1	A STATE-OF-THE-ART REVIEW ON INDOOR AIR POLLUTION AND THE
2	POTENTIAL OF BIOTECHNOLOGIES FOR INDOOR AIR PURIFICATION
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16	Abstract
17	Indoor air pollution has traditionally received less attention than outdoors pollution despite
18	indoors pollutant levels are typically twice higher, and people spend 80-90 % of their life in

1 n increasing air-tight buildings. More than 5 million people die every year prematurely from 19 illnesses attributable to poor indoor air quality, which also causes multi-millionaire losses due 20 to reduced employee's productivity, material damages and increased health system expenses. 21 Indoor air pollutants include particulate matter, biological pollutants and over 400 different 22 chemical organic and inorganic compounds, whose concentrations are governed by several 23 outdoor and indoor factors. Prevention of pollutant is not always technically feasible, so the 24 implementation of cost-effective active abatement units is required. Up to date no single 25 physical-chemical technology is capable of coping with all indoor air pollutants in a cost-26

effective manner. This problem requires the use of sequential technology configurations at the 27 28 expenses of superior capital and operating costs. In addition, the performance of conventional physical-chemical technologies is still limited by the low concentrations, the diversity and the 29 variability of pollutants in indoor environments. In this context, biotechnologies have emerged 30 as a cost-effective and sustainable platform capable of coping with these limitations based on 31 the biocatalytic action of plants, bacteria, fungi and microalgae. Indeed, biological-based 32 purification systems can improve the energy efficiency of buildings, while providing additional 33 aesthetic and psychological benefits. This review critically assessed the state-of-the-art of the 34 indoor air pollution problem and prevention strategies, along with the recent advances in 35 36 physical-chemical and biological technologies for indoor pollutants abatement.

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Keywords: air pollutant; biotechnology; indoor air quality; prevention strategy; physicalchemical technology; volatile organic compound.

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# 41 Highlights:

42 • A state-of-the-art review on the global problem of poor indoor air quality

• Typology, sources and health risks of the most common indoor pollutants are revised

• Measures for prevention and abatement of indoor pollutants are carefully examined

• Evaluation of the performance of traditional physical-chemical methods

• Assessment of innovative and sustainable systems based on biotechnologies

### 47 Abbreviation list

- 48 AC Activated carbon
- 49 BF Biofilter
- 50 BTF Biotrickling filter
- 51 BTEX Benzene, toluene, ethylbenzene and xylene
- 52 CADR Clean Air Delivery Rate
- 53 DMS Dimethyl sulfide
- 54 EBRT Empty bed residence time
- 55 EPA Environmental Protection Agency
- 56 ETS Environmental tobacco smoke
- 57 HCHO Formaldehyde
- 58 HVAC Heating, Ventilation and Air Conditioning
- 59 IAQ Indoor air quality
- 60 IEQ Indoor Environmental Quality
- 61 LCM Carbon microspheres
- 62 LCNT Carbon nanotubes
- 63 MERV Minimum efficiency reporting value
- 64 MOF Metal oxide framework
- 65 NTP Non-thermal plasma
- 66 PAN Polyacrylonitrile
- 67 PBTF Plant-based biotrickling filter
- 68 PBDEs Polybrominated diphenyl ethers
- 69 PCO Photocatalytic oxidation
- 70 PDMS Polydimethylsiloxane
- 71 PE Polyethylene
- 72 PAHs Polycyclic aromatic hydrocarbons

- 73 PM Particulate matter
- 74 PP Polypropylene
- 75 Ppm<sub>v</sub> Volumetric parts-per-million
- 76 PU Polyurethane
- 77 PVC Polyvinylchloride
- 78 REACH Registration, Evaluation and Authorization of Chemicals system
- 79 RH Relative humidity
- rGO Reduced graphene oxide
- 81 TPPB Two-liquid phase partitioning bioreactor
- 82 TVOC Total volatile organic compounds
- 83 UV Ultraviolet radiation
- 84 VICs Volatile inorganic compounds
- 85 Vis Visible radiation
- 86 VOCs Volatile organic compounds
- 87 WHO World Health Organization
- 88 ZIF Zeolitic imidazole framework

#### 89 **1.** The indoor air quality problem

Air pollution and human exposure to low-quality air is nowadays the most critical 90 environmental threat to public health worldwide according to the World Health Organization 91 (WHO) (World Health Organization, 2016). Recent reports have indicated that, at a global 92 scale, 1 out of 10 deaths are attributable to air pollution. Indeed, air pollution alone caused 93 approximately 5.5 million deaths in 2013 in the world (World Bank and Institute for Health 94 Metrics and Evaluation, 2016). At a European level, recently updated data have ascribed more 95 than 500,000 premature deaths in 2016 to long-term exposure to polluted air, from which 96 412,000 corresponded to particulate matter (PM), 71,000 to NO<sub>2</sub>, and 15,000 to O<sub>3</sub> (European 97 98 Environmental Agency, 2019). These effects will be even more critical due to the continuous growth of major cities. Indeed, about 90% of the population worldwide lives in urban areas 99 exposed to air quality levels that exceed the WHO guidelines (World Health Organization, 100 101 2016), which are typically based on PM pollution.

The latest studies on human exposure to indoor pollution revealed that indoor environments 102 could be at least twice as polluted as outdoor environments (European Comission, 2003). 103 Indeed, the air in an urban street with average traffic might actually be cleaner than the air in a 104 105 living room. Traditionally, indoor air pollution has received significantly less attention than 106 outdoor air pollution, especially in highly industrialized or dense traffic areas. However, the 107 threats posed by a long-term exposure to indoor air pollution have become more apparent in recent years (European Environment Agency, 2013) as buildings are progressively sealed 108 109 against the outside environment to obtain heating and cooling energy cost savings. Many buildings rely entirely on mechanical ventilation to recirculate indoor air with a greatly reduced 110 outdoor air dilution level, leading to the accumulation of indoor pollutants. A recent 111 commission report estimated that nearly 3 billion people worldwide are daily exposed to poor 112 indoor air quality (IAQ) caused by the use of solid fuels for cooking, heating and lighting. This 113 report concluded that household air pollution is a major contributor to global figures for 114

morbidity and mortality, with major effects on respiratory and cardiovascular systems (Royal 115 College of Physicians, 2016). Additionally, the report highlighted the significant contribution 116 of poor IAQ to the total number of premature deaths. At a global level, the WHO estimated that 117 each year 4.3 million people die prematurely from illnesses attributable to indoor air pollution 118 119 (World Health Organization, 2014), almost all in low and middle income countries, with pollutants produced by indoor combustion processes, such as cooking or heating, as the main 120 121 cause of deficient IAQ. Moreover, the prevalence of health problems related with IAQ such as sick building syndrome, building related illness or multiple chemical sensitivity has been 122 extensively reported (Burge, 2004). Although the levels of indoor air pollutants and their 123 124 associated symptoms may decrease in new or recently renovated buildings during the first half 125 year, poor IAQ and its associated symptoms have been reported over years (INSHT, 2015).

In this context, human exposure to indoor air (houses, workplaces, public buildings or means 126 of transport) exceeds 80 % in developed countries and 85-90 % in Europe (European 127 Comission, 2003) (Figure 1). Likewise, the National Human Activity Pattern Survey in the US 128 revealed that an adult spends 86% of his time indoors, to which a further 6% must be added as 129 time spent inside vehicles or public transport (Marć et al., 2018). There has been a significant 130 131 global shift in the economy from the manufacturing sector towards the service and knowledge-132 based sectors, which operate in indoor office environments (Al Horr et al., 2016). Poor IAQ has been classified as a priority problem affecting children's health, while it is considered as one of 133 the largest national environmental threats by the US-EPA (Guieysse et al., 2008). 134

Besides being responsible for many respiratory illnesses, allergies and even cancerous diseases, poor IAQ has also a noteworthy economic impact since it damages valuable objects from libraries, archives or museums, and may reduce employee's productivity in working places by 10-15 % (Cincinelli et al., 2016) (Figure 1). The costs associated to air pollution in the WHO European Region, including exposure to both outdoor and indoor air, amounted to 1.431\$ trillions in 2010, from which 42.9\$ billion corresponded to Spain (WHO Regional Office for Europe, 2015). If only IAQ is considered (in terms of mortality, loss of productivity, and
illnesses and sick leaves), recent studies have calculated annual expenses of up to 20,000
million € in France (ANSES et al., 2014).



In 2010, the WHO published the first specific IAQ guidelines (World Health Organization and 148 WHO Regional Office for Europe, 2010), which have been since adopted by other organizations 149 in several countries. Whereas these guidelines represented the first attempt to establish a general 150 IAQ standard, only a few priority indoor air pollutants with known adverse health effects were 151 included. Indeed, an exhaustive quantitative regulation comprising all physical, chemical, 152 biological and particulate exposures is rather challenging due to the inherent complexity of 153 154 indoor air pollution and the lack of information concerning the long-term health effects of each pollutant. In this context, indoor air pollutants include particulate matter, biological pollutants 155 (allergens, bacteria, mold, fungi, spores, etc.), physical agents (temperature, electromagnetic 156 157 fields) and over 400 different chemical compounds, mainly volatile organic and inorganic 158 compounds (VOCs and VICs, respectively). More recently, organic emerging compounds emitted from plastics, pharmaceuticals or personal care products have been detected in indoor 159 160 environments. In addition to the wide spectrum of indoor pollutants, their occurrence and concentration depend on many factors such as outdoors air quality, the socio-economic 161 development of the country, type of activities, occupancy, ambient conditions, season of the 162 year, etc. Moreover, pollutant sources and emission rates rapidly change over time (Luengas et 163 164 al. 2015): endogenous sources include permanent (building materials, adhesives, paints, 165 varnishes, etc.) and occasional sources (furniture, cleaning and disinfection products, cooking, personal care products, human metabolism, etc.), while outdoor pollutants intrusion clearly 166 depends on human activities (road traffic, industry, etc.). Additionally, secondary pollutants 167 might be produced by indoor gas-phase reactions from other compounds present in indoor air 168 (Hubbard et al., 2005; SCHER, 2007). 169

Unfortunately, there is limited information over IAQ due to the great number of pollutants and sources and their variability. Significant efforts must be undertaken in order to better understand the effects of indoor air pollutants on human health and to design *ad-hoc* regulations to protect human health (Royal College of Physicians, 2016). A better understanding of the type and

concentration of indoor air pollutants will facilitate the design of effective control strategies 174 both in terms of prevention and active abatement. Since prevention must be considered as the 175 first step for improving IAQ, actions have been recently implemented to reduce or eliminate 176 certain pollutant sources. For instance, smoking in workplaces and most public places is 177 prohibited in Europe and many countries in the world, and the use of harmful construction 178 materials such as asbestos or products containing hazardous components like halogenated 179 pesticides is widely restricted (EU Council, 2008). However, pollution control at source is only 180 possible when sources are known, while new substances are constantly detected and classified 181 as hazardous. Therefore, prevention or minimization of indoor pollutant emissions is commonly 182 183 insufficient, technically non-viable, or economically unfeasible (Guieysse et al. 2008, Luengas 184 et al. 2015). Moreover, according to the Directive 2010/31/EU (European Parliament, 2010), EU Member States have committed to build nearly zero energy buildings by the end of 2020, 185 while enhancing energy performance during major renovations of existing buildings or 186 retrofitting of building elements. New designs devoted to effective energy savings involve 187 airtight, well-insulated and sealed constructions, which substantially reduces natural 188 ventilation. This increase in building air tightness will cause a severe impact on the levels of 189 gas and particulate indoor air pollutants, increasing their concentration as a result of the reduced 190 191 ventilation rates (Broderick et al. 2017). In this context of potential conflict between energy efficiency measures and enforcement of IAQ standards, the development and optimization of 192 versatile technologies for in-situ indoor air purification is increasingly needed. 193

