

Recent Advances in Biological Systems for Improving Indoor Air Quality

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Abstracts

Studies on human exposure to indoor air pollution reveal that indoor environments could be at least twice as polluted as outdoor environments. Indoor air pollution has not received as much attention than outdoor air pollution, despite an adult spending now most of the time indoors as a result of the global shift in the economy from the manufacturing sector towards the service and knowledge-based sectors, which operate in indoor office environments.

Additionally, the health threats caused by a long-term exposure to indoor air pollution have become more apparent over the last decades as buildings are progressively sealed against the outside climate conditions to obtain heating and cooling energy cost savings and in response to stricter safety guidelines. Currently there is not a single technology that can efficiently provide a complete and satisfactory purification of indoor air. Biological systems for improving indoor air quality are promising, but challenges need to be considered to properly address the bioavailability of low pollutant concentrations, guarantee microbial safety, and incorporate CO₂-removal. This study presents the recent research advances in biological

27 indoor air purification methods as a ‘green’ alternative to physical-chemical methods, with
28 emphasis on current challenges and opportunities it can provide for improving Indoor
29 Environment Quality, building energy cost savings and improvements on indoor comfort and
30 well-being.

31

32 **Keywords:** Indoor Air Quality, Indoor Environmental Quality, Biofiltration, Membrane
33 Bioreactor, Capillary Bioreactor, Photobioreactor.

34

35 **1. INTRODUCTION**

36 *1.1 The indoor air quality problem*

37 The indoor concentration of air pollutions is almost always higher than the outdoor
38 concentration of air pollutions because outdoor-sourced contaminated air enters indoor
39 occupied spaces and combines with indoor-sourced pollutants (European Commission, 2003;
40 EPA, 2020). Indoor air pollution has not been acknowledged as outdoor air pollution has been,
41 especially in highly industrialized or dense traffic areas. However, the health threats of indoor
42 air pollutants caused by their long-term exposure have become more apparent over the last
43 decades as buildings are progressively sealed against the outside climate conditions to obtain
44 heating and cooling energy cost savings (EEA 2019) and as a result also of stricter safety
45 guidelines. Modern buildings cultivate higher indoor air pollutant concentrations because they
46 increasingly rely on mechanical ventilation with greatly reduced outdoor air ingress.

47 At a global level, the World Health Organization (WHO) estimated that each year 3.8 million
48 people die prematurely from illnesses ascribed to indoor air pollution, much of this due to
49 cooking or heating, which represents 7.7% of the global mortality (WHO 2018). For most
50 European countries the economic cost to society of household air pollution is significant in
51 terms of gross domestic product (WHO 2015), with for example annual expenses of up to

52 20,000 million € in France (Anses 2014). Moreover, health problems such as respiratory
53 illnesses, allergies and even cancerous diseases associated with poor Indoor Air Quality (IAQ)
54 are compounded by sick building syndrome (Burge 2004). Additional to health impacts, poor
55 IAQ has been shown to reduce workplace productivity by 10-15% (Cincinelli et al. 2016).
56 Between 80% (developed countries) and 90% (EU-28) of the average 250 million liters of air
57 a person breathes during their life (about 10,000 liter per day) is sourced from indoor sources
58 (houses, workplaces, schools, shopping centers, public buildings or means of transport (Royal
59 College of Physicians, 2016). Similarly, the USA Human Activity Pattern Survey found that
60 an average adult spends 86% of their time indoors and an additional 6% inside vehicles or
61 public transport (Maré et al. 2018). IAQ has been classified as a priority concern for children's
62 health (EU Environmental Agency 2019) and one of the USA's largest environmental threats
63 (Guieysse et al. 2008).

64 Indoor air pollutants include particulate matter, bioaerosols and over 400 different chemical
65 compounds, mainly volatile organic compounds (VOCs) for instance formaldehyde, BTEX
66 (benzene, toluene, ethylbenzene and xylene) and trichloroethylene, and inorganic compounds
67 (VICs) for instance carbon monoxide (CO), carbon dioxide (CO₂), nitrogen oxides (NO_x) and
68 ozone (O₃). Pollutant sources and emission rates may rapidly change over time (Luengas et al.
69 2015). Indoor pollutant sources include permanent sources (building materials, carpets, paints,
70 varnishes, etc.) and occasional sources (furniture, cleaning and disinfection products, cooking,
71 personal care products, tobacco smoke etc.), while outdoor pollutants intrusion mostly depends
72 on human activities (road traffic, industry, etc.) (Hubbard et al. 2005). Table 1 summarizes the
73 most relevant indoor air pollutants, their typical sources and commonly used measurement
74 methods. Recorded indoor air pollutant concentrations and their physical-chemical properties
75 are summarized elsewhere (Gonzalez-Martin et al., 2021).

