

1 **Methodology for determining indoor air quality and identifying the origin of odour**  
2 **episodes in indoor environments through dynamic VOC sampling and TD-GC/MS**  
3 **analysis**

4  
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14  
15 **Abstract.** A methodology for identifying volatile organic compounds (VOC) and  
16 determining air quality in indoor air has been developed. The paper standardizes the  
17 procedure of sampling and analysis for low concentration VOC air samples in buildings.  
18 The methodology is based in the activation of pump samplers by the inhabitants of the  
19 dwelling when they perceive odorous and/or discomfort episodes. Glass multi-sorbent  
20 tubes (Carbotrap, Carbopack X and Carboxen-569) are connected to the pump samplers  
21 for the retention of VOC. The analysis is performed by Automatic Thermal Desorption  
22 (ATD) coupled with GC/MS. Hence, an accurate determination of all VOC present in  
23 samples can be done. This methodology can be applied in cases of sick building  
24 syndrome (SBS) evaluation, in which building occupants experience a series of varied  
25 symptoms that appear to be linked to time spent in the building. Chemical pollutants  
26 concentrations (e.g. VOC) are among the factors that have been described to contribute  
27 to SBS. To exemplify the methodology described, a qualitative determination of all  
28 VOC present and an evaluation of their concentrations were done in a dwelling where  
29 the occupants experienced the SBS symptoms. Higher TVOC values were found in  
30 episodes in indoor air ( $1.33 \pm 1.53 \text{ mg m}^{-3}$ ) than in outdoor air ( $0.71 \pm 0.46 \text{ mg m}^{-3}$ ).  
31 The concentrations of some individual VOC (ethanol, acetone, isopropanol, 1-butanol,  
32 acetic acid, acetonitrile and 1-methoxy-2-propanol) were also higher than the expected  
33 for a standard dwelling. Further research in the activities realized in the adjacent  
34 buildings was done to determine the origin of the major VOC. The external source of

35 VOC was found to be a not declared activity of storage and manipulation of solvents  
36 located at the bottom of a contiguous building.

37 **Keywords:** indoor air, thermal desorption, TVOC, VOC, sick building syndrome

38

### 39 **Introduction**

40 Most indoor air pollution comes from sources inside the building, such as  
41 adhesives, carpeting, wood products or cleaning products, which may emit VOC  
42 (Knöppel *et al.*, 1989; Salthammer, 1997; Hodgson *et al.*, 2000; Jensen *et al.*, 2001;  
43 Zhang and Xu, 2002). However, the outdoor air that enters the building through poorly  
44 located air intake vents, windows and other openings can also be the main source of  
45 indoor air VOC pollution for some contaminants, for example, pollutants from motor  
46 vehicle exhausts, plumbing vents, building exhausts (kitchen and bathroom exhausts)  
47 and combustion products from a nearby garage (EC, 1989; Møhlhave, 1991, EC, 1997;  
48 Møhlhave, 1999; Weschler and Shields, 2000; Zuraimi *et al.* 2003; Weisel *et al.*, 2005).  
49 Therefore, if an appropriate ventilation rate with good quality outdoor air is applied to  
50 the dwelling, VOC concentrations could be maintained in an acceptable level (Holcomb  
51 and Seabrook, 1995; Weschler and Shields, 2000; Zuraimi *et al.*, 2003). If adequate  
52 ventilation is applied and VOC values continue high, probably strong local sources of  
53 these compounds are located inside or near the building (Weschler *et al.*, 1990).

54 The knowledge of the adverse effects on health associated to indoor air VOC is  
55 nowadays very limited (Johansson, 1999; Edwards *et al.*, 2001; Venn *et al.*, 2003),  
56 knowing much less about the health risks from polluted indoor air than from outdoor  
57 contamination (Jones, 1999). Several human exposure studies have been carried out  
58 (Otto *et al.*, 1990; Møhlhave, 1991, Wolkoff, 1995; Andersson *et al.*, 1997; Pappas *et al.*,  
59 2000; Møhlhave, 2001); however, the VOC concentrations experimented were relatively  
60 high and far from the usual dwelling concentrations, making difficult the extrapolation  
61 between the results obtained and the real effects of indoor air VOC concentrations  
62 (Holcomb and Seabrook, 1995; Wolkoff *et al.*, 1997). The evaluation of health issues  
63 caused by complex VOC mixtures is also difficult because their effects may be additive,  
64 synergistic, antagonistic or even independent for each other, making hardly predictable the  
65 toxicological consequences of VOC exposition to human wellness (Møhlhave *et al.*,  
66 1997; Johansson, 1999). On the other hand, the wide range of sampling strategies and  
67 Total VOC (TVOC) concentration calculation methods used in the literature often make  
68 TVOC published data not comparable (Møhlhave, 1992; EC, 1997; Møhlhave *et al.*, 1997;

69 Wolkoff *et al.*, 1997; Johansson, 1999). The European Commission has recommended a  
70 definition of TVOC and a procedure for its sampling and analysis (EC, 1997), focusing  
71 in the main indoor VOC families, such as aliphatic hydrocarbons, aromatic  
72 hydrocarbons, terpenes, alcohols, aldehydes, ketones, halocarbons and esters. In the  
73 same way, the international standard ISO 16000-6 describes a way of measuring TVOC.  
74 TVOC has been recommended to be used as a screening tool, due to it does not have  
75 biological relevance, and is not recommended for making definitive conclusions about  
76 indoor air quality (Andersson *et al.*, 1997). However, it has some useful applications as  
77 an indicator of the presence of VOC indoors (Mølhave, 1992; Mølhave, 2000; Wolkoff  
78 and Nielsen, 2001; Mølhave, 2003).

79         Although some epidemiological studies have been inconsistent or their results  
80 have not been confirmed, that does not mean that VOC are not a possible source of  
81 discomfort complaints for people exposed to them, increasing their possibility of  
82 experiencing negative health effects in the future. People repetitively exposed to long-  
83 term relatively high levels of VOC, specially the subgroup of population more sensitive  
84 to VOC concentrations found indoors, can modulate their physiological response to a  
85 given VOC (Hummel *et al.*, 1999; Wolkoff and Nielsen, 2001), and achieve multiple  
86 chemical sensitivity (MCS) (Levy, 1997; Shorter, 1997). For this reason, legislated  
87 guideline recommendations should be established for VOC concentrations in indoor air.  
88 In the interim period, VOC indoor concentrations should be maintained as low as  
89 reasonably achievable (ALARA), to diminish the organic pollution load in indoor  
90 environments (EC, 1997; Mølhave, 2003).

91         VOC indoor concentrations have been related to the sick building syndrome  
92 (SBS). The cause of SBS is probably multifactorial and it is not usually accompanied by  
93 any organic lesion or physical sign, and generally is diagnosed by exclusion (EC, 1989).  
94 The symptomatology of SBS is varied and complex, but nasal, ocular, oropharyngeal,  
95 cutaneous and general (tiredness, headache, etc.) manifestations are the five symptoms  
96 most common. The causes of SBS remain unknown but some factors have been  
97 described to contribute to SBS, such as outdoor and indoor chemical contaminants (e.g.  
98 VOC), biological contaminants and inadequate ventilation (WHO, 1982; WHO, 1986,  
99 EC, 1989). VOC, defined as having a boiling point that ranges between 50°C and 260°C  
100 (WHO, 1989), are a highly diverse class of chemical contaminants, and between 50 and  
101 300 compounds may be found in non-industrial indoor air environments (Mølhave,  
102 1992; Johansson, 1999).

103           The aim of this study was to establish a methodology to evaluate external VOC  
104 sources that contribute to indoor environments pollution. To exemplify the  
105 methodology, a characterisation of the indoor air quality of a dwelling with a history of  
106 occupant complaints, determining exhaustively the VOC present in the dwelling and  
107 evaluating the concentrations of the most important VOC, was done. The dwelling  
108 occupants experienced symptoms that resembled the SBS symptoms, such as headache,  
109 eyes, tongue and throat irritation, anxiety, irritability, tiredness, fatigue and poor  
110 concentration. The symptoms appeared variably over time, generally related to percept  
111 odour and nuisance. The symptoms decreased or disappeared when the inhabitants were  
112 away from the building. In addition to these symptoms, a mild neutropenia was  
113 observed in one of the inhabitants of the dwelling in the period 2005-2006, with values  
114 of absolute neutrophil counts (ANC) ranging from 1,306-1,319 cells per mm<sup>3</sup>. Before  
115 starting the complaints, in year 1998, this inhabitant presented values of ANC of 2,540  
116 cells per mm<sup>3</sup>. Neutropenia is a decrease in circulating neutrophils in the peripheral  
117 blood, and abnormal ANC values are below 1,500 cells per mm<sup>3</sup>. The potential causes  
118 of neutropenia are varied and complex, but acquired neutropenia can be caused by  
119 exposition to drugs and chemicals, such as industrial solvents (Williams *et al.*, 1977).

120           Twenty-two samples were taken during a 3-month period to evaluate the VOC  
121 concentrations found in the dwelling. The study was initially designed to determine if  
122 VOC concentrations were higher than the expected for a standard dwelling, if so, to  
123 identify the possible sources and other factors that might be responsible for the kind of  
124 VOC found and that could contribute to the elevated concentrations determined.

125           This study is, in our knowledge, the only exhaustive evaluation done in a  
126 Spanish dwelling with sick building syndrome complaints by the occupants. Generally,  
127 indoor air studies have been carried out in Northern European countries, where the  
128 temperatures are lower and problems related to indoors are affected by poor ventilation.  
129 In our work we compare the results obtained with other European studies and apply  
130 evaluation criterions to these results, principally criterions established by the  
131 Environment Institute-European Commission. These comparisons permitted us to  
132 establish that the source of the chemical contaminants was not located inside the flat  
133 studied and that the high concentrations of VOC came from an external activity.

