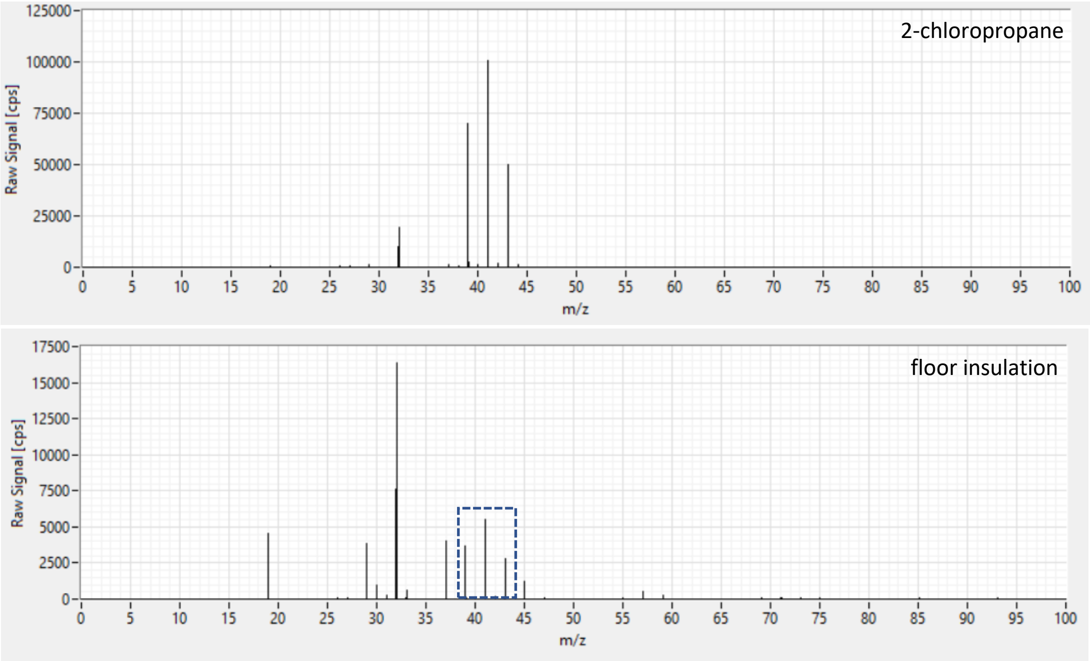

Figure A6: Spectra of the PTR-QiToF-MS analysis of pure 2-chloropropane headspace (top) and floor insulation Kingspan Kooltherm K3 (bottom). Both spectra are corrected with the spectrum of a blank Nalophan bag filled with only N_2. The protonated product ions of 2-chloropropane (m/z = 39.023 ((C_3H_2)H^+), 41.039 ((C_3H_4)H^+), 43.055 ((C_3H_6)H^+)) are clearly present in the floor insulation sample. (m/z = 19.018 = ((H_2O)H^+); 29.013 = (N_2)H^+; 31.990 = O_2^+; 37.029 = (H_3O^+.H_2O)).

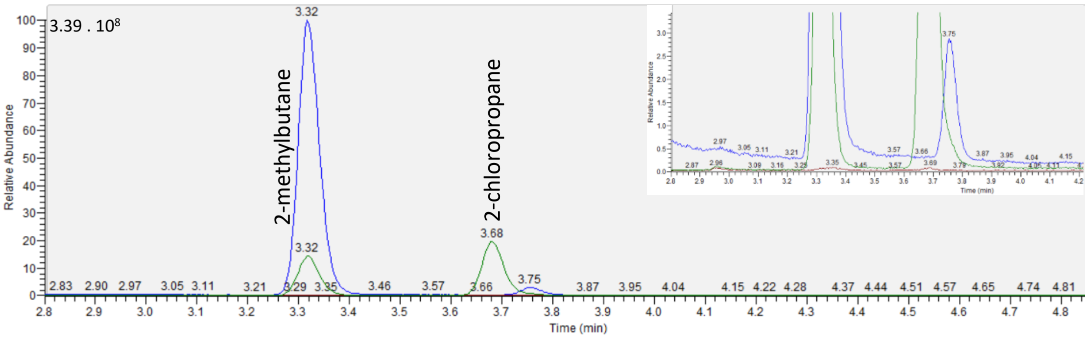

Figure A7: Chromatogram (retention time 2.8-4.9 min) of the TD-GC-MS analysis of headspace of the floor insulation (green), wall insulation (blue), and the rubber floor top layer (red). At RT 3.32 min, 2-methylbutane was measured in the headspace of the wall and floor insulation; at RT 3.68 min, 2-chloropropane was measured in the headspace of the floor insulation. The peak at RT 3.75 min represents n-pentane.

