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Malak Rizk, Marie Verrièle, Sébastien Dusanter, Coralie Schoemaeker, Stéphane Le Calvé, et al.. Fast sorption measurements of volatile organic compounds on building materials: Part 1 – Methodology developed for field applications. *Building and Environment*, 2016, 99, pp.200-209. 10.1016/j.buildenv.2015.12.017 . hal-02303729

HAL Id: hal-02303729

<https://hal.science/hal-02303729>

Submitted on 2 Oct 2019

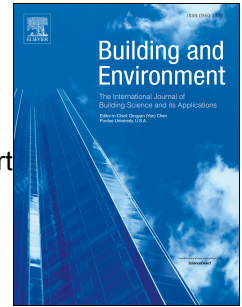
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Accepted Manuscript

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PII: S0360-1323(15)30215-8

DOI: [10.1016/j.buildenv.2015.12.017](https://doi.org/10.1016/j.buildenv.2015.12.017)

Reference: BAE 4348

To appear in: *Building and Environment*

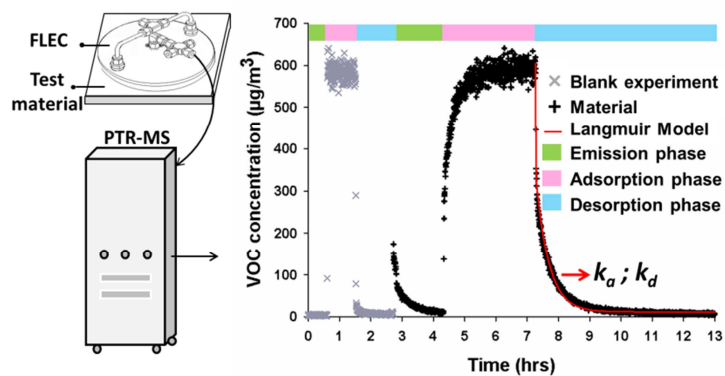
Received Date: 22 September 2015

Revised Date: 14 December 2015

Accepted Date: 20 December 2015

Please cite this article as: Rizk M, Verrièle M, Dusanter S, Schoemaeker C, Le calve S, Locoge N, Fast sorption measurements of volatile organic compounds on building materials: Part 1 - Methodology developed for field applications, *Building and Environment* (2016), doi: 10.1016/j.buildenv.2015.12.017.

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Graphical abstract

1 Fast sorption measurements of volatile organic 2 compounds on building materials: Part 1 - Methodology 3 developed for field applications

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16 **Abstract**

17 Several physicochemical processes occurring within buildings are key drivers of indoor
18 concentrations of Volatile Organic compounds VOCs. Many models and experimental studies
19 have been proposed to predict VOCs concentration indoors given these processes. However,
20 there is a lack of representative data in literature to present gas-surface interaction in order to
21 validate mathematical models. This work is divided in two parts and aims to develop and
22 validate a method to perform fast measurements of VOC sorption parameters on the field by
23 coupling a Field and Laboratory Emission Cell (FLEC) to a Proton Transfer Reaction-Mass
24 Spectrometer (PTR-MS). In the part 1 of the work, sorption coefficients of aromatic
25 compounds on a gypsum board and vinyl flooring were investigated at ppb levels to test and
26 evaluate the proposed methodology. Sorption coefficients in the range of 0.03-1.88 m.h⁻¹ for
27 k_a and 2.04-17.32 h⁻¹ for k_d were successfully measured within a (0.5-8 hours) for the two
28 materials. Robustness tests highlight that the determination of sorption coefficients does not
29 depend on operating conditions. While sorption coefficients for the gypsum board were
30 measured with a PTR-MS time resolution of 20 seconds, the vinyl flooring material required
31 measurements at a higher time resolution of 2 seconds due to its lower sorption properties.
32 Limits of applicability assessed for this method indicate that sets of sorption parameters (k_a ,
33 k_d) of (0.01 m.h⁻¹; 0.01 h⁻¹) and (0.09 m.h⁻¹; 0.09 h⁻¹) can be measured with an accuracy better
34 than 10% at time resolutions of 2 and 20 seconds respectively.

35 **Keywords**

36 Field and laboratory emission cell (FLEC), Proton transfer reaction-mass spectrometer (PTR-
37 MS), Sorption, VOCs, Building materials.

38 1. Introduction

39 Volatile organic compounds VOCs are the main pollutants in indoor environment, which
40 present a strong impact on human health, comfort and productivity [1-3]. Several
41 physicochemical processes occurring within buildings are key drivers of indoor
42 concentrations of VOCs [4]. These processes include heterogeneous and gas-phase chemical
43 reactions, air exchange with outdoor and gas-surface interactions. A good understanding of
44 these processes is important to develop efficient strategies to reduce human exposure to
45 indoor air pollution. The available methods developed in literature to study the concentrations
46 of VOCs indoors, given the above-mentioned processes fall into two categories, modeling
47 approaches and experimental investigation under well-controlled environments.

48 Under the first category, several models have been developed to simulate the VOC
49 emission/sorption by indoor surfaces. As far as the sorption process is concerned, some
50 models considered the adsorption (k_a) and desorption (k_d) rates [5-7] while, other accounted
51 for the diffusion coefficient (D_m) in the building material and represents the sorption with a
52 partitioning coefficient (K) between the air and the material through a diffusion-controlled
53 mass transfer model proposed by Little [8]. Later Deng [9] proposed an improved model that
54 considers for the convective mass transfer coefficient (h_m) through the boundary layer present
55 on the surface of a material as well as the diffusion and the partitioning coefficients. For the
56 emission process, the diffusion-controlled mass transfer model proposed by Little [8]
57 promoted the development of the emissions mass transfer model such as the model of Xu [10]
58 applied for single-layer building material, the model of Lee [11] applied for porous materials
59 and the model of [12, 13] applied for multilayer building materials. A general model was
60 developed by Xiong [14] to characterize both emission and sorption process in ventilated and
61 airtight chambers taking into account the convective mass transfer coefficient, the diffusion
62 coefficient (D_m) and the partitioning coefficient (K). All these models are presented in the
63 reviews of Liu [13] and a comparison between some of the typical models is given elsewhere
64 [15]. Nevertheless, all the mentioned-above models consider only the gas-surface interactions
65 and neglect the chemical reactions that can take place indoors in both homogenous and
66 heterogeneous phases. Therefore, some studies [16, 17] proposed indoor air quality models
67 which accounts for the effect of ventilation, deposition of inorganic species on surfaces,
68 emission and photochemistry reactions with a consideration for a perfect mixing of the air
69 inside simulated room. However, these photochemistry models neglects the sink behavior of
70 surfaces.

