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# Evaluation of the Seasonal Variation of VOC Surface Emissions and Indoor Air Concentrations in a Public Building with Bio-Based Insulation

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### 15 Abstract

16 The use of bio-based insulation materials is widely spreading in buildings. Due to their 17 organic load, they can be an important source of Volatile Organic Compounds (VOCs). This 18 study is among the first that evaluates the spatial and seasonal in-field VOC surface 19 emissions from bio-based and conventional building structures as a whole, in a French public 20 building insulated with wood wool. In addition to surface emissions, measurements of VOC 21 concentrations in indoor air were taken. Results showed that a spatial difference (up to 5 22 times) in VOC emissions was observed due to the inhomogeneity of the surface. Moreover, 23 the cardinal orientation of building structures with the same constitution induced a 24 difference (up to a factor 30) in emission rates due to the exposure to different 25 hygrothermal conditions. The variation in temperature and relative humidity between 26 seasons led to higher summer VOC emissions and indoor air concentrations. In addition, 27 indoor VOC concentrations were shown to be higher at night compared to daytime due to 28 the decreased ventilation rate. Furthermore, an interesting approach was developed in this 29 study to have a primary overview of the impact of surface emissions on indoor VOC levels. 30 Results confirmed that the three bio-based walls have no significant specific VOC emissions 31 at high rates compared to the floor and the ceiling. Bio-based insulations showed no impact 32 on microbial indoor air concentrations during the two seasons. Moreover, no detected VOCs 33 could be attributed to microbial development as they were also emitted from building 34 materials.

### 35 Keywords

36 Bio-based insulations, VOC, in-field emissions, indoor air, seasonal campaigns

37 Highlights

- Spatial, temporal, and seasonal variations affect in-field measured VOC emissions.
- VOC emissions rates from bio-based and conventional structures are comparable.
- No specific VOCs are exclusively emitted from bio-based surfaces.
- Emissions from building structures contribute moderately to VOC indoor air levels.
- No direct link between VOCs and microbial development could be evidenced.

#### 43 **1. Introduction**

44 Indoor air pollution was traditionally overlooked compared to outdoor air pollution. 45 However, studies showed that the level of some organic pollutants is two to four times 46 higher indoors than outdoors [1]. Nowadays, indoor air quality (IAQ) receives large public 47 attention as people spend most of their times indoors, which has been exacerbated during 48 the COVID crisis [2]. During this time, they are exposed to indoor pollutants which can lead 49 to adverse health effects. Volatile organic compounds (VOC) are widely spread indoor 50 pollutants. These compounds can originate from anthropogenic activities, infiltration of 51 outdoor air, or indoor products emissions [3].

52 The indoor sources of VOC emissions can be mainly divided into two categories: short-term, 53 intensively emissive sources, such as cooking activities, air fresheners, and cleaning products 54 [4], and long-term emissive sources, like building materials and furniture [5], [6]. The latter 55 are considered an important source of VOCs. Molhave et al. found, in their Danish twin apartment study, that the concentration of total VOCs (TVOC) in an unoccupied apartment 56 one year after construction was equal to 0.5 mg.m<sup>-3</sup>, due to emissions from building 57 58 materials and furniture [7]. Several European regulations, labels, schemes, and directives 59 were established to classify building and construction materials based on their VOC 60 emissions in order to minimize human's indoor exposure to these compounds. Among them 61 is the French label issued in 2011 by the Ministry of Ecology [8], the German Committee for 62 Health-related Evaluation of Building Products (AgBB) [9], M1 Finnish label, Indoor Climate 63 Label (ICL), Natureplus, and the Blue Angel label [10]. Similarly, the European directive 64 2004/42/CE, was issued in 2004 to reduce air pollution by limiting the total VOC content in 65 certain paints and consumer products [11]. In 1988, the European directive 89/106/EEC was 66 founded with the aim of harmonizing the test methods, the declaration of performance 67 values, and the conformity assessment methods of construction products between member 68 states of the European Union [12]. Later on, this directive was replaced by the regulation 69 305/2011/EU in 2011 [13].

In France, approximately 20 million buildings were built before the introduction of the thermal regulations in 1975, and these buildings account for more than 75% of energy consumption in the housing sector [14]. However, environmental awareness and fights against global warming led to the first thermal regulations in 1975, imposing thermal insulation in walls and the regulation of heating systems with the aim of reducing energy consumption. In this context, and as part of the Grenelle Environment agreements, a French law was issued in 2009 stating that the primary energy consumption in buildings should be reduced by 38% by the year 2020 [15]. It also engaged to renovating 400 thousand dwellingsper year starting from the year 2013.

79 The construction or demolition of buildings as well as the production and use of building 80 materials account for high carbon dioxide emissions. Therefore, the French Ministry of 81 Ecology has issued new environmental regulation to be applied from January 2022 aiming to 82 reduce the carbon footprint of new buildings over their estimated 50-year life cycle [16]. 83 Hence, finding new building materials, especially insulation materials, which enable the 84 decrease of energy consumption and greenhouse gas emissions is necessary. The use of low 85 carbon materials and reuse of recycled materials are effective approaches to reduce carbon 86 dioxide emissions by up to 28% at the construction site [17]. The use of natural materials 87 and in particular bio-based materials, is also found to reduce the embodied energy and 88 carbon dioxide impact of construction [18], [19]. As a result, the use of bio-based insulation 89 materials has been spreading widely due to their lower production energy and enhanced 90 insulation properties [20]. In 2016, bio-based insulations represented already 60% of the 91 available bio-sourced products on the French market, illustrating the growth of this sector 92 [21]. However, taking into consideration the inherent carbon-based chemical structure of 93 these materials, bio-based insulations are a potential source of VOC emissions and could 94 potentially contribute to IAQ deterioration. Moreover, being composed of natural fibers, 95 bio-based insulation materials could serve as a substrate for the development of 96 microorganisms, such as fungi, under highly humid conditions [22]. Upon microbial 97 development, fungi could emit microbial VOCs (mVOCs) [23].

Several studies evaluate VOC emissions from bio-based insulations in test chambers. Their
results showed that mainly acetic acid, aldehydes, alcohols, and aliphatic hydrocarbons are
the main VOCs emitted from hemp, flax, cellulose, and wood wool insulation materials [5],
[18], [24]–[27]. When comparing VOC emissions between bio-based and conventional
building materials, bio-based materials are generally classed as low-emitting [18], [28].
However, in some other studies, wood insulations have shown to have the highest TVOC and
aldehydes emissions compared to other building materials [19], [26], [29].

105 However, most of the existing studies that characterize VOC emissions from bio-based 106 materials, particularly insulation, are based on laboratory tests employing test chambers or 107 emission cells to characterize VOC emissions from individual materials that are part of 108 different building structures (walls, ceiling, and floor). However, until today, and to our 109 knowledge, there are only a few studies that report the on-site emission of VOCs from a 110 building structure as a whole (wall or floor) [30], [31]. Therefore, the original objective of this 111 study is to characterize the in-situ surface emissions of the major VOCs emitted from the 112 building structures as an entity, especially those containing bio-based insulation, which could 113 help to have a better perspective on IAQ. Moreover, since emissions from building materials 114 and thus building structures can be greatly affected by the change in temperature and 115 relative humidity [32]-[35], it is interesting to evaluate the seasonal variation on VOC 116 emissions.

117 In this context, two seasonal, summer and winter, field campaigns took place at a public 118 building (town hall) in the North of France. The choice of the building was mainly based on two factors. The first was that the selected building should contain bio-based insulation materials to answer the implemented approach of the project in study. The second criterion was that the targeted building should be publicly accessible, as these buildings are less affected by occupant-specific activities (such as cooking and cleaning), and since a wider public can be impacted by IAQ in these buildings.

124 This paper presents in its first part the spatial and seasonal variation of VOC surface 125 emissions from several building structures, in addition to the comparison of VOC emissions 126 from building structures including bio-based insulation to conventional ones. The second 127 part of this paper discusses the contribution of bio-based and conventional surface 128 emissions to indoor air concentrations in a real-world building, an understudied scale for 129 VOC measurements. This paper also includes indoor air microbial measurements in order to 130 estimate the potential contribution of microorganism to VOC emission, more specifically 131 mVOCs.