76

77 **Table 1: Common Indoor Air Pollutants**

Indoor Air Pollutants	Typical Pollutant Sources¹	Common Measurement Methods²
Particle Matter (PM _{2.5} and PM ₁₀)	Indoor sources: ovens, heaters or stoves, fireplaces and tobacco smoke. Outdoor anthropogenic sources: combustion processes, industry and traffic. Outdoor natural sources: dust from sand or sea salt, pollen or fire ash.	Real-time direct reading instrument; light scattering airborne particle counter
Bioaerosols	Pets, mould, insects, sick occupants, (de)humidifiers or improper air filters (and may be attached to Particle Matter).	Impactor (air sampler directed onto a growth surface intending microbial colony enumeration) or metagenomic techniques
Volatile Organic Compounds (VOCs)		
Formaldehyde	Resins, glues, paints, paper products, cosmetics, electronic equipment, cleaning agents and fabrics. Construction materials such as insulation foams and wooden-based materials in floorings or furniture (note that emissions from some of these materials, e.g. plywood, usually decay within several weeks after installation).	ISO 16000-3, ASTM D5197, NIOSH 2016, EPA TO-11 (or 11A) or EPA Method IP-6 (or 6A).
BTEX	Indoor sources: combustion devices, tobacco smoke, construction materials (polymeric furnishings, carpets, paints, wooden furniture, resins, coatings and adhesives), cosmetics, cleaning products and pharmaceuticals. Outdoor sources: traffic and industrial activity.	Tenax sorbent followed by thermal desorption and GC-MS or MS-FID as per ISO 16000-6, ASTM D5197 or EPA TO-17
Trichloroethylene	Lubricants, varnishes, paint removers, adhesives and typewriter correction fluids and some bleach household products and other cleaning agents.	(and includes Total VOC)
Terpenes	Perfumery products, deodorizers and cleaning products, air fresheners, deodorants, fragrances and shampoos. Terpenes can also originate from furniture or flooring made from pine wood.	
Volatile Inorganic Compounds (VICs)		
Carbon monoxide (CO)	Indoor sources: defective cooking and heating devices, fireplaces, tobacco smoke and vehicle gases from attached garages. Outdoor sources: dense traffic or high industrialized districts.	Real-time direct reading instruments
Nitrogen oxides (NO _x)	Indoor sources: gas appliances like stoves, ovens or heaters. Outdoor sources: power generation, industries and traffic.	
Ozone (O ₃)	Photocopier machines, laser printers and other electronic devices with high voltage. Outdoor	

	sources: photochemical reactions in the presence of VOC, NO _x and UV light.	
Carbon dioxide (CO ₂)	Occupants producing CO ₂ as well as fireplaces and some cooking and heating devices.	
Radon	Radon is a radioactive gas that is released through the decay of radium in soils and rocks and enters indoor air spaces of buildings or other enclosed locations.	Real-time devices using alpha-particle sensitive material

78 ¹ (WHO 2015; (Rösch et al. 2014)

79 ² (International WELL Building Institute, 2019)

80

81

82 ***1.2 Prevention and Treatment of Indoor Air Pollution***

83 Maintaining a healthful indoor environment is increasingly important as buildings are
84 progressively sealed to obtain energy cost savings in building heating, ventilation and air
85 conditioning (HVAC) or due to safety regulations. A comprehensive understanding of indoor
86 air pollutants (type, concentration and variability in space and time) is relevant for the
87 development of effective control strategies both in terms of prevention and active abatement.
88 Prevention should be considered as the first step for improving IAQ and therefore, some
89 measures have recently been implemented to eliminate certain pollutant sources. The European
90 Directive 99/77/EC restricted harmful construction materials (e.g. asbestos) and products
91 containing hazardous components (e.g. halogenated pesticides) and workplace and public
92 places now commonly ban smoking in many countries. Such control at the source is achievable
93 when sources are known, whereas new hazardous substances are recurrently detected. It is
94 therefore technically difficult and economically exorbitant to completely prevent indoor air
95 pollutants at all time (Guieysse et al. 2008; Luengas et al. 2015). Additionally, increasing
96 concern with greenhouse gas emissions has led many countries to commit to zero energy
97 buildings and to enhanced energy performance during major renovations of existing buildings
98 (European Directive 2010/31/EU). New building design standards such as Passivhaus involve
99 well-insulated and sealed construction, which removes or reduces natural ventilation,

100 increasing the risk of gas and particulate indoor air pollutants accumulation (Broderick et al.
101 2017). This conflict between energy efficiency and IAQ standards points to an increased need
102 for development of effective in-situ indoor air purification systems.