71 Concerning the experimental investigations, only laboratory studies have provided adsorption
72 (k_a) and desorption (k_d) rate coefficients for VOCs on surfaces, to describe the role of these
73 interactions on VOCs indoors concentrations. The experimental procedure is based on the
74 exposure of sample materials, inside an emission test chamber, to artificially polluted
75 atmospheres containing several ppm of VOCs [7, 18-20]. The mixing ratios of VOCs used in
76 these experiments are several orders of magnitude higher than those observed in indoor
77 environments at ppb levels [21] and are then not representative. These works were conducted
78 considering either a single organic compound and a single material [7, 18, 22, 23] or mixtures
79 of VOCs and combinations of materials [5, 24, 25]. To extract the parameters of interest,

80 experimental concentration profiles, usually measured by gas chromatography techniques, are
81 fitted by a relevant model, more or less complex as already presented above, depending on the
82 processes considered (sorption processes, diffusion into the material, and diffusion through
83 the boundary layer formed above the material surface) and the type of material (homogenous
84 or porous media). .

85 However, when sorption coefficients derived from chamber experiments was incorporated in
86 indoor air quality (IAQ) models , a significant differences as high as a factor of 9 [6] was
87 shown between modeled concentrations and ambient observations made in test houses. This
88 disagreement may be due to sorption coefficients for real indoor surfaces that are different
89 than those measured during laboratory experiments [6] because the implementation of a
90 material in a real environment and its aging due to varying environmental conditions can
91 affect its sorption properties compared to those observed in laboratory chambers. In addition,
92 the use of inappropriate models to describe real situations can leads to this disagreement as
93 already highlighted by Xu [10] .

94 Given the studies mentioned above, there is actually a need to:

- 95 • Develop a model that account for all physical and chemical processes occurring indoors
96 such as emission, outdoor input, chemical reactivity and also sorption on the surface of
97 building materials and,
- 98 • Validate proposed models using more representative experimental data from real
99 environment to determine the influence of each process on the VOCs concentration
100 indoors.

101 Consequently, a research project called MERMAID (Mesures Expérimentales Représentatives
102 et Modélisation Air Intérieur Détaillée / Representative Experimental Measurement-Indoor
103 Air Detailed Model) was designed to address the leaks found in literature as already
104 mentioned above [26]. The originality of the MERMAID project is the combination between
105 an experimental approach based on several types of measurements for VOCs, inorganic gases,
106 particulate matter and reactive species to a modeling approach based on a detailed indoor air
107 quality model called INCA-Indoor, including the physical and chemical processes.
108 Concerning the sorption processes, the INCA-indoor model proposed by Mendez [27] treats
109 the sorption of VOCs on the surface of building materials through the adsorption (k_a) and
110 desorption (k_d) coefficients represented as a function of the mass transfer coefficient (h_m) in
111 the boundary layer. However, the INCA-Indoor model neglects the diffusion or the mass
112 transfer in the inner of the material since several inputs parameters are needed which is
113 difficult to determine on the field (effective diffusion coefficients, initial concentration in the
114 materials...etc) and which also requires computational efforts to solve diffusion equations.
115 Nevertheless, the emission rate coefficient is taken into account to represent the mass transfer
116 in the inner of the material. To meet the objective of the MERMAID project, this study aims
117 to develop a methodology suitable to perform in-situ fast measurements of VOC sorption
118 coefficients in indoor environments and under real conditions. Derived experimental
119 parameters will be useful as data inputs to INCA-indoor air quality model.

120 This work aims to develop a method to determine in a fast and simple way the sorption
121 parameters on the surface of a material present in indoor environment. Therefore this work is
122 divided in three steps:

123 (1) Develop a new device based on a field and laboratory emission cell (FLEC) coupled to a
124 proton transfer reaction mass spectrometer (PTRMS);

125 (2) Validate the proposed methodology during laboratory experiments to assess the feasibility
126 of fast in-situ measurements and the limitations of the method applicability; and

127 (3) Compare this method to the traditional emission test chamber method used usually in
128 literature to perform sorption measurements.

129 Only the findings of the first two steps are presented in this part of the study and the
130 comparison with the traditional method is presented in the part 2 which includes a discussion
131 on the usage of the experimental data in the INCA-Indoor model. Therefore, in this first part
132 of the work the sorption properties of a mixture of aromatic VOCs at ppb levels were
133 investigated on an unpainted gypsum board and vinyl flooring presenting contrasting sorption
134 behavior [18]. These VOCs are considered as ubiquitous compounds in indoor environments
135 and presents a large fraction of total VOC concentrations [21, 28-30]. Moreover, BTEX were
136 chosen to test the proposed methodology, because they can be easily purchased in certified
137 cylinders and are not prone to memory effects on sampling materials.

138 **2. Materials and Methods**

139 **2.1. *Chemicals***

140 For all the experiments, the BTEX mixture was provided by Air products and contained the
141 following VOC in the mixing ratio of 2 for benzene, 6 for toluene, 1 for ethyl benzene, 2 for
142 p-xylene and 1 for o-xylene.