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195 2. Main pollutants and sources

The most important indoor air pollutants investigated include particulate matter, volatile inorganic compounds and volatile organic compounds. Particulate matter is typically classified as a function of its size, but the most reported value corresponds to the concentration of particles with a mean diameter lower than 2.5 micrometers (PM<sub>2.5</sub>). Carbon dioxide (CO<sub>2</sub>), carbon

monoxide (CO), nitrogen oxides  $(NO_x)$  and ozone  $(O_3)$  rank among the most studied inorganic 200 201 compounds. VOCs include a wide group of organic gas pollutants with a low boiling point (ranging from 50/100°C to 240/260°C) and low vapor pressure ubiquitous in indoor 202 environments (Luengas et al., 2015). This group is continuously growing as new materials are 203 being used in construction and interior design (Figure 2). Despite the large number of pollutants 204 present in indoor environments, only a few of them are typically measured and used as 205 206 surrogates in order to simplify data acquisition. These surrogate compounds are selected based on their human risk potential or as representative of a more extended group of compounds with 207 similar behaviour. Sampling sites where environmental tobacco smoke (ETS) is present are 208 209 typically omitted in most monitoring projects since ETS is so dominant over other indoor contaminant sources that results are not reliable (Jantunen et al., 2011). 210

Research projects investigating indoor air pollution are mainly focused on public and private 211 212 buildings. These environments are very different in terms of type of pollutants and concentration ranges. Public indoor environments comprise office buildings, schools, shopping 213 centers, libraries, etc., while private indoor spaces involve apartment buildings or detached 214 dwellings. Some studies have investigated the levels of pollutants before and after building 215 216 renovation, remodeling or refurbishment, which entailed the implementation of new energy-217 saving concepts and therefore a higher air tightness and accumulation of indoor pollutants. The typical sources (Figure 2), health risks and common concentrations of the most relevant indoor 218 pollutants are here discussed. These pollutants, including PM, VICs (CO, NO<sub>x</sub>, O<sub>3</sub>) and VOCs 219 220 (benzene, toluene, ethylbenzene, xylenes, naphthalene, formaldehyde, trichloroethylene, αpinene, limonene), have been selected based on their occurrence in indoor spaces or their 221 222 concerns in terms of human health (Table 1).

Compound	CAS number	Molecular weight <b>(g/mol)</b>	Boiling point (°C at 1 atm)	Vapor pressure (mmHg at 25°C)	Water solubility (mg/L at 25°C)	Environmental risks <sup>a</sup>	Henry constant (mol/m <sup>3</sup> ·Pa)
СО	630-08-0	28.0	-191.7	>35 atm	26.8 at 20°C		9.64E-06
NO <sub>2</sub>	10102-44-0	46.0	21.0	720	Reacts		1.20E-04
O <sub>3</sub>	10028-15-6	48.0	-111.7	>1 atm	570 at 20°C	H400; H410	1.10E-04
Benzene	71-43-2	78.1	78.8	101	940.0	H412; P273	1.70E-03
Toluene	108-88-3	92.1	110.6	27.7	320.0	H412	1.50E-03
Ethylbenzene	100-41-4	106.2	136.2	9.21	110.0	H412	1.30E-03
o-xylene	95-47-6	106.2	145.9	5.99	120.0	H412; P273	2.08E-03
m-xylene	108-38-3	106.2	140.6	7.61	99.0	H412; P273	1.37E-03
p-xylene	106-42-3	106.2	139.6	7.94	100.0	H412	1.48E-03
Naphthalene	91-20-3	128.2	221.5	0.159	140.0	H400; H410	2.20E-02
Formaldehyde	50-00-0	30.0	-19.5	3460	$1.98 \cdot 10^5$		3.20E+01
TCE	79-01-6	131.4	87.2	72.4	390.0	H412; P273	9.50E-04
α-pinene	80-56-8	136.2	157.9	3.5	8.9	H411	2.12E-04
Limonene	138-86-3	136.2	175.4	1.54	3.4	H400; H410; P273	6.27E-04

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<sup>a</sup>Code: H400: very toxic to aquatic life; H410: very toxic to aquatic life with long lasting effects; H411: toxic to aquatic life with long lasting effects;

H412: harmful to aquatic life with long lasting effects; P273: avoid release to the environment

**Table 1**. Physical/chemical properties of the most relevant indoor air pollutants.





Particulate matter consists of solid particles suspended in air able to enter the respiratory tract, 229 which mainly occurs when particle size is below 2.5 micrometers. Natural sources include 230 suspended dust from sand or sea salt, pollen or fire ash, while anthropogenic sources are 231 associated to combustion processes, industry, agriculture and transportation. PM can also be 232 originated by reactions between ozone and some VOCs (e.g. terpenes) (Carazo Fernández et 233 al., 2013; European Environment Agency, 2017; Royal College of Physicians, 2016; SCHER, 234 2007; US Environmental Protection Agency, 1995). Fibers, such as fiber glass and asbestos, 235 can also be included in this group (Carazo Fernández et al., 2013; Leung, 2015; Royal College 236 of Physicians, 2016). Outdoor PM enters indoor environments through ventilation, while indoor 237 238 sources include combustion-based appliances like ovens, heaters or stoves, and also tobacco smoke and fireplaces (Carazo Fernández et al., 2013; European Environment Agency, 2017; 239 Royal College of Physicians, 2016; SCHER, 2007). When biomass is used as a fuel, indoor 240 241 levels of PM can exceed those of polluted cities. Long-term exposure to particulate matter can provoke conditions from several respiratory and cardiovascular problems such as eye, nose, 242 throat and bronchial irritation and asthma to fibrosis, anthracosis and lung cancer (Carazo 243 Fernández et al., 2013; Jantunen et al., 2011; Leung, 2015; SCHER, 2007; US Environmental 244 Protection Agency, 1995). Kitchens and bedrooms may exhibit PM2.5 concentrations in the 245 range of 25-1526 and 13-27  $\mu$ g/m<sup>3</sup>, while concentrations of 10-44 and 3-23  $\mu$ g/m<sup>3</sup> have been 246 recorded in offices and schools, respectively. Shopping complexes also present high 247 concentrations of PM<sub>2.5</sub> (74-164 µg/m<sup>3</sup>) (Armendáriz-Arnez et al., 2010; Barmparesos et al., 248 249 2018; Broderick et al., 2017; Canha et al., 2017; Datta et al., 2017; Mandin et al., 2017; Pokhrel et al., 2015; Sidhu et al., 2017; Singh et al., 2016; Vasile et al., 2016; Yip et al., 2017). 250 Carbon monoxide is a colorless, odorless, tasteless, non-irritant toxic gas emitted as a result of 251 incomplete combustion processes. CO is usually produced from fossil fuels in power 252 generation, road transportation and heating and cooking devices. However, this pollutant is also 253

emitted by natural sources, accounting for one third of the total emissions. The main indoor

sources of CO are tobacco smoke, defective cooking and heating devices, fireplaces and vehicle 255 256 gases from attached garages. Outdoor air exchange in dense traffic and parking areas or highly industrialized districts also mediates the intrusion of CO in indoor environments (Carazo 257 Fernández et al., 2013; Kotzias et al., 2005; Leung, 2015; Royal College of Physicians, 2016; 258 Singh et al., 2016; US Environmental Protection Agency, 1995; World Health Organization 259 and WHO Regional Office for Europe, 2010). Inhalation is the main entrance to human body 260 of CO, which is able to bind reversibly to hemoglobin with an affinity 200-250 times higher 261 than oxygen. CO toxic effects are thus based on the interference with O<sub>2</sub> transportation through 262 the human body resulting in tissue hypoxia. The extent of health damage depends on the 263 264 concentration and duration of exposure, and include cardiovascular, respiratory and neurological problems. CO can even cause death under high acute exposure (>1000 ppmv) or 265 long-term exposure (Kotzias et al., 2005; Leung, 2015; US Environmental Protection Agency, 266 267 1995; World Health Organization and WHO Regional Office for Europe, 2010). Concentrations of 0.5-17 ppm<sub>v</sub>, 0.9-3.8 ppm<sub>v</sub> and up to 8.5 ppm<sub>v</sub> have been recorded in kitchens, bedrooms and 268 shopping areas, respectively (Broderick et al., 2017; Canha et al., 2017; Sidhu et al., 2017; 269 Singh et al., 2016; Vasile et al., 2016; Yamamoto et al., 2014; Yip et al., 2017; Zielinska et al., 270 271 2012).

272 Nitrogen monoxide (NO) and nitrogen dioxide (NO<sub>2</sub>) are the most common and important nitrogen oxides. These pollutants are generated by combination of oxygen and nitrogen during 273 combustion in power generation industries, transportation and heating systems. NO is produced 274 275 in a larger extent and is further oxidized to NO<sub>2</sub> in the presence of oxygen, ozone or VOCs (European Environment Agency, 2017; World Health Organization and WHO Regional Office 276 277 for Europe, 2010). The most frequent indoor sources of NO<sub>x</sub> are gas appliances like stoves, ovens or water heaters. ETS and fireplaces can also contribute to NO<sub>x</sub> emissions (Carazo 278 Fernández et al., 2013; Kotzias et al., 2005; Leung, 2015; Luengas et al., 2015; Royal College 279 of Physicians, 2016; Singh et al., 2016; US Environmental Protection Agency, 1995; World 280

Health Organization and WHO Regional Office for Europe, 2010). Infiltration from outdoor air 281 strongly influences indoor levels, in particular within short distance from roadways or high-282 density industrial areas. Inhaled NO<sub>2</sub> is converted to nitric acid in the lungs, with the associate 283 damage to cells and to the immune system. Inhalation can cause respiratory malfunctioning 284 when other pathologies are previously present (limit for protection of asthmatic of 200  $\mu$ g/m<sup>3</sup> 285 1-h average;  $\approx 2000 \text{ }\mu\text{g/m}^3$  impact normal individuals). NO<sub>2</sub> can cause also eve irritation by 286 direct contact (Carazo Fernández et al., 2013; Kotzias et al., 2005; Leung, 2015; US 287 Environmental Protection Agency, 1995; World Health Organization and WHO Regional 288 Office for Europe, 2010). Concentrations of 5-17  $\mu$ g/m<sup>3</sup>, 16-18  $\mu$ g/m<sup>3</sup> and up to 30  $\mu$ g/m<sup>3</sup> have 289 290 been measured in houses, offices and shopping areas, respectively (Broderick et al., 2017; Langer et al., 2015; Mandin et al., 2017; Prasauskas et al., 2016; Singh et al., 2016; Vasile et 291 al., 2016). 292

