1 Volatile Organic Compounds removal by means of a felt-based

2 living wall to improve indoor air quality

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12	
13	Abstract

14 Currently, the population spends most of the time in indoor environments, which makes 15 Indoor Air Quality (IAQ) very important for health and comfort. As vegetation can act as a 16 biofilter capturing air pollutants, this study aims to assess the effectiveness of a living wall 17 module in the removal of the Total Volatile Organic Compounds (TVOCs) for IAQ 18 improvement. An airtight glass chamber was used to release contaminants, monitoring the TVOCs both with the chamber empty (control) and with a small Fytotextile[®] living wall 19 module planted with Nephrolepis exaltata L. A substantial reduction of TVOCs was 20 21 observed when the living wall was inside the chamber. In few hours, TVOCs levels were 22 reduced below the recommended limit (following Spanish regulations). More tests are 23 recommended considering different plant species and other variables related to the IAQ.

Keywords: vertical greening systems; Indoor environment; TVOCs; n-Hexane;
 Nephrolepis exaltata; Fytotextile.

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27 **1. Introduction**

Atmospheric pollution is nowadays a serious problem in the world, having a negative impact on human health. Unfortunately, also the indoor atmosphere has worsened in the last years and it is not usually monitored. People spend much of their time indoors, which makes indoor air quality (IAQ) very important for their health and comfort. For instance, in north America it was estimated that adults spend 80-90% of their time inside a building (Wetzel and Doucette, 2015).

34 The sick building syndrome (SBS) describes a situation in which the occupants of a 35 building experience acute health- or comfort-related effects (Joshi, 2008), being the chemical 36 contaminants one of its main causes. Indoor air can be contaminated by many kinds of 37 pollutants from different sources, both anthropogenic (Qiu et al., 2014) or natural (Guenther, 2013). Among them, volatile organic compounds (VOCs) are major contributors of indoor 38 39 air pollution (Teiri et al., 2018). VOCs are emitted from a variety of different sources, like 40 paints, paint strippers, fuels, cleaning supplies, building materials or also from external 41 sources (e.g., vehicle exhausts) (Cheng et al., 2018; Joshi, 2008; Zhu et al., 2020). The World 42 Health Organization confirmed that some VOCs are carcinogenic, damage the nervous and 43 circulatory system (Zhu et al., 2020) and cause allergies, skin irritation and respiratory 44 diseases (Sahu et al., 2017). There are several VOCs commonly found in indoor 45 environments and each of them has different hazardous effects. For example, benzene is 46 confirmed as a human carcinogen while n-hexane, heptane and octane can affect the central 47 nervous system (Srivastava et al., 2000).

48 There are conventional methods for removing VOCs, including adsorption, thermal or 49 catalytic combustion, photocatalytic and biological methods. Currently, available techniques 50 prove to be inefficient for treating indoor air pollution at very low concentrations (Guieysse 51 et al., 2008). Among the methods usually employed to improve IAQ, ventilation is the most 52 common. However, it can have some drawbacks as the outdoor air used may also contain 53 contaminants which will be brought inside. The ventilation system itself could also be a 54 significant source of emissions (EPA, n.d.). Additionally, the energy costs will rise due to the 55 higher air conditioning needs required to heat or cool the intake air. Conversely, there are 56 suitable biological technologies with the potential to detoxify organic compounds, such as 57 some plants that efficiently and cost-effectively remove contaminants from the air (Sriprapat 58 and Thiravetyan, 2013).

59 Many studies confirmed that ornamental plants are able to accumulate VOCs and reduce 60 their concentration in the indoor environments (Aydogan and Montoya, 2011; Hörmann et 61 al., 2017; Liu et al., 2007; Parseh et al., 2018; Soreanu et al., 2013; Teiri et al., 2018; Wetzel 62 and Doucette, 2015; Yang et al., 2009). For example, ivy (Hedera helix L.) is considered as a 63 bioprotective species (Sternberg et al., 2010) and is capable of absorbing atmospheric particles, 64 as well as certain species of Nephrolepis, Epipremnum and Spathiphyllum genus(Wolverton 65 and Wolverton, 1993). An hydroculture of Syngonium podophyllum was also able to remove 66 indoor VOCs (Irga et al., 2013). Plants uptake VOCs by stomatal absorption into the 67 mesophyll and by sorption to leaves and plant growth media (Keymeulen et al., 1997; Orwell 68 et al., 2004). The cuticle adsorbs VOCs as dry or wet deposition and stomatal conductance 69 and certain metabolic processes affect VOCs exchange (Seco et al., 2007). Also rhizosphere 70 microbial activity plays an important role to reduce VOC, and some experiments 71 demonstrated that it was an important mechanism for VOC reduction (Kim et al., 2008; 72 Orwell et al., 2004; Wood et al., 2002).