134

## 135 **1 Materials and methods**

### 136 **1.1 Chemicals and materials**

137 VOC standards are purchased as commercial neat chemicals from Aldrich  
138 (Milwaukee, WI, USA), Merck (Darmstadt, Germany) and Fluka (Buchs,  
139 Switzerland), with purity of not less than 98%. Perkin Elmer glass tubes (Pyrex, 6  
140 mm external diameter, 90 mm long), unsilanised wool and Carbotrap (20/40 mesh),  
141 Carbopack X (40/60 mesh) and Carboxen-569 (20/45 mesh) adsorbents are obtained  
142 from Supelco (Bellefonte, PA, USA).

### 143 **1.2 Sampling strategy**

144 Custom packed glass multi-sorbent cartridge tubes (Carbotrap 20/40, 70 mg;  
145 Carbopack X 40/60, 100 mg and Carboxen-569 20/45, 90 mg) have been optimized for  
146 the retention of VOC in air samples (Ribes *et al.*, 2007). A comparative study was done  
147 between the adsorbent Tenax TA, recommended by the European Commission (EC,  
148 1997) and the international standard ISO 16000-6, and the above-mentioned  
149 multisorbent-tube, containing Carbotrap, Carbopack X and Carboxen-569. The  
150 performance evaluation between the two kinds of sampling adsorbent methods was  
151 done in real samples. Important differences were observed between the sampling  
152 efficiency for the most volatile fraction of VOC, giving better qualitatively and  
153 quantitatively responses the multisorbent-tube in respect to Tenax TA (Fig. 1)  
154 (Berenguer *et al.* 2002).

155 In order to take the samples, the tube cartridges are connected to remote-  
156 controlled specially-designed LCMA-UPC pump samplers ( $90\text{-}120\text{ mL min}^{-1}$ ) equipped  
157 with inert sampling line and high precision total volume measurement. Other  
158 characteristics include 10 calibration flow levels, high flow stability, very low  
159 breakthrough values, inexistent tube contamination during pre-activation processes and  
160 the possibility of remote activation through radiofrequency or mobile phone (Roca,  
161 2006).

162 The strategy for the evaluation of VOC occurrence in a dwelling starts with a  
163 first phase of 24-hours samplings. This first phase has been designed to determine  
164 qualitatively the type of VOC present in indoor air. The second control phase is  
165 based on integrated odour episodes sampling in several points of the dwelling. The  
166 odour episodes are sampled when medium and high odour intensity and nuisance is  
167 percept by the inhabitants of the dwelling, being this part of the study dedicated to  
168 characterise and determine the episodic concentrations of VOC. The remote-  
169 controlled LCMA-UPC pump samplers allow the dwelling occupants to turn on and  
170 turn off the samplers when an odorous and/or discomfort episode occurs. The third

171 and last phase of the sampling design simulates a working timetable, from 8 a.m. to  
172 14 p.m. in the morning (6 hours). Sampling flows applied are of 90 mL min<sup>-1</sup> for  
173 control periods higher than 6 hours and 120 ml min<sup>-1</sup> for episodic controls.

174 A minimisation of sampling costs is achieved with this control program, as  
175 the activation of the sampler by the inhabitants of the building in the very moment  
176 of the occurrence of odorous episodes diminishes the number of samples necessary  
177 to obtain reliable data for the study.

### 178 **1.3 Analytical instrumentation**

179 Analysis of VOC is performed by Automatic Thermal Desorption (ATD)  
180 coupled with capillary Gas Chromatography (GC)/ Mass Spectrometry Detector  
181 (MS), using a Perkin Elmer ATD 400 (Perkin Elmer, Boston, Massachusetts, USA)  
182 and a Thermo Quest Trace 2000 GC (ThermoQuest, San Jose, California, USA)  
183 fitted with a Thermo Quest Trace Finnigan MS. The methodology is described  
184 elsewhere (Ribes *et al.*, 2007). Briefly, thermal desorption of the sampling tubes is  
185 carried out at 300°C with a flow rate of 50 mL min<sup>-1</sup> for 10 min (primary  
186 desorption), during which time the eluted VOC are swept from the tube to a  
187 cryofocusing trap (containing approximately 15 mg of Tenax TA and 15 mg of  
188 Carbotrap) maintained at -30°C, applying a flow split of 4 mL min<sup>-1</sup>. After a  
189 primary desorption, the cold trap is rapidly heated from -30°C to 300°C (secondary  
190 desorption) and then maintained at this temperature for 10 min. During the  
191 secondary desorption, VOC are submitted to a flow split of 7 mL min<sup>-1</sup> and are  
192 injected onto the capillary column (DB-624, 60 m x 0.25 mm x 1.4 µm, provided by  
193 J&W, Folsom, CA, USA) via a transfer line heated at 200°C. The column oven  
194 temperature starts at 40°C for 1 min, increases to 230°C at a rate of 6°C min<sup>-1</sup> and  
195 then is maintained at 230°C for 5 min. Helium (99.999%, Air Liquide) carrier gas  
196 flow in the analytical column is approximately 1 mL min<sup>-1</sup> (1.4 bar).

197 Mass spectral data are acquired over a mass range of 20-300 amu. A 6 min  
198 solvent delay time is applied for standards analysis to avoid saturation of mass  
199 spectrometer detector.

200 Qualitative identification of VOC is based on the match of the retention times  
201 and the ion ratios of the target quantification ions and the qualifier ions (Xcalibur  
202 1.2 validated software package). The majority of the identified VOC are quantified,  
203 establishing as a criterion their abundance in indoor air, toxicity or request in TVOC

204 analysis (WHO, 1989). Quantification of field samples is conducted by the external  
205 standard method with nine point calibration. Standards are prepared in methanol and  
206 injected at 30°C on the multisorbent-tubes under an inert Helium gas flow (100 mL  
207 min<sup>-1</sup>) using a conventional gas chromatograph packed column injector. Tube  
208 loading lasts not less than 5 min. Almost all VOC exhibit linearity ranges (ng) from  
209 3 to 4 orders of magnitude. Ethanol, carbon disulfide, methylethylketone and  
210 benzene exhibit the wider linear dynamic ranges (around 10<sup>6</sup> ng). Most of the VOC  
211 show repeatibilities (% relative standard deviation values) ≤ 25% that accomplish  
212 EPA performance criteria (US EPA, 1999).

213 Extreme precautions have been established for quality assurance, such as  
214 automatic daily tuning performed by the Xcalibur software (version 1.2 Thermo  
215 Quest) at  $m/z = 69, 219$  and  $502$ , and control of air leaks ( $m/z = 4, 18$  and  $28$ ).  
216 Toluene has been chosen as reference standard, the retention of this compound is  
217 established to be between ±0.33 min of the most recent calibration, and toluene area  
218 response changes (at 100 ng in tube) should be within ±40% between sample and  
219 most recent valid calibration. Blank samples are taken during sampling and storage  
220 (Ribes *et al.*, 2007).

#### 221 **1.4 TVOC value calculation**

222 TVOC value is calculated following the European Union recommended  
223 procedure (EC, 1997) and Mølhav *et al.* 1997 recommended method. The main and  
224 more toxic compounds found in our studies and the minimum number of compounds to  
225 be analysed suggested by the methods are quantified individually. The rest of  
226 compounds are quantified by the response factor of toluene. The summation of all that  
227 values gives us the TVOC value.

#### 228 **1.5 Case study**

229 VOC were dynamically sampled during a 3-month period (May to July 2006)  
230 in several locations of a dwelling during 24-hour, 6-hour controls and odour  
231 episodes (Table 1). The odour episodes were sampled when medium and high odour  
232 intensity and nuisance was percept by the occupants. Generally the nuisance was  
233 accompanied by a fresh sensation of dense air and a perfumed or combustion odour.  
234 When the episodes occurred, the occupants started to feel sick, basically due to the  
235 irritation of their mucous membranes and eyes, and headache and difficulty to  
236 concentrate were accentuated. The episodes were quite variable, from 5 minutes to

237 2-3 hours. Usually they were located between 10 a.m. and 14 p.m. in the morning  
 238 and 16 p.m. and 21 p.m. in the evening. The dwelling is located in the first floor of a  
 239 6 floor building in a moderate population density district with medium traffic flow  
 240 in Barcelona city. The flat size is 120 m<sup>2</sup>, having 4 bedrooms, 2 bathrooms, a  
 241 kitchen and a dinning room. Only one bedroom, one bathroom and the dinning room  
 242 have street external windows. Inside exhausts ventilate the kitchen and the other  
 243 bathroom, and 3 bedrooms are ventilated trough an inside patio (Fig. 2). The flat is  
 244 not provided with air conditioning and ventilates trough open doors and windows.  
 245 However, ventilation rates are thought to be adequate and indoor air is thought being  
 246 well mixed. Natural ventilation is expected to have maximum air exchange rates  
 247 when opening the windows and doors while outdoor conditions are favourable, as  
 248 well as will depend on wind speed, the position of open windows and doors relative  
 249 to each other and prevailing winds (Godish, 1997; Godish, 2001). The dwelling has  
 250 not been decorated within the previous 7 years and did not contain newly purchased  
 251 furniture during the air sampling. Two people (non smokers) and a cat live in the  
 252 flat.