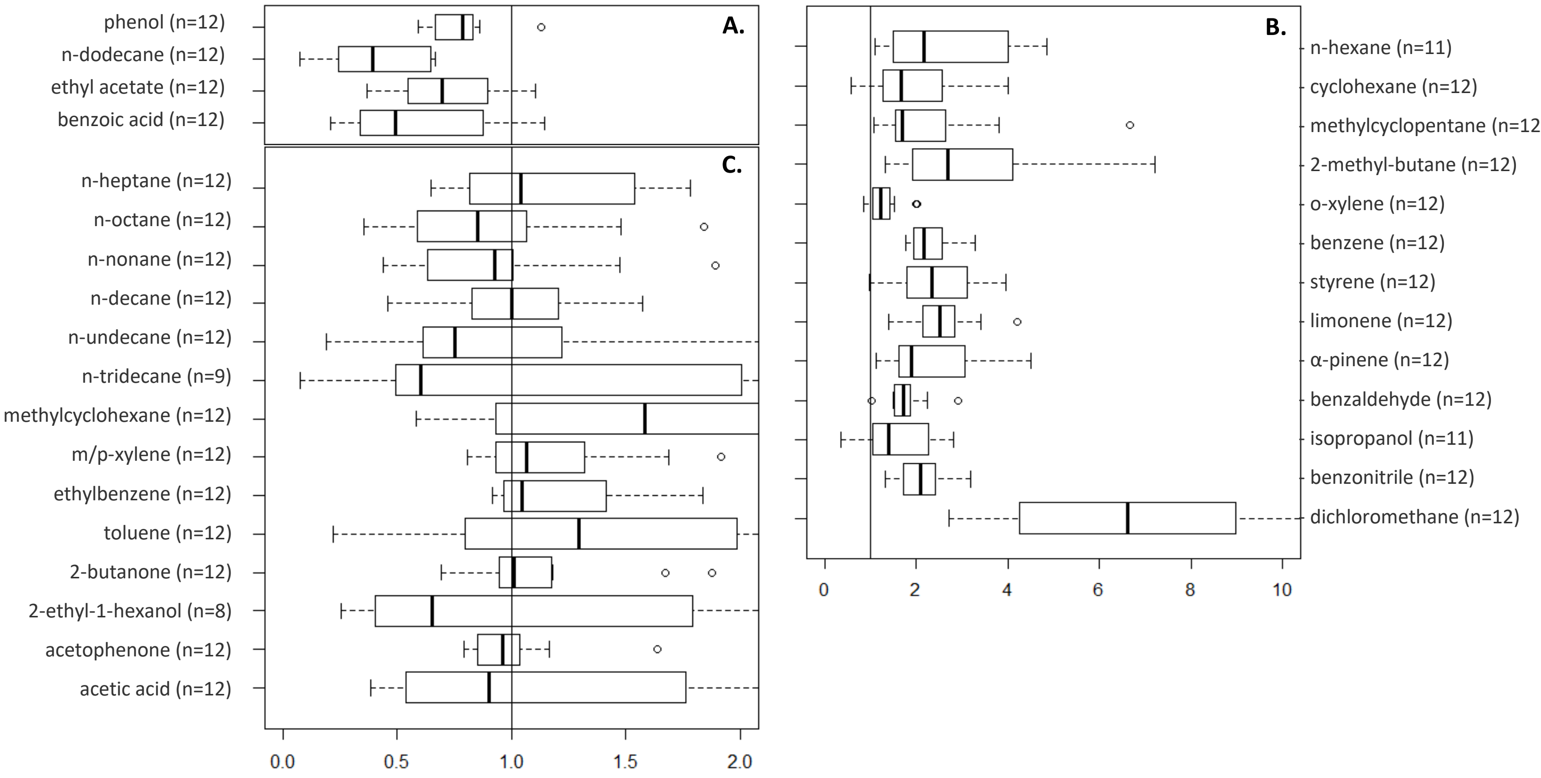

Figure A8: Outdoor concentration ratios January/September. (A.) VOCs with the 75th percentile below one; (B.) VOCs with the 25th percentile higher than one; (C.) 'one' between the 25th and 75th percentile. A and C are enlargements of a ratio from zero to two; B is an enlargement of a ratio from zero to ten. The boxes shown represent the 25, 50 (median) and 75 percentiles.