293 Benzene is an organic volatile compound with a characteristic aromatic odor and widely used in industry as a solvent and in the synthesis of several plastics, resins and pharmaceuticals. 294 Outdoor air intrusion contributes to indoor levels, with benzene originated by traffic road and 295 industrial activity (European Environment Agency, 2017; Jantunen et al., 1998; World Health 296 297 Organization and WHO Regional Office for Europe, 2010). Indoor sources of benzene include 298 ETS, combustion devices and construction materials such as polymeric furnishings, carpets, paints, solvents and wooden furniture (Campagnolo et al., 2017; Carazo Fernández et al., 2013; 299 Jantunen et al., 1998; Kotzias et al., 2005; Royal College of Physicians, 2016; SCHER, 2007; 300 Singh et al., 2016; World Health Organization and WHO Regional Office for Europe, 2010). 301 Benzene (and other pollutants) emissions from these materials decay with time, thus levels are 302 much higher in new or renovated buildings. Health risks for acute intoxication are low (e.g. 303 headaches for 5-h exposure to 150-500 mg/m<sup>3</sup>), although benzene is a proven carcinogenic and 304 genotoxic chemical under chronic exposure. Benzene can also induce blood illnesses and 305 neurological and reproductive problems (Carazo Fernández et al., 2013; Kotzias et al., 2005; 306

Leung, 2015; World Health Organization and WHO Regional Office for Europe, 2010). No safe level of exposure is recommended. The highest level of benzene has been recorded in shopping areas (2.5-48  $\mu$ g/m<sup>3</sup>) followed by offices (1.4-5.5  $\mu$ g/m<sup>3</sup>), homes (0.7-4.4  $\mu$ g/m<sup>3</sup>) and schools (0.5-3  $\mu$ g/m<sup>3</sup>) (Broderick et al., 2017; Dodson et al., 2008; Du et al., 2015; Edwards et al., 2001; Geiss et al., 2011; Kotzias et al., 2009; Langer et al., 2015; Rösch et al., 2014; Singh et al., 2016; Xu et al., 2016; Zielinska et al., 2012).

Toluene is also an aromatic compound used in several industrial processes and emitted also by 313 power generation industries and traffic (Campagnolo et al., 2017; Jantunen et al., 1998; Kotzias 314 et al., 2005; Leung, 2015; Royal College of Physicians, 2016). Toluene, present in gasoline, 315 316 paints, resins, adhesives, cosmetics and coatings, is one of the most common indoor air pollutants and the most abundant among BTEX (benzene, toluene, ethylbenzene and xylene). 317 Indoor sources are infiltration from outdoor air, tobacco smoke, combustion devices and a 318 variety of household products (Carazo Fernández et al., 2013; Jantunen et al., 1998; Kotzias et 319 al., 2005; Royal College of Physicians, 2016; SCHER, 2007; Singh et al., 2016). Toluene is not 320 a confirmed carcinogenic substance, and therefore no long-term health effects have been 321 studied yet. Exposure to toluene increases risks of developing asthma and other respiratory 322 323 conditions, while short and long-term exposure can affect central nervous system (Kotzias et al., 2005). Shopping complexes present the highest toluene concentrations (15-164  $\mu$ g/m<sup>3</sup>), 324 followed by offices (6-32  $\mu$ g/m<sup>3</sup>) and homes (3-20  $\mu$ g/m<sup>3</sup>). The presence of toluene in schools 325 is marginal (1.8 µg/m<sup>3</sup>). Concentrations are typically higher in new or recently renovated 326 327 buildings as a result of the new installed materials (Broderick et al., 2017; Dodson et al., 2008; Du et al., 2015; Edwards et al., 2001; Geiss et al., 2011; Langer et al., 2015; Mandin et al., 328 2017; Rösch et al., 2014; Singh et al., 2016; Xu et al., 2016; Zhong et al., 2017; Zielinska et al., 329 2012). 330

Ethylbenzene is a colorless liquid compound at ambient temperature, with a strong gasoline-like scent. Sources of pollution include plastics, paints, adhesives and other products where

ethylbenzene is used as a solvent during manufacturing. Ethylbenzene is also emitted during 333 combustion processes such as traffic (ethylbenzene is included in gasolines), heating devices 334 and cooking (Campagnolo et al., 2017; Jantunen et al., 1998; Leung, 2015; Singh et al., 2016). 335 Acute effects at relatively high concentrations include eye and throat irritation and dizziness. 336 No long-term effects have been described yet in humans. Ethylbenzene has been classified as a 337 potential human carcinogen by the International Agency for Cancer Research (ATSDR, 2007). 338 Concentrations of ethylbenzene up to 16  $\mu$ g/m<sup>3</sup> have been detected in offices and shopping 339 complexes. On the other hand, homes and schools exhibit maximum concentrations of 8 and 340 0.2 µg/m<sup>3</sup>, respectively (Broderick et al. 2017; Dodson et al. 2008; Edwards et al. 2001; Rösch 341 342 et al. 2014; Singh et al. 2016; Xu et al. 2016; Zhong et al. 2017; Zielinska et al. 2012).

343 Xylene is a colorless liquid at room temperature with an aromatic odor present as three isomers: ortho, meta and para (o-, m- and p-, respectively). Xylene is used as a solvent in industry and 344 as an additive in gasoline. Indoor sources of xylenes include paints, adhesives, dyes, polymers, 345 cleaning products and pharmaceuticals. Outdoor air is also a source of indoor pollution if dense 346 traffic or industrial facilities using xylene are nearby (Campagnolo et al., 2017; Jantunen et al., 347 1998; Kotzias et al., 2005; Leung, 2015; SCHER, 2007; Singh et al., 2016). An acute exposure 348 to xylene can cause eye and throat irritation, headache and nausea. Long-term exposure is 349 350 associated with problems in the respiratory, gastrointestinal and central nervous systems, lungs, kidney, heart and reproductive system. Xylene is a suspected carcinogenic chemical, associated 351 with an increased risk of leukemia, non-Hodgkin's lymphoma and colon/rectum cancer 352 (Kotzias et al., 2005). Shopping areas, with levels ranging from 1.3 to 74  $\mu$ g/m<sup>3</sup>, and offices, 353 with levels of 2.2 to 16  $\mu$ g/m<sup>3</sup>, rank among the most polluted sites. Maximum xylene 354 concentrations of 3.1 and 0.3  $\mu$ g/m<sup>3</sup> have been measured at homes and schools (Broderick et al. 355 2017; Dodson et al. 2008; Edwards et al. 2001; Geiss et al. 2011; Langer et al. 2015; Mandin 356 et al. 2017; Singh et al. 2016; Vasile et al. 2016; Xu et al. 2016; Zhong et al. 2017; Zielinska et 357 358 al. 2012).

Naphthalene is a white crystalline solid at ambient temperature, with a characteristic aromatic 359 odor (mothballs), widely used as a raw material in the chemical industry. The main emission 360 source is traffic as a result of the inherent presence of naphthalene in gasoline and diesel. Indoor 361 sources include tobacco smoke, defective combustion devices, herbicides, rubber materials and 362 insect repellents (usually contaminating clothes) (Kotzias et al., 2005; Royal College of 363 Physicians, 2016; World Health Organization and WHO Regional Office for Europe, 2010). 364 Outdoor air is a mayor source in high-traffic and industrial areas. Inhalation is the principal 365 naphthalene intake mechanism in humans, although accidental ingestions of mothballs have 366 been reported. Acute naphthalene intoxication can induce hemolytic anemia and cataracts. No 367 368 long-term adverse health effects or evidence of human carcinogenicity or genotoxicity have been reported (Kotzias et al., 2005; World Health Organization and WHO Regional Office for 369 Europe, 2010). Naphthalene concentrations ranging from 3 to 26  $\mu$ g/m<sup>3</sup> have been recorded at 370 371 homes (Du et al. 2015; Xu et al. 2016; Zhong et al. 2017).

Formaldehyde (HCHO) is a colorless, odorous, highly reactive gas emitted during incomplete 372 reaction of hydrocarbons in industries, traffic and other processes. Formaldehyde is also 373 produced by oxidation of other VOCs with ozone or radiation (Campagnolo et al., 2017; Kotzias 374 375 et al., 2005; US Environmental Protection Agency, 1995; World Health Organization and WHO 376 Regional Office for Europe, 2010). This chemical is widely used in resins, glues, paints, paper products, cosmetics, electronic equipment, cleaning agents or fabrics. Formaldehyde is present 377 in construction materials such as insulation foams of wooden-based materials employed in 378 379 floorings or furniture. However, emissions from these materials (e.g. plywood, particleboard or fiberboard) usually decay within several weeks (Carazo Fernández et al., 2013; Jantunen et al., 380 1998; Kotzias et al., 2005; Royal College of Physicians, 2016; World Health Organization and 381 WHO Regional Office for Europe, 2010). Formaldehyde is rapidly absorbed by the respiratory 382 or gastrointestinal system. Acute exposure effects include odor, irritation, headache and 383 eczema. Formaldehyde is a known carcinogenic and genotoxic chemical, whose long-term 384

exposure can originate nasopharyngeal cancer and myeloid leukemia (Kotzias et al., 2005; US Environmental Protection Agency, 1995; World Health Organization and WHO Regional Office for Europe, 2010). The level of exposure in homes, offices and schools is comparable (7.7-30, 8-17, 9-17  $\mu$ g/m<sup>3</sup>) (Broderick et al., 2017; Canha et al., 2017; Dodson et al., 2008; Geiss et al., 2011; Kotzias et al., 2009; Langer et al., 2015; Mandin et al., 2017; Prasauskas et al., 2016; Zhong et al., 2017; Zielinska et al., 2012).