The use of vegetation as a means of improving IAQ is, at the same time, a solution to the greater energy consumption of other devices and techniques of air purification, in addition to the psychological effect and other benefits that the presence of vegetation has on the building users.

77 However, the use of enough vegetation indoors in order to produce a significant result 78 in improving indoor air quality (IAQ) is not always easy due to the availability of space. For 79 this reason, building integrated vegetation systems such as living walls (Pérez-Urrestarazu et 80 al., 2015) offer a solution. Living walls are greening systems that allow plants to root all over 81 a vertical surface. Therefore, they enable the possibility of introducing more vegetation 82 indoors, using the walls to grow more plants in less space with a high density and including 83 different species (Gunawardena and Steemers, 2019). There are different systems and 84 construction methods (Radić et al., 2019), being the felt-based systems one of the most 85 employed.

Living walls offer many benefits, such as aesthetical, noise reduction or thermal performance improvement (Ghazalli et al., 2019), but the relation between living walls and IAQ improvement through phytoremediation has received an increased interest recently (Irga et al., 2018). For instance, it has been proved than living walls contribute to remove particulate matter (Ottelé et al., 2010). Several studies proved the ability of living walls to reduce VOCs concentration, though it was evaluated in a single pass (Mikkonen et al., 2018; Pettit et al., 2019, 2018; Torpy et al., 2018).

93 The aim of this work was to investigate the potential of a living wall module to remove 94 volatile organic compounds (measured as TVOCs) during extended exposure for IAQ 95 improvement.

96

97 2. Materials and Methods

For this study, the contaminants were released in a sealed glass chamber with 0.128 m³ of volume (0.8 m length; 0.4 m width; 0.4 m height) and their concentration was monitored both with the chamber empty (control) and with the living wall inside (Figure 1). A felt-based system (Fytotextile[®], Terapia Urbana, S.L., Seville, Spain) (Pérez-Urrestarazu et al., 2019) was used to build the living wall. Two small modules, 0.49 m width by 0.36 m height, were specifically prepared for the experiment, forming a grid of 2 by 3 pockets in which the plants were inserted. The living wall modules displaced a volume of 0.029 m³ inside the chamber.



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106 Figure 1. Photograph showing the experimental setup: (1) Fytotextile; (2) Nephrolepis 107 exaltata; (3) Temperature sensor; (4) TVOCs sensor; (5) portable fan; (6) grow lamp. 108 The plant species selected for the present work was *Nephrolepis exaltata* L., known as the 109 sword fern or Boston fern. It is an ornamental herbaceous plant that has hairy leaves with 110 serrated margins and a deep green colour. It is suitable to indoor cultivation, so it is commonly 111 used in living walls. Three plants were planted in each module (two in the upper row and one 112 in the middle pocket of the lower row) by inserting the rootball (with a substrate composed by 113 a mixture of coconut fibre and peat) into the pockets of the Fytotextile modules. They were 114 maintained during eight months in order to ensure the complete development of the roots 115 throughout the felt of the living wall. As the development of the plants was different, so was 116 the total biomass of each of the two modules. Right after the test exposing the plants to the 117 contaminant, both fresh and dry weight were measured using an AH-300 precision scale (I.C.T,

S.L., La Rioja, Spain). The total leaf area (cm²·plant⁻¹) was determined by means of an LI3100 Leaf Area Meter (Li-Cor, Nebraska, USA).