103 Active purification units can be installed to lower or eliminate hazardous levels of indoor air
104 contaminants. Mechanical and electronic filtration as well as adsorption and ozonation
105 comprise most systems commercially available. These physical-chemical technologies have
106 been traditionally installed as portable units or as part of the central heating and ventilation
107 system (Luengas et al. 2015; González-Martín et al. 2021). However, these systems still present
108 multiple drawbacks. For instance, the simplest and most used method for PM removal is
109 mechanical filtration, which is based on circulating air through a fibrous material that retains
110 particulate pollutants. However, frequent filter replacements are required to maintain the
111 capture efficiency and prevent the re-emission of particulate pollutants. On the other hand,
112 electrical filtration attracts and retains negatively charged particles on a plate of opposite
113 polarity. Unfortunately, by-products such as ions, ozone or other compounds may be generated
114 during electronic filtration (Luengas et al. 2015; Hubbard et al. 2005). Adsorption involved the
115 retaining of pollutants on a surface and happens because all molecules employ attractive forces,
116 especially molecules at the exterior of solid materials (e.g. pore walls of activated carbon), and
117 these surface molecules seek to adhere to other molecules. The exact nature of the bonding
118 depends on the details of the species involved and the adsorbent materials such as activated
119 carbon, zeolites, alumina, silica gel and polymers. The adsorption process may be a
120 combination of a physical processes, such as the weak van der Waals forces, or chemical
121 processes, with the characteristic of covalent bonding (Luengas et al. 2015). Regular
122 replacement of adsorbent materials is required to prevent pollutants re-emission and to
123 maintain efficiency levels. Potentially harmful microorganisms can be also accumulated and
124 re-emitted. Finally, electronic ozonation relies on the generation of O₃ from ambient O₂ by

125 high-voltage discharge or UV radiation. However, Luengas et al. (2015) also found that despite
126 the abatement efficiencies of electric ozonisers are being superior to other physical-chemical
127 methods, VOCs and VICs can react with O₃ (a strong oxidant) during ozonisation and form
128 hazardous secondary pollutants. In addition, health issues may arise from potentially toxic
129 indoor levels of O₃, which has a typical exposure limit of only 0.1 ppm_v for 8 hours (Luengas
130 et al. 2015; Hubbard et al. 2005; Chen et al. 2005)

131 This paper presents the recent research findings on biological indoor air purification methods
132 as a 'green' alternative to physical-chemical methods for improving indoor air quality, with
133 emphasis on the recent advances, current challenges, and opportunities for further
134 development.

135

136 **2 BIOLOGICAL AIR PURIFICATION METHODS**

137 ***2.1 Biological Processes Relevant to Biological Indoor Air Purification***

138 Biological air purification methods eliminate or transform gas pollutants through the action of
139 microorganisms or plants, for which the pollutants serve as an energy and/or carbon source for
140 the built-up of new cell material (cellular replication) and cell maintenance. The pollutant
141 biodegradation processes involved are generally enzymatic oxidative, where microorganisms,
142 primarily bacteria, are the main catalyst. Biodegradation of air pollutants typically takes place
143 by mixed microbial communities thriving in a complex ecosystem (Kennes C, Veiga, 2013;
144 Soreanu and Dumont, 2020). The microorganisms are mostly embedded in a biofilm, a moist
145 film of cells confined within an exopolysaccharide matrix that adheres to a surface, protects
146 microbial communities from extreme or changing conditions and enhances communication
147 (e.g. quorum sensing) among them. The mixed microflora living in the biofilm consists of
148 pollutant degraders, competitors and predators, which typically have significantly different
149 properties from free-living bacteria of the same species such as an increased resistance to