Trichloroethylene (TCE) is a volatile chemical with sweet chloroform odor, widely produced 391 by the industry, and used as solvent and cleaning agent (vapor degreasing of metals). TCE can 392 be present in contaminated waters and soils, which could indirectly contribute to indoor levels. 393 394 Indoor sources include products where TCE is used as solvents such as lubricants, varnishes, paint removers, adhesives and typewriter correction fluids. TCE can be also present in some 395 bleach household products and other cleaning agents (Jantunen et al., 1998; Singh et al., 2016; 396 397 World Health Organization and WHO Regional Office for Europe, 2010). Human exposure to this VOC occurs by dermal absorption, ingestion and inhalation. Exposure to TCE is often 398 intermittent, as the products that contain this pollutant are not constantly used. Acute exposure 399 to TCE affects the central nervous system at concentrations of  $\sim 270 \text{ mg/m}^3$ , causing a reduced 400 sensorial capacity. TCE is a carcinogenic chemical under long-term exposure, and a chronic 401 exposure may cause liver, kidney and bile duct cancer and non-Hodgkin's lymphoma. 402 Therefore, no safe threshold is recommended due to its carcinogenicity (World Health 403 Organization and WHO Regional Office for Europe, 2010). TCE concentrations ranging from 404 0.3 to 0.6  $\mu$ g/m<sup>3</sup> are typically recorded at homes, and from 0.6 to 4.7  $\mu$ g/m<sup>3</sup> in shopping 405 complexes (Dodson et al., 2008; Edwards et al., 2001; Mandin et al., 2017; Singh et al., 2016; 406 Xu et al., 2016). 407

Alpha-pinene is a colorless liquid with a strong pine scent. This terpene is naturally present in
plants and a common constituent of essential oils. Pinene is used as a solvent in some paints
and waterproof substances, and can be found in perfumery products, deodorizers and cleaning

products. Pinene emissions can also originate from wooden materials like furniture or flooring, 411 412 particularly those made from pine wood (Campagnolo et al., 2017; Kotzias et al., 2005; Luengas et al., 2015; Royal College of Physicians, 2016). No significant pinene concentrations are found 413 outdoors, except in forest areas. On the other hand, limited data of pinene health risks is 414 available. While acute exposure to high concentration can produce irritation and inflammation, 415 no chronic exposure risks have been reported yet. Health risks might be expected when ozone 416 or other reactive radicals are present, due to their high reactivity with pinene and other 417 unsaturated molecules (Hubbard et al., 2005; Kotzias et al., 2005; Leung, 2015; SCHER, 2007). 418 These reaction by-products would produce eye and upper airways irritation in a more extended 419 420 way than pinene itself. The largest pinene concentrations are found at homes (11-32  $\mu$ g/m<sup>3</sup>) and libraries (10-30  $\mu$ g/m<sup>3</sup>), while offices and schools exhibit concentrations up to 6.3 and 1.5 421  $\mu g/m^3$ , respectively (Dodson et al. 2008; Edwards et al. 2001; Geiss et al. 2011; Langer et al. 422 423 2015; Mandin et al. 2017; Rösch et al. 2014; Xu et al. 2016; Zhong et al. 2017).

Limonene is a natural terpene present as two isomers (d- and l-), which have citric and pine 424 scent, respectively. Limonene is used in a wide variety of household products like cleaning 425 agents, resins, air fresheners, deodorants, fragrances and shampoos (Campagnolo et al., 2017; 426 427 Kotzias et al., 2005; Luengas et al., 2015; Royal College of Physicians, 2016). This terpene is 428 also found in food as an additive due to its citric odor and flavor. Limonene intake occurs by 429 inhalation or ingestion. This pollutant has low acute toxicity, no symptoms further than eye or skin irritation have been described. Chronic health effects for limonene have not been studied, 430 431 and there is no evidence of carcinogenicity or genotoxicity. Heath risks are associated to ambient reaction with ozone or reactive radicals, which form byproducts such as aldehydes, 432 carboxylic acids and peroxides, responsible of irritation and odor annoyance at low 433 concentrations (Hubbard et al., 2005; Kotzias et al., 2005; Leung, 2015; SCHER, 2007). 434 Maximum concentrations of 32, 19 and 11  $\mu$ g/m<sup>3</sup> have been recorded at homes, offices and 435 schools, respectively (Dodson et al., 2008; Du et al., 2015; Edwards et al., 2001; Geiss et al., 436

2011; Langer et al., 2015; Mandin et al., 2017; Rösch et al., 2014; Xu et al., 2016; Zhong et al.,
2017).

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# 440 **3. Prevention of indoor air pollutant emission**

Among the different approaches to control indoor air quality, prevention of pollutant formation 441 and emission rank first in terms of cost-effectiveness. Several strategies to prevent the emission 442 and to decrease the concentration of gas pollutants in indoor environments have been proposed. 443 Overall, ventilation is the easiest measure to prevent the accumulation of indoor pollutants. 444 Indoor pollutant concentrations typically decrease when increasing outdoor air exchange rate 445 446 unless outdoor pollutant concentrations are high (such as in areas with intense traffic or 447 industrial activities). Mechanical ventilation systems introduce outdoor fresh air into the building, which dilutes the concentration of indoor air pollutants. Some sources can be sealed 448 or removed by professionals, like asbestos and other insulation fibers (Chenari et al. 2016; 449 Kotzias et al. 2005; Leung 2015; Luengas et al. 2015). Smoking bans are also a very effective 450 measure since tobacco smoke is a source of multiple harmful chemicals in elevated 451 concentrations. The suppression of tobacco smoke in indoor places has resulted in consistent 452 IAQ improvements (Kotzias et al., 2005; US Environmental Protection Agency, 1995). Finally, 453 454 an adequate control of the relative humidity and temperature is crucial to control the emissions of organic pollutants from indoor materials. In this context, air conditioning systems can control 455 the temperature and relative humidity, potentially using smart systems to automatize 456 457 ventilation or heating depending on the indoor atmosphere (high CO<sub>2</sub> levels, low temperatures) or occupants' activities (Chenari et al. 2016; Luengas et al. 2015; Zhou et al. 2017). 458

459 Many indoor air pollutants, including PM, BTEX, NO<sub>x</sub> or CO, are emitted during combustion 460 processes at homes, such as cooking or heating. Increased ventilation rates and the reduction of 461 these combustion gases from cooking stoves, boilers or fireplaces prevent indoor pollutant 462 accumulation. Combustion devices in buildings must be regularly checked and maintained to

prevent malfunctioning and emissions of harmful pollutants to the indoor atmosphere (Carazo 463 Fernández et al., 2013; Leung, 2015; SCHER, 2007; US Environmental Protection Agency, 464 1995). Highly polluting fuels like biomass or kerosene need to be replaced by more efficient 465 fuels such as natural gas or electricity at homes, although there is an increasing popularization 466 of biomass in developed countries due to its higher sustainability. This effect was demonstrated 467 in rural kitchens in Nepal, where emissions of PM2.5 from different fuels were investigated. A 468 remarkable improvement in IAQ when using electricity and liquified petroleum gas (PM2.5 469 levels of 80 and 101  $\mu$ g/m<sup>3</sup>, respectively) compared to wood and rice husk (PM<sub>2.5</sub> levels of 630 470 and 656  $\mu$ g/m<sup>3</sup>, respectively) (Pokhrel et al., 2015). In addition, improved cooking stoves have 471 472 been proved to reduce emissions of PM and CO (Armendáriz-Arnez et al., 2010; Sidhu et al., 2017; Yip et al., 2017). Other combustion sources, such as candles or scented incense, may also 473 contribute to a reduced IAQ (Kotzias et al., 2005). 474

475 Some VOCs are emitted from common construction materials. The manufacture of some wooden-based materials, like plywood or fiberboard, often implies the utilization of resins and 476 varnishes that contain formaldehyde and other VOCs. Paints, glues, plastics and other 477 construction materials include VOCs (usually BTEX) as solvents or additives. Some of these 478 479 emissions can be prevented by using low-emitting materials like improved plastics and paints 480 (phenol resins instead of urea resins, polyurethane coatings, etc.) and solid wood or old furniture (Carazo Fernández et al. 2013; Environmental Protection Agency 1995; Kotzias et al. 2005; 481 Wei et al. 2015). 482

Occupancy of buildings should be avoided during several weeks after construction or renovation, as pollutant emissions from these sources typically decrease in time until reaching low emission steady state levels. Indeed, pollutant concentrations were found higher even four months after renovation (Broderick et al., 2017). Similarly, emissions of VOCs from paints, adhesives, cleaning products and fuels can be partially mitigated by properly sealing and storing these liquid materials and minimizing storage periods to prevent leakages and emissions (US
Environmental Protection Agency, 1995).

A wide variety of indoor pollutants originate from household and personal care products. Some compounds like pinene and limonene are used as scents in cleaning products, deodorants, cosmetics and even in food (Carazo Fernández et al., 2013; Kotzias et al., 2005; SCHER, 2007).
Pollutant emissions can be partially avoided by using household products according to manufacturer instructions (good ventilation, no mixing, being aware of warnings). In addition, unused or nearly empty containers should not be stored for long periods and must be disposed safely (Environmental Protection Agency 1995; Wei et al. 2015).

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## 498 4. Physical-chemical technologies for indoor air treatment

The intrusion or generation of indoor air pollutants cannot always be reduced or suppressed in 499 500 a cost-effective or technically feasible manner. In these scenarios, active abatement units can be installed in order to lower or eliminate the levels of indoor air contaminants. Traditionally, 501 these devices consisted of physical-chemical technologies such as filters or ozonisers installed 502 as part of a central heating and ventilation system or operated as portable units (Luengas et al., 503 2015). Real-time sensing has been applied to these devices to optimize their performance. These 504 505 sensors on-line monitor ambient conditions (temperature, humidity, concentrations of key pollutants) and activate the abatement units based on the need of the occupants and their 506 activities, with the subsequent energy savings (Guyot et al., 2018; Kumar et al., 2016; Tran et 507 508 al., 2017; Yang et al., 2019). Today, the market of physical/chemical technologies for indoor air treatment is dominated by mechanical and electronic filtration, adsorption and ozonation 509 510 (Tables 2 and 3).

Design parameters	Air flow (m <sup>3</sup> /h)	Single pass efficiency (%)	Reference				
Mechanical filtration							
Multilayer (glass + synthetic fibers) non-woven filter (≈ MERV 16 filter) filter installed in HVAC system 5.08cm depth	2.5 m/s	Black Carbon: 88% / Ultrafine PM: 86% PM(0.3-2.5 μm): 91% / PM(>2.5 μm): 88%	(Polidori et al., 2013)				
MERV16 filter + 12 gas-phase filters + particle postfilter Dimensions: 0.41m <sup>2</sup> ×1.95m (portable)	1362	Black Carbon: 90% / Ultrafine PM: 94% PM(0.3-2.5 μm): 92% / PM(>2.5μm): 75%	(Polidori et al., 2013)				
PVC/PU fibers (8 w/w%) on filter paper support Membrane: 100 cm <sup>2</sup> , 20.72 g/m <sup>2</sup>	1.91 / 5.01	PM (0.3-0.5 μm): >99.5%	(Wang et al., 2013)				
Polyester filter with sericin coating	145	PM(2.5,10 µm): ≈56%	(Verma et al., 2019)				
ZIF8-SiO2 nanofiber composite membrane (2h growing time) Dimensions: Ø30mm disc (7.07cm <sup>2</sup> )	0.05	Soft smoke PM: > 99.95%	(Zhu et al., 2019)				
Double layer nonwoven filter: PP-g-DMAEMA (Dimethylaminoethylmethacrylate)-rGO (reduced graphene oxide) (6.3%) + PP-g-DMAEMA sheet Filter: 25g/m <sup>2</sup>	19.2	65% (PP-DMAEMA-rGO) 76.4% (PP-DMAEMA) 72.2% (Double Layer)	(Tang et al., 2020)				
	Electronic filtrat	ion					
Ion-spray with carbon fiber electrodes (charger and plates) Charger outside-mounted (emit ions to ambient air) Dimensions: 120×185×85mm <sup>3</sup>	120	PM (0.3μm; laboratory): 47.8% PM (0.3μm; real chamber): 39.8%	(Sung et al., 2019)				
Electrostatic precipitator based on corona discharge and parallel plates	120	PM (0.3/0.5/1/3): 67.7/67.7/40.6/14.5%	(Zeng et al., 2020)				