132

### 133 **2. Materials and Methods**

#### 134 2.1. Selection of the Building

The town hall was renovated in 2014, with wood wool as the insulation material, which is 135 good for our study to prevent the overestimation of VOC emissions from new building 136 137 materials. The wedding hall, a semi-detached building, was selected to conduct this study. 138 The choice of the hall was based on the presence of the greatest number of surfaces 139 containing bio-based building materials compared to the other surfaces. Moreover, it was 140 also less often occupied, which made it more accessible for conducting the desired 141 experiments, and thus IAQ in this hall was less impacted by occupants' activities. The bio-142 based structures of the hall consisted of the floor and three outer walls insulated with wood wool, in addition to apparent wood structures (beam). The surface area of the floor was 143 about 85 m<sup>2</sup> for a room volume of 441 m<sup>3</sup>. Two seasonal field campaigns were carried out in 144 145 this building to evaluate VOC surface emissions and indoor air concentrations. The summer 146 campaign was conducted from 8 to 19 July 2019 while the winter campaign was carried out 147 from 28 January to 3 February 2020.

148 Figure 1 shows a schematic representation of this room. It included three walls exposed to 149 the exterior and constituted, from the exterior to the interior, of brick, followed by a layer of 150 120 mm thick wood wool, a vapor barrier, another 60 mm layer of wood wool, and two 18 151 mm plasterboards covered with a layer of painting, as shown in figure 2. These three walls 152 were oriented southeast (SE), southwest (SW), and northwest (NW). The northeast-oriented 153 wall (wall 1) in addition to half of the northwest-oriented wall (wall 2) were interior walls 154 with lighter insulation with rockwool of 45 mm thickness. The floor was made up of floating 155 parquet insulated by wood wool flakes (220 mm) in between. The ceiling was composed of 156 prefabricated sandwich panels insulated with 180 mm polyurethane foam and covered with 157 18 mm plasterboard. On the SE side, the roof was equipped with solar panels. The hall also 158 showed a number of wood beams from the roof structure. It also contained some tables, a 159 group of chairs, and some plants.

However, the three walls (SE, SW, and NW) containing wood wool will be considered the only bio-based surfaces in this study in order to facilitate the comparison of VOC emissions between similar building structures. In addition, the total surface area of the used bio-based insulation in the three walls is greater than that of the floor and the beam compared to the volume of the hall, and thus VOC emissions from bio-based materials are expected to be more remarkable from these walls rather than from the other bio-based surfaces.

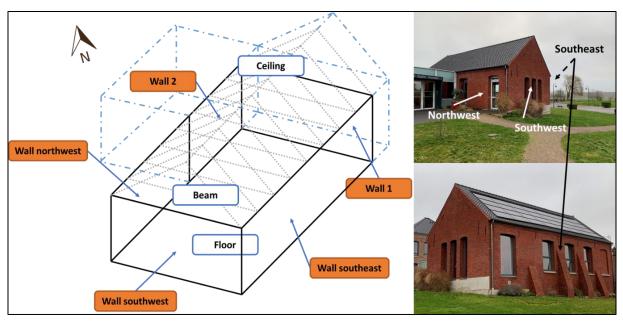




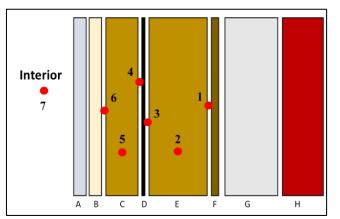
Figure 1. A schematic representation of the marriage hall at the town hall

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2.2. Monitoring the Temperature and Relative Humidity in the Bio-based Walls

Since surface emissions can be greatly affected by the change in temperature and relative humidity, 7 sensors (HMP 60 Vaisala) were installed for one year inside the SE wall to monitor the change in temperature (T) and relative humidity (RH) at different spots of the structure (sensors 1 to 6) and inside the wedding hall (sensor 7) (figure 2).



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Figure 2. The installation of 7 T and RH sensors, indicated by numbers 1-7, inside the
 southeast wall. A and B: plasterboards, C: 60 mm wood wool, D: vapor barrier, E: 120 mm
 wood wool, F: air blade, G: concrete block, and H: brick

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#### 179 2.3. VOC Surface Emissions

180 *2.3.1.* Instrumentation

181 Two FLECs (Field and Laboratory Emission Cells) were used to characterize VOC emissions 182 from the eight different surfaces as indicated in figure 1: the three exterior walls (SE, SW, 183 and the exterior part of the NW wall), two interior walls (wall 1 and part of the NW wall, 184 referred to as wall 2), the floor, the ceiling, and a beam. Once installed on a surface, the 185 FLECs were supplied with a flux of 500 mL min<sup>-1</sup> zero air provided by a zero-air generator 186 (Clained Brezza, Italy) at ambient temperature and 50% relative humidity (provided by a 187 system of bubblers).

188

#### 189 *2.3.2. Sampling*

190 Two surfaces were characterized simultaneously per day. Active sampling occurred during 191 daytime (from approximately 9h-17h) 24 hours after the installation of the FLEC on the 192 surface, following the European standard ISO 16000-10 [36]. Sampling was done 193 simultaneously using 2,4-dinitrophenylhydrazine (DNPH) cartridges, for carbonyl 194 compounds, and Tenax TA tubes, for the other emitted VOCs. Before sampling, the Tenax TA 195 tubes were conditioned at 250 °C under a nitrogen flow for at least 8 hours. After sampling, 196 the Tenax TA tubes were tightly closed and kept at ambient temperature while the DNPH 197 cartridges were stored in the freezer (- 21 °C) until analysis, which was realized at the end of 198 each campaign. The sampling flow rate and time were adjusted to have a sampling volume 199 equal to 36 L. Potential breakthrough of VOCs on the adsorbents was considered by using 200 two adsorbent supports in series, for both DNPH and Tenax TA sampling. Table 1 201 summarizes the sampling from each surface with the number of collected samples as well as 202 the sampling dates and hours during both seasons.

Sampling of the five different walls took place at about 1.5 m height. However, as shown in
table 1, sampling of the SE wall occurred at two different heights (0.5 and 1.5 m) during the
summer campaign to evaluate the spatial effect on VOC emissions.

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## Table 1. A summary of the number of samples collected from each surface and theircorrespondent sampling dates and hours; n is the number of collected samples

Surface		Summ	er		Winter						
_	n	Date	Sampling time	n	Date	Sampling time					
Beam	1	08/07/2019	14h – 17h	3	29/01/2020	15h – 00h					
Floor	2	08/07/2019	11h – 17h	2	01/02/2020	09h – 15h					
Wall 1	2	10/07/2019	11h – 17h	3	03/02/2020	09h – 17h					
Wall 2	1	10/07/2019	14h – 17h	3	03/02/2020	08h – 17h					
Wall SW	1	12/07/2019	14h – 17h	3	29/01/2020	15h – 00h (+1)					
Wall SE-1.5 m	5	13-14/07/2019	04h - 00h (+1)	2	01/02/2020	09h – 15h					

Wall SE-0.5 m	3	14/07/2019	22h - 07h (+1)	-	-	-
Wall NW	3	16/07/2019	02h - 14h	3	28/01/2020	08h – 17h
Ceiling	1	16/07/2019	14h – 17h	3	28/01/2020	08h – 17h

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Moreover, in order to evaluate VOC concentration in indoor air and characterize the impact of surface emissions on indoor air VOC levels, a 24-hour sampling (corresponding to 8 x 3 hsamples) of indoor ambient air took place, during both seasons. A field blank was collected every sampling day by opening a DNPH cartridge or Tenax TA tube for few seconds and then tightly closing it.

215

#### 216 2.3.3. Sample Analysis

DNPH cartridges were first eluted by 3 mL acetonitrile (velocity =  $1.5 \text{ mL min}^{-1}$ ) and then 217 analyzed using an HPLC-UV (High Performance Liquid Chromatography coupled to Ultra 218 219 Violet Detector, Dionex Ultimate 3000, Thermo Scientific U.S.A.) equipped with an Acclaim 220 RSLC Carbonyl column (2.2 µm, 2.1 x 150 mm, Thermo Scientific, U.S.A.). Tenax TA tubes 221 were analyzed using a TD-GC-MS/FID system (Thermal Desorption Gas Chromatography, 222 Clarus 680 - Mass Spectrometry and Flame Ionization Detection, Clarus SQ 8T, Perkin Elmer, 223 U.S.A.) with a CP-Sil 5CB column (60 m x 0.25 mm x 1 µm, Agilent U.S.A.). A description of 224 the used TD-GC-MS/FID and HPLC-UV methods is presented in tables S1 and S3, respectively, 225 of the supporting information.