253

254 Table 1. Control samples from May to July 2006

Ref.	Sample Type	Date	Sampling Point	Volume (L)
A1	24-hour	23-24/5/06	Point A	125.14
A2	24-hour	24-25/5/06	Point A	133.96
A3	24-hour	25-26/5/06	Point A	130.96
A4	24-hour	26-27/5/06	Point A	119.58
A5	Episodes	28/5/06	Point A	17.10
C1	24-hour	23-24/5/06	Point C	145.71
C2	24-hour	24-25/5/06	Point C	114.55
C3	24-hour	25-26/5/06	Point C	111.84
C4	24-hour	26-27/5/06	Point C	118.98
C5	Episodes	28/5/06	Point C	17.00
BE1	Episodes	9-12/6/06	Points B, E	57.01
D1	Episodes	7-11/6/06	Point D	95.80
D2	Episodes	13-19/6/06	Point D	48.80
BE2	Episodes	13-19/6/06	Points B, E	49.16
BE3	Episodes	20-26/6/06	Points B, E	55.20
D3	Episodes	20-26/6/06	Point D	47.67
D4	6-hours	4/7/06	Point D	36.10
F1	6-hours	4/7/06	Point F	57.85
BE4	6-hours	4/7/06	Point B, E	35.68

255

256 Six sampling points were distributed among the dwelling near the VOC  
 257 potential emission sources (Fig. 2). Point A: inside patio 1 (outdoors), where three  
 258 exhausts (the building water, gas and electricity cupboards ventilation exhausts)



259 coming from downstairs were located at 20 cm from the pavement. Point B: kitchen  
260 (kitchen exhaust). Point C: bathroom (bathroom exhaust). Point D: inside patio 2  
261 (outdoors). Point E: dinning room. Point F: terrace (outdoors). Only air sampled in  
262 Point F came from street outdoors. Air sampled in points A and E came from interior  
263 patios, and although it is considered outdoor air, it is mixed in a less extent than  
264 point F air, probably presenting VOC concentrations less dispersed than in the street.  
265 The different points were distributed trough the flat as slightly different sensations  
266 were experimented by the occupants in each part of the dwelling. Twenty-two  
267 control samples (eight 24-hours samples, eight episode samples and six 6-hours  
268 samples) were taken during the period from the 23<sup>rd</sup> of May to the 4<sup>th</sup> of July of  
269 2006 (Table 1).

270 Volumes sampled ranged between 17 and 145 litres. The indoor air was  
271 supposed to be mixed and outdoor meteorological conditions were similar during the  
272 sampling in the three different phases of the study, being the values of temperature,  
273 humidity and pressure in the range of 21-28°C, 47-65% and 1015-1022 hPa,  
274 respectively. According to that, indoor VOC concentrations should not be influenced by  
275 external factors.

276

## 277 **2 Results and discussion**

### 278 **2.1 Individual compounds**

279 One hundred and thirteen VOC have been identified qualitatively in the samples  
280 analysed. A 40% of these compounds had already been identified in several worldwide  
281 studies as major indoor air VOC (Holcomb and Seabrook, 1995) (Table 2). Alkanes,  
282 aromatic hydrocarbons, esters, alkenes, carboxylic acids and alcohols are the main  
283 compounds found in the majority of samples (Fig. 3). Alkanes and aromatic  
284 hydrocarbons contribute nearly to a half of the compounds determined in the samples.  
285 24-hours, 6-hours and episode samples show similar compound distributions (Fig. 4);  
286 however, concentrations of dichloromethane, toluene, DL-limonene, *m+p*-xylenes and  
287 *NN*-dimethylformamide are higher in 24-hours control samples.

288 Ethanol, acetone, isopropanol, 1-methoxy-2-propanol, 1-butanol, acetic acid,  
289 *p*-dichlorobenzene, toluene, *m+p*-xylenes and acetonitrile are the main compounds  
290 found in all samples. In 24-hours samples, the values range between 30-98, 30-63,  
291 11-46, 28-147 and 62-139  $\mu\text{g Nm}^{-3}$  for ethanol, acetone, isopropanol, toluene and  
292 acetic acid, respectively (Table 3). As it was expected, in episode samples the values

293 are higher, ranging between 49-2,787, 41-142, 19-125, 14-95 and 19-211  $\mu\text{g Nm}^{-3}$   
 294 for ethanol, acetone, acetic acid, 1-butanol and 1-methoxy-2-propanol, respectively  
 295 (Table 3).

296

297 Table 2. Identified VOC in the qualitative analysis of the studied samples and their  
 298 Odour Threshold ( $\text{mg m}^{-3}$ ), TLV/420 ( $\mu\text{g m}^{-3}$ ) and relevant associated R-phrases.

Compound	CAS#	Outdoor		Outdoor air (street)	Odour Threshold <sup>c</sup> ( $\text{mg m}^{-3}$ )	TLV/420 ( $\mu\text{g m}^{-3}$ )	R-phrases
		air (Interior patios)	Indoor air				
$\alpha$ -Pinene <sup>a</sup>	80-56-8	x	x	x	230		
$\beta$ -Pinene <sup>a</sup>	127-91-3	x	x	x	8,900		
1,1,2-Trimethylcyclohexane	7094-26-0		x				
1,1,3-Trimethylcyclohexane	3073-66-3		x				
1,2,4-Trimethylbenzene <sup>a</sup>	95-63-6	x	x	x	140	298	20-36/37/38
1,3,4-Trimethylbenzene	95-63-6	x				298	
1,3,5-Trimethylbenzene <sup>a</sup>	108-67-8	x	x	x	180	298	37
1,3-Pentadiene	504-60-9	x	x	x			
1,8-Cineole	470-82-6	x			7.5		
1-Butanol <sup>a</sup>	71-36-3	x	x	x	0.48	148	22-37/38-41-67
1-Heptene	592-76-7	x	x		37		
1-Hexene	592-41-6	x	x			417	
1-Methoxy-2-propanol	107-98-2	x	x	x	37,000	655	
2,2,4,4,6,8,8-Heptamethylnonane	4390-04-9		x				
2,2,4,6,6-Pentamethylheptane	13475-82-6	x	x				
2,2,5,5-Tetramethylhexane	1071-81-4		x				
2,4-Dimethylheptane	2213-23-2		x				
2,6-Dimethyloctane	2051-30-1	x	x	x			
2-Bromoheptane	1974-04-5	x	x	x			
2-Butanol	78-92-2	x			285,000	733	36/37-67
2-Butoxyethanol	111-76-2	x	x		0.5	555	20/21/22-36/38
2-Ethyl-1-butanol	97-95-0	x	x	x	0.3		
2-Hexene	592-43-8	x	x				
2-Methyl-1-butene	563-46-2	x	x	x			
2-Methyl-1-heptene	15870-10-7	x					
2-Methyl-1-pentene	763-29-1	x	x	x			
2-Methyl-2-butene	513-35-9	x	x	x			
2-Methylhexane <sup>a</sup>	591-76-4	x	x	x			38-65-67
2-Methylpentane	107-83-5	x	x	x		4,267 <sup>b</sup>	
2-Methylpentanol	105-30-6	x	x	x	0.1		
3,5-Dimethyloctane	15869-93-9	x					
3-Ethylhexane	619-99-8	x	x				
3-Methylheptane	589-81-1	x	x	x			
3-Methylhexane	589-34-4	x	x	x			
3-Methylpentane	96-14-0	x	x	x		4,267	
4-Methyl-2-pentanone <sup>a</sup>	108-10-1	x	x	x	140	495	20-36/37-66
4-Methyl-2-pentene	4461-48-7	x					
4-Methyloctane	2216-34-4	x					
Acetic acid	64-19-7	x	x	x	90	60	35
Acetone <sup>a</sup>	67-64-1	x	x	x	1,000	2,876	36-66-67
Acetonitrile	75-05-8	x	x	x	285,000	81	20/21/22-36
Benzaldehyde	100-52-7	x	x	x	10		22
Benzene <sup>a</sup>	71-43-2	x	x	x	1,500	4	45-46-36/38-48/23/24/25-65
Benzoic acid, 2-[(trimethylsilyl)oxy]-,	3789-85-3	x	x				