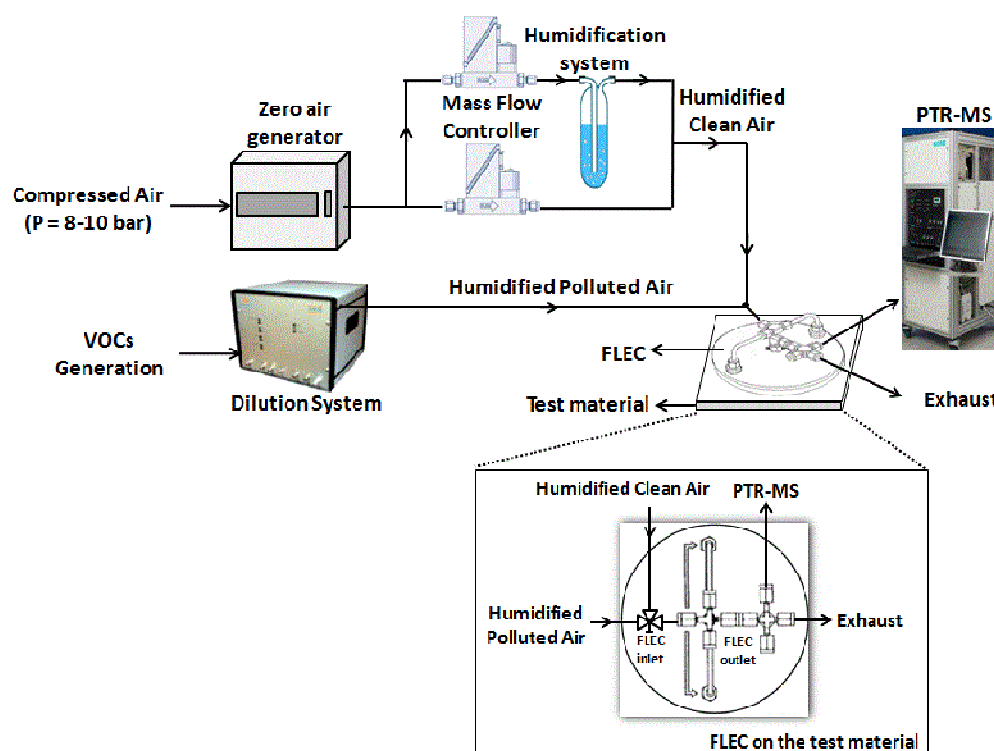
143 **2.2. *Test materials***

144 An unpainted 12.5-mm thick gypsum board and a 2.4-mm thick piece of vinyl flooring were
145 used. Several samples of each material were stored at 4°C for three months before the
146 experiments to ensure that replicate measurements are performed on samples having the same
147 aging history.

148 **2.3. *Experimental setup***

149 The setup used in this study is based on coupling a FLEC (*Chematec*) and a high resolution
150 PTR-MS (*PTR-ToFMS, Kore technology*) and presented in Figure 1. The FLEC inlet is
151 connected to two gas generation systems using a three-way valve. The first generation system
152 is composed of a dry zero air generator (*Claind*) and a humidificator made of a water bubbler
153 and mass flow controllers (*MKS*). This system is used to supply the FLEC with humid clean
154 air at constant flow rate (200-500 ml.min⁻¹) and stable relative humidity (50±5 % at 23±2°C).
155 The second generation system is made of a VOC cylinder connected to a dilution system (*Gas
156 Calibration Units - Ionicon Analytik*), which is used to dilute the VOC mixture at a constant

157 relative humidity of 50%. The FLEC's outlet is connected to the PTR-MS to quantify VOC
 158 concentrations exiting the cell. An exhaust is left at atmospheric pressure to prevent a pressure
 159 build-up in the FLEC apparatus.



160
 161 **Figure 1. Experimental setup for measuring adsorption and desorption coefficients of VOCs on**
 162 **building materials.**

163 The FLEC can be easily handled and fixed on surfaces and the PTR-MS instrument was
 164 designed for easy and safe transportation. In fact, the deployment of PTRMS instruments has
 165 been demonstrated during field campaigns in ambient air [31] and for indoor studies [32].
 166 This setup exhibits reasonable dimensions to be used during intensive field campaigns for
 167 research purposes in buildings but is not proposed as a tool for quick indoor air diagnostics
 168 due to its high price and high weight.

169 **FLEC.** The FLEC[®] is a portable tool commercialized by CHEMATEC, which can be
 170 exposed on flat and non-porous surfaces to investigate gas-surface interactions. Technical
 171 details have been provided elsewhere [33]. When the cell is exposed on a flat material, it
 172 covers a surface area of 177 cm², leading to a loading factor of 506 m².m⁻³. It has a small
 173 internal volume (35 mL), which allows conducting fast sorption experiments (a few hours)
 174 compared to laboratory emission test chambers of several liters (a few days).

175 **PTR-MS.** The PTR-MS was chosen as a fast analytical tool to accurately monitor the
 176 fast variations of VOC concentrations at the sub ppb level [34] with time resolutions in the
 177 range of 2-20 seconds. As shown in the result section, the measurement time resolution is a
 178 critical parameter to correctly describe fast sorption processes taking place at the material
 179 surface inside the FLEC apparatus. The PTR-MS technique has been described in detail
 180 elsewhere [34]. As proposed by De Gouw [34], signals of protonated VOCs are normalized to
 181 H₃O⁺ and H₃O⁺(H₂O) to correct for a small humidity dependence of the PTR-MS response.

182 $\text{H}_3\text{O}^+(\text{H}_2\text{O})$ is an ionic cluster that is formed when humid samples are analyzed. Detected
183 VOC signals are then converted to concentrations ($\mu\text{g}\cdot\text{m}^{-3}$) using calibration coefficients
184 determined experimentally for each targeted species. In the following, ethylbenzene and o/p-
185 xylene are referred as C8-aromatics due to their detection as a sum of isobaric compounds
186 with the PTR-MS. Calibration coefficients were measured before each experiment using zero
187 air and a standard mixture of VOCs provided by IONICON. Response factors measured over
188 a period of 3 months indicate a good stability of the PTR-MS instrument with a relative
189 standard deviation for each species in the range 9-10%. In addition, the stability of the PTR-
190 MS response over the time of a sorption experiment (only a few hours) is estimated to be
191 better than 3% from differences observed between subsequent calibrations performed every 2-
192 3 days. VOCs background signals measured using zero air were used to estimate detection
193 limits (LOD) as three times the standard deviation on the zero measurements. Measured
194 LODs are less than 3.4, 4.7 and $11 \mu\text{g m}^{-3}$ for a time resolution of 20, 10 and 2 seconds
195 respectively and which is low enough to measure accurately the concentrations used in this
196 study (See Table 1- [15]).

197 **2.4. Experimental protocols**

198 First of all, a blank experiment is carried out on a Pyrex glass before each sorption experiment
199 on a tested material and using the same procedure. This experiment, referred as “No sink” in
200 the following, allows evaluating sorption processes on internal surfaces of the FLEC
201 apparatus and the Teflon tubing. A sorption experiment involves a 3-step procedure as
202 described in the following. The FLEC is first exposed on a material and supplied with
203 humidified zero air. When concentrations reach relatively steady state, the FLEC is supplied
204 with humid air containing targeted VOCs. This second phase is named “adsorption phase” as
205 illustrated in Figure 2. During this step, VOCs concentrations increase until an equilibrium is
206 reached where the concentrations are equal to those registered for the adsorption phase
207 already performed on the Pyrex glass. Once the VOCs concentrations are stable (Figure 2:
208 t_{eq}), humidified zero air is provided to the cell instead of the VOCs mixture. This third phase
209 is named “desorption phase”. During this step, VOC concentrations decrease until steady
210 concentrations similar to those reached in the end of the first phase, are observed (Figure 2).
211 Standard operating conditions were defined as an air temperature of $23 \pm 2^\circ\text{C}$ and a relative
212 humidity of $50 \pm 5 \%$ for all experiments described in this study.