226 Calibrations were run before each HPLC and GC analysis. The 20 Carbonyls analyzed by HPLC 227 were quantified using their individual calibration coefficients. These include formaldehyde, 228 acetaldehyde, propanone, acrolein, propanal, butanal, crotanaldehyde, benzaldehyde, 229 pentanal, isopentanal, tolualdehyde (o-, m-, and p-), methylpropenal, methyl vinyl ketone, 230 methyl ethyl ketone, glyoxal, methylglyoxal, hexanal, and 2,5-dimethylbenzene. For the TD-231 GC-FID quantification a calibration was performed with authentic standards of benzene in 232 addition to the eight VOCs (toluene, ethylbenzene, xylene, styrene, 2-butoxyethanol, tetrachloroethylene, 1,2,4-trimethylbenzene, and 1,4-dichlorobenzene) targeted by the 233 234 French label [8]. Other detected VOCs in addition to TVOCs were quantified by FID as 235 toluene equivalent. The emission of TVOCs from building materials is also standardized by 236 the French label [37]. There exist several methods for calculating TVOCs [38]; however, in 237 this study the ER of TVOCs was determined based on the method provided by the European 238 Standard ISO 16000-6, where they are defined as the sum of VOCs ranging between C<sub>6</sub> and 239 C<sub>16</sub> quantified by TD-GC-FID as toluene equivalent [39]. The limit of detection (LOD) of the 240 different VOCs used for calibration in GC and HPLC methods are shown in tables S2 and S4, 241 respectively.

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#### 243 2.4. Characterizing Micro-Organism Levels in Indoor Air

The microbial concentrations in indoor air were measured during winter and summer using a Microbial Air Monitoring System (MAS-100). Seven agar culture media were chosen to represent the fungal and bacterial populations. Measurements were taken from spots next to the walls with bio-based insulation: next to the NW wall in summer and from the same spot and another one next to the SW wall in winter. Moreover, sampling occurred in the morning and the afternoon during both seasons. The sampling flow rate of the MAS-100 was set at 100 mL min<sup>-1</sup> and the sampling time at 5 min. Then, the petri dishes containing the agar media were incubated for 4 to 5 days at 25 °C after sampling and before counting the colony-forming units (CFU).

253

#### 254 **3. Results and Discussion**

#### 255 3.1. VOC Surface Emissions

256 VOC surface emission will be presented in terms of emission rate (ER,  $\mu$ g m<sup>-2</sup> h<sup>-1</sup>), calculated 257 based on the following equation:

$$ER = \frac{C \times Q}{S}$$
(1)

where C is the determined VOC concentration measured in the sampling flow collected from the FLEC placed on each surface ( $\mu g m^{-3}$ ), Q is the air flow rate into the FLEC and is equal to 0.03 m<sup>3</sup> h<sup>-1</sup>, and S is the tested surface area of the building structure, which is equal to the surface area of the FLEC (0.0177 m<sup>2</sup>).

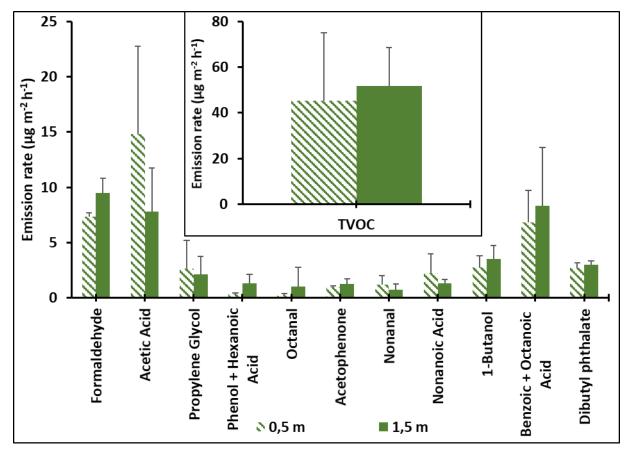
17 VOCs in addition to TVOCs were chosen to be presented in this study based on the importance of their surface emission rate ( $ER \ge 1 \ \mu g \ m^{-2} \ h^{-1}$  from at least one characterized surface during summer or winter). However, since VOC emissions were weak in winter compared to summer (discussed in parts 3.1.3 and 3.1.4), only summer emissions will be discussed in the following two parts (3.1.1 and 3.1.2) due to the increased uncertainty in quantifying compounds at low emission rates (close to the limit of quantification, LOQ).

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#### *3.1.1. Spatial Comparison of VOC Surface Emissions in Summer*

270 Figure 3 shows the summer surface emissions of the major VOCs from the SE wall at two different heights (0.5 and 1.5 m) measured on two consecutive days (13 and 14/07/2019). 271 272 The ratio (R) of individual VOCs emission rates from the same building structure at two 273 different spots can be variable, ranging from 1 to 5. For most of the compounds, in addition to TVOCs, emissions at 0.5 and 1.5 m were approximately equivalent (R  $\approx$  1). However, this 274 difference is higher (R up to 5 times) for the weakly emitted VOCs (ER < 1  $\mu$ g m<sup>-2</sup> h<sup>-1</sup>), such as 275 octanal and the coelution of phenol and hexanoic acid, since the uncertainty in quantifying 276 277 the detected compounds increases as we are close to LOQ, as previously mentioned. This 278 slight variation can be explained by the non-uniform variation of temperature and humidity 279 within the different layers of the wall structure or the inhomogeneity of the surface leading 280 to heterogeneity in VOC surface adsorption and desorption.

To eliminate the effect of the hygrothermal variation within the wall on VOC emissions at different heights, the variations in T and RH were monitored within the SE wall (sensors 1 to 6 in figure 2) and in the room (sensor 7 in figure 2) during the sampling days at 0.5 and 1.5 284 m. As shown in figure 4, the T and RH were very similar between the two heights at each 285 measurement point in the wall, and almost stable between the two sampling days, indicating 286 that the variability in T and RH cannot explain the observed variation of VOC emissions from 287 the same surface at two different heights. Another probable hypothesis is the 288 inhomogeneity of the surface which might have an effect on the diffusivity of VOCs from the 289 bulk of the material to the surface [40], and can therefore result in heterogenous VOC 290 emissions from different parts of the same surface. Thus, the variation in VOC emissions 291 should be considered when interpreting the obtained results.



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Figure 3. VOC surface emissions from the southeast wall at two different heights: 0.5 and
 1.5 m; the error bars represent one standard deviation of 3 and 5 measurements at 0.5 m
 and 1.5 m, respectively

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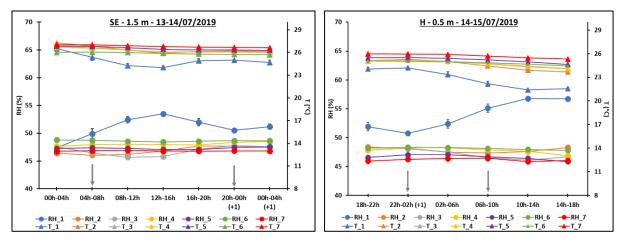


Figure 4. The change in temperature (▲, right axis) and relative humidity (●, left axis) within
 the wall-oriented Southeast (SE) during the sampling days: 13-14/07/2019 at 1.5 m and 14 15/07/2019 at 0.5 m. The sensors are placed within the wall in order from 1 (exterior) to 7
 (inside the wedding hall). Each presented value is the average of 24 measurements. The
 sampling period during these days is indicated by arrows on the graphs

302

#### 303 3.1.2. VOC Surface Emissions from the three Bio-based Walls in Summer

304 Surface emissions from the three bio-based walls in the wedding hall were compared to 305 evaluate the effect of wall orientation, and thus different hygrothermal conditions, on VOC 306 emissions. Table 2 shows the major VOC emissions from the three walls that contain wood 307 wool as insulation material during summer.

308 In general, although emission rates vary largely during summer, the same emission profiles 309 can be seen in the three walls, with a dominance of oxygenated VOCs. Overall, for the three 310 walls, the largest emissions are measured for formaldehyde, acetic acid, the co-eluted 311 benzoic and octanoic acids, and nonanoic acid. These compounds can be emitted by the wood wool insulation present in the walls [41]. In the study of Maskell et al., wood fiber 312 313 emitted high concentrations of acetic acid [18]. This VOC may come from the cleavage of 314 acetyl groups of the hemicellulose, while aldehydes mainly originate from the oxidative 315 decay of cellulose and lignin present in the wood [42].