trimethylsilyl ester								
Benzothiazole	95-16-9	x	x	x				
Butanoic acid	107-92-6	x	x		0.35			34
Butyl acetate <sup>a</sup>	123-86-4	x	x	x	7,700	1,726		66-67
Carbon disulfide <sup>a</sup>	75-15-0	x	x	x	180	7		36/38-48/23-62-63
Chloroform <sup>a</sup>	67-66-3	x	x	x	650,000	117		22-38-40-48/20/22
Cyclohexane <sup>a</sup>	110-82-7	x	x	x	35,600	833		38-65-67
Cyclohexanone	108-94-1	x	x		880	195		68-65-67
Dichloromethane <sup>a</sup>	75-09-2	x	x	x	4,100	421		40-48/20
DL-Limonene <sup>a</sup>	7705-14-8	x	x	x	1,700			38-43
Ethanol	64-17-5	x	x	x	154,000	4,564		
Ethyl acetate <sup>a</sup>	141-78-6	x	x	x	623,000	3,493		36-66-67
Ethylbenzene <sup>a</sup>	100-41-4	x	x	x	10	1,052		20
Ethylcyclobutane	4806-61-5	x	x	x				
Ethylcyclohexane	1678-91-7		x					
Ethylhexanoic acid	149-57-5	x	x			12		63
Formic acid	64-18-6	x			2,000	24		35
Hexamethylethane	594-82-1	x	x					
Hexamethylcyclotrisiloxane	541-05-9	x	x	x				
Hexanoic acid	142-62-1	x	x	x	20			
Isobutyl isobutyrate	97-85-8		x					
Isooctane	540-84-1	x	x	x				38-65-67
Isopropanol <sup>a</sup>	67-63-0	x	x	x	8,000	1,190		36-67
Isopropylbenzene <sup>a</sup>	98-82-8		x		650	595		37-65
Methyl acetate	79-20-9	x	x	x	22,000	1,469		36-66-67
Methylcyclobutane	598-61-8	x	x					
Methylcycloheptane	4126-78-7	x	x	x				
Methylcyclohexane <sup>a</sup>	108-87-2	x	x	x		3,888		38-65-67
Methylethylketone <sup>a</sup>	78-93-3	x	x	x	2,900	1,429		36-66-67
<i>m</i> -Ethyltoluene <sup>a</sup>	620-14-4	x	x	x				
<i>m</i> -Xylene <sup>a</sup>	108-38-3	x	x	x	540	1,052		20/21-38
<i>N,N</i> -Dimethylformamide	68-12-2	x	x		140	71		61-20/21-36
Naphthalene <sup>a</sup>	91-20-3	x	x	x	7	126		22-40
<i>n</i> -Decane <sup>a</sup>	124-18-5	x	x	x	11,300			
<i>n</i> -Dodecane <sup>a</sup>	112-40-3	x	x		11,800			
<i>n</i> -Heptanal	111-71-7	x	x	x	64			
<i>n</i> -Heptane <sup>a</sup>	142-82-5	x	x	x	165,000	3,969		38-65-67
<i>n</i> -Hexadecane <sup>a</sup>	544-76-3	x	x		0.5			
<i>n</i> -Hexane <sup>a</sup>	110-54-3	x	x	x	107,000	426		38-48/20-62-65-67
<i>n</i> -Nonanal <sup>a</sup>	124-19-6	x	x	x	20			
<i>n</i> -Nonane <sup>a</sup>	111-84-2	x	x	x	60,000	2,540		
<i>n</i> -Octanal	124-13-0	x	x		0.01			
<i>n</i> -Octane <sup>a</sup>	111-65-9	x	x	x	71,000	3,395		38-65-67
<i>n</i> -Pentadecane <sup>a</sup>	629-62-9	x	x	x				
<i>n</i> -Pentanal	110-62-3	x	x		92	426		
<i>n</i> -Pentane	109-66-0	x	x	x	92	4,286		65-66-67
<i>n</i> -Propylbenzene <sup>a</sup>	103-65-1	x	x	x	14,400			
<i>n</i> -Tetradecane <sup>a</sup>	629-59-4	x	x		5,000			
<i>n</i> -Tridecane <sup>a</sup>	629-50-5	x	x		42,000			
<i>n</i> -Undecane <sup>a</sup>	1120-21-4	x	x	x	9,600			
Octamethylcyclotetrasiloxane	556-67-2	x	x	x				62
Octanoic acid	124-07-2		x		600			
<i>o</i> -Ethyltoluene <sup>a</sup>	611-14-3	x	x	x				
<i>o</i> -Xylene <sup>a</sup>	95-47-6	x	x	x	770	1,052		20/21-38
<i>p</i> -Dichlorobenzene <sup>a</sup>	106-46-7	x	x	x	730	290		36-40
<i>p</i> -Diethylbenzene	25340-17-4	x	x					
Pentanoic acid	109-52-4	x	x	x	0.00015			34
<i>p</i> -Ethyltoluene	622-96-8	x	x	x				
Propyl acetate	109-60-4	x	x		0.2	2,024		36-66-67
<i>p</i> -Xylene <sup>a</sup>	106-42-3	x	x	x	520	1,052		20/21-38
Styrene <sup>a</sup>	100-42-5	x	x	x	12	205		20-36/38
<i>tert</i> -Butylethylether	637-92-3	x	x	x				
<i>tert</i> -Butylmethylether <sup>a</sup>	1634-04-4	x	x			436		38
Tetrachloroethylene <sup>a</sup>	127-18-4	x	x	x	8,300	407		40
Toluene <sup>a</sup>	108-88-3	x	x		3,800	183		38-48/20-63-65-67
Trichloroethylene <sup>a</sup>	79-01-6	x	x	x	3,900	129		45-36/38-67

Trichlorofluoromethane <sup>a</sup>	75-69-4	x	x	x	28,000
299	<sup>a</sup> VOC identified in several worldwide indoor air studies (Holcomb and Seabrook, 1995); <sup>b</sup> As hexane				
300	isomers; <sup>c</sup> van Gemert, 1999				
301					
302					

303 Table 3. Individual VOC concentrations ( $\mu\text{g m}^{-3}$ ) in the quantified samples

REFERENCE	A1 – A4	C1 – C4	A5	C5	D1	BE1	D2	BE2	BE3	D3	D4	BE4	F1
SAMPLE TYPE	24-hour	24-hour	Ep <sup>a</sup>	Ep	Ep	Ep	Ep	Ep	Ep	Ep	6-h	6-h	6-h
SAMPLING POINT	A	C	A	C	D	B, E	D	B, E	B, E	D	D	B, E	F
ALKANES													
<i>n</i> -Hexane	2.47 – 13.06	0.99 – 7.59	2.65	4.20	1.15	1.06	1.07	1.12	0.84	1.18	3.00	0.77	0.58
<i>n</i> -Heptane	5.53 – 11.76	1.80 – 26.24	3.44	3.60	2.70	3.71	2.20	5.95	1.85	3.38	8.59	2.20	1.73
<i>n</i> -Octane	2.91 – 3.69	1.06 – 2.96	1.48	1.51	0.83	1.29	0.90	1.09	0.67	0.78	0.76	0.63	0.41
<i>n</i> -Nonane	0.28 – 15.14	0.27 – 4.18	11.63	8.01	3.51	7.45	3.38	2.78	3.78	3.10	2.60	3.67	1.54
<i>n</i> -Decane	7.23 – 20.30	1.74 – 9.05	6.60	2.98	n.d.	4.59	1.95	1.50	2.54	2.02	4.27	2.23	0.79
<i>n</i> -Undecane	1.18 – 1.70	0.53 – 1.93	1.82	1.07	0.36	1.08	1.23	0.75	2.01	1.40	0.54	1.17	0.14
<i>n</i> -Dodecane	2.40 – 7.75	0.82 – 6.24	n.d. <sup>b</sup>	n.d.	0.16	0.50	0.81	0.57	1.88	1.29	0.29	0.98	n.d.
2-Methylpentane	<loq <sup>c</sup>	<loq	<loq	<loq	<loq	<loq	<loq	<loq	<loq	<loq	<loq	<loq	<loq
3-Methylpentane	3.50 – 4.32	1.01 – 8.45	1.96	2.27	1.06	0.62	1.00	1.25	0.81	1.24	3.96	0.95	0.81
<i>n</i> -Tridecane	0.30 – 2.72	0.92 – 2.65	2.18	2.09	0.62	1.88	0.96	0.60	1.08	0.62	0.32	0.84	n.d.
<i>n</i> -Tetradecane	0.29 – 1.27	0.88 – 1.48	2.46	1.83	0.23	0.87	0.75	0.39	0.39	0.43	0.35	0.65	n.d.
<i>n</i> -Pentadecane	0.48 – 15.82	0.37 – 4.77	12.37	4.08	2.42	4.22	3.63	2.13	4.00	2.11	1.66	1.93	0.57
<i>n</i> -Hexadecane	2.48 – 5.23	1.18 – 2.32	5.74	2.34	0.75	1.73	1.65	0.82	1.91	1.09	0.64	0.78	n.d.
Cyclohexane	0.81 – 3.59	0.34 – 1.90	0.65	0.70	0.40	0.28	0.40	0.58	0.30	0.45	1.28	0.54	0.37
Methylcyclohexane	1.58 – 3.30	0.79 – 2.52	0.94	1.18	0.66	0.72	0.63	0.75	0.70	0.70	1.19	0.63	0.41
ALCOHOLS													
Ethanol	38.28 – 79.53	29.54 – 98.30	622.39	2786.97	49.24	75.95	149.59	101.91	94.68	89.83	286.95	443.72	106.88
Isopropanol	30.66 – 46.06	11.29 – 31.02	15.94	17.89	33.26	8.15	6.98	35.07	54.71	5.43	259.96	75.72	28.77
1-Butanol	14.67 – 24.17	27.95 – 55.60	42.79	95.23	15.29	35.80	50.13	22.60	20.52	14.20	12.40	31.47	7.19
3-Ethyl-1-Hexanol	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
GLYCOLS													
2-Methoxyethanol	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1-Methoxy-2-propanol	18.22 – 59.91	8.53 – 19.22	210.48	134.80	22.47	30.37	36.01	18.75	41.59	28.93	36.70	378.47	11.79
2-Ethoxyethanol	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2-Buthoxyethanol	1.38 – 60.83	0.66 – 21.34	n.d.	0.79	0.85	2.20	1.58	0.72	1.07	0.70	n.d.	n.d.	n.d.
2-Buthoxyethoxyethanol	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
ALDEHYDES													
Butanal	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Pentanal	5.36 – 11.16	5.05 – 12.32	3.60	5.78	2.03	3.43	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Hexanal	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Heptanal	1.82 – 2.91	2.02 – 2.50	2.32	2.50	0.88	3.06	1.50	0.78	1.57	0.87	0.47	1.59	0.30
<i>n</i> -Nonanal	2.82 – 5.13	7.01 – 13.34	19.18	19.66	1.54	12.45	12.27	4.95	11.13	8.47	2.95	8.50	0.66
Benzaldehyde	0.81 – 14.35	3.05 – 13.13	8.35	5.16	3.92	2.14	1.52	5.78	2.11	4.28	3.71	1.43	2.62
Formaldehyde	<sup>d</sup>	-	-	-	-	-	-	-	-	-	94.90	108.20	8.10
KETONES													
Acetone	31.76 – 62.51	36.04 – 58.92	86.93	69.23	70.95	94.65	85.24	141.75	66.00	41.13	114.10	132.48	116.97