120 There are a variety of analytical methods for monitoring VOCs. They can be measured 121 separately, or together as Total Volatile Organic Compounds (TVOCs). This means that a 122 single compound is considered to be representative of the VOC mixture (Mølhave et al., 123 1997). To monitor VOCs separately, several types of detectors can be used, such as the flame-124 ionization detector (FID), the photo-ionization detector (PID), the electron capture detector 125 (ECD), the mass spectrometry detector (MSD), the photo-acoustic sensor (PAS), electronic 126 noses and others (Bicchi and Maffei, 2012). However, a simple, low-cost and portable option 127 is the use of calibrated gas sensors for TVOCs. These instruments are based on semi-selective electronic sensors that react chemically, optically, conductively or electrochemically to 128 recognize a gas pattern (Oh et al., 2011). In our case, a PCE-VOC 1 gas detector (PCE Ibérica 129 130 S.L., Albacete, Spain) was used for the direct measurement of Total Volatile Organic Compounds (TVOCs) (in a range from 0 to $10 \text{ mg} \cdot \text{m}^{-3}$). 131

Air temperature inside the chamber was also monitored by means of a HOBO Pro TempHR U23-001 (Onset Computer Corp., Bourne, Massachusetts, USA) sensor.

Direct-reading detectors are generally calibrated with one single compound (e.g. a hydrocarbon such as n-hexane or toluene), so the signal obtained from a mixture of VOCs is expressed as concentration equivalents of this compound (regardless of the composition of the mixture) (Mølhave et al., 1997). In this study, two different initial volumes (50 and 70 µl) of n-hexane, frequently detected indoors (Rösch et al., 2014), were used to increase the TVOCs level inside the chamber.

140 Tests without plants and different initial n-hexane concentrations were carried out before 141 introducing the living wall modules in order to check the airtightness of the chamber and record 142 the maximum level of TVOCs obtained for each concentration used. The duration of the tests 143 varied between 6 and 72 hours. In each one, the desired volume of n-hexane was introduced in 144 the chamber with a micropipette. Then, the chamber was closed and sealed to prevent any gas 145 interchange with the exterior. Three tests were performed for each of the scenarios considered: 146 empty chamber - 50 μ l, empty chamber - 70 μ l, module 1 - 50 μ l, module 1 - 70 μ l, module 2 147 - 70 μ l. At the end of each experiment, the chamber was opened and aerated in other to prepare 148 it for the next test.

The air inside the chamber was mixed by means of a small portable fan connected to an external battery. Artificial lighting was provided by a CF-UT01 LED Grow lamp (Panda Grow, Shenzhen, China), located over the chamber (facing the vegetation in an angle of 10° with respect to the horizontal position), with a 15-h light cycle. The vegetation received an average illuminance of 6828 Lux. The temperature range in which the tests were performed was 17.9-23.4 °C.

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156 **3. Results**

In the experiment without vegetation (empty chamber), the mean TVOCs level (average of all the values obtained in each of the three test for the same initial volume of contaminant) was 5.69 mg m⁻³ and 7.51 mg m⁻³ for 50 and 70 μ l of n-hexane, respectively (Table 1).

Table 1. Total volatile organic compounds (TVOCs, mean ± standard deviation) in the
empty chamber scenario with two different initial volumes of n-hexane.

n-hexane	Duration	$TVOC_0 (mg m^2)$	
(µl)	(h)	³)	TVOC _s (mg m ⁻³)
50	23.3	1.80	5.26 ± 0.17
50	7.67	1.90	6.28 ± 0.14

50	7.5	1.50	5.53 <u>+</u> 0.15
70	21.6	1.10	7.33 <u>+</u> 0.48
70	22	1.40	7.74 <u>+</u> 0.43
70	44.2	1.80	7.45 <u>+</u> 0.28

In the experiments with plants, initial TVOCs values (TVOC₀) before the introduction of the contaminant were in the range of 0.7 to 2.2 mg m⁻³ (Table 2). Once the n-hexane was released, the maximum TVOCs value (TVOC_{max}) did not reach the average value obtained for each initial volume of n-hexane in the empty-chamber tests. In fact, for a lower initial concentration of n-hexane, the average maximum TVOC value was 4.07 mg m³, while in the case of 70 μ l, it was 6.47 mg m³. This represented a difference of 1.62 and 1.04 mg m³ for 50 and 70 μ l of n-hexane, respectively.

Table 2. Results of the experiment with the living wall inside the chamber for twodifferent initial volumes of n-hexane and two values of biomass (module 1 and module 2).