316 The difference in the emission of the most emitted VOCs and TVOCs varies from 1 to 5 times 317 between the walls; however, this difference can increase up to 30 times for the minor VOCs  $(ER < 1 \mu g m^{-2} h^{-1})$  due to the increased uncertainty associated to quantifying VOCs near 318 LOQ. Moreover, the highest emissions for most of the quantified VOCs and TVOCs were 319 320 observed for the SW wall, followed by the SE wall, and the NW wall. In the morning, the SE 321 wall is exposed to the sun, but at the beginning of the afternoon and until the end of the 322 day, the wall-oriented SW becomes fully exposed to the sun. The NW wall is located in the 323 shade, with almost no sun exposure throughout the day. This difference in exposure may 324 explain the higher emissions from the walls oriented SW and SE compared to the NW wall, 325 as higher temperatures can induce higher emissions of VOCs [32]–[34]. The relatively high 326 TVOC emission rate from the SW wall was driven by, in addition to the major emitted VOCs, species with ER  $\leq 1 \mu \text{g m}^{-2} \text{h}^{-1}$  which are thus not shown in this list. This wall also shows high 327

328 formaldehyde emissions, which might point towards a local difference in this wall or 329 anthropogenic contamination.

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# Table 2. VOC surface emission rates (μg m<sup>-2</sup> h<sup>-1</sup>) of the 8 surfaces at the wedding hall during summer

VOC	SE	SW	NW	Wall 1	Wall 2	Floor	Beam	Ceiling
Formaldehyde	9.5	30.8	5.8	9.4	26.7	17.0	9.6	29.7
Propanal	<lod< td=""><td>1.7</td><td><lod< td=""><td><lod< td=""><td>1.6</td><td><lod< td=""><td>0.5</td><td>1.8</td></lod<></td></lod<></td></lod<></td></lod<>	1.7	<lod< td=""><td><lod< td=""><td>1.6</td><td><lod< td=""><td>0.5</td><td>1.8</td></lod<></td></lod<></td></lod<>	<lod< td=""><td>1.6</td><td><lod< td=""><td>0.5</td><td>1.8</td></lod<></td></lod<>	1.6	<lod< td=""><td>0.5</td><td>1.8</td></lod<>	0.5	1.8
Pentanal	<lod< td=""><td>1.2</td><td><lod< td=""><td><lod< td=""><td>1.3</td><td>0.6</td><td><lod< td=""><td>1.7</td></lod<></td></lod<></td></lod<></td></lod<>	1.2	<lod< td=""><td><lod< td=""><td>1.3</td><td>0.6</td><td><lod< td=""><td>1.7</td></lod<></td></lod<></td></lod<>	<lod< td=""><td>1.3</td><td>0.6</td><td><lod< td=""><td>1.7</td></lod<></td></lod<>	1.3	0.6	<lod< td=""><td>1.7</td></lod<>	1.7
Tolualdehyde	1.3	2.1	0.8	1.2	2.2	1.0	2.5	1.8
Furfural	0.2	1.3	0.2	0.1	0.6	0.4	<lod< td=""><td>1.4</td></lod<>	1.4
Octanal	1.0	1.7	0.1	0.1	0.4	0.7	0.4	2.4
Nonanal	0.8	2.8	0.6	0.6	0.8	2.4	0.3	3.4
Acetic Acid	7.8	3.7	7.7	1.8	2.6	29.0	0.3	0.9
Phenol + Hexanoic Acid	1.3	1.6	0.4	0.2	1.3	1.6	2.6	3.3
Benzoic + Octanoic Acids	10.4	5.9	5.0	14.5	1.3	8.5	0.3	15.6
Nonanoic Acid	1.3	4.0	1.2	2.4	1.5	1.1	4.1	14.1
1-Butanol	3.5	0.6	2.3	1.2	0.5	1.3	0.1	0.6
2-Ethyl-1-hexanol	0.7	2.9	0.4	0.7	0.9	1.0	0.3	9.7
Propylene Glycol	2.1	1.6	1.6	<lod< td=""><td>1.5</td><td>1.0</td><td>0.2</td><td>1.3</td></lod<>	1.5	1.0	0.2	1.3
Acetophenone	1.3	0.5	0.6	1.3	0.8	2.1	0.1	0.6
Dimethylglutarate	0.4	2.2	0.3	0.4	1.3	0.6	8.7	6.2
Dibutyl Phthalate	3.0	0.1	2.2	4.3	0.4	4.2	7.7	1.8
ТVОС	51.7	73.1	37.2	61.4	25.2	101.0	41.3	130.8

340 <LOD: Analytical limit of detection (for carbonyls by HPLC-UV and for all the other VOCs by TD-GC-FID), TVOC:

341 The sum of all VOCs quantified by GC-FID between C6 and C16 as equivalent toluene

#### 343 3.1.3. VOC Emissions from the different Surfaces

Tables 2 and 3 sum up the VOC seasonal emissions from the 8 characterized surfaces in the wedding hall during summer and winter, respectively. Even if concentration in this part and the previous part (3.1.2.) are on the summer VOC emissions, it is worth mentioning that winter emissions were more homogeneous among the three walls compared to summer emissions. This can be related to the higher temperatures during summer, which may lead to

<sup>342</sup> 

higher temperature variability within the different parts of the wall, and thus can increasethe inhomogeneity in VOC emissions.

351 For each of the VOCs, the highest emission rate from any of the characterized surfaces is 352 indicated in bold in both tables (tables 2 and 3). The highest emission rates of most of the 353 individual VOCs (10 out of 17), as well as for TVOCs, were from the ceiling during both 354 seasons. Wall 2 showed the lowest TVOC emissions in winter and summer (8.1 and 25.2 µg  $m^{-2} h^{-1}$ , respectively). This might be explained here again by the exposure to the sun, where 355 356 the ceiling is the most exposed, and likely the surface undergoing the largest variations of 357 temperature during the day, while wall 2 is not directly exposed to the sun, as it is fully interior to the building. The strongest emitted VOCs from all surfaces cumulated are 358 formaldehyde and acetic acid, in winter (23 and 17.9  $\mu$ g m<sup>-2</sup> h<sup>-1</sup>, resp.) as well as in summer 359 (138.5 and 53.8  $\mu$ g m<sup>-2</sup> h<sup>-1</sup>, resp.). The co-eluted benzoic and octanoic acids as well as the co-360 361 elution of phenol and hexanoic acid also show high emission rates, but this needs to be 362 relativized as these represent the sum of two individual emission rates.

363 It is interesting to compare the VOC emissions from the three bio-based structures (SE, SW, 364 and NW walls) to emissions from the other structures (wall 1, wall 2, floor, beam, and 365 ceiling). When comparing the VOC emissions from the most emissive surface (ceiling) to 366 those from the bio-based walls in summer, we observe that emissions are generally lower, as 367 illustrated by the lower TVOC emission rates, despite the relatively high TVOC values for the 368 SW wall, as discussed in the previous section. Hence, for individual VOCs, emission rates are 369 comparable or lower for most compounds. However, emissions are higher from the bio-370 based walls for some VOCs, notably propylene glycol and 1-butanol. These two compounds 371 are however also emitted by the other surfaces, and the difference in emission rate is relatively small (max. 2.2  $\mu$ g m<sup>-2</sup> h<sup>-1</sup>). It is hard to attribute specific VOCs, except acetic acid, 372 as emission fingerprints from wood wool insulation material during only 24-hour surface 373 374 measurement. This is due to multiple factors including the thickness of the building 375 structure, and thus the diffusion time needed by the VOC to arrive at the surface, and the 376 probable existing contamination (infiltration of outdoor air or anthropogenic activities), that 377 can result in the adsorption of VOCs on these surfaces and thus their detection upon 378 measurement.

379 VOC emissions from the different surfaces remain weak, even in summer, when compared 380 to the EU-LCI (European Lowest Concentration of Interest) guide on the emission of VOCs 381 from building materials [43]. For example, the highest emission of formaldehyde from the 382 SW wall (30.8  $\mu$ g m<sup>-2</sup> h<sup>-1</sup> equivalent to 17.9  $\mu$ g m<sup>-3</sup>) is less than the recommended LCI value 383 (100  $\mu$ g m<sup>-3</sup>). Similarly, the emission rate of acetic acid from the floor (29.0  $\mu$ g m<sup>-2</sup> h<sup>-1</sup> 384 equivalent to 16.9  $\mu$ g m<sup>-3</sup>) is far below the guide value (1200  $\mu$ g m<sup>-3</sup>).