Methylethylketone	2.98 – 12.40	1.09 – 5.38	2.59	3.42	0.82	0.80	n.d.	n.d.	2.93	4.29	9.58	3.23	1.57
Cyclohexanone	0.53 – 1.37	0.37 – 0.46	0.47	0.43	0.17	0.28	0.26	0.14	0.26	0.15	n.d.	n.d.	n.d.
Methylisobutylketone	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Acetophenone	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
HALOCARBONS													
Dichloromethane	4.95 – 19.79	12.52 – 21.40	3.53	8.63	0.98	2.45	2.22	0.91	2.12	1.48	3.25	16.15	0.52
Chloroform	16.81 – 15.28	0.72 – 1.40	3.94	3.18	0.24	0.41	0.30	0.27	0.30	0.32	0.66	0.18	0.19
Trichloroethylene	1.67 – 9.46	1.43 – 8.96	1.12	2.23	0.51	0.76	0.45	0.38	1.18	1.98	2.00	0.37	0.56
Tetrachloroethylene	2.33 – 13.12	3.78 – 9.75	1.33	2.90	1.49	1.99	2.08	1.84	1.73	2.13	2.16	1.38	0.97
<i>p</i> -Dichlorobenzene	1.37 – 2.52	16.08 – 48.22	2.30	20.15	13.65	23.72	15.76	4.62	16.46	11.38	1.07	8.27	0.24
Trichloronitromethane	0.29 – 2.76	0.61 – 1.78	1.47	1.49	2.92	3.65	1.99	0.28	0.62	1.03	4.23	1.56	1.67
1,1,1-Trichloroethane	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
ESTERS													
Methyl acetate	2.99 – 5.25	1.54 – 4.14	1.15	1.58	15.63	0.72	0.89	0.73	0.81	1.23	13.40	23.75	8.47
Ethyl acetate	14.10 – 36.05	3.94 – 11.72	9.97	14.97	2.56	3.28	6.39	3.77	2.94	3.70	6.61	2.71	1.52
Butyl acetate	13.30 – 24.47	6.75 – 24.78	15.03	11.75	1.75	3.21	5.23	3.51	5.29	5.84	1.86	2.57	0.80
Iso-propylacetate	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2-Ethoxyethyl acetate	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
TXIB (Texanol iso-butirate)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
AROMATIC													
HYDROCARBONS													
Benzene	8.71 – 27.7	3.77 – 10.36	2.61	3.85	2.52	1.65	1.50	2.09	1.02	1.74	2.83	0.54	0.49
Toluene	41.90 – 147.29	27.85 – 130.55	29.54	32.61	22.49	21.47	25.52	50.95	20.01	33.36	33.20	24.44	14.40
Ethylbenzene	8.00 – 23.78	3.68 – 5.01	6.17	8.85	3.39	2.79	3.41	4.90	4.65	6.44	3.52	2.09	1.84
<i>m+p</i> -Xylene	30.08 – 40.63	10.04 – 30.45	9.36	12.76	13.88	4.58	7.20	15.45	10.03	15.96	8.07	4.12	3.47
Styrene	1.55 – 3.27	0.32 – 1.40	0.70	0.76	0.39	0.28	0.22	0.28	1.00	1.23	0.74	0.79	0.50
1,2,4-Trimethylbenzene	13.06 – 15.49	1.40 – 17.10	4.22	2.93	2.99	2.62	2.43	3.68	3.05	3.22	2.17	2.13	1.39
<i>m</i> -Ethyltoluene	3.98 – 11.26	0.86 – 4.00	2.19	1.66	1.27	1.12	1.21	1.75	1.51	1.81	1.09	1.05	0.72
1,3,5-Trimethylbenzene	2.28 – 5.17	0.70 – 2.82	1.88	1.51	1.07	1.06	1.14	1.94	n.d.	0.93	0.66	0.47	0.36
<i>n</i> -Propylbenzene	0.02 – 0.03	0.01 – 0.02	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02
Naphtalene	0.03 – 0.08	0.03 – 0.05	0.05	0.04	0.04	0.03	0.03	0.04	0.04	0.04	0.03	0.04	0.01
Tetrahydrofuran	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2-Pentylfuran	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
<i>o</i> -Ethyltoluene	1.38 – 2.99	0.30 – 1.41	1.00	0.63	0.39	0.49	0.44	0.55	0.57	0.59	0.33	0.41	0.24
<i>p</i> -Ethyltoluene	0.42 – 2.42	0.67 – 1.07	0.49	0.68	0.40	0.64	0.46	0.40	0.77	0.72	0.28	0.40	0.13
ACIDS													
Acetic acid	62.11 – 114.20	64.47 – 138.99	91.45	124.67	59.91	43.30	22.86	19.25	42.89	83.47	69.81	207.84	38.09
Pentanoic acid	5.28 – 7.01	3.24 – 9.58	n.d.	n.d.	n.d.	11.28	n.d.	n.d.	n.d.	n.d.	1.04	4.85	0.87
Hexanoic acid	9.71 – 12.83	7.77 – 24.96	n.d.	n.d.	1.83	23.40	15.75	4.39	25.16	4.20	1.94	14.61	2.05
Octanoic acid	n.d. – 49.75	n.d. – 29.47	n.d.	n.d.	0.60	n.d.	n.d.	n.d.	38.81	n.d.	n.d.	2.58	n.d.
TERPENES													
$\alpha$ -Pinene	6.15 – 15.20	5.36 – 8.74	4.39	7.34	1.48	2.80	2.16	1.41	2.62	1.75	0.37	1.63	0.12
DL-Limonene	30.28 – 32.52	3.46 – 9.90	8.31	20.87	1.12	3.30	2.58	1.67	3.11	1.43	0.50	6.04	0.23
$\beta$ -Pinene	0.57 – 1.72	1.81 – 2.02	1.69	4.96	1.04	2.07	1.52	0.91	2.05	1.32	0.67	6.22	0.21

3-Carene	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
<b>AMIDES</b>													
<i>N,N</i> -Dimethylformamide	14.15 - 37.40	n.d - 0.22	2.6	n.d.	n.d.	11.74	7.24	n.d.	13.32	n.d.	n.d.	n.d.	n.d.
<b>ORGANOSULFURS</b>													
Carbon disulfide	0.49 - 2.10	0.77 - 2.14	0.82	1.46	12.22	0.44	1.5	2.01	28.79	10.54	76.91	23.35	15.22
<b>ORGANONITROGENATES</b>													
Acetonitrile	0.20 - 0.33	0.07 - 0.47	0.45	1.23	0.27	0.05	0.05	0.07	0.36	0.26	4.34	397.38	21.51
ΣOTHER <sup>e</sup>	55.62 - 97.38	40.54 - 101.66	122.62	160.16	43.86	65.08	60.89	71.87	53.61	48.62	155.42	52.21	44.06
<b>TVOCs<sup>f</sup></b>	<b>534.5 - 1269.2</b>	<b>329.8 - 1078.4</b>	<b>1399.4</b>	<b>3631.6</b>	<b>427.2</b>	<b>539.7</b>	<b>559.1</b>	<b>552.7</b>	<b>604.2</b>	<b>464.8</b>	<b>1250.3</b>	<b>2012.9</b>	<b>453.1</b>

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<sup>a</sup>Episodes; <sup>b</sup>Not detected; <sup>c</sup>Below limit of quantification; <sup>d</sup>Not determined; <sup>e</sup>Remaining compounds identified and quantified by the response factor of toluene; <sup>f</sup>Total Volatile Organic Compounds;

305 In non-industrial established buildings the concentrations of individual VOC  
306 are usually below  $5 \mu\text{g m}^{-3}$ , with only a few compounds exceeding  $50 \mu\text{g m}^{-3}$ , such  
307 as *m+p*-xylenes, toluene, DL-limonene, acetone and ethanol (Brown *et al.*, 1994;  
308 Johansson, 1999). The concentrations of some VOC found in this study (ethanol,  
309 isopropanol, 1-butanol, acetonitrile and 1-methoxy-2-propanol) are higher than the  
310 expected, being generally over  $50 \mu\text{g Nm}^{-3}$  and with a maximum of more than 2,500  
311  $\mu\text{g Nm}^{-3}$  in the case of ethanol (Table 3).

312 On the other hand, several studies in different worldwide dwellings expose  
313 that the most abundant VOC found in homes are toluene, benzene, ethylbenzene,  
314 *m+p*-xylenes, 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene, DL-limonene, *p*-  
315 dichlorobenzene, trichloroethylene, tetrachloroethylene, *n*-decane, chloroform, *n*-  
316 hexanal and *n*-nonanal (Krause *et al.*, 1987; Wallace *et al.*, 1987; Lee *et al.*, 2002).  
317 In addition to that, in American office buildings acetone and 2-butoxyethanol also  
318 have been detected in relevant abundance (Griman *et al.*, 1999). These above  
319 mentioned compounds have been detected in the studied dwelling (Table 2; Table 3),  
320 however, are not the most abundant. The compounds found in highest concentrations  
321 in all episode and 6-hours samples of this study (ethanol, isopropanol, 1-butanol, 1-  
322 methoxy-2-propanol and acetonitrile), as it has been said before, are not expected  
323 being detected in such high concentration in a dwelling, as their release is not  
324 originated by the common sources of VOC in indoor air homes (Jones, 1999), like  
325 cleaning products, floor waxes, paints or vehicle exhausts (Weschler *et al.*, 1990).  
326 The highest concentrations of these compounds are registered in episode periods,  
327 which coincide with odour detection episodes and expression of symptoms of SBS  
328 by the inhabitants of the dwelling.