213 The feasibility of measuring sorption parameters with the FLEC/PTR-MS coupling was first
214 investigated by repeating the same experiment on a gypsum board and a piece of vinyl
215 flooring (FLEC inlet flow rate = $400 \text{ mL}\cdot\text{min}^{-1}$; PTR-MS time resolution = 20 s). The VOCs
216 concentrations used for the adsorption phase were closed to $565 \mu\text{g}\cdot\text{m}^{-3}$ for toluene, $434 \mu\text{g}\cdot\text{m}^{-3}$
217 for C8 aromatics, and $160 \mu\text{g}\cdot\text{m}^{-3}$ for benzene. The VOC steady-state-concentrations
218 measured before the sorption experiment by exposing the material to humidify zero air, are
219 negligible and represent less than 3% of C_e . A second set of experiments was carried out on
220 the gypsum board to test the robustness of this method. It consisted in assessing (1) the
221 repeatability on 5 different samples (of the same board); (2) the repeatability of 4 tests
222 performed consecutively on the same sample; (3) the influence of VOCs concentrations

223 (threefold variation); and (4) the influence of different inlet flow rates (threefold variation)
 224 keeping VOCs concentrations unchanged. Experimental conditions are listed in Table 1.

225 **Table 1. Experimental conditions used to perform tests of robustness on an unpainted gypsum**
 226 **board at a PTR-MS time resolution of 20sec.**

Test	Flow rate (mL.min ⁻¹)			Air exchange rate (h ⁻¹)			Specific flow rate per unit of surface [m ³ /(h.m ²)]			Concentration (µg.m ⁻³)		
										Benzene	C8 aromatics	Toluene
R (n=5)	300			514			1.0			106	289	377
M (n=4)	300			514			1.4			106	289	377
F (n=3)	300	400	500	514	686	857	1.0	1.4	1.7	106	289	377
C (n=3)	400			686			1.4			479	868	1131
										319	434	565
										106	289	377

227 *n*: number of experiments; *R*= Repeatability tests performed on different samples of the same board; *M*=
 228 Multiple successive measurements (replicates on the same sample); *F*= Inlet flow rate tests performed using
 229 three different air flow and *C* = Concentrations tests performed using three different concentrations.

230 3. Theory and data treatment

231 The data analysis yields values for C_{se} the mass of VOCs in the sink per unit area of the
 232 material at equilibrium ($\mu\text{g.m}^{-2}$), k_a the adsorption rate constant (m.h^{-1}), k_d the desorption rate
 233 constant (h^{-1}) and K_e (k_a/k_d) the equilibrium constant (m) described in the following.

234 3.1. Determination of C_s

235 Based on the experimental procedure described above, the mass of VOCs adsorbed on the
 236 material is in steady state at time t_{eq} . The total mass adsorbed or desorbed by a material
 237 corresponds to the area between the sorption curves (FLEC exposed on a material) and the
 238 “no sink” curve (FLEC exposed on pyrex), for the adsorption and the desorption phases
 239 respectively (Figure 2).

240 Equations (1) and (2) were used to calculate the absorbed and the desorbed mass of VOC
 241 respectively.

$$242 \quad C_{se} = \int_{t_0}^{t_{eq}} \frac{C_g \times F}{A} dt \quad (1)$$

$$243 \quad C_{se} = \int_{t_{eq}}^{t_f} \frac{C_g \times F}{A} dt \quad (2)$$

244 where, F is the flow rate at the FLEC's inlet ($\text{m}^3.\text{h}^{-1}$), A the surface area of the material
 245 covered by the cell ($177 \times 10^{-4} \text{ m}^2$), and t the exposure time to the VOCs mixture (h).

246 3.2. Determination of k_a and k_d

247 The model used to derive sorption parameters from experimental data is the Tichenor model
 248 based on the Langmuir theory [7]. This model is based on the assumption that the
 249 concentration of a species in bulk air is homogeneous and proportional to the surface
 250 concentration. However, this model neglects the diffusion (D_m) inside the material and the
 251 mass transfer coefficient (h_m) between the surface of the material and the bulk air. Despite
 252 these limits, this model is suitable for this work to determine k_a and k_d coefficients, for the
 253 reasons detailed below. Zhang [35] studied the flow field in the FLEC cavity and calculated
 254 the local Sherwood number Sh_L for the different flow rates (186 to 509 mL.min⁻¹). According
 255 to this work, the mass transfer coefficient was calculated in the FLEC cavity for the different
 256 flow rates used in Table 1 (See Figure 1 in [36]). The flow field in the FLEC is laminar
 257 because the Reynolds number Re varies between 1.4 and 2.3 for 316 mL/min and 509 mL/min
 258 respectively ($Re < 2000$ for laminar flow). The mass transfer coefficient is very high on the
 259 FLEC periphery and decreases in the center to reach a value varying between 1.4 and 2.3
 260 m.h⁻¹. The mass transfer coefficient h_m , is always higher than the specific flow rate per unit of
 261 surface (Table 1) which means that the that any substance present on the surface of the
 262 material is immediately diffused in the transverse direction of the flow by molecular diffusion
 263 as already highlighted by Zhu [37]. Therefore, the effect of the mass transfer coefficient is not
 264 significant on the concentrations in the FLEC cavity and the concentration measured in the
 265 FLEC is considered uniform in the entire cavity (C_g) as Zhang [35] confirmed previously by
 266 conducting experimental and numerical studies of the fluid flow and the convective mass
 267 transfer coefficients in a FLEC. They concluded that the air becomes nearly saturated shortly
 268 after it begins to flow on the emission surface, due to the small spacing between the cap and
 269 the bottom surface. Concerning the diffusion in the inner of the material, this parameter is
 270 neglected in this work and supposed to be slower than the instantaneous adsorption on the
 271 surface of the material, since the FLEC-PTRMS method is dedicated to fast in-situ
 272 measurements and the INCA-indoor model neglects the diffusion in the inner of the material.
 273 It worth to note that further works will use a model that considers the diffusion coefficient in
 274 the inner of the material to determine the diffusion coefficient and this point will be discussed
 275 later in this paper.

276 Giving the reasons above, the concentration variations (dC_g/dt and dC_s/dt) observed in a
 277 chamber depends on the adsorption (k_a) and desorption (k_d) coefficients, as shown in the
 278 following equations:

$$279 \quad \frac{dC_g}{dt} = NC_{in} - NC_g - k_a C_g L + k_d C_s L \quad (3)$$

$$280 \quad \frac{dC_s}{dt} = k_a C_g - k_d C_s \quad (4)$$

281 where, N is the air exchange rate (h⁻¹), C_{in} the VOC concentration at the chamber inlet
 282 performed ($\mu\text{g.m}^{-3}$), C_g the gas-phase VOC concentration inside the chamber ($\mu\text{g.m}^{-3}$), k_a the
 283 adsorption rate constant (m.h⁻¹), k_d the desorption rate constant (h⁻¹), C_s the surface
 284 concentration ($\mu\text{g.m}^{-2}$), and L the loading factor ($\text{m}^2.\text{m}^{-3}$).