										RR	
						TR	R _{1/4}	R_1	\mathbf{R}_3	(mg	
\mathbf{V}_0		D	$TVOC_0$	TVOC _{max}	TVOC _f	(mg	(mg m ⁻	(mg	(mg	m ⁻³	RE
(µl)	Mod	(h)	(mg m ⁻³)	(mg m ⁻³)	(mg m ⁻³)	m ⁻³)	3)	m ⁻³)	m ⁻³)	h ⁻¹)	(%)
50	1	6.2	1.7	3.9	3.4	0.5	0.3	0.4	0.7	0.08	12.8
50	1	23.3	1.4	4.3	2.7	1.6	0.9	1.0	1.1	0.07	37.2
50	1	70.8	1.3	4.0	0.5	3.5	0.7	0.6	0.5	0.05	87.5

¹⁶²

70	1	21.2	1.3	6.9	1.5	5.4	1.6	2.2	2.3	0.25	78.3
70	1	23.0	2.2	7.2	3.2	4.0	1.2	1.8	2.0	0.17	55.6
70	1	69.7	0.7	6.1	0.4	5.7	1.5	1.8	1.8	0.08	93.4
70	2	23.6	0.9	5.4	2.7	2.7	1.1	1.1	0.9	0.11	50.0
70	2	28.7	1.6	6.6	3.7	2.9	1.7	2.1	2.1	0.10	43.9
70	2	71.8	1.1	6.6	1.5	5.1	1.9	2.3	2.1	0.07	77.3

173 V₀: volume of n-hexane employed; Mod: number of the module used; D: duration of the test; TVOC: total
174 volatile organic compounds; TVOC₀: TVOCs concentration before adding the n-hexane; TVOC_{max}: maximum
175 TVOCs value after adding the n-hexane; TVOC_f: TVOCs value at the end of each test; TR: Total TVOCs
176 Reduction; R_{1/4}: TVOCs reduction after 15 min; R₁: TVOCs reduction after 1 h; R₃: TVOCs reduction after 3
177 h. RR: TVOCs reduction rate; RE: reduction efficiency.

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179 Obviously, the duration of the experiment was an important factor, as the highest 180 reductions were found in the longer tests (more than 5 mg m³ for tests 70-h long). However, 181 the TVOCs reduction rate was, in average, 0.07 mg m⁻³ h⁻¹ for lower initial concentrations, 182 being 0.17 and 0.1 mg m⁻³ h⁻¹ for 70 μ l (in module 1 and 2, respectively). 183 Table 3 shows the main parameters obtained in both living wall modules tested. Module

184 2 nearly doubled the fresh weight with respect to module 1, being the leaf area 2.6 times
185 higher in module 2.

186

187

188 Table 3. Total biomass (fresh and dry weight) and leaf area in each of the modules

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exposed to n-hexane

Plant characteristics Module 1 Module 2

Total fresh weight (g)	300.01	582.88
Aerial part	90.43	215.34
Roots	209.58	367.54
Total dry weight (g)	41.91	110.11
Aerial part	16.87	53.31
Roots	25.04	56.8
% Water	86.03	81.11
Aerial part	81.34	75.24
Roots	88.05	84.55
Fresh weight per plant (g)	108.88±43	214.13±94
Total leaf area (cm ²)	3070.6	8043.7
Mean area per leaf (cm ²)	35.7	69.3
Number of leaves	86	116

Figure 2 shows the evolution of TVOCs in the first 5 hours for three scenarios: (a) Module 1- 50 μ l, (b) Module 1- 70 μ l, (c) Module 2- 70 μ l. In all the cases, the highest reduction occurs in the first 15 minutes, with a very fast reduction response. TVOCs levels continue going down at a lesser rate until approximately 30 minutes after the release of the contaminant. Afterwards, TVOCs levels stabilize or diminish in a much slower rate.





Figure 2. Evolution of TVOCs (first six hours) for the three replications in each scenario: a)
50 µl with module 1. b) 70 µl with module 1. c) 70 µl with module 2. Each of the
replications is represented with a different marker type. The horizontal orange line denotes
the baseline (average level TVOCs in the corresponding empty chamber) scenario.

4. Discussion and conclusions

The results presented in this work show how the vegetation of a living wall can help to improve IAQ by removing VOCs. Several authors have suggested that no harmful byproducts are produced with the plant uptake and degradation of VOCs, just CO2, organic and amino acids (Kim et al., 2008; Soreanu et al., 2013). According to Kvesitadze et al. (2009), contaminants degradation proceeds to standard cell metabolites or mineralization. The plant cells not only avoid their toxic action but also utilize its carbon, nitrogen, and other atoms for intracellular biosynthetic and energetic needs.