329 The rest of compounds analysed in the study (mainly aromatic hydrocarbons,  
330 halocarbons, alkanes and terpenes) are found in the same range of concentrations in  
331 great part of all sampling points, and presented concentrations in the same order of  
332 magnitude of other indoor air studies carried out in homes in USA, The Netherlands,  
333 U.K., Canada, Germany, Finland, Korea and Hong Kong (Hartwell *et al.*, 1984; Gupta  
334 *et al.*, 1984; Lerbet *et al.*, 1986; Pellizzari *et al.*, 1986; Krause *et al.*, 1987; Wallace *et al.*,  
335 1987; Montgomery and Kalman, 1989; Chan *et al.*, 1990; Proctor *et al.*,  
336 1991; Holcomb and Seabrook, 1995; Koistainen, 1995; Baek *et al.*, 1995; Lee *et al.*,  
337 2002; Guo *et al.*, 2004). Hence, the general distribution of VOC in indoor air is the  
338 standard for an established dwelling, however, several compounds are detected in



339 higher concentrations than the expected (e.g. ethanol, 1-butanol, 1-methoxy-2-propanol)  
340 and other compounds detected are not commonly found in homes (e.g. acetonitrile).

341 Indoor air samples show a similar VOC composition to outdoor air samples, as  
342 outdoor air is an important source of indoor air pollution, and its composition is usually  
343 reflected indoors (Johansson, 1999). In outdoor air, ethanol, 1-methoxy-2-proanol and  
344 1-butanol show a gradient of concentrations in points A>D>F, being the concentrations  
345 higher in the interior patios, probably due to a less mixing and diffusive activities in  
346 these locations in respect to street outdoor air. Point A, corresponding to interior patio 1,  
347 shows the highest concentrations, being ethanol, 1-methoxy-2-propanol and 1-butanol  
348 from 6 to 18 times more concentrate than the street outdoor air (Point F).

349 In indoors, Point C (corresponding to the bathroom) is the site with the  
350 highest concentrations of the above-mentioned compounds, followed by Points B  
351 and E (Table 3). However, the rest of compounds analysed in Points C, B and E  
352 show concentrations in the range of a standard dwelling (Holcomb and Seabrook,  
353 1995). In Point C, ethanol indoor/outdoor ratio is in the range 5-19 in respect to the  
354 interior patios, and 26 in respect to point F. Similar ratios are observed for 1-  
355 methoxy-2-propanol and 1-butanol. Point C presents the highest concentrations of  
356 the dwelling for these compounds probably as a cause of an important lack of  
357 ventilation. Its space is reduced and the only possibility of ventilation comes trough  
358 the bathroom exhaust. On the other hand, Points B and E (dinning room and kitchen)  
359 can ventilate trough the large windows of the terrace.

360 These high indoor/outdoor ratios indicate that there are also indoor building  
361 sources that influence the dwelling air quality, as the street outdoor air  
362 concentrations cannot contribute significantly to indoor ethanol, 1-methoxy-2-  
363 propanol and 1-butanol concentrations, even with many openings and inadequate  
364 ventilation rates. However, the interior patios could contribute to these high indoor  
365 concentrations, as their air concentrations are much higher than the street outdoor  
366 air. The known activities around the dwelling neither explained the presence of these  
367 concrete VOC nor their quite high concentrations. The possible sources of these  
368 compounds remained unknown and might have an internal origin, being probably  
369 released from industrial processes developed in the nearness of the studied flat, and  
370 entering the dwelling trough the interior patios and the kitchen and bathroom  
371 exhausts.

372 An exhaustive revision of the activities realized in the adjacent buildings of

373 the studied dwelling, conducted by the environmental municipality services and the  
374 police, allowed the finding of a not declared manipulation and storage facility of  
375 industrial solvents. However, a monitoring in this facility was not possible as an  
376 investigation was being conducted. One year later (December 2007), a monitoring  
377 was done in the studied dwelling, showing lower levels of the most important VOC  
378 detected in the first sampling period and the total absence of acetonitrile.

379 The main health effects linked to human exposure to ethanol, 1-methoxy-2-  
380 propanol, 1-butanol and acetonitrile through inhalation are sore throat, cough,  
381 respiratory system irritation, eye redness, headache, poor concentration capacity and  
382 nausea (International Chemical Safety Cards (WHO/IPCS/ILO)). These symptoms  
383 coincide with the suffered by the dwelling inhabitants; probably being the relatively  
384 high concentrations of these concrete compounds one of the major causes of their  
385 discomfort and nuisance perception. On the other hand, several of the determined  
386 compounds are harmful, irritant, and toxic and may cause sensitization or even  
387 cancer, as their associated R-phrases show (Table 2, Fig. 5, Fig. 6).

388 From all identified compounds in the dwelling that have an associated R-  
389 phrase (Table 2), a 23% emit vapours that may cause drowsiness and dizziness, a  
390 14% are irritating to skin, a 13% are harmful and may cause lung damage, a 9% are  
391 irritating to eyes and a 9%, the repeated exposure to them may cause skin dryness  
392 (Fig. 5). On the other hand, from all identified compounds those have a combined R-  
393 phrase associated (Table 2), a 25% are irritating to eyes and skin, a 15% are harmful  
394 by inhalation and in contact with skin, and a 15% are a danger to health by  
395 prolonged exposure to them by inhalation (Fig. 6).

396 In addition to that, the concentrations determined of some of them (Table 3)  
397 are above the odour threshold (Table 2). 1-Butanol, 2-butoxyethanol, *n*-hexadecane,  
398 and pentanoic acid concentrations have been always detected above the odour  
399 threshold concentration. Other compounds such as benzaldehyde, ethylbenzene and  
400 hexanoic acid only overpass the concentration odour threshold in 24-hour samples.  
401 In Table 2, the value of TLV/420 is shown for several of the determined compounds.  
402 TLV/420 is commonly used to establish maximum concentration limits in urban  
403 outdoor air in a 24-hour period for non-carcinogenic compounds. The factor 420 is  
404 an uncertainty factor that takes into account the varied physiological status of people  
405 (Repetto, 1997). In the present case, acetic acid overpasses in 24-hour controls and  
406 in some episode samples its value TLV/420.

## 407 2.2 Total Volatile Organic Compounds

408 Higher TVOC values are found in episodes (ranging from 520 to 3,632  $\mu\text{g Nm}^{-3}$ )  
409  $\text{Nm}^{-3}$ ) and working timetable samples (6-hour) (2,013  $\mu\text{g Nm}^{-3}$ ) in indoor air than in  
410 terrace and interior patios outdoor air (ranging from 453 to 1,399  $\mu\text{g Nm}^{-3}$ ) (Table  
411 3). The individual compounds with major contribution in TVOC are ethanol,  
412 acetone, isopropanol, 1-butanol, 1-methoxy-2-propanol and acetonitrile, as it has  
413 been discussed before. TVOC concentrations are generally found in the same order  
414 of magnitude in samples taken in the same day at different dwelling locations (Table  
415 1, Talbe 3), indicating that the most important contribution to punctual  
416 concentrations of VOC in indoor air are odour episodes above room location in the  
417 dwelling. However, Point C presents high TVOC concentrations probably due to its  
418 low level of ventilation.

419 TVOC values found in European standard dwellings in several studies ranged from  
420 40 to 1,050  $\mu\text{g m}^{-3}$  (Table 4). In addition to this, Seifert (1990) estimated a target  
421 indoor air quality guideline value based on two empirical field studies in German  
422 and Dutch homes (WHO, 1989; Lebret *et al.*, 1986; Krause *et al.*, 1987; Seifert and  
423 Abraham, 1982), suggesting that TVOC concentrations in indoor air should not  
424 exceed 300  $\mu\text{g m}^{-3}$ . Generally, TVOC calculated values do not include very volatile  
425 organic compounds ( $< 0^\circ\text{C} < \text{Boiling point} < 50^\circ\text{C}$ ), and only include the range of  
426 compounds obtained in the analytical window between hexane and hexadecane on a  
427 non-polar column (ISO 16000-6). In these cases, compounds such as ethanol and  
428 acetone are not included in the TVOC value. However, as in the present study these  
429 compounds do have been included, all indoor samples analysed overtake the  
430 previously mentioned guideline value, indicating that the indoor VOC  
431 concentrations are higher than the expected for a European standard dwelling. If  
432 ethanol and acetone were removed form the calculated TVOC value, obtained values  
433 would be in the range of an European standard dwelling. Hence, only several  
434 compounds are the responsible for the annoyance and discomfort that the occupants  
435 of the dwelling suffer.

436

437 Table 4. TVOC concentrations in European dwellings

TVOC ( $\mu\text{g m}^{-3}$ )	Country	Date	Reference
250	Sweden	1981/1982	Mølhave, L., 2000

400	Germany	1985/1986	Krause, C. et al., 1987
40-235	Finland	1995	Kostiainen, R. et al., 1995
200-500	United Kingdom	1996	Brown & Crump, 1996
85-1050	Sweden	2000	Bornehag & Stridh, 2000
330-3630	Spain	2006	This study

438

439 In addition to that, Mølhave (1991) suggested four exposure ranges of  
 440 TVOC: comfort range ( $<0.2 \text{ mg m}^{-3}$ ), multifactorial exposure range ( $0.2\text{-}3 \text{ mg m}^{-3}$ ),  
 441 discomfort range ( $3\text{-}25 \text{ mg m}^{-3}$ ) and toxic range ( $>25 \text{ mg m}^{-3}$ ). Indoor air results  
 442 obtained in this study suggest that the occupants of the dwelling are in a  
 443 multifactorial exposure range, where they can suffer mucous and skin irritation and  
 444 general discomfort. This discomfort may increase if exposures of other compounds  
 445 interact with the exposures of VOC (Mølhave, 1991).