Therefore, we can conclude that the bio-based walls do not emit specific VOCs at high rates, and that even if emissions from the floor and ceiling are higher than emissions from the other surfaces, no alerting VOC surface emissions were observed in the selected building.

388

390 Significant seasonal difference was observed for almost all individual VOC and TVOC 391 emissions. In general, winter VOC emissions were relatively weak compared to summer 392 emissions. A ratio (R) of the VOC summer emission rate to winter emission rate is calculated 393 in table 3. Most emission rates from all surfaces were higher in summer than in winter. Only 394 three emission rates show a ratio < 1, and thus a decrease during summer. For each of them, 395 very low emission rates (close to LOQ) in summer lead to relatively large differences with 396 winter measurements; therefore, these ratios should thus be interpreted with care. For the 397 other VOCs, the ratio between summer and winter ranges from 1 and up to 87 times 398 between summer and winter. In this study, the placement of the FLEC at the same evaluated 399 position of each surface during both seasons remains approximate, and thus low variations, 400 ranging from 1 to 5 times between winter and summer, can be attributed to spatial 401 variations, as previously seen in part 3.1.1. On the other hand, the higher emission rates 402 observed in summer for the other VOCs (such as formaldehyde and nonanoic acid emitted 403 from the ceiling) can be explained by the change in meteorological conditions between 404 summer and winter. During the summer campaign, the average measured indoor 405 temperature and relative humidity in the wedding hall were 26 °C and 46%, respectively, 406 while they were equal to 22 °C and 45%, respectively, in winter. The relative humidity inside 407 the hall seems to be stable during winter and summer; however, this is not the case within 408 the different parts of the wall, where a higher variation in RH was observed during the 409 winter campaign compared to summer (Figures S1 and S2). Therefore, the change in the 410 temperature and relative humidity, especially higher temperatures, leads to higher VOC 411 emissions [32]–[34].

			Wi	nter Surf	ace Emis	sions			Ratio (R)							
VOC	SE	SW	NW	Wall 1	Wall 2	Floor	Beam	Ceiling	SE	SW	NW	Wall 1	Wall 2	Floor	Beam	Ceiling
Formaldehyde	1.9	2.2	2.9	1.1	3.4	6.2	3.7	1.6	5	14	2	9	8	3	3	19
Propanal	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>0.9</td><td><lod< td=""><td>0.5</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>3</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>0.9</td><td><lod< td=""><td>0.5</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>3</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td>0.9</td><td><lod< td=""><td>0.5</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>3</td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td>0.9</td><td><lod< td=""><td>0.5</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>3</td></lod<></td></lod<></td></lod<>	<lod< td=""><td>0.9</td><td><lod< td=""><td>0.5</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>3</td></lod<></td></lod<>	0.9	<lod< td=""><td>0.5</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>3</td></lod<>	0.5	-	-	-	-	-	-	-	3
Pentanal	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>0.7</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>2</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>0.7</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>2</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>0.7</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>2</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>0.7</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>2</td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td>0.7</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>2</td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td>0.7</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>2</td></lod<></td></lod<>	<lod< td=""><td>0.7</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>2</td></lod<>	0.7	-	-	-	-	-	-	-	2
Tolualdehyde	0.5	<lod< td=""><td><lod< td=""><td>0.5</td><td>1.2</td><td>1.3</td><td><lod< td=""><td><lod< td=""><td>3</td><td>-</td><td>-</td><td>2</td><td>2</td><td>1</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td>0.5</td><td>1.2</td><td>1.3</td><td><lod< td=""><td><lod< td=""><td>3</td><td>-</td><td>-</td><td>2</td><td>2</td><td>1</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<>	0.5	1.2	1.3	<lod< td=""><td><lod< td=""><td>3</td><td>-</td><td>-</td><td>2</td><td>2</td><td>1</td><td>-</td><td>-</td></lod<></td></lod<>	<lod< td=""><td>3</td><td>-</td><td>-</td><td>2</td><td>2</td><td>1</td><td>-</td><td>-</td></lod<>	3	-	-	2	2	1	-	-
Furfural	<lod< td=""><td><lod< td=""><td><lod< td=""><td>0.1</td><td>0.1</td><td>0.1</td><td><lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>1</td><td>6</td><td>4</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td>0.1</td><td>0.1</td><td>0.1</td><td><lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>1</td><td>6</td><td>4</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td>0.1</td><td>0.1</td><td>0.1</td><td><lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>1</td><td>6</td><td>4</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<>	0.1	0.1	0.1	<lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>1</td><td>6</td><td>4</td><td>-</td><td>-</td></lod<></td></lod<>	<lod< td=""><td>-</td><td>-</td><td>-</td><td>1</td><td>6</td><td>4</td><td>-</td><td>-</td></lod<>	-	-	-	1	6	4	-	-
Octanal	<lod< td=""><td><lod< td=""><td>0.1</td><td>0.2</td><td>0.1</td><td><lod< td=""><td>0.2</td><td>0.8</td><td>-</td><td>-</td><td>1</td><td>1</td><td>4</td><td>-</td><td>2</td><td>3</td></lod<></td></lod<></td></lod<>	<lod< td=""><td>0.1</td><td>0.2</td><td>0.1</td><td><lod< td=""><td>0.2</td><td>0.8</td><td>-</td><td>-</td><td>1</td><td>1</td><td>4</td><td>-</td><td>2</td><td>3</td></lod<></td></lod<>	0.1	0.2	0.1	<lod< td=""><td>0.2</td><td>0.8</td><td>-</td><td>-</td><td>1</td><td>1</td><td>4</td><td>-</td><td>2</td><td>3</td></lod<>	0.2	0.8	-	-	1	1	4	-	2	3
Nonanal	<lod< td=""><td>0.1</td><td>0.2</td><td>0.5</td><td>0.3</td><td><lod< td=""><td>0.4</td><td>1.4</td><td>-</td><td>28</td><td>3</td><td>1</td><td>3</td><td>-</td><td>1</td><td>2</td></lod<></td></lod<>	0.1	0.2	0.5	0.3	<lod< td=""><td>0.4</td><td>1.4</td><td>-</td><td>28</td><td>3</td><td>1</td><td>3</td><td>-</td><td>1</td><td>2</td></lod<>	0.4	1.4	-	28	3	1	3	-	1	2
Acetic Acid	0.6	0.3	1.0	0.6	0.4	0.5	0.4	0.4	13	12	8	3	7	58	1	2
Phenol + Hexanoic Acid	1.0	0.5	1.1	0.9	0.5	1.7	1.8	4.6	1	3	0.4	0.2	3	1	1	1
Benzoic + Octanoic Acids	2.4	0.9	2.3	1.2	0.9	2.5	2.9	4.8	4	7	2	12	1	3	0.1	3
Nonanoic Acid	0.8	0.5	0.7	0.5	0.4	0.3	1.3	2.0	2	8	2	5	4	4	3	7
1-Butanol	0.1	0.1	0.3	0.3	0.2	0.1	0.1	0.1	35	6	8	4	3	13	1	6
2-Ethyl-1-hexanol	0.2	0.1	0.2	0.1	0.1	0.2	0.2	4.9	4	29	2	7	9	5	2	2
Propylene Glycol	0.7	<lod< td=""><td>0.3</td><td>0.3</td><td>0.3</td><td>0.3</td><td>0.1</td><td>1.1</td><td>3</td><td>-</td><td>5</td><td>-</td><td>5</td><td>3</td><td>2</td><td>1</td></lod<>	0.3	0.3	0.3	0.3	0.1	1.1	3	-	5	-	5	3	2	1
Acetophenone	0.1	0.2	0.2	0.3	0.3	0.1	0.1	0.3	13	3	3	4	3	21	1	2
Dimethylglutarate	0.1	<lod< td=""><td>0.1</td><td><lod< td=""><td><lod< td=""><td>0.1</td><td>0.1</td><td>1.3</td><td>4</td><td>-</td><td>3</td><td>-</td><td>-</td><td>6</td><td>87</td><td>5</td></lod<></td></lod<></td></lod<>	0.1	<lod< td=""><td><lod< td=""><td>0.1</td><td>0.1</td><td>1.3</td><td>4</td><td>-</td><td>3</td><td>-</td><td>-</td><td>6</td><td>87</td><td>5</td></lod<></td></lod<>	<lod< td=""><td>0.1</td><td>0.1</td><td>1.3</td><td>4</td><td>-</td><td>3</td><td>-</td><td>-</td><td>6</td><td>87</td><td>5</td></lod<>	0.1	0.1	1.3	4	-	3	-	-	6	87	5
Dibutyl Phthalate	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td></lod<></td></lod<>	<lod< td=""><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td><td>-</td></lod<>	-	-	-	-	-	-	-	-
TVOC	16.8	12.1	14.5	12.2	8.1	14.2	20.9	51.7	3	6	3	5	3	7	2	3