285 The model used to fit the experimental observations is characterized by equations (3) and (4).
 286 Analytical solutions given in equations (5) and (6) can be derived for the desorption phase
 287 using the following initial conditions at t_{eq} : $t = 0$; $C_g(0) = C_e$ and $C_s(0) = C_{se} = C_e(k_a/k_d)$.

$$288 \quad C_g(t) = \frac{C_e[(N - r_2)e^{-r_1 t} - (N - r_1)e^{-r_2 t}]}{r_1 - r_2} \quad (5)$$

$$289 \quad C_s(t) = \frac{C_e k_a [r_1 e^{-r_2 t} - r_2 e^{-r_1 t}]}{k_d (r_1 - r_2)} \quad (6)$$

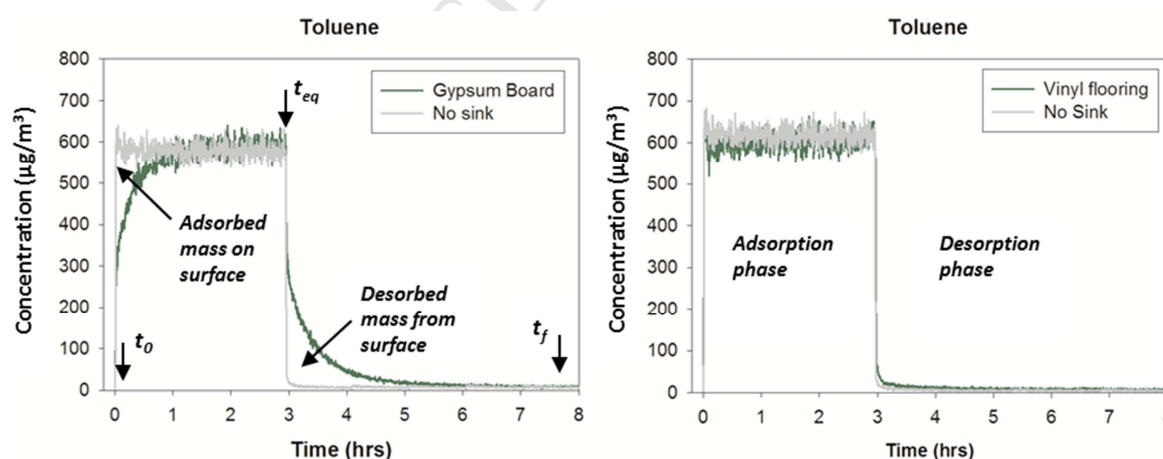
$$290 \quad \text{where, } r_{1,2} = \frac{(N + k_a L + k_d) \pm [(N + k_a L + k_d)^2 - 4Nk_d]^{1/2}}{2}$$

291 To determine k_a and k_d , the concentration time profiles $C_g(t)$ measured from t_{eq} to the end of
 292 the test (desorption phase) is fitted using equation (5) and a non-linear least square regression
 293 procedure in SigmaPlotTM. C_e is the average concentration observed in the gas-phase at the
 294 end of the adsorption phase. N , L and C_e are experimental parameters so that they are
 295 constrained during the fit.

296 4. Results

297 4.1. Technical feasibility

298 Time-resolved concentration profiles obtained for toluene are shown in Figure 2 for two
 299 experiments performed on a gypsum board and a piece of vinyl flooring. Concentration
 300 profiles of other compounds are given elsewhere (see Figure 2 in [15]) and exhibit the same
 301 behavior.

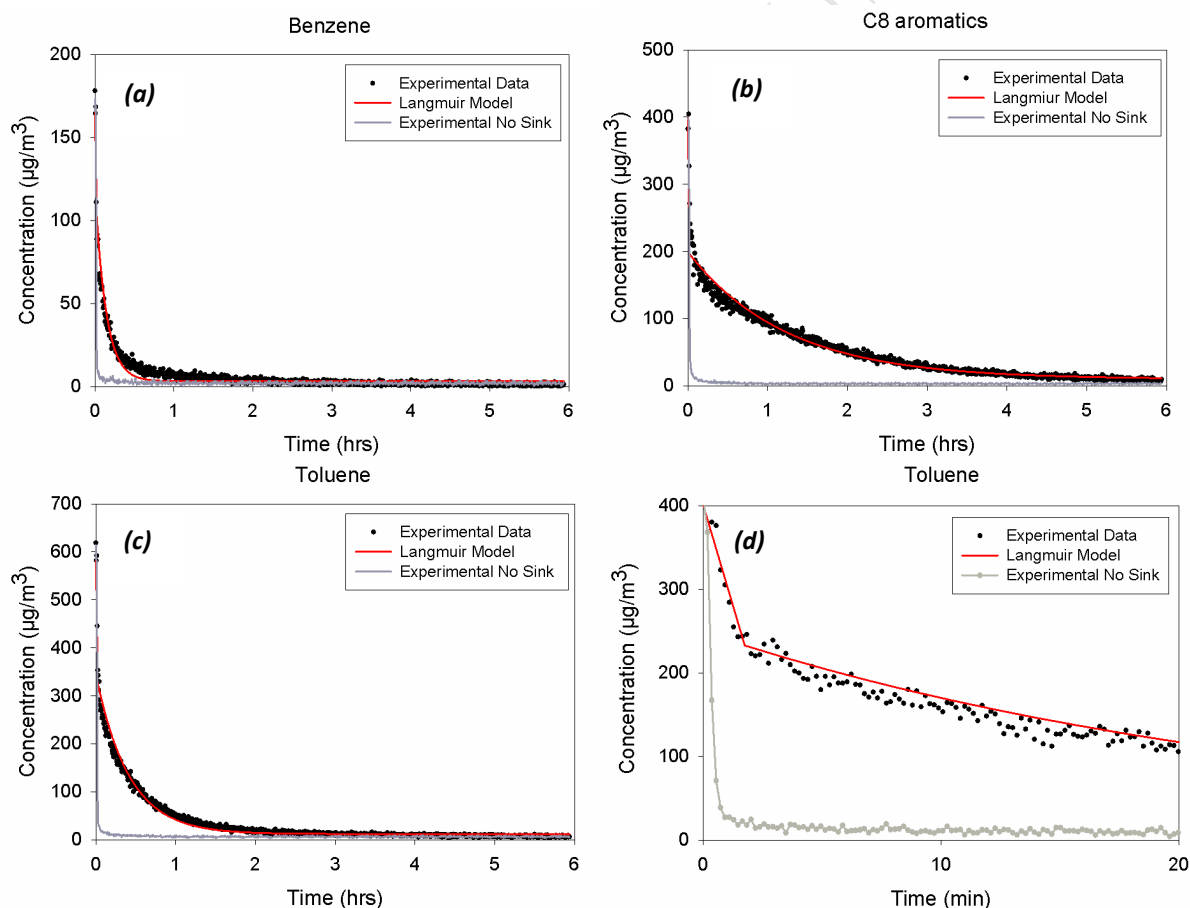


302
 303 **Figure 2. Concentration profiles measured during sorption experiments on a gypsum board and a**
 304 **piece of vinyl flooring for benzene, C8 aromatics and toluene. The “no sink” plot is also shown for**
 305 **comparison.**