446 It can be concluded that there is an indoor pollution, which diminishes the  
 447 indoor air quality and triggers the discomfort sensation among the dwelling  
 448 occupants, coming from several major compounds, mainly ethanol, acetone, acetic  
 449 acid and 1-methoxy-2-propanol, that alters the VOC family distributions expected  
 450 for a standard dwelling (Seifert, 1990). These compounds are used as solvents in a  
 451 wide range of industrial activities.

452 The chemical analysis method used, the results obtained in this study and the  
 453 comparison of values previously determined in worldwide dwellings not affected by  
 454 external industrial activities, has shown to be enough to point out that the origin of  
 455 VOC indoor concentrations was external. Besides, the present study has served as a  
 456 basis for an environmental authority action in order to solve a problem of odorous  
 457 and nuisance episodes in a dwelling caused by high concentrations of VOC in its  
 458 indoor air.

459

### 460 **3. Conclusions**

461 The methodology proposed in the present work, which is focused in evaluating  
 462 external VOC sources that contribute to indoor environments pollution, has been  
 463 demonstrated to be useful in a case study carried out in an urban dwelling.

464 The study, based in air quality control systems through TD-GC/MS, determined  
 465 qualitatively and quantitatively a relevant number of VOC in a Spanish dwelling where  
 466 the occupants seemed to experience sick building syndrome symptoms. The main  
 467 compounds detected in the studied dwelling (ethanol, isopropanol, 1-butanol, acetic  
 468 acid, acetonitrile and 1-methoxy-2-propanol) are not the most abundant VOC found in

469 worldwide homes, and their concentrations are generally higher than the expected  
470 (generally over 50  $\mu\text{g Nm}^{-3}$ ). TVOC concentrations are also higher than the expected for  
471 a standard dwelling, mainly due to the contribution to TVOC of the above mentioned  
472 main compounds. The highest concentrations of VOC are registered in episodic periods,  
473 which coincide both with odour detection episodes and expression of symptoms of SBS  
474 by the inhabitants of the dwelling. High indoor/outdoor ratios for some compounds  
475 (ethanol, 1-methoxy-2-propanol and 1-butanol) indicate that there are also indoor  
476 building sources that influence the dwelling air quality, as the street outdoor  
477 concentrations cannot contribute significantly to the indoor concentrations of these  
478 concrete compounds. They are probably released from industrial processes developed in  
479 the nearness of the studied flat, entering the dwelling through the interior patios and the  
480 kitchen and bathroom exhausts. This theory has been eventually confirmed with the aid  
481 of the environmental inspection services of the city town hall and the police, which  
482 found a not declared management and storage industrial solvent facility in the bottom of  
483 an adjacent building. Until this activity was discovered, it remained unknown to the  
484 neighbours.

485

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488

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### **List of figure captions**

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Fig 1. TD-GC/MS chromatograms of a simultaneous sample using Tenax TA and multi-sorbent (Carbotrap, Carbopack X and Carboxen-569) adsorbents for its sampling.

Fig 2. Potential emission sources (black arrows) and sampling points in the studied dwelling.

Fig 3. VOC familiar distribution for all compounds identified qualitatively. Percentage of compounds of each family in respect to all compounds identified.

Fig 4. GC chromatograms of 24-hour and episode samples. Correspondent VOC reference numbers are listed in the adjacent table.

Fig 5. Distribution of compounds with R-phrases associated. Percentage of identified compounds having concrete R-phrases.

Fig 6. Distribution of compounds with combined R-phrases associated. Percentage of identified compounds having combined R-phrases.

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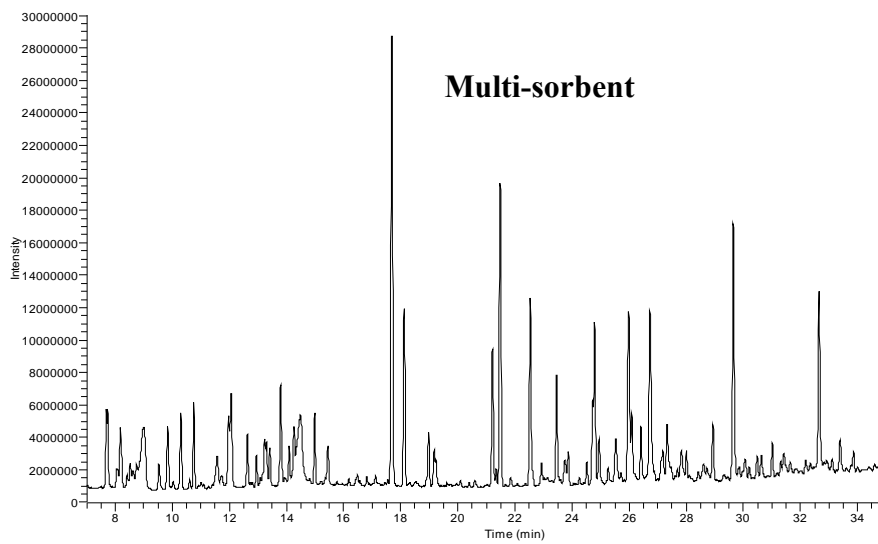
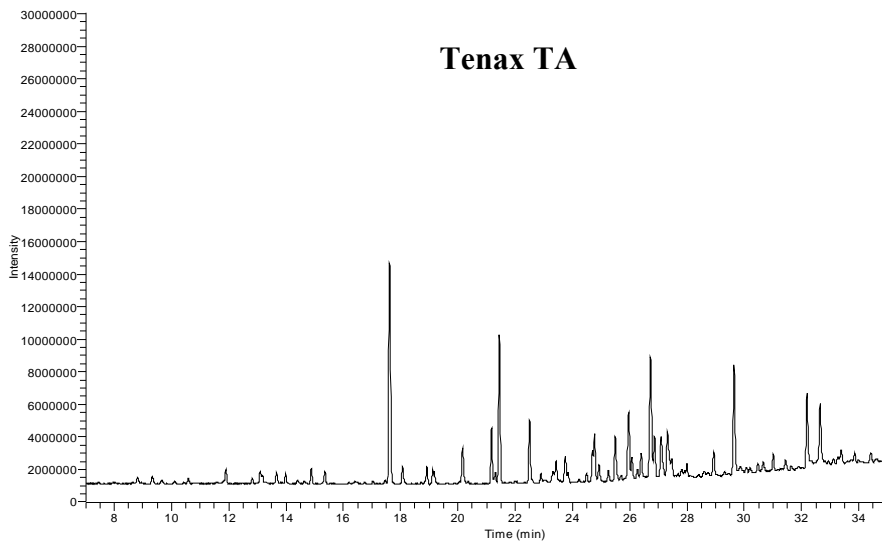
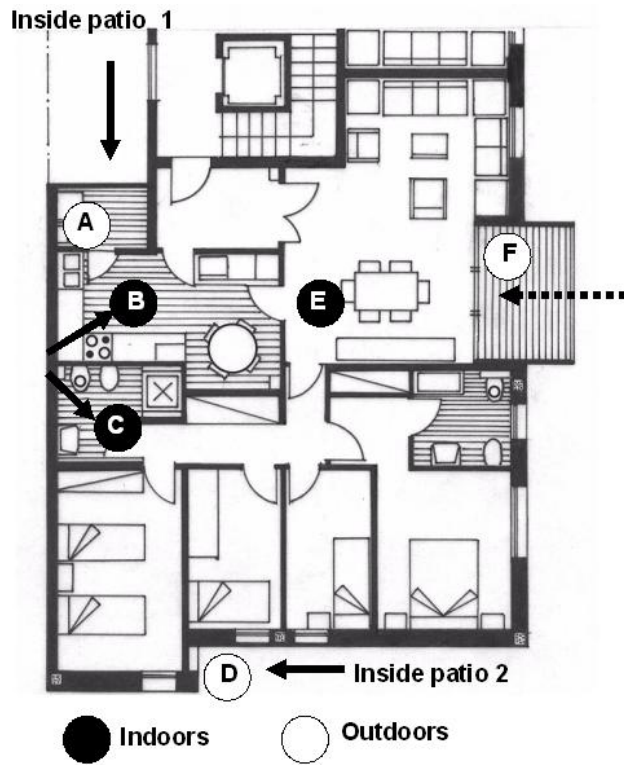


Figure 1



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Figure 2.