**Table 2**. Performance of physical-chemical technologies for the removal of particulate matter.

Design parameters	Air flow (m <sup>3</sup> /h)	Pollutant Removal Capacity	Single Pass Efficiency (%)	Reference			
Adsorption							
Indoor passive panels (gypsum based; different treatments) Dimensions: 0.089 m <sup>2</sup>	0.2	HCHO: 40-140 $\mu$ g/m <sup>2</sup> ·h Toluene: 30-210 $\mu$ g/m <sup>2</sup> ·h	-	(Zuraimi et al., 2018)			
Granular activated carbon Ø46mm, 150mm tube ; 7.0 g of adsorbent Thermal desorption (300°C)	0.06	-	Benzene: 81.5-91.6 Toluene: 86.6-100 Ethylbenzene: 91.6-99.2 Xylene: 89.9-100	(Jo and Yang, 2009)			
Porous ferrihydrite/SiO <sub>2</sub> composite (different aging times) ; 0.27g of adsorbent in Ø140mm petri	-	HCHO: 6.30-8.11 mg/g 6mg/g after 7 cycles		(Xu et al., 2013)			
Polyester filter with sericin coating	145	-	Benz: 2.96 ; Tol: 2.96 Et-Benz: 3.71 ; Xyl: 4.18	(Verma et al., 2019)			
Carbon nanotubes (LNCT) and carbon microspheres (LCM) Dimensions: Ø16mm, 90mm ; Adsorbent: 3g	0.001	Tol / Et-Benz / <i>o</i> -xyl LCNT: 52.2, 78.0, 102 mg/g LCM: 47.1, 64.0, 62.7 mg/g	Tol / Et-Benz / <i>o</i> -xyl LCNT: 57.8, 47.5, 61.8 LCM: 51.7, 53.5, 52.6	(Srivastava et al., 2019)			
Alkaline-treated carbon nanotubes (LNCT) and carbon microspheres (LCM) Dimensions: Ø16mm, 90mm ; Adsorbent: 2g	0.001	Tol / Et-Benz / <i>o</i> -xyl LCNT: 1821, 964, 1076 mg/g LCM: 1093, 895, 996 mg/g	Tol / Et-Benz / <i>o</i> -xyl LCNT: 84.0, 83.3, 82.6 LCM: 82.5, 83.4, 74.8	(Srivastava et al., 2019)			
High-grade activated carbon filter (portable)	510		HCHO: 0.6 ; Toluene: 32.0 n-decane: 40.0 Tetrachloroethylene: 31.3	(Chen et al., 2005)			
Activated carbon prefilter (portable)	569		HCHO: 0.2 ; Toluene: 7.8 n-decane: 12.5 Tetrachloroethylene: 6.0	(Chen et al., 2005)			

Carbon-zeolite mixture impregnated with potassium iodide (29.5 kg)	272		HCHO: 4.0 ; Toluene: 19.4 n-decane: 31.3 Tetrachloroethylene: 33.1	(Chen et al., 2005)		
Activated charcoal filter (portable)	225		HCHO: 0.4 ; Toluene: 5.6 n-decane: 6.0 Tetrachloroethylene: 5.2	(Chen et al., 2005)		
Non-woven polyester filter (2 layers) impregnated with activated carbon (portable)	340		HCHO: 1.5 ; Toluene: 26.0 n-decane: 62.0 Tetrachloroethylene: 22.5	(Chen et al., 2005)		
Granular activated carbon + KMnO4-impregnated alumina (in-duct)	-		HCHO: 1.4 ; Toluene: 3.5 n-decane: 3.4 Tetrachloroethylene: 3.3	(Chen et al., 2005)		
Photocatalytic oxidation						
Indoor passive panels + UV-Vis light (TiO <sub>2</sub> -wood flooring; wallpaper; fabric) Dimensions: 0.089 m <sup>2</sup>	0.2	Toluene: 59-120 μg/m <sup>2</sup> ·h ; 58-70 μg/m <sup>2</sup> ·h ; <15 μg/m <sup>2</sup> ·h		(Zuraimi et al., 2018)		
Roofing tiles (three types) and corrugated sheets containing TiO <sub>2</sub> (143cm <sup>2</sup> ; 12.1-46.4 mg/cm <sup>2</sup> and 90cm <sup>2</sup> ; 22.2 mg/cm <sup>2</sup> , respectively)	-	Toluene (RT / CS): 308-512 μg/m <sup>2</sup> ·h at RH47% 248 μg/m <sup>2</sup> ·h at RH47%	Toluene (RT /CS): 28.2-62.9 at RH47% ; 78±2% at RH1%22.7 at RH47%	(Demeestere et al., 2008)		
UV light (253.7 nm) TiO <sub>2</sub> converter (+MERV11 filter) (in-duct)	720		Benz: 0.58; Tol: 0.58; Et-Benz: 0.50; Xyl:0.32; EtOH: 0.19%; n-hex: 0.26; HCHO: 0.08	(Kadribegovic et al., 2011)		
UV light (253.7 nm) TiO <sub>2</sub> converter (+MERV11 filter) (in duct)	3600		O3 (UV off ): <2 O3 (UV on): 15	(Kadribegovic et al., 2011)		
ZnO nanorod-wrapped PTFE nanofiber antibacterial membrane with Ag nanoparticles Ø30mm (7.07cm <sup>2</sup> ) ; 0.1mm thick	Bacteria: 0.06 HCHO: 0.013	НСНО: 1989 µg/m <sup>2</sup> ·h	Bacteria: >99 HCHO: ≈60	(Feng et al., 2017)		

Cellulose/Polyester/Polyamide filter coated with P25 TiO <sub>2</sub> /SiO <sub>2</sub> as catalyst (in duct) Filter: 400cm <sup>2</sup> , 250 µm thick, 17g/m <sup>2</sup> TiO <sub>2</sub>	25.8	Toluene: 15687 $\mu$ g/m <sup>2</sup> ·h n-decane: 15071 $\mu$ g/m <sup>2</sup> ·h TCE: 1646.9 $\mu$ g/m <sup>2</sup> ·h	Toluene: 8.63 n-decane: 5.76 TCE: 0.17	(Héquet et al., 2018)
VUV lights + MnO <sub>x</sub> /TiO <sub>2</sub> on Ni foam catalyst Photocatalyst: 834cm <sup>2</sup> , 8mm thick	120		TVOC (UV off/on): 50.14 / 74.86	(Zeng et al., 2020)
Polyacrylonitrile (PAN)-TiO <sub>2</sub> filter at different designs (simple membrane ; monolith ; truncated cone ; corrugated) ; Properties (cm <sup>2</sup> , g/cm <sup>2</sup> ): 30, 0.33 ; 198, 0.48 ; 136, 0.23 ; 121, 1.01	9.5	Acetaldehyde Membrane: 2300 mg/m <sup>2</sup> ·h Monolith: 2228 mg/m <sup>2</sup> ·h Trunc. cone: 2300 mg/m <sup>2</sup> ·h Corrugated: 3414 mg/m <sup>2</sup> ·h	Acetaldehyde Membrane: 5.36% Monolith: 4.43% Truncated cone: 5.36% Corrugated: >4.99%	(Roso et al., 2019)
Two honeycomb monoliths coated with TiO <sub>2</sub> as catalyst + 3 UV lamps (in-duct)	-		HCHO: 1.6 ; Toluene: 1.8 n-decane: 2.5 Tetrachloroethylene: 0.7	(Chen et al., 2005)
SiO <sub>2</sub> /TiO <sub>2</sub> -coated glass fiber tissue 6.5g/m2 SiO <sub>2</sub> + 6.5g/m2 TiO <sub>2</sub> Ø76mm in-coated tube	2		Benzene: ≈30 Propionic acid: ≈49.8	(Zadi et al., 2020)
	Non-	-thermal plasma		
Post-catalytic plasma with different catalysts (Pt/Al <sub>2</sub> O3, Cu-Mn/TiO <sub>2</sub> , Fe <sub>2</sub> O <sub>3</sub> +MnO <sub>2</sub> , CuO+MnO <sub>2</sub> ) and energy densities (J/L) Ø42mm tube; 15g of catalyst	0.6		Toluene (all catalyst): >90 at RH0%, 2.5J/L Toluene (Pt/Al2O3): 39-61 at RH30-72%, 10J/L O3 (Pt/Al2O3): ≈90 at all RH	(Van Durme et al., 2009)
Dielectric barrier discharge NTP Dimensions: Ø12mm, 20mm tube (exposure time of 0.06 / 0.12s)	Bacteria: 0.75 Fungi: 1.71		Bacteria: >95 Fungi: 85-98	(Liang et al., 2012)
Dielectric barrier discharge NTP 6.5g/m <sup>2</sup> SiO2 + 6.5g/m <sup>2</sup> TiO <sub>2</sub> Ø76mm in-coated tube	2		Benzene: 29-52 Propionic acid: 10.3-34.7	(Zadi et al., 2020)

Hybrid methods						
Activated carbon (Jo et al.) + PCO device Inner surface Ø20mm tube, 0.5mg/cm <sup>2</sup> of TiO <sub>2</sub> Thermal desorption (300°C)	0.06		Benzene: 95.0-98.7 Toluene: 97.5-100 Ethylbenzene: 98.3-100 Xylene: 96.6-100	(Jo and Yang, 2009)		
Plasma deodorization unit + Activated charcoal filter (portable)	408		HCHO: 1.2 ; Toluene: 6.7 n-decane: 9.6 Tetrachloroethylene: 5.8	(Chen et al., 2005)		
Electronic cell + Activated carbon postfilter (portable)	544		HCHO: 0.3 ; Toluene: 2.0 n-decane: 8.1 Tetrachloroethylene: 1.5	(Chen et al., 2005)		
High intensity UV lamp + Photocatalytic semiconductor + Pleated activated carbon filter (in-duct)	-		HCHO: 3.0 ; Toluene: 42.1 n-decane: 44.8 Tetrachloroethylene: 46.6	(Chen et al., 2005)		
ZIF8-SiO <sub>2</sub> nanofiber composite membrane Dimensions: Ø30mm disc (7.07cm <sup>2</sup> )	0.013	HCHO: 36.04 µg/m <sup>2</sup> ·h; 48.87mg/g	HCHO: ≈80	(Zhu et al., 2019)		
Dielectric barrier discharge NTP + UV light + SiO <sub>2</sub> /TiO <sub>2</sub> -coated glass fiber tissue $6.5g/m^2$ SiO2 + $6.5g/m^2$ TiO2 Ø76mm in-coated tube	2		Benzene: 58-90 Propionic acid: 63.8-94.8	(Zadi et al., 2020)		
Electrostatic precipitator + Vacuum-UV lights + MnO <sub>x</sub> /TiO <sub>2</sub> on Ni foam catalyst + MnO <sub>2</sub> catalyst for O <sub>3</sub> removal MnO <sub>x</sub> /TiO <sub>2</sub> catalyst: 834cm <sup>2</sup> , 8mm thick	120		TVOC: 4.04 No O <sub>3</sub> generation	(Zeng et al., 2020)		
MOF TiO <sub>2</sub> /UiO-66-NH <sub>2</sub> composites + UV light Toluene exp: 100mg of cat. on 250-mesh screen Acetaldehyde exp: 100mm, Ø6mm tube	0.06	Toluene (25/75/90%w TiO <sub>2</sub> ): 106.7/164.4/142.0 mg/g CH <sub>3</sub> CHO: (75%w TiO <sub>2</sub> ): 275.3 mg/g	Toluene (25/75/90%w TiO <sub>2</sub> ): 47.2/72.7/62.8 CH <sub>3</sub> CHO: (75% w TiO <sub>2</sub> ): 70.74	(Zhang et al., 2020)		