211 Other researchers reached similar results, though using different substances as 212 contaminants and working with different plant species (Abdo et al., 2019; Mikkonen et al., 213 2018; Pettit et al., 2019, 2018). For example, Pettit et al. (2018) carried out an experiment 214 involving a small living wall module planted with Nephrolepis exaltata with different 215 substrates and obtained a removal efficiency (which they defined as the difference between 216 air pollutant concentration within the duct with and without the application of the biofilter 217 treatments) up to 87 % (using ethyl acetate as VOC contaminant) and 60 % (using benzene) 218 in a single pass of air. In our case, the air was in contact with the plant during a long period, 219 so the resulting removal efficiency was in accordance to the total duration of the test and the 220 initial concentration of pollutants, ranging from low values (13%) for short periods and low concentration to much higher values (77-93 % after 70 hours). Therefore, the time during 221 222 which there is contact between the air and the plants is clearly a key factor, obtaining higher 223 VOCs reductions in longer periods.

The initial level of contaminants is also an important variable, since the greater reductions were observed when the maximum TVOCs value at the beginning of the test was higher. On the other hand, results showed that the total plant biomass of the living wall module did not have the expected influence in the TVOCs reduction obtained. In fact, for similar test durations, the living wall module with a higher biomass of plants (module 2)
entailed lower reductions (an average of 57.1 % in reduction efficiency) than the one with a
lower plant development (module 1) (showing an average reduction efficiency of 75.8 %).

Spanish regulations (UNE 171330-2:2014) consider as 'discomfort range' the TVOCs values between 3 and 25 mg m⁻³. In all the test performed, initial TVOCs levels were over 3 mg m⁻³, but the final TVOCs values were below the suggested threshold or very close to it, confirming the effectiveness of the living wall used to increase IAQ.

235 The improvement of the IAQ also entails a potential economic impact, though it is very 236 difficult to quantify because the methodologies and models used for the estimation are not 237 completely reliable and well developed (Clausen et al., 2003). When measuring the costs of 238 indoor air pollution, it is important to look further than just the main effects on health (Duflo 239 et al., 2008). If the reduced working efficiency or job-related productivity losses are 240 computed, the economic impact increases (Brooks and Davis, 1992). Brooks and Davis 241 (1992) estimated that the annual per-employee productivity loss attributed to IAQ problems in the United States was 3 % (14 minutes day⁻¹), plus an added 0.6 % because sick days. Also, 242 243 in the United States, the yearly potential productivity increase due to the reduction of 244 respiratory infection cases was projected to result in US\$ 7–23 billion, while a reduction of 245 sick building syndromes (SBS) could yield around US\$ 10-20 billion. The impact of an 246 improved working efficiency was estimated in US\$ 12-125 billion (Fisk and Rosenfeld, 247 1997).

The use of indoor living walls as a mean for improving IAQ can reduce or complement the ventilation requirements of buildings (Pérez-Urrestarazu et al., 2016), hence decreasing the costs (energy and maintenance) associated to air conditioning. Therefore, the economic impact of a living wall as a measure to enhance IAQ would be higher (even considering installation and maintenance costs) that the obtained with ventilation. It is important to note that living walls entail other benefits (e.g., aesthetical, psychological and environmental
(Pérez-Urrestarazu et al., 2017, 2015)) which also influence the economic value of these
greening systems.

256 In this kind of studies, it is difficult to generalize the results since they depend on many 257 different factors such as plant species, temperature, light intensity, growing media, and VOC 258 (identity, concentration, potential mixture effects) (Dela Cruz et al., 2014). In addition, 259 experiments carried out in chambers with controlled environments are difficult to be 260 transferred to 'real life' situations (Han and Ruan, 2020; Hörmann et al., 2017). Also, the 261 reliability of the device used for the TVOC measurement (compared with other widely 262 accepted detection techniques) could be considered as a limitation of the study. Therefore, it 263 is very complicated to compare the conclusions obtained with the different experiments 264 performed by other authors. Also, the number of plants involved, their biomass, leaf area, 265 root volume and other variables will affect the final results. However, despite the actual 266 removal efficiency obtained, in all the cases, a reduction in VOCs has been reported thanks 267 to the living wall. Nevertheless, more studies about this topic are required in order to better 268 comprehend the potential of vegetation for the IAQ improvement.

269

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