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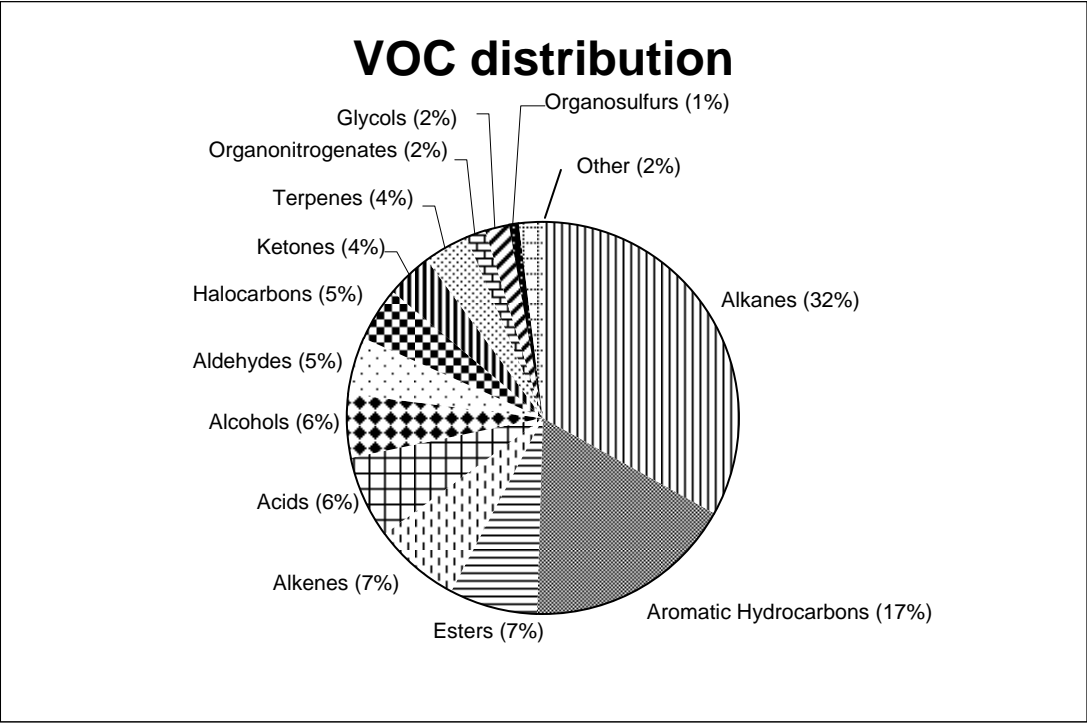
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Fig. 3

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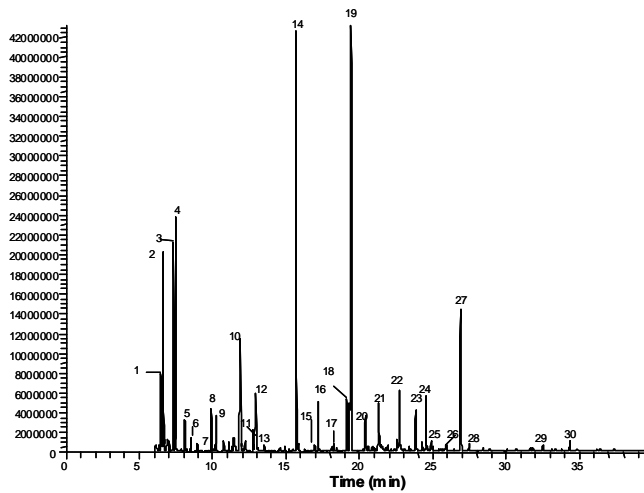
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Number	Compound
1	<i>n</i> -Pentane
2	Ethanol
3	Acetone
4	Isopropanol
5	Isohexane
6	3-Methylpentane
7	<i>n</i> -Hexane
8	<i>tert</i> -Butylmethylether
9	Methylethylketone + Ethyl acetate
10	Acetic acid + Benzene
11	1-Butanol
12	1-Methoxy-2-propanol
13	Methylcyclohexane
14	Toluene
15	Hexamethylcyclotrisiloxane
16	Butyl acetate
17	<i>N,N</i> -Dimethylformamide
18	Ethylbenzene
19	<i>m</i> -Xylene + <i>p</i> -Xylene
20	<i>o</i> -Xylene + Styrene
21	2-Butoxyethanol
22	<i>n</i> -Undecane
23	<i>p</i> -Ethyltoluene
24	Limonene
25	<i>p</i> -Dichlorobenzene
26	<i>n</i> -Dodecane
27	Benzoic acid, 2-[(trimethylsilyl)oxy]-, trimethylsilyl ester
28	<i>n</i> -Nonanal
29	Benzothiazole
30	<i>n</i> -Pentadecane

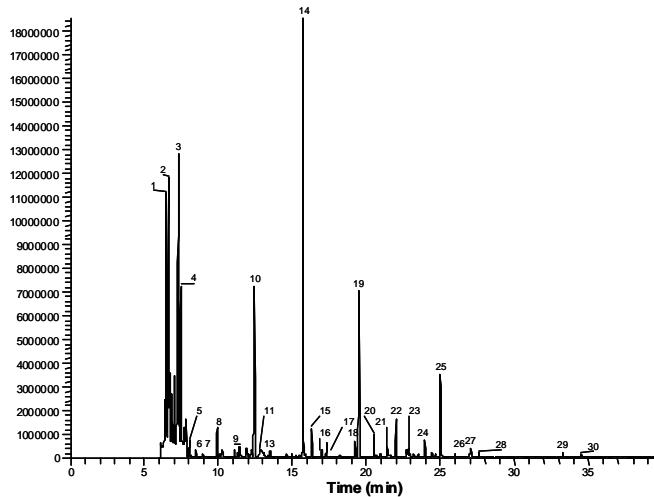
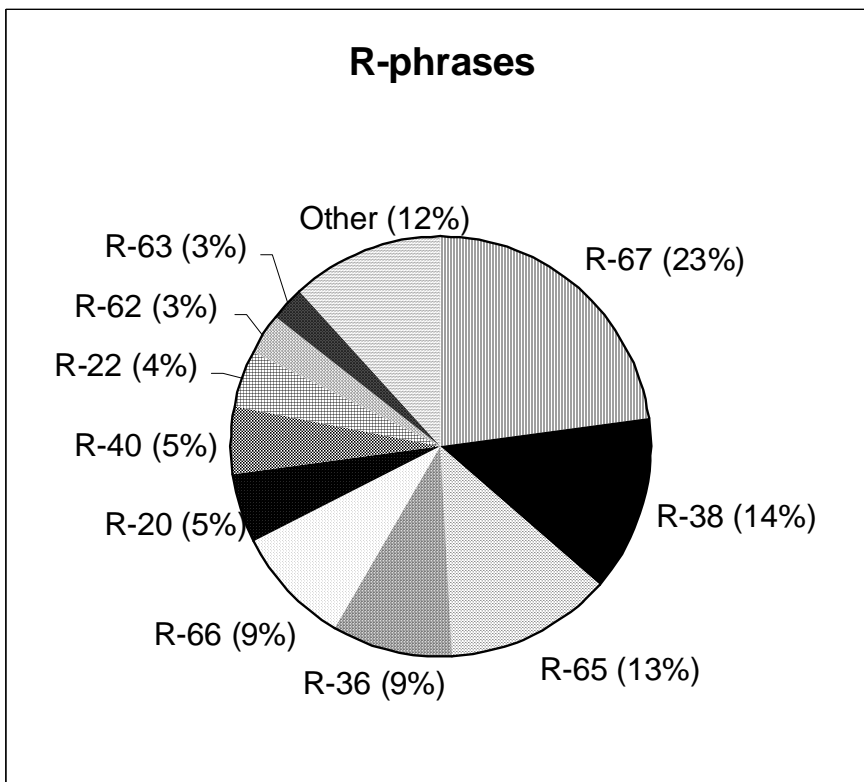


Figure 4.

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- R-67: Vapours may cause drowsiness and dizziness
- R-38: Irritating to skin
- R-65: Harmful: may cause lung damage if swallowed
- R-36: Irritating to eyes
- R-66: Repeated exposure may cause skin dryness or cracking
- R-20: Harmful by inhalation
- R-40: Possible risk of cancer
- R-22: Harmful if swallowed
- R-62: Possible risk of impaired fertility
- R-63: Possible risk of harm to the unborn child

Figure 5



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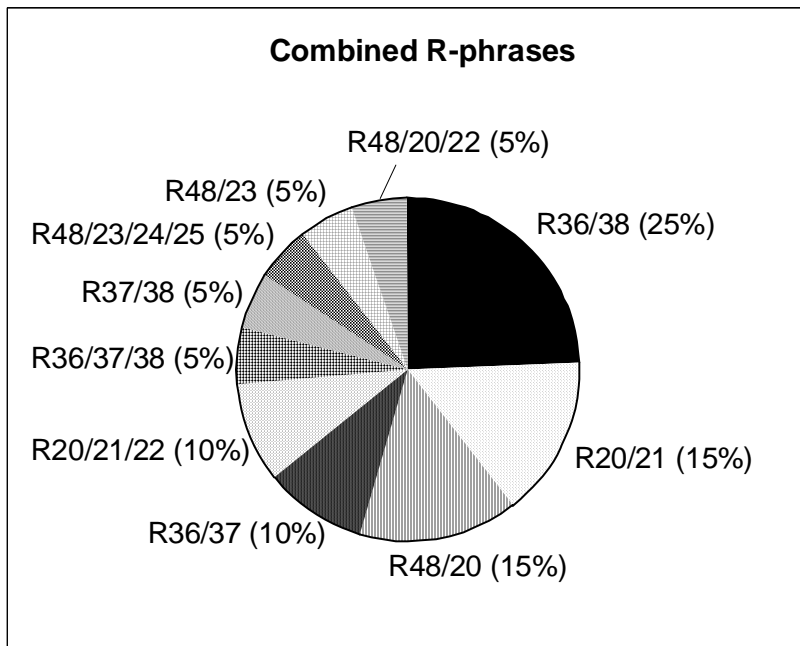
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R36/38: Irritating to eyes and skin

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R20/21: Harmful by inhalation and in contact with skin

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R48/20: Harmful: danger of serious damage to health by prolonged exposure through inhalation

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R36/37: Irritating to eyes and respiratory system

856

R20/21/22: Harmful by inhalation, in contact with skin and if swallowed

857

R36/37/38: Irritating to eyes, respiratory system and skin

858

R37/38: Irritating to respiratory system and skin

859

R48/23/24/25: Toxic: danger of serious damage to health by prolonged exposure through inhalation, in contact with skin and if swallowed

861

R48/23: Toxic: danger of serious damage to health by prolonged exposure through inhalation

862

R48/20/22: Harmful: danger of serious damage to health by prolonged exposure through inhalation and if

863

swallowed

864

865 Figure 6.