Double layer nonwoven filter: PP-g-DMAEMA (Dimethylaminoethylmethacrylate)-rGO (reduced graphene oxide) (6.3%) + PP-g-DMAEMA sheet ; Filter: 25g/m <sup>2</sup>	0.18	Benzene / Toluene / Xylene PP-DMAEMA-rGO: 51.4 / 50.6 / 48.9 mg/g PP-DMAEMA: <5mg/g each Double layer: 39.1 mg/g overall	(Tang et al., 2020)
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**Table 3**. Performance of physical/chemical technologies for the removal of VOCs.

Mechanical filtration, based on the forced circulation of air through a fibrous material where 516 pollutants are captured, is the simplest and most popular method for PM removal. PM removal 517 efficiencies depend on several factors such as the type and material of the filter or the air flow 518 (Harriman et al., 2019; Luengas et al., 2015). These filters need regular replacements to 519 maintain the capture efficiency, prevent the re-emission of pollutants and to avoid the growth 520 of microorganisms on the organic matter trapped in the filter material (Guievsse et al., 2008). 521 Single pass efficiencies higher than 90% are typically achieved in conventional filters, even at 522 high areal flow rates of 3240 m<sup>3</sup>/m<sup>2</sup>h (Polidori et al., 2013). New materials such as zeolites 523 (Zhu et al., 2019) or polymers (Tang et al., 2020; Wang et al., 2013) have been tested as 524 525 effective filters for particulate matter (Table 2).

Electronic filtration is based on the attraction of negatively charged particles to a plate with 526 opposite polarity, where particles are retained. Two commercial devices are available: 527 electrostatic precipitators ionizing pollutant particles and ion generators dispersing into the air 528 ions that will subsequently attach to pollutants. This technology exhibits higher capital and 529 operating costs than mechanical filtration (particularly ion generators) and requires an active 530 removal of the particles accumulated on the plates. Electronic filtration also entails the 531 generation of harmful pollutants such as ions, ozone or other compounds from VOCs ionization 532 533 (Hubbard et al., 2005; Luengas et al., 2015). The efficiencies achieved by electronic filters are lower than those provided by mechanical filtration, and range between 14.5% and 67.7% for 534 different PM size (Zeng et al., 2020) (Table 2). 535

Adsorption consists of the capture of organic and inorganic volatile pollutants on the surface of an adsorbent material. The most popular materials are activated carbon and zeolites, although alumina, silica gel or polymers are also used (Jo and Yang, 2009; Kim and Ahn, 2012; Luengas et al., 2015). These adsorbent materials can be even incorporated in construction materials, which are easy to integrate in interior surfaces (Cros et al., 2012; da Silva et al., 2017; Gall et al., 2011; Zuraimi et al., 2018). A high relative humidity and the inherent variability of pollutant

levels in indoor environments decrease the efficiency of adsorption filters. In addition, the 542 adsorption specificity of the material with one pollutant may inhibit the efficient adsorption of 543 other gas pollutants. These systems can also accumulate microorganisms hazardous to human 544 health. Adsorbent materials need to be periodically replaced or regenerated upon saturation to 545 maintain the efficiency and prevent re-emission, thus increasing operating costs (Guieysse et 546 al., 2008; Luengas et al., 2015). VOC removal efficiencies in adsorption devices are very 547 heterogeneous and dependent on pollutant hydrophobicity (Chen et al., 2005). Nevertheless, 548 removal rates of 90% are achieved for BTEX using activated carbon (Jo and Yang, 2009) and 549 carbon nanocomposites (Srivastava et al., 2019) as adsorbent materials. Removal capacities up 550 to 140 µg/m<sup>2</sup>·h (Zuraimi et al., 2018) or 8.11 mg/g (Xu et al., 2013) have been reported for 551 formaldehyde. Likewise, Srivastava et al. achieved retention capacities of 1821, 964 and 1076 552 mg/g for the removal of benzene, toluene and xylene, respectively (Table 3). Overall, 553 554 adsorption technologies are considered the best physical/chemical technology to remove indoor VOCs. 555

Ozonisers generate O3 molecules from ambient O2 by high-voltage discharge or UV radiation . 556 O<sub>3</sub> is a strong oxidant that can react with VOCs and VICs present in indoor air. Although the 557 558 levels of some VOCs may be reduced, the abatement efficiencies of ozonisers are not superior 559 to their physical/chemical counterpart (Luengas et al., 2015). In addition, health problems may arise from high indoor levels of ozone or by hazardous secondary pollutants produced by the 560 reaction of O<sub>3</sub> with VOCs, particularly with terpenes (Chen et al., 2005; Hubbard et al., 2005; 561 562 Luengas et al., 2015). The performance of ozonisers has not been evaluated in this review as they are not recommended for indoor air treatment due to their above-mentioned health risks. 563

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565 UV photolysis relies on the degradation of VOCs, bacteria, fungi and viruses by ultraviolet 566 radiation. Indeed, this technology constitutes an effective method for sterilization at ambient 567 temperature and pressure (De Robles and Kramer, 2017). However, UV radiation is harmful for human health and only some air pollutants are effectively destroyed. The high cost of lamp acquisition and intensive energy consumption rank among the main disadvantages of this technology. In addition, undesired compounds such as ozone or secondary organic pollutants might be released to indoor air as a result of the unspecific action of UV radiation (Chen et al., 2005; Guieysse et al., 2008; Luengas et al., 2015). The performance of stand-alone photolysis units was not herein evaluated since this technology is commonly implemented as photocatalytic oxidation.

Photocatalytic oxidation (PCO) utilizes catalytic semiconductors to create reactive radicals 575 from ambient O<sub>2</sub> and radiation (generally UV), which further react with indoor air pollutants. 576 577 TiO<sub>2</sub> is the most popular catalyst due to its low cost, non-toxicity, biocompatibility and effectiveness. Other metallic oxides and sulfides (SnO<sub>2</sub>, ZnO, CdS) are also used as catalysts 578 (Chen et al., 2005; Mo et al., 2009). This technology can cope with a larger spectrum of 579 580 pollutants than photolysis (Guieysse et al., 2008). The generation of harmful radicals and secondary organic aerosols, along with the high costs of catalyst media replacement, limit the 581 widespread use of this technology (Chen et al., 2005; Luengas et al., 2015). Single pass 582 efficiencies in photocatalytic devices are pollutant and catalyst specific. For instance, Feng et 583 al. (2017) reported a removal efficiency of 60% for formaldehyde (Feng et al., 2017), while 584 Zeng et al. (2020) reported a 75% removal of TVOC (Zeng et al., 2020). The areal elimination 585 capacity is an important design parameter in PCO, with values as high at 3414 mg/m<sup>2</sup> h reported 586 for acetaldehyde (Roso et al., 2019) (Table 3). 587

Plasma consists of a cloud of highly ionized gas containing electrons, positive ions, oxidants and free radicals able to degrade organic pollutants, precipitate particulate matter and kill pathogens (Luengas et al., 2015). Non-thermal plasma (NTP) is generated at ambient temperature by high voltage discharges (usually corona discharge) at the expenses of a high energy consumption. Similarly to UV or photocatalytic oxidation, plasma mediates the generation of O<sub>3</sub> and other by-products or oxidation intermediates that may be as dangerous for human health as the parent pollutants (Chen et al., 2005; Luengas et al., 2015). NTP can achieve
removal efficiencies of up to 90% for VOCs (Van Durme et al., 2009) and microorganisms
(Liang et al., 2012), although operation under real conditions (i.e. high humidity and low
pollutant concentrations) could reduce its abatement performance (Table 3).

Membrane separation is based on the transfer of indoor air pollutants to another fluid through 598 a semipermeable membrane. This technology has been consistently proven at high pollutant 599 concentrations, but its efficiency at the low levels of pollutants found in indoor air is not 600 guaranteed (Guieysse et al., 2008). Mechanical resistance and pressure drop, and therefore 601 operational costs, are higher compared to other technologies. In addition, VOCs captured on 602 603 the other side of the membrane must be appropriately managed or destroyed in a separate unit (Guieysse et al., 2008; Luengas et al., 2015). Membrane separation is still a developing 604 technology in indoor air treatment. 605

606 Overall, there is no single physical-chemical technology capable of coping with all indoor air pollutants in a cost-effective manner, which requires the use of sequential technology 607 configurations at the expenses of superior capital and maintenance costs. Mechanical filtration 608 is typically installed to eliminate particulate matter prior to other abatement technology 609 (Luengas et al., 2015). The broad nature of VOCs and VICs requires tandems of gas pollutant 610 611 abatement technologies. For instance, Jo and Yang (2009) reported removal efficiencies of BTEX of ~100% using a granular activated carbon adsorbent coupled with a PCO-TiO<sub>2</sub> unit 612 (Jo and Yang, 2009). Likewise, Zhu et al. (2019) (Zhu et al., 2019)(Zhu et al., 2019)achieved 613 614 a removal efficiency of 80% for formaldehyde, with an adsorption capacity of 48.9 mg/g (gram of adsorbent) (Zhu et al., 2019). Zhang et al. (2020) recorded capacities of up to 164.4 mg/g for 615 toluene and 275.3 mg/g for acetaldehyde (Zhang et al., 2020), while a capacity of 39.1 mg/g 616 has been reported by Tang et al. (2020) for benzene, toluene and xylene (Tang et al., 2020) 617 (Table 3). 618

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### 620 5. Biological Based Purification Methods