#### Table 3. VOC surface emission rates ( $\mu g m^{-2} h^{-1}$ ) of the 8 surfaces at the wedding hall during winter in addition to the ratio (R) of the summer 412 413 to the winter emission rate

414 < LOD: Analytical limit of detection (for carbonyls by HPLC-UV and for all the other VOCs by TD-GC-FID), TVOC: The sum of all VOCs quantified by GC-FID between C6 and C16

415 as equivalent toluene

#### 416 *3.2. Indoor Air Concentrations*

#### 417 3.2.1. Seasonal Variation

27 VOCs, in addition to TVOCs, were found to have a concentration higher than 1  $\mu$ g m<sup>-3</sup> in 418 the indoor air of the wedding hall during summer and winter. The detected compounds 419 belonged to different chemical families including aromatic hydrocarbons, alcohols, terpenes, 420 421 and carboxylic acids, and a majority of ketones and aldehydes (54%). Table 4 shows the 422 average concentrations of the 8 measurements taken over 24h for both seasons. The 423 concentrations of TVOCs and almost all individual VOCs, except propanone, were two to six 424 times higher in summer than in winter. These findings are coherent with the study of 425 Mandin et al. [44], where they observed significantly higher concentrations of almost all 426 detected VOCs in indoor air in summer compared to winter.

The measured ventilation rate in the wedding hall during the summer and winter campaigns 427 was 217 and 173 m<sup>3</sup> h<sup>-1</sup>, respectively. Therefore, for equal source strength, lower 428 concentrations can be expected during the summer, due to the dilution from this increased 429 430 ventilation rate [45]. Propanone is the only compound showing an increase in winter 431 concentrations. This is in coherence with the study of Missia et al. [25], which showed that 432 the indoor air concentration of propanone is three times higher in winter than in summer. 433 This compound is usually widely emitted by household products [46]. Toluene and xylene 434 show insignificant seasonal variation compared to other VOCs. Even if in this study we did 435 not measure VOC concentrations in outdoor air, according to literature, these two 436 compounds mainly come from the infiltration of outdoor air [47]. On the other hand, for 437 aldehydes, the strongest increases in concentrations over the summer are seen for the 438 lighter aldehydes ( $\leq C_6$ ). The observed higher concentrations can come from indoor source 439 emissions [48], which increase with increasing indoor temperatures. Moreover, due to 440 higher outdoor ozone concentrations in summer, the secondary formation of aldehydes 441 upon the reaction of ozone with indoor VOCs can also lead to an increase in the indoor 442 concentration of these compounds, and in particular formaldehyde [39]. Such increase has 443 been observed previously and was explained by the oxidation of terpenes by ozone [44], 444 [48]–[50]. In this case, the observed indoor concentration of terpenes might be expected to 445 be lower in summer than in winter. However, this is not the case here since the 446 concentration of  $\alpha$ -pinene and 3-carene was three times higher in summer than in winter. 447 Thus, the indoor sources of terpenes in summer are stronger than their oxidation reaction 448 with ozone. Moreover, the source of the detected aldehydes indoors is probably dominated 449 by emissions from indoor building materials, especially wood wool, as illustrated previously 450 in this study, showing the domination of surface-emission sources compared to gas-phase 451 sources in indoor air [51]. Acetic acid shows the strongest increase over summer (seven 452 times higher), but overall its concentration remains relatively low.

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#### Table 4. The average concentration ( $\mu g m^{-3}$ ) of VOCs measured in the indoor air of the 457 wedding hall during summer and winter. Standard deviations are calculated from 8 458 459 measurements taken during 24 hours

Family	voc	Summer (µg m⁻³)	Winter (µg m <sup>-3</sup> )	Ratio of summer to winter
Aromatic	Toluene	1.3 ± 0.5	0.7 ± 0.2	2
Hydrocarbons	m+p-Xylene	$1.1 \pm 0.1$	0.6 ± 0.04	2
	Acetic Acid	2.2 ± 1.2	0.3 ± 0.2	7
Organic Acids	Benzoic + Octanoic Acids	10.1 ± 1.1	4.0 ± 0.8	3
Organic Acius	Phenol + Hexanoic Acid	9.8 ± 2.6	3.7 ± 0.8	3
	Nonanoic Acid	$1.1 \pm 0.1$ $0.6 \pm 0.04$ $2.2 \pm 1.2$ $0.3 \pm 0.2$ cids $10.1 \pm 1.1$ $4.0 \pm 0.8$ acid $9.8 \pm 2.6$ $3.7 \pm 0.8$ $5.5 \pm 1.0$ $1.2 \pm 0.2$ $55.4 \pm 10.4$ $15.3 \pm 9.6$ $38.7 \pm 8.5$ $15.4 \pm 18.8$ $3.6 \pm 0.7$ $0.6 \pm 0.4$ $3.3 \pm 0.4$ $0.6 \pm 0.4$ $3.5 \pm 0.6$ $0.8 \pm 0.5$ $5.8 \pm 1.5$ $1.1 \pm 0.7$ $2.8 \pm 1.2$ $1.3 \pm 0.4$ $4.8 \pm 1.5$ $1.0 \pm 0.6$ $16.1 \pm 3.5$ $2.6 \pm 2.0$ $2.7 \pm 0.8$ $1.9 \pm 0.9$ $5.0 \pm 1.0$ $3.0 \pm 1.2$ $2.1 \pm 1.5$ $1.3 \pm 0.6$	1.2 ± 0.2	5
	Formaldehyde	55.4 ± 10.4	15.3 ± 9.6	4
	Acetaldehyde	38.7 ± 8.5	15.4 ± 18.8	3
	Propanal	3.6 ± 0.7	<b>0.6</b> ± 0.6	6
	Butanal	3.3 ± 0.4	0.6 ± 0.4	6
	Benzaldehyde	3.5 ± 0.6	0.8 ± 0.5	4
Aldehydes	Pentanal	5.8 ± 1.5	1.1 ± 0.7	5
	Furfural	2.8 ± 1.2	1.3 ± 0.4	2
	Methylglyoxal	4.8 ± 1.5	1.0 ± 0.6	5
	Hexanal	16.1 ± 3.5	2.6 ± 2.0	6
	Octanal	2.7 ± 0.8	1.9 ± 0.9	1
	Nonanal	5.0 ± 1.0	3.0 ± 1.2	2
Alaabala	1-Butanol	2.1 ± 1.5	1.3 ± 0.6	2
Alcohols	2-Ethyl-1-hexanol	5.1 ± 2.0	2.1 ± 0.6	2
	Propanone	26.7 ± 3.7	45.9 ± 43.2	0.6
Ketones	Methyl Vinyl Ketone	3.1 ± 0.9	0.7 ± 0.4	4
	Methyl Ethyl Ketone	2.8 ± 0.6	2.2 ± 1.1	1
Torponac	3-Carene	3.2 ± 2.0	1.1 ± 0.8	3
Terpenes	α-Pinene	4.7 ± 3.6	1.6 ± 1.1	3
Halogenated VOC	Tetrachloroethylene	13.1 ± 8.2	<lod< td=""><td>-</td></lod<>	-
	TVOC	101.7 ± 20.4	52.2 ± 9.9	2

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#### 461 3.2.2. **Temporal Variation**

462 The variation of VOC concentrations was monitored during 24 hours in both summer and 463 winter. Figure 5 illustrates the 24-hour variation of the indoor air concentrations for TVOC 464 and selected VOCs during summer and winter campaigns. The hours are presented in the 465 figure according to the sampling order during both seasons.