306 The concentration profiles indicate that the duration of a sorption experiment is about (0.5-8
 307 hours), which is faster than that observed for regular chamber experiments, usually few days
 308 [7, 18]. The experimental no sink profile obtained using Pyrex glass indicate that VOC
 309 sorption on the FLEC internal walls is negligible since no difference can be observed with the

310 theoretical no sink profile obtained according to the equation $C_g(t) = C_e e^{-Nt}$. In addition,
 311 these results figure out different sorption behaviors for the 2 materials as seen from the total
 312 mass adsorbed on the material at equilibrium and the time needed to reach this equilibrium.

313 Similar concentration profiles are observed for vinyl flooring and pyrex, suggesting low
 314 sorption properties for this material. As a consequence, experiments discussed below to
 315 characterize the proposed methodology were only performed using the gypsum board, which
 316 exhibit significant sorption properties. Values of k_a and k_d were evaluated for the gypsum
 317 board by fitting Equation (4) to concentration profiles acquired during the desorption phase as
 318 described in the theory and data treatment section. Obtained values for k_a and k_d are
 319 respectively (3.2 m.h^{-1} - 24 h^{-1}) for benzene, (1.3 m.h^{-1} - 1.6 h^{-1}) for C8 aromatics and (1.7 m.h^{-1} -
 320 5.5 h^{-1}) for toluene. The modeled curves shown in Figure 3 are in good agreement with
 321 experimental observations because the correlation coefficient R^2 is very close to 1 (0.96 for
 322 benzene, 0.97 for C8 aromatics and 0.97 for toluene). This good agreement is also observed
 323 for the first minutes of the experiments (Figure 3 (d)) contrary to some previous studies made
 324 using emission chambers [7, 38].



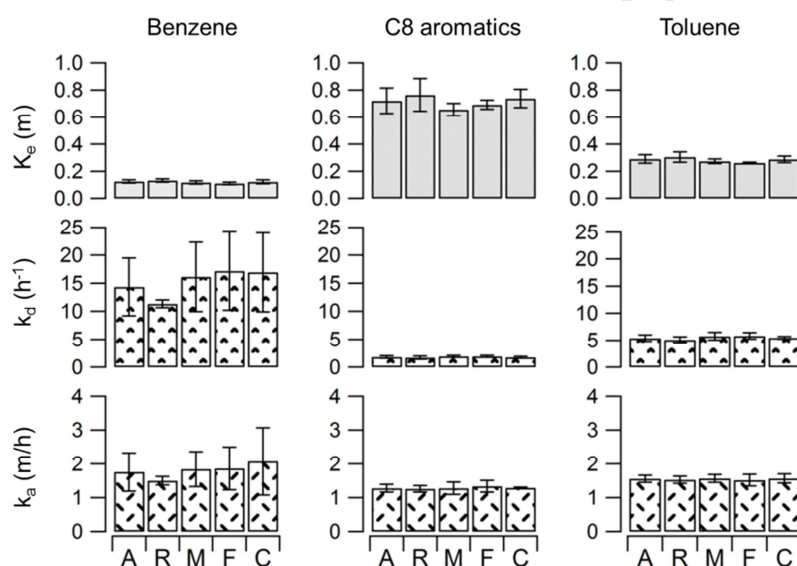
325

326

327 **Figure 3. Analysis of the desorption phase for the gypsum board: Benzene (a), C8 aromatics (b),**
 328 **and toluene (c, d). The “no sink” curve is also shown in grey for comparison. The first minutes of**
 329 **the toluene decay are expended in panel (d) to evaluate the “best-fit” curve.**

330 4.2. Robustness

331 Figure 4 shows mean values of k_a , k_d and K_e measured for the targeted VOCs for the Gypsum
 332 board and using experimental conditions described above in Table 1. The first set of
 333 experiments made on different samples of the same gypsum board (R in Figure 4) indicates a
 334 good repeatability for the 5 tests, with RSD (1σ) values lower than 17% for k_a , k_d , and K_e .
 335 Four adsorption/desorption cycles (M in Figure 4) made successively on the same sample also
 336 show a good repeatability for K_e , k_a , and k_d , with RSD values lower than 12%, except for
 337 benzene for which the RSD reaches 38% for k_d . The high uncertainty seen on the
 338 determination of benzene sorption parameters is due to the fact that the benzene profile is
 339 close to that obtained on pyrex glass, having negligible sorption effect. The uncertainty on the
 340 determination of low sorption values will be discussed later in this work. Tests performed at
 341 different VOCs concentrations (C in Figure 4), as well as different inlet flow rates (F in
 342 Figure 4), give consistent results for sorption parameters with RSD values lower than 12% for
 343 C8 aromatics and toluene but around 40% for benzene. These results highlight that the
 344 determination of sorption coefficients does not depend on operating conditions such as the
 345 VOCs concentration used, the inlet flow rate, and the number of measurements made
 346 previously on the investigated material.



347

348 *Figure 4. Average results for k_a , k_d and K_e measured on the gypsum board using a mixture of*
 349 *aromatic compounds (A=All experiments; R=Repeatability experiments; M = Multiples adsorption-*
 350 *desorption experiments on the same piece of material; F = different air flow rates at the FLEC*
 351 *inlet; C = different concentrations of VOCs). Error bars correspond to 1 standard deviation.*

352 5. Discussion

353 5.1. Method evaluation

354 Based on the results shown above, the FLEC/PTR-MS coupling appears as a promising setup
 355 to measure sorption coefficients of VOCs on building materials within a few hours (1-16
 356 hours). Compared to common test chamber experiments usually conducted over 27-600 hours
 357 (i.e. over 1-24 days) [18-20, 38, 39], the experimental time is reduced by a factor of 54-75.
 358 This method exhibits also a good repeatability and the determination of sorption coefficients
 359 is independent on gas concentration or the number of tests investigated on the same sample.

360 In addition, derived parameters are independent on flow rates conditions, which validated the
 361 choice of the Langmuir model to extract sorption parameters from experiments performed
 362 using an enclosure where the gas-phase concentration above the material is homogenous
 363 under the conditions of this study.