The performance of conventional physical-chemical methods for indoor air purification is 621 hampered by the low concentrations, the diversity and the variability of VOCs in indoor 622 environments. However, these limitations represent an opportunity for biological based 623 purification systems. Biotechnologies are based on the action of microorganisms or plants, 624 which are able to eliminate or transform the gas pollutants by utilizing them as energy and/or 625 carbon source for either cell maintenance or replication (Guieysse et al., 2008). Overall, indoor 626 air pollutant biodegradation relies on the action of oxidative enzymes, which function at 627 ambient pressure and temperature without the need of additional chemicals. In botanical 628 629 technologies, both plant and soil microenvironment are responsible of pollutant degradation. 630 On the other hand, gas-water contactors are based on the biocatalytic action of suspended or immobilized microorganisms, which bio-convert air pollutants into CO<sub>2</sub>, H<sub>2</sub>O and new 631 microorganisms. These microorganisms need to be integrated by diverse, versatile and adaptive 632 microbial communities of bacteria, fungi, microalgae or yeasts able to remove simultaneously 633 a wide spectrum of pollutants at variable concentrations without generating undesirable by-634 products (Soreanu, 2016). Microorganisms are typically present as biofilms, which consist of a 635 636 moist layer of cells adhered to the surface of a support material. Biofilms provide a protected 637 environment to microorganisms and support a direct contact between the gas pollutants and cells, which enhances pollutant mass-transfer, a commonly limiting parameter in gas-phase 638 bioreactors (Guieysse et al., 2008; Soreanu, 2016; Soreanu and Dumont, 2020). 639

Biotechnologies have been successfully developed for industrial applications, where varying concentrations of multiple gas pollutants from different sources are cost-effectively treated (Estrada et al., 2011; Kennes and Veiga, 2013; Soreanu and Dumont, 2020; Van Groenestijn and Kraakman, 2005). However, the potential of biotechnologies for indoor air purification remains unexplored due to the particular and challenging conditions of indoor environments (Pacheco-Torgal et al., 2015). Mass-transfer limitations and the rapidly changing

concentrations of a large spectrum of gas pollutants are the main drawbacks associated with 646 647 indoor air treatment. Additionally, a cost-competitive CO<sub>2</sub> removal and the control of biological hazards need to be addressed prior in-house implementation of biotechnologies. Biological-648 based purification systems for indoor air would improve the energy efficiency of buildings, 649 while providing additional benefits in indoor environmental quality such as aesthetic and 650 psychological improvements (Guieysse et al., 2008; Moya et al., 2019). As indoor environments 651 652 are often condensed spaces, biotechnologies need to be compact and aesthetically integrated units with high effectiveness treating large air flowrates. In this context, new configurations of 653 bioreactors such as membrane, capillary or two-phase reactors have been successfully tested to 654 655 maximize the mass-transfer of gas pollutants (Kraakman et al., 2011; Muñoz et al., 2012; Soreanu, 2016). Alternatively, botanical biofiltration could have additional economic, 656 environmental and social benefits, including remarkable psychological impacts of 'greening' 657 658 the indoor space with plants. In addition, plants are able to effectively decrease CO<sub>2</sub> concentration, which has been proved to impact on human decision-making performance 659 (Satish et al., 2012). 660

661 The most common technologies for indoor air treatment are discussed below. Furthermore, 662 some examples of these purification methods and their principal operational characteristics are 663 shown in Table 4 and in Figure 3.

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Description of the Technology	Air flow <b>(m³/h)</b> EBRT <b>(min)</b>	Removal Efficiency (%) Elimination capacity (mg/m <sup>3</sup> ·h)	Inlet concentration Inlet load	Reference			
Botanical technologies							
Active potted plant: <i>Epipremnum aureum</i> (2) Filter bed: 0.35×0.2×0.15 m <sup>3</sup> , granular activated carbon + shale pebbles (1:1 mixture; Ø5 mm) 5.1 m <sup>3</sup> chamber	50 m <sup>3</sup> /h	HCHO: 19.9-32.7% at RH55-90%	HCHO: 824±15 μg/m <sup>3</sup>	(Wang et al., 2014)			
Biowall: <i>Philodendron scandens</i> (18) + <i>Philodendron scandens</i> 'Brazil' (13) + <i>Asplenium</i> <i>antiquum</i> (19) + <i>Syngonium podophillum</i> (13) 1.5 m <sup>2</sup> module; 30 m <sup>3</sup> chamber Support: inorganic media + activated carbon	50 m <sup>3</sup> /h	Butanone: 55.1-58.5%	Butanone: 39.4-44.8 μg/m <sup>3</sup>	(Torpy et al., 2018)			
		Biofilters					
Compost/perlite (75:25) as packing (8.5L) 120 cm, Ø10 cm PVC reactor Activated sludge as inoculum	1.57-0.53 min	H <sub>2</sub> S: >99% Toluene: >99.9% Butanone: >98.9%	H <sub>2</sub> S: 23.6-43.3 mg/m <sup>3</sup> Toluene: 0.40-0.60 mg/m <sup>3</sup> Butanone: 4.3-6.3 mg/m <sup>3</sup>	(Lebrero et al., 2011)			
Compost as packing 2L PVC column (53 cm, Ø8.3 cm) Irrigation flow rates: 30/120 mL/day Activated sludge as inoculum	0.72/0.30 min	MeSH: >98%; $\approx$ 1.2 g/m <sup>3</sup> ·h Toluene: >98%; $\approx$ 0.22 g/m <sup>3</sup> ·h α-pinene: >98%; $\approx$ 0.25 g/m <sup>3</sup> ·h n-hexane: 96-98%; $\approx$ 0.20 g/m <sup>3</sup> ·h	MeSH: 410-980 mg/m <sup>3</sup> ·h Toluene: 69-164 mg/m <sup>3</sup> ·h $\alpha$ -pinene: 76-182 mg/m <sup>3</sup> ·h n-hexane: 63-150 mg/m <sup>3</sup> ·h	(Lebrero et al., 2014)			
Compost/pozzolan (80:20) as packing (3.8L) 0.25 m, Ø0.1 m reactor Activated sludge as inoculum	0.38 min	Toluene: ≈100%; ≈23.9 mg/m <sup>3</sup> ·h p-xylene: ≈65-100%; ≈14 mg/m <sup>3</sup> ·h	Toluene: 0-25 mg/m <sup>3</sup> ·h p-xylene: 0-22.5 mg/m <sup>3</sup> ·h	(Luengas et al., 2017)			
<i>Pseudomonas putida</i> embedded in calcium alginate gel (granules); 30 cm, Ø6 cm column (4 beds); Liquid flow: 1.23 L/day	0.09 m <sup>3</sup> /h	HCHO: 68.6-93.5%; 25-158.4 mg/m <sup>3</sup> ·h	$\approx$ 0.2-1.34 mg/m <sup>3</sup>	(Lu et al., 2012)			

PU foam as packing (2L, 1cm <sup>3</sup> cubes); PVC column (53 cm,Ø8.3 cm); Liquid flow: 1.5 m/h Activated sludge as inoculum	0.07-0.72 min	MeSH: >97%; $\approx 1.2 \text{ g/m}^3 \cdot \text{h}$ Toluene: >97%; $\approx 0.22 \text{ g/m}^3 \cdot \text{h}$ $\alpha$ -pinene: >97%; $\approx 0.25 \text{ g/m}^3 \cdot \text{h}$ n-hexane: 88-95%; $\approx 0.20 \text{ g/m}^3 \cdot \text{h}$	MeSH: 410-980 mg/m <sup>3</sup> ·h Toluene: 69-164 mg/m <sup>3</sup> ·h $\alpha$ -pinene: 76-182 mg/m <sup>3</sup> ·h n-hexane: 63-150 mg/m <sup>3</sup> ·h	(Lebrero et al., 2014)		
PU foam as packing (48 cubes; 4×4×4 cm <sup>3</sup> ) Acrylic 8×8×76 cm <sup>3</sup> reactor Liquid flow: 0.3/0.5 L/min Activated sludge as inoculum	0.12/0.30 m <sup>3</sup> /h 1.53/0.61 min	VOCs (normalized carbon content) 78-87%; 1.2-9.3 mg <sub>carbon</sub> /m <sup>3</sup> ·h	1.5-10.7 mg <sub>carbon</sub> /m <sup>3</sup> ·h	(Saucedo- Lucero and Revah, 2018)		
PE Ralu-rings (1.5×1.5 cm <sup>2</sup> ) as packing (60 cm high); 100 cm, 48L reactor; Liquid flow: 1.2 m <sup>3</sup> /h Enriched culture inoculum	3.0-9.0 m <sup>3</sup> /h 0.96-0.32 min	Ethanol: 100%; 5-22 g/m <sup>3</sup> ·h DMS:10-100%; 0.65-6.6 g/m <sup>3</sup> ·h Optimal 95% (low gas flux)	Ethanol: 5-22 g/m <sup>3</sup> ·h DMS: 2.8-8.5 g/m <sup>3</sup> ·h	(Bak et al., 2017)		
	Membra	ine blotechnologies				
Porous acrylonitrile support (50μm) + PDMS layer (0.3μm); 40 cm <sup>2</sup> membrane in 200×5×2 mm <sup>3</sup> (8mL) compartments; Liquid flow: 0.03 L/min Activated sludge + hydrophobic bacteria as inoculum (+ silicon oil);	1.0-0.12 min	Acetone: 95-100% Toluene: 93-100% n-hexane: 7-28% Limonene: 88-100%	Acetone: 0.1-10.2 mg/m <sup>3</sup> ·h Toluene: 0.2-13.4 mg/m <sup>3</sup> ·h n-hexane: 0.1-5.6 mg/m <sup>3</sup> ·h Limonene: 0.1-9.6 mg/m <sup>3</sup> ·h	(Lebrero et al., 2013)		
Commercial hollow-fiber membrane reactor (10600 PDMS fibers, Ø190 μm; V: 300 mL; A: 8300 cm <sup>2</sup> ); Liquid flow: 0.2 L/min; Activated sludge as inoculum	1.4-0.27 min	MeSH: 8.5-98%; $\approx 0.58 \text{ g/m}^3 \cdot \text{h}$ Toluene: 67-99%; $\approx 0.11 \text{ g/m}^3 \cdot \text{h}$ $\alpha$ -pinene: 70-99%; $\approx 0.12 \text{ g/m}^3 \cdot \text{h}$ n-hexane: 38-58%; $\approx 0.09 \text{ g/m}^3 \cdot \text{h}$	MeSH: 210-1103 mg/m <sup>3</sup> ·h Toluene: 35-185 mg/m <sup>3</sup> ·h $\alpha$ -pinene: 39-205 mg/m <sup>3</sup> ·h n-hexane: 32-169 mg/m <sup>3</sup> ·h	(Lebrero et al., 2014)		
Hybrid technologies						
Biofilter + Adsorption system Pyrex column (23 cm, Ø10 cm) packed with compost + Activated carbon adsorption column (2.9 cm, Ø2.5 cm column)	0.5 m <sup>3</sup> /h 0.22 min (BF) 0.002 min (AC)	Toluene: 88.2-98.2% (BF) Toluene: 84.9-99.9% (BF+AC)	Toluene: 17-52 μg/m <sup>3</sup> (4.2-14 mg/m <sup>3</sup> ·h) Peaks of 733±55 μg/m <sup>3</sup> (200±20 mg/m <sup>3</sup> ·h)	(Hort et al., 2014)		