466 The indoor air VOC concentrations were found to be higher during the night time for TVOC and all the presented VOCs, except for the coeluted benzoic and octanoic acids and 467 nonanoic acid. Maximum VOC concentrations were measured between 21h30 - 3h30 in 468

469 summer and 1h30 - 7h30 in winter. During the day, the opening of doors to enter the hall 470 contributes to the increase in the air exchange rate, leading to further dilution of indoor air, 471 and thus a decrease in VOC indoor air concentrations [52]. On the contrary, the air exchange 472 rate inside the wedding hall is decreased during the nocturnal period, leading to higher 473 indoor air concentrations of the VOCs originating from indoor emissions.

On the other hand, the indoor air concentration of nonanoic acid was almost stable during hours in both summer and winter. Similarly, the indoor air concentration of benzoic and octanoic acids was almost stable in summer; however, it was found to decrease during the night hours (19h30-7h30) in winter. As this is a coelution, it is impossible to evaluate the individual contributions and therefore too uncertain to emit a hypothesis on drivers of this changing temporal behavior between seasons.

In general, the indoor air concentrations of VOCs are higher at night compared to daytime;
however, it is still difficult to precisely predict the individual temporal behavior of each
detected VOC due to the multitude of influencing factors, including VOC sources,
hygrothermal variation, the change in ventilation rates, anthropogenic activities, etc.

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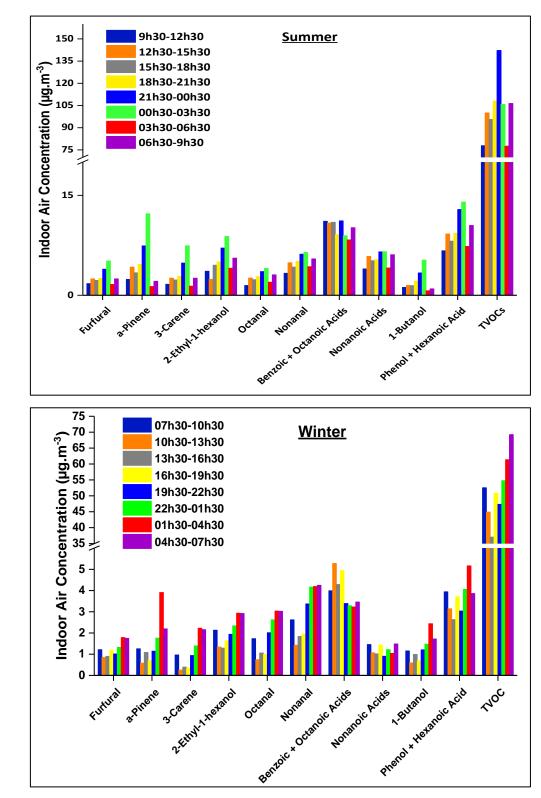
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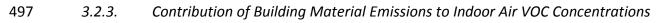


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Figure 5. The temporal variation during 24 hours in the VOC indoor air concentrations of
 the wedding hall of the town hall during the a) summer and b) winter campaigns

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The indoor air concentration of each VOC emitted from the building structures of the wedding hall can be estimated by applying the following equation [25], [53]:

$$C_{e,i} = \frac{1}{Vn} A_i ER_i \quad (2)$$

Where  $C_{e,i}~(\mu g~m^{\text{-}3})$  is the estimated VOC indoor concentration contributed by the  $i^{\text{th}}$ 500 material, V is the volume of the wedding hall which is equal to 441  $m^3$ , n ( $h^{-1}$ ) is the air 501 exchange rate,  $A_i$  (m<sup>2</sup>) is the surface area of the i<sup>th</sup> material (of only building structures 502 evaluated in our study), and ER<sub>i</sub> ( $\mu$ g m<sup>-2</sup> h<sup>-1</sup>) is the VOC emission rate from the i<sup>th</sup> material. 503 The air exchange rate in the wedding hall was measured to be 0.4 and 0.5  $h^{-1}$  in winter and 504 505 summer, respectively. Based on the dimensions of the room, the surface area of each building structure was approximately calculated: 27.4 m<sup>2</sup> for wall 1 and the SW wall, 22.6 m<sup>2</sup> 506 for wall 2 and the NW wall, 45.2 m<sup>2</sup> for the SE wall, 85.7 m<sup>2</sup> for the floor, and 107.1 m<sup>2</sup> for 507 the ceiling. The surface area of the beam was difficult to calculate due to the lack of 508 509 dimensions; therefore, the contribution of this surface to VOC indoor air concentration will 510 not be considered in this part. Then, the relative contribution of each material to the 511 measured indoor air concentrations can be calculated by dividing C<sub>e,i</sub> by the measured 512 indoor concentrations for each VOC (C<sub>in</sub>).

Table 5 sums up the estimated contributions of each surface to the indoor air concentrations for each VOC detected both in ambient air and FLEC measurements (previously shown in tables 2 and 3). In addition, figure S4 of the SI shows the concentrations of the major VOCs quantified in indoor air and the corresponding surface emission rates from the eight characterized surfaces during both seasons.

518 Three trends can be observed when comparing the total estimated VOC indoor air 519 concentration ( $\Sigma C_{e,i}$ ) to the measured indoor air concentration ( $C_{in}$ ): i) approximately equal, ii) higher C<sub>in</sub>, or iii) lower C<sub>in</sub>. For almost equal C<sub>in</sub> and ΣC<sub>e,i</sub>, such as for 2-ethyl-1-hexanol and 520 521 1-butanol in summer, this can be since the emission of these compounds from the 7 building 522 structures is probably the only source of these compounds indoors. Nevertheless, when C<sub>in</sub> is higher than  $\Sigma C_{e,i}$ , sources other than emissions from the seven characterized building 523 structures are expected to contribute to the indoor air concentration of some VOCs (like 524 525 furfural, nonanal, phenol and hexanoic acid, and formaldehyde). Such sources may include 526 emissions from furniture and consumer materials, secondary emissions due to indoor 527 chemical reactions, infiltration of outdoor air, or even limitations related to emission 528 measurements using the FLEC [54].  $\Sigma C_{e,i}$  was higher than  $C_{in}$  mainly for the quantified organic 529 acids (acetic, benzoic coeluted with octanoic, and nonanoic acids). This can also be, as 530 previously mentioned, due to indoor chemical reactions that may reduce these VOC indoor 531 concentrations, in addition to the FLEC measurement limitations which may increase 532 measurement uncertainties. Moreover, benzoic, octanoic, and nonanoic acids may be 533 classified as semi-volatile compounds (SVOCs) since they have a low vapor pressure and high 534 boiling temperature [55], [56]. Therefore, they can be adsorbed after emission on the 535 different existing surfaces in the hall, and thus are less detected in indoor air.

536 When interpreting the estimated individual contribution of each surface to VOC indoor air 537 concentrations, the calculations show that the ceiling, followed by the floor, are the main 538 contributors to the indoor air VOC concentrations during summer and winter for almost all 539 VOCs and TVOCs (marked in bold in tables 2 and 3), with contributions exceeding sometimes 540 100%, as these surfaces have the highest surface area and have previously shown the 541 highest VOC surface emissions (tables 2 and 3). In general, the contributions of the five walls 542 to indoor air concentrations were relatively weak in winter and summer, except for acetic 543 acid. VOC emissions from the SE wall contribute relatively more, which can be explained by 544 its higher surface area. In winter, contributions from these surfaces were less than 20% for 545 TVOCs and all individual VOCs, except for the SE wall that showed a high contribution of 546 acetic acid (59%). Similarly, the summer contributions were also low ( $\leq$  20%), but once again 547 the SE wall showed a high contribution of acetic acid (74%) in addition to acetophenone 548 (34%) and 1-butanol (34%). Care should be taken however to not conclude from this first 549 comparison that bio-based walls contribute more to indoor VOC concentrations than the 550 other surfaces, as the conventional walls present smaller surface areas than the SE wall. 551 When comparing the contributions of building structures with bio-based insulation to 552 conventional structures of the same surface area, i.e. the SW wall to wall 1 and the NW wall 553 to wall 2, no significant difference ( $\leq$  10%) is observed during the two seasons, except for 554 acetic acid and acetophenone. These results support the previously made hypothesis that a 555 part of the acetic acid indoor air concentration comes from the wood wool used for 556 insulation in the three bio-based walls [42].