364 The applicability of the Langmuir isotherm was verified by investigating the partitioning of
 365 VOCs between the gas and adsorbed phases when the equilibrium is reached. The relationship
 366 between C_{se} (calculated from equations 1 and 2) and C_e (experimental measurements) for the
 367 Gypsum board and target VOCs, shows a linear trend with a correlation coefficient of 0.92,
 368 0.80 and 0.94 respectively for benzene, C8 aromatics and toluene (See Figure 3 in [15]).
 369 According to the relation ($C_{se}=C_e \times K_e$), the slope of the regression line should be equal to K_e .
 370 The slope determined for benzene (0.16), C8 aromatics (0.66) and toluene (0.29) is in
 371 excellent agreement with the average K_e value derived from 15 measurements presented in
 372 Figure 4. This linear relationship confirms that Langmuir equilibrium can be applied at the
 373 concentrations used in this study ($106\text{-}1131 \mu\text{g}\cdot\text{m}^{-3}$).

374 In order to test the robustness of the fitting procedure, multiple fits have been performed for
 375 several sorption experiments by varying the end-time of desorption phase (Table 2). All
 376 desorption phases were performed for 450 minutes and then were analyzed for three different
 377 time durations of 450, 300, and 200 minutes.

378 *Table 2. Comparison between k_a and k_d values derived from the fitting procedure applied to the*
 379 *gypsum board. All experimental measurements were made for duration of 450 minutes.*
 380 *Mathematical fits were performed using an end-time set at 450, 300, and 200 minutes of the*
 381 *desorption phase. Errors correspond to 1σ standard deviation. Number of replicates = 5.*

Time of desorption phase (min)	Benzene		C8 aromatics		Toluene	
	k_a (m.h ⁻¹)	k_d (h ⁻¹)	k_a (m.h ⁻¹)	k_d (h ⁻¹)	k_a (m.h ⁻¹)	k_d (h ⁻¹)
450	1.8±0.57	16±5.2	1.3±0.11	2,0±0.24	1.6±0.11	6.2±0.59
300	1.6±0.16	12±1.1	1.3±0.10	1.8±0.32	1.6±0.05	5.2±0.35
200	1.7±0.24	15±3.4	1.2±0.13	2.1±0.35	1.6±0.04	5.8±0.69

382
 383 This test indicates that determinations of k_a and k_d are robust for these 3 experimental
 384 durations. Deviations between individual calculated values and mean values (of all sorption
 385 experiments) are within the experimental RSD displayed in Figure 4 (10-17 %). This result
 386 also indicates that the time of a sorption experiment, including adsorption and desorption
 387 phases, can be adjusted to less than 7 hours for this type of material.

388 5.2. Limitations of applicability

389 While the setup described in this study is suitable for fast measurements of sorption
 390 coefficients in laboratory and indoor environments, this setup exhibits some drawbacks. On
 391 the first hand, it is important to avoid high leakage between the cell and the material. For
 392 homogenous and flat materials, the cell can be directly exposed on the surface. However,
 393 porous or rough surfaces cannot be sampled directly and need to be introduced in a sub unit
 394 (cylindrical container) made of aluminum, with the FLEC exposed on the top of the container.
 395 Even if this method is destructive in the case of porous materials, it still exhibits the

396 advantage of reducing the measurement time compared to test chambers. In addition,
 397 measurements of sorption coefficients require a fast analytical tool such as the PTR-MS
 398 instrument used in this study. In addition, PTR-MS instruments can only measure a limited
 399 number of VOCs present in indoor environments and some species such as formaldehyde
 400 cannot be measured due to a proton affinity that is only slightly higher than the water proton
 401 affinity [34]. Further studies should focus on coupling the FLEC apparatus to other fast
 402 analyzers, such as a formaldehyde analyzer to study interactions of this important indoor
 403 pollutant with indoor surfaces. Fast analytical tools that are currently under development in
 404 several laboratories, designed to measure different compounds, may offer a more compact and
 405 lighter alternative than the PTR-MS.

406 On the other hand, the Langmuir model used to extract sorption parameters neglects the
 407 diffusion inside material which is an important key parameter for indoor air quality models.
 408 Nevertheless, Jorgensen [40] used previously the Langmuir model and a diffusion model to
 409 analyze sorption experiments performed in emission test chambers. They found that the
 410 adsorption and desorption coefficients still unchanged between the two models even if the
 411 diffusion models determined a supplementary diffusion coefficients. Further works will be
 412 conducted to analyze experimental data obtained by the FLEC-PTRMS method, using a
 413 model that accounts for the diffusion in the inner of the material, to determine the VOCs
 414 diffusion coefficient.

415 In order to determine the lower values of sorption parameters that could be measured with this
 416 method, measurements made on a piece of vinyl flooring exhibiting sorption properties close
 417 to that observed for a Pyrex glass, were investigated at different PTR-MS time resolutions.
 418 Indeed, an adsorption experiment made at a time resolution of 20 seconds showed that it was
 419 not possible to extract sorption coefficients from the desorption phase using equation (5) due
 420 to a fast decrease of VOCs concentrations (over a few minutes). Additional experiments were
 421 performed at resolution times of 10 and 2 seconds. These experiments showed that k_a and k_d
 422 values could only be extracted from measurements performed at the fastest time resolution of
 423 2 seconds (Table 3).

424 *Table 3. Average values of k_a , k_d , and K_e for a piece of vinyl flooring at a PTR-MS time resolution of*
 425 *2 seconds. Number of replicates = 5.*

VOC	Toluene			C8 aromatics			Benzene		
Sorption parameters	k_a (m.h ⁻¹)	k_d (h ⁻¹)	K_e (m)	k_a (m.h ⁻¹)	k_d (h ⁻¹)	K_e (m)	k_a (m.h ⁻¹)	k_d (h ⁻¹)	K_e (m)
Average	0,06	11	0,01	0,18	17	0,01	0,03	8,3	0,01
Std	0,02	5,2	0,01	0,03	5,2	0,01	0,02	9,0	0,01
RSD (%)	28	46	26	18	30	16	57	109	119

426 RSD values calculated from 5 replicates range from 15-110% and are higher than values
 427 determined for the gypsum board due to higher noise levels on the PTR-MS measurements at
 428 faster time resolutions (Table 3). These results highlight that large uncertainties are associated
 429 to the determination of weak sink effects. However, materials exhibiting very weak sink
 430 effects will have minor influences on the variation of VOCs concentrations in indoor
 431 environments.