Biofilter + Adsorption system 25 cm, Ø10 cm reactor; Compost/pozzolan (80:20) packing (1.26L) + Activated carbon (1.26L) column Activated sludge as inoculum	0.13-0.04 min	Toluene: $\approx 100\%$ (at 45 mg/m <sup>3</sup> ·h), up to 80 mg/m <sup>3</sup> ·h (BF); $\approx 100\%$ , up to 187 mg/m <sup>3</sup> ·h (AC) p-xylene: $\approx 100\%$ (at 32 mg/m <sup>3</sup> ·h) up to 32 mg/m <sup>3</sup> ·h ((BF); $\approx 100\%$ , up to 63 mg/m <sup>3</sup> ·h (AC)	Toluene: 0-235 mg/m <sup>3</sup> ·h p-xylene: 0-63 mg/m <sup>3</sup> ·h	(Luengas et al., 2017)
Biofilter + Adsorption system 25 cm, Ø10 cm reactor; Compost/pozzolan (80:20) packing (1.26L) + Activated carbon (1.26L) column Activated sludge as inoculum	0.13-0.04 min	Toluene: 100% at 45 mg/m <sup>3</sup> ·h (BF); 100% at 187 mg/m <sup>3</sup> ·h (AC) p-xylene: 100% at 32 mg/m <sup>3</sup> ·h (BF); 100% at 63 mg/m <sup>3</sup> ·h (AC)	Toluene: 0-235 mg/m <sup>3</sup> ·h p-xylene: 0-63 mg/m <sup>3</sup> ·h	(Luengas et al., 2017)
Bag filter + Photocatalytic reactor + BTF Bag filter: 90 cm, Ø60 cm reactor PC: 45×45×100 cm <sup>3</sup> ; 10 UV lamps; 20 Ni foam plates (21.5×25.0 cm <sup>2</sup> ) coated with TiO <sub>2</sub> BTF: 220 cm, Ø40 cm reactor; ceramic beads/ Raschig rings (50:50) packing; 6×12cm bed layers	200 m <sup>3</sup> /h	TVOC: 84% (4+65+15%) PHAs: 87% (18+63+6%) PBDEs: 94% (80+11+3%) % for Bag filter, BTF and PC, respectively	TVOC: 48-920 μg/m <sup>3</sup> PHAs: 0.94-1.2 μg/m <sup>3</sup> PBDEs: 0.85-4.8 μg/m <sup>3</sup>	(Chen et al., 2016)

**Table 4**. Performance of biological technologies for the removal of VOCs.

Phytoremediation is an established method for the treatment of polluted soils and waters. The 668 application of botanical systems to indoor environments has been shown effective for removing 669 VOCs and PM (Liu et al., 2007; Pacheco-Torgal et al., 2015; Torpy et al., 2014; Wood et al., 670 2006; Yang et al., 2009). Botanical technologies can be divided into passive (e.g. potted plants) 671 or active (plant-based biotrickling filters, PBTFs) systems. Passive systems rely on the diffusion 672 of gas pollutants, which is a slow process at the low concentrations typically present in indoor 673 environments. Active systems incorporate mechanical ventilation devices to increase the 674 availability of the gas pollutants. In PBTFs, the polluted air is forced to flow though the aerial 675 parts and roots of hydroponic plants to maximize the removal of pollutants. Additional benefits 676 677 of these active systems such as decreasing indoor temperature, increasing relative humidity or acting as passive acoustic insulation have been identified (Moya et al., 2019). While CO<sub>2</sub>, SO<sub>x</sub>, 678 NO<sub>x</sub> and O<sub>3</sub> appear to be taken up directly through the stomates of plants during daylight, VOCs 679 680 are mostly degraded by microorganisms present in the rhizosphere. A symbiotic relationship between plants and microorganisms may be achieved (Moya et al., 2019; Oh et al., 2011; 681 Pacheco-Torgal et al., 2015; Torpy et al., 2014; Yang et al., 2009). The performance of 682 botanical technologies is still lower than that of other conventional technologies (Table 4). 683

Alike phytoremediation, biofiltration is a well-known and proven method for the removal of 684 685 gas pollutants, which can be engineered in multiple configurations (Figure 3). In biofilters (BFs), microorganisms are immobilized in a packing media, traditionally an organic material 686 like compost, irrigation is intermittent, and moisture and nutrients are retained by the biofilm 687 688 and the packing media. BFs are the preferred option for the treatment of hydrophobic pollutants based on the occurrence of a direct contact between the biofilm and the air emission. 689 690 Bioscrubbers consist of two separate units, where pollutants are initially transferred in a packedbed or mist absorption unit from the gas phase to an aqueous phase, which is recirculated to a 691 second unit where microorganisms growing in suspension biodegrade the dissolved pollutants. 692 693 This configuration provides high abatement efficiencies for hydrophilic pollutants. Biotrickling

filters (BTFs) are stand-alone units with packing media, typically inorganic materials with large 694 695 surface area supporting biofilm growth, with a solution of nutrients continuously trickling down over the packing. BTFs are effective for the treatment of hydrophilic pollutants and typically 696 operate at lower gas residence time than BF or bioscrubbers (Soreanu, 2016; Soreanu and 697 698 Dumont, 2020). These systems are well-established technologies for air pollution control in many industrial applications, providing an effective abatement performance, reliability and low 699 700 operating costs (Estrada et al., 2011; Kennes and Veiga, 2013; Soreanu and Dumont, 2020; Van Groenestijn and Kraakman, 2005). Table 4 highlights the performance of BFs and BTFs 701 702 operating at the concentration range commonly found in indoor air. Removal efficiencies higher than 95 % at gas residence times lower than 95 s have been reported in BFs and BTFs for a 703 wide range of pollutants. 704

Membrane bioreactors are based on the separation of gas and liquid phase by a membrane. A 705 706 nutrient solution, containing the pollutant degrading microorganisms, is continuously recirculated at one side of the membrane, while the contaminated air flows (usually counter-707 708 currently) on the other side. An attached biofilm might be formed on the aqueous side of the membrane. The pollutants diffuse from the gas side through the membrane and are biodegraded 709 710 by the microorganisms in the biofilm or in the bulk cultivation medium (Soreanu and Dumont, 711 2020). Despite exhibiting high investment costs, membrane bioreactors bring some innovations in the field of indoor air purification. First, the air stream and microorganisms in liquid phase 712 are physically separated, which allows a better control of operational conditions, including 713 714 biofilm thickness and nutrient supply, and prevents the drying of the biofilm. Secondly, the risk of an undesired stripping of microorganisms is avoided and the excess of humidity in the 715 716 purified air is limited. Finally, the mass-transfer of gas pollutants is improved due to the affinity and permeability of the membrane, especially in the case of hydrophobic pollutants (Kumar et 717 al., 2008; Studer and von Rohr, 2008). Membrane bioreactors have been tested for the treatment 718 of high VOC concentration, and their performance in indoor air applications remains 719

unexplored. Promising removal efficiencies have been obtained by Lebrero et al. (2013; 2014),

although the degradation of highly hydrophobic pollutants like n-hexane is still problematic.

722 (Lebrero et al., 2014, 2013).

Capillary bioreactors are a new approach on the abatement of gas pollutants. Capillary 723 bioreactors are engineered as parallel straight microchannels, with a maximum internal 724 diameter of 5 mm in air-water reactors (Kreutzer et al., 2005b). A segmented flow (or Taylor 725 726 flow) is desired inside the capillaries, which consists of alternating sections of gas and liquid (air bubbles and liquid slugs) that flow co-currently either upwards or downwards. The mass-727 transfer of gas pollutants is enhanced by the internal circulation within liquid slugs, which 728 729 increases the contact between the gas and the liquid phase. This configuration improves the gas-730 liquid mass-transfer (much higher than conventional stirred systems), while pressure drop remains low (Kraakman et al., 2011; Kreutzer et al., 2005b, 2005a; Rocha-Rios et al., 2013). 731 732 This technology could achieve better results than traditional bioreactors for a given power input (Kreutzer et al., 2005a). Although capillary bioreactors are promising solutions for indoor air 733 purification, some problems such as the long-term maintenance of a robust Taylor flow needs 734 to be solved (Kreutzer et al., 2005a). In this context, some innovative designs have been 735 736 proposed, such as a capillary microbioreactor where pollutants are pre-concentrated to 737 guarantee an excellent performance of the Taylor flow bioreactor, (López De León et al., 2019). Microalgae fix CO<sub>2</sub> during photosynthesis using light as energy source while releasing O<sub>2</sub> to 738 the ambient air. While plants can also perform photosynthesis, algae are much more effective 739 740 in this process because of the simpler cell structure and higher light utilization efficiencies (Brennan and Owende, 2010; Soreanu and Dumont, 2020). Algae photobioreactors may be a 741 742 feasible solution for indoor environments with elevated CO<sub>2</sub> levels such as offices, schools or shopping centers. Some species of microalgae are also able to use hydrocarbons from the 743 environment to obtain carbon and energy (heterotrophic metabolism), and even exhibit a mixed 744 745 metabolism depending on the environmental conditions. Microalgae systems have been widely studied in recent decades for biofuel or food production and wastewater treatment, where
operational conditions are far from those encountered in indoors environments (Anbalagan et
al., 2017; Mendoza et al., 2013; Niederwieser, 2015; Soreanu and Dumont, 2020). However,
there are few studies assessing the performance of algal photobioreactors as facades for indoor
air purification as well as energy recovery systems (Araji and Shahid, 2018; Wilkinson et al.,
2017).

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## 753 Conclusions

The problem of exposure to indoor air pollution has become more apparent in recent years, as 754 755 people spend more than 80% of the time indoors and buildings are progressively sealed against the environment to obtain heating and cooling energy savings. Recent studies have shown that 756 poor indoor air quality is a major contributor to global health problems and causes multi-757 758 millionaire losses. Currently, there is not a single physical-chemical technology that can efficiently address all challenges related to indoor air purification. Biological based purification 759 systems are promising solutions to overcome the limitations of indoor air treatment, such as 760 diversity and hydrophobicity of pollutants, variability of concentrations, etc. Additionally, these 761 762 systems need to assure humidity control of purified air, prevent microorganism releases and 763 incorporate CO<sub>2</sub> removal. The accomplishment of all these features would lead to 'green' energy efficient systems able to improve IAQ. Some hybrid solutions integrating 764 biotechnologies with physical-chemical systems have been proposed to overcome these 765 limitations, where technologies complement each other to cover a wider range of action. 766

As modern building regulations are based on energy savings, which reduces air exchange and increases the concentration of indoor pollutants, future research should focus on the progressive development of bio-based systems. Hence, biotechnologies have the potential to successfully deal with the problem of poor IAQ, although more investigation is necessary to develop cost-

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- effective systems that can be integrated in indoor spaces combining optimal performance withacceptable aesthetics.
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