557 Although several studies have examined the levels of indoor air pollutants or have characterized emission measurements in laboratories, in-situ studies linking the VOC 558 559 concentrations to their indoor sources are still rather limited [25], [30], [31]. This is mainly 560 due to the multiplicity of VOC indoor sources and the consequent time and cost difficulty 561 related to identifying and characterizing emissions from all these sources [54]. Despite all 562 limitations, this approach is interesting since it gives a primary overview of the impact of 563 VOC surface emissions from building structures, especially bio-based structures, on indoor 564 air concentrations. It allowed us to observe that no significant indoor air concentrations are 565 attributed to emissions from bio-based surfaces, except for acetic acid, which is considered a 566 fingerprint VOC for emissions from wood wool.

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# 576 Table 5. The estimated indoor air concentrations (μg m<sup>-3</sup>) and the respective contribution (%) of VOC emissions from the 7 building structures 577 present in the wedding hall to VOC indoor air levels

		Summer									Winter							
voc	_	Contribution (%)							_	$\sum_{n=7}^{n=7}$			Cont	ribut	ion (%	%)		
	C <sub>in</sub> (µg m⁻³)	$\sum_{1}^{L} C_{e,i}$ (µg m <sup>-3</sup> )	Wall 1	Wall 2	SE	sw	NW	Floor	Ceiling	C <sub>in</sub> (µg m⁻³)	$\sum_{1}^{1} C_{e,i}$ (µg m <sup>-3</sup> )	Wall 1	Wall 2	SE	sw	NW	Floor	Ceiling
Acetic Acid	2.2	15.0	10	12	74	21	36	518	20	0.3	1.0	36	20	59	18	49	94	94
Phenol + Hexanoic Acid	9.8	2.9	0	1	3	2	0	6	16	3.7	2.6	4	2	7	2	4	22	76
Benzoic + Octanoic Acids	10.1	16.2	18	1	21	7	5	33	75	4.0	4.3	5	3	15	4	7	30	73
Nonanoic Acid	5.5	8.6	5	3	5	9	2	8	125	1.2	1.4	6	4	17	6	7	12	101
Formaldehyde	55.4	31.3	2	5	4	7	1	12	26	15.3	7.1	1	3	3	2	2	20	6
Propanal	3.6	1.3	0	5	0	6	0	0	25	0.6	0.7	10	4	8	0	6	71	50
Furfural	2.8	1.1	0	2	1	6	1	6	24	1.3	0.1	1	1	0	0	0	4	0
Octanal	2.7	1.9	1	1	7	8	0	10	43	1.9	0.2	2	1	0	0	1	0	26
Nonanal	5.0	3.3	1	2	3	7	1	19	33	3.0	0.4	3	1	0	1	1	0	29
Acetophenone	0.8	1.7	20	10	34	8	8	103	37	0.2	0.3	19	16	10	13	10	20	74
2-Ethyl-1-hexanol	5.1	5.8	2	2	3	7	1	8	91	2.1	0.3	1	1	2	1	1	5	144
1-Butanol	2.1	2.0	7	2	34	4	11	24	14	1.3	0.3	4	2	2	1	3	4	5
TVOC	101.7	136.4	7	3	10	9	4	39	62	52.2	30.5	4	2	8	4	4	13	60

578 C<sub>in</sub> is the measured indoor air concentration by active sampling in the wedding hall.

579  $\sum_{1}^{n} C_{e,i}$  is the sum of the estimated indoor concentrations contributed from surface emissions from the different building structures (C<sub>e;i</sub>)

#### 580 3.3. Micro-organisms and Microbial VOC (mVOC) Concentrations in Indoor Air

581 The results from the microbial analysis obtained during the two measurement campaigns 582 are grouped in table 6. The concentration values show that the concentrations of microbial 583 aerosols in the air of the wedding hall are below the guide values presented by Brauner et al. 584 [57], who stipulates that the concentration of viable molds and bacteria in the indoor air of 585 non-industrial indoor locations in the European Union should be less than 2000 CFU m<sup>-3</sup>. The 586 measured indoor microbial concentrations from the same spot were up to 5 times higher in 587 summer compared to winter which is coherent with other studies reported in literature [58]. 588 Winter microbial aerosol concentrations measured in the morning, next to the NW wall, 589 were approximately equal to concentrations taken at the same time near the SW wall, 590 indicating that the indoor air of the wedding hall is quite homogeneous or mixed in the case 591 of xerophilic fungi (measured with Dichloran-Glycerol (DG18) Agar).

592 Many of the detected VOCs in indoor air (table 4), such as acetic acid, octanal, nonanal, 2-593 ethyl-1-hexanol, and 1-butanol, are classified as mVOCs based on the mVOC database [59]. 594 However, these compounds were also emitted from the building structures (tables 2 and 3). 595 Therefore, it is impossible to attribute these compounds solely to microbial development.

596

Table 6. The concentrations of microbial aerosols (CFU m<sup>-3</sup>) measured during the summer
 and winter campaigns in the indoor air of the wedding hall of the town hall.
 Measurements were done next to the northwest (NW) and southwest (SW) walls

Media	Sun	nmer	Winter							
	NW-Morning	NW-Afternoon	NW-Morning	NW-Afternoon	SW-Morning					
Ordinary Agar	340	200	102-116	42-68	122					
R2A Agar	148	78	90-110	68-98	98					
Chapman Agar	50	44	8-16	8-10	18					
MEA Agar	ND*	ND	70-72	56-106	68					
Czapek Agar	322	184	52-88	32	50					
DRBC Agar	42	28	4-46	12-22	12					
DG18 Agar	366	188	100-106	16-34	52					

\*ND: Non-determined; R2A: Reasoner's 2A; MEA: Melt-Extract Agra; DRBC: Dichloran Rose Bengal Chloramphenicol; DG18: Dichloran-Glycerol

602

### 603 **4. Conclusion**

604 This study characterizes the seasonal in-field VOC emissions from building structures as a 605 whole in public buildings. The measurements undertaken in two different seasons, winter 606 and summer, allowed us to evaluate the changes in VOC emissions from structures 607 containing bio-based insulation materials (wood wool) in contrasted conditions, and to 608 compare VOC emissions from bio-based and conventional building structures. Observed 609 emission rates from the different construction surfaces were then compared with VOC levels 610 measured in indoor air. Potential microbial development on bio-based surfaces was 611 explored.

612 A comparison of VOC emissions between samples taken at different locations of the same 613 surface showed a pronounced heterogeneity with emission rates changing up to a factor of 614 5. Moreover, a difference in VOC emissions ranging between 1 to 30 times was observed 615 among the three bio-based walls with a similar building structure. This difference was 616 explained by the exposure of the walls to different hygrothermal conditions. Part of this 617 variation could be related to spatial heterogeneity. Acetic acid emitted from these bio-based 618 surfaces seems to be the only quantified compound to consider as a fingerprint VOC for 619 emissions from wood wool. In the future, this compound should receive particular attention 620 if wood wool was intensively used.

In addition, VOC emissions from the eight different characterized surfaces (bio-based and conventional) were also compared. Results showed that the two conventional surfaces, ceiling and floor, were the most emissive surfaces. However, in general, emissions from the different building structures, especially the three bio-based walls, remained well below sanitary recommendations. VOC levels varied largely between summer and winter, with summer emissions being 1 to 87 times higher than those in winter due to the difference in hygrothermal conditions between the two seasons.

628 Indoor air concentrations followed a similar seasonal trend, and were 1 to 6 times higher in 629 summer compared to winter, with part of these levels driven by the increase in surface 630 emissions in summer. The calculated contributions of the emissions of the different surfaces 631 to ambient VOC concentrations showed that:

- 632 Conventional surfaces with the highest VOC emissions and surface area (floor and ceiling) showed the highest contribution to indoor VOC concentrations.
- Bio-based surfaces showed a relatively small contribution to indoor air levels (< 20%</li>
  for most VOCs) compared to the floor and ceiling.
- This comparison gives a first estimation of the relative contributions of surface emissions,especially bio-based ones, to indoor air concentrations.
- 638 No VOC emissions could be attributed specifically to microbial development since the 639 detected mVOCs were also emitted by the characterized building structures.

640 In light of the results obtained in this study, comparison of in-field VOC emission rates with 641 emissions rates from laboratory-tested materials is not always straightforward. Our 642 understanding of the underlying mechanisms explaining these differences is fundamental, 643 especially to be integrated in modeling experiments. As an ongoing part of this project, a 644 comparison of the in-field results to laboratory VOC emissions from the individual materials 645 constituting each building structure is planned.

646

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