432 As mentioned above, faster measurements lead to higher noise levels on measured signals,
 433 and thus lead to a degradation of the measurement precision. Therefore, the best conditions
 434 correspond to a compromise between temporal resolution and noise levels. To determine the
 435 best operating conditions of the proposed setup, a set of curves, characteristic of the
 436 desorption phase, was simulated at 3 different time resolution (2, 10 and 20 seconds) using
 437 equation (4), an air exchange rate of 514 h^{-1} and several values of adsorption (k_a) and
 438 desorption (k_d) coefficients. To reduce the number of couples for k_a and k_d , the same value
 439 was used for both parameters. The simulations were also performed using experimental values
 440 measured for the vinyl flooring at a PTR-MS time resolution of 2 seconds. Equation (4) was
 441 fitted on the simulated curves using Sigma Plot to check whether the mathematical fit is
 442 capable of retrieving the values of k_a and k_d used to generate the curves. The accuracy of the
 443 fit was calculated as the difference observed between values of k_a and k_d used to simulate the
 444 curves and values determined by fitting the curve as shown in Table 4.

445 **Table 4. Accuracy determined by simulation for low values of k_a and k_d for three PTR-MS temporal**
 446 **resolutions.**

Temporal resolution (sec)	k_a (m h^{-1})	k_d (h^{-1})	Accuracy k_a (%)	Accuracy k_d (%)
2	0.01	0.01	9.89	10.00
	0.02	0.02	4.71	5.00
	0.10	0.10	0.10	0.10
	0.01	0.10	0.001	0.60
	0.10	0.01	1.20	1.00
	0.03	17.00	0.33	0.24
10	0.07	0.07	6.22	8.71
	0.09	0.09	2.51	4.33
	0.10	0.10	1.63	3.00
20	0.09	0.09	5.39	10.11
	0.10	0.1	3.63	7.30
	0.20	0.20	0.02	0.95

447 An accuracy lower than 10% is considered as satisfactory to accurately determine k_a and k_d . A
 448 close inspection of Table 4 indicates that temporal resolutions of 20 and 10 seconds are fast
 449 enough to accurately measure adsorption and desorption parameters (k_a ; k_d) as low as
 450 (0.09m.h^{-1} ; 0.09h^{-1}) and (0.07m.h^{-1} ; 0.07h^{-1}), respectively. Faster measurements (2 seconds
 451 time resolution) are needed to determine lower values down to $k_a=0.01\text{m.h}^{-1}$ and $k_d=0.01\text{h}^{-1}$. It
 452 is important to note that these results only provide an estimation of the lower values of k_a and
 453 k_d that can be measured at a specific time resolution with the FLEC-PTRMS coupling since
 454 combinations of different values of k_a and k_d are usually observed. Experimental values
 455 derived for the vinyl flooring were also tested and they prove that more resolute
 456 measurements are necessary to accurately determine sorption coefficients in this range. In
 457 addition, these simulations indicated that the determination of low values of k_a is more
 458 impacted by the temporal resolution than k_d because the value of k_a is mainly determined by
 459 using the first minutes of the experimental measurements.

460 5.3. Literature comparison

461 Won [6] measured sorption coefficients for a gypsum board exposed to toluene and
 462 ethylbenzene. Tichenor et al. [7] also report sorption coefficients of toluene for the same type

463 of material. To the best of our knowledge, there is no measurement of sorption coefficients for
 464 benzene available in the literature. Sorption coefficients measured in this study are in the
 465 same order of magnitude of previous studies and show a difference by a factor of 2-3 times
 466 for K_e parameter (Table 5). However, a difference is also reported between previous works for
 467 the same type of material with ethylbenzene. For the vinyl flooring the difference is more
 468 important. This difference can be due to the composition of the tested material as reported by
 469 Won [41]. The aging of the surface, or its physicochemical properties may also introduce
 470 some variability in the results as observed by Meininghaus [42]. From the other hand, this
 471 difference can be due to the inappropriate use of the Tichenor model for emission test
 472 chambers used for sorption experiments. The effect of the mass transfer due to the presence of
 473 a boundary layer on the surface of the tested material can have an important effect on the
 474 determination of sorption parameters. The companion paper (part 2 of this work), will present
 475 a comparison using an emission test chamber and the same type of material and the same
 476 VOCs mixture. Therefore, different type of models will be used to determine sorption
 477 parameters with a consideration for the effect of the mass transfer coefficient and the effect of
 478 the boundary layer.

479 **Table 5. Comparison of literature values of K_e for a gypsum board with results from this**
 480 **study.**

VOCs	k_a (m.h ⁻¹)	k_d (h ⁻¹)	K_e (m)	References
Benzene	1.8 ± 0.57	14 ± 5.2	0.12 ± 0.01	This work
	0.21	1.7	0.12	[6]
Toluene	1.6 ± 0.11	5.4 ± 0.59	0.29 ± 0.03	This work
	0.21	0.87	0.24	[6]
Ethylbenzene	0.45	1.50	0.30	[7]
	1.3 ± 0.11	1.8 ± 0.24	0.72 ± 0.09	This work
C8 aromatics				

481 **6. Conclusions**

482 The proposed experimental approach, based on a FLEC-PTRMS coupling, has been used
 483 successfully to measure sorption coefficients of aromatic compounds on different types of
 484 materials. This method presents the advantage to be transportable and non-destructive method
 485 for flat and homogeneous material to perform fast measurements in real environments and
 486 under indoor conditions. Moreover, this new setup exhibits the advantage of reducing the
 487 measurement time to 0.5-8 hours compared to emission test chambers that require several
 488 days of measurements and allows measuring low sorption values. The FLEC/PTR-MS
 489 coupling allows measuring low sorption values and will be used for in-situ measurements in
 490 energy efficient buildings. Sorption coefficients determined on the field will be then used as
 491 inputs for a new indoor air quality model (INCA-indoor) that takes into account the boundary
 492 layer effect and the mass transfer coefficient, to predict VOCs concentrations in indoor
 493 environments (see companion paper). The FLEC-PTRMS method will be compared in the
 494 part 2 of this work, to the traditional emission test chamber method used previously in
 495 literature to measure sorption parameters only on the laboratory scale. Further works, will use
 496 a more complex model to take into account for the diffusion inside building materials.

497 **Acknowledgments**

498 The authors would like to thank the French Environment and Energy Management Agency
499 ADEME (Agence De l'Environnement et de la Maîtrise de l'Energie) for their financial support
500 in this study, through the MERMAID project (PRIMEQUAL Program).

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Highlights

- Development of a new methodology to measure *in-situ* VOCs sorption on building materials
- Evaluation of sorption parameters under real conditions within few hours
- Measurement of low sorption properties with 10% of accuracy
- Sorption parameters are useful to IAQ models to predict VOCs concentration indoors