150 extreme contaminant concentrations. Both bacteria and fungi are microorganisms capable of
151 biodegrading common indoor air pollutants, while photosynthetic microorganisms such as
152 cyanobacteria and microalgae (referred to herein as microalgae) can also fix CO₂ (Cervera and
153 Gomez, 2015). Bacteria typically display relatively high growth and pollutant biodegradation
154 rates, high tolerance to toxicity and often require a neutral pH (6-8) and high water-activities
155 (> 0.9) (Hernández et al. 2010). In contrast, fungi are more tolerant to low moisture contents,
156 low pH and nutrient limiting conditions. While bacteria remove most efficiently hydrophilic
157 pollutants from the air, fungi perform better with hydrophobic compounds. On the other hand,
158 microalgae can convert CO₂ into biomass through oxygenic photosynthesis, while also being
159 capable of improving indoor air quality by degrading hydrocarbons as microalgae can have
160 autotrophic (photosynthetic), heterotrophic and/or mixotrophic metabolisms.

161 The large variability in type and concentration of indoor air pollutants, because of the periodic
162 occurrence of pollution events (cleaning/polishing, use of air fresheners, cooking, painting,
163 smoking, etc.) and the random introduction of new pollution sources such as new electronic
164 devices, furniture, etc., requires treatment using microorganisms with a large functional
165 versatility and robustness. In this context, metabolically versatile large-genome
166 microorganisms should play a key role on biological indoor air treatment. The large genome
167 of these microorganisms includes many accessory genes encoding active substrate transport,
168 environmental sensing, multiple catabolism, stress response and secondary metabolisms, which
169 confers them with the ability to carry out many non-essential activities related to substrate
170 accession and stress response. These properties are critical to colonize and survive in complex
171 and variable environments (Guieysse and Wuertz 2012). The size of the genome is typically a
172 respectable indicator of metabolic adaptability in bacteria, since the genome of prokaryotes
173 holds a low quantity of non-coding genes and its coding density is rather constant. Bacteria
174 with a genome size > 5 Mbp are often considered large-genome microorganisms, which

175 correspond typically to aerobic mesophilic bacteria. Large genomes typically host a large and
176 effective portfolio of enzymes capable of sensing, accessing and simultaneously metabolizing
177 a broad range of pollutants at trace level concentrations ($\mu\text{g-ng L}^{-1}$) which would be particularly
178 relevant for indoor air treatment.

179

180 ***2.2 Biological Based System Design Configurations***

181 Indoor applications often limit the size of the indoor air purifier. Plant-based (botanical)
182 biofiltration has received recently increased attention due to its economic, environmental and
183 social benefits, including psychological impacts by botanic plants in an indoor space. The
184 relatively large air purifier to room volume ratio typically required for current botanical indoor
185 air systems restrains the development and implementation of biological air purification
186 systems (Guieysse et al. 2008). Therefore, the design of compact indoor air purifiers with high
187 effectiveness is a challenge for biological based systems to establish a healthy indoor
188 environment, which requires a more complete treatment of air; that is not only VOCs, but all
189 other pollutants including CO₂, PM, CO and NO_x.

190 Different system design configurations or combinations of different biological treatment
191 approaches (Figure 1) may overcome this constraint and potentially other challenges such as
192 mass transfer limitations, bioavailability, guaranteed microbial safety, control of indoor air
193 relative humidity, and CO₂-removal, while being simple and robust enough to provide long-
194 term sustainable economic functionality with minimal maintenance.

195 **<FIGURE 1>**

196

197 **Plant Based Systems** - The treatment of pollutants using plants (phytoremediation) is mature
198 and often applied for remediation of contaminated soil and water polluted with organic
199 pollutants such as hydrocarbons. Biotreatment of indoor air using potted plants has been

200 extensively studied and all plants tested were shown to be capable of removing VOCs from
 201 indoor air (Wolverton 1997; Wood et al. 2006; Liu et al. 2007; Yang et al. 2009; Irga et al.
 202 2013; Pacheco-Torgal et al. 2015). A comprehensive overview of the research on botanic plants
 203 related to indoor air quality is provided by Irga et al. (2018). While potted plants are considered
 204 passive systems that depend on the diffusion of pollutants (relatively slow for the indoor air
 205 pollutant low concentrations, especially in spaces without forced air circulation), active
 206 biotreatment systems use active ventilation (fans) to improve the removal capacity. Plant-based
 207 biotrickling filters (PBTfFs) are active biotreatment systems containing hydroponic plants
 208 growing in vertical panels that eliminate maintenance difficulty related to potted plants in soil
 209 as further discussed by Soreanu et al. (2013). Although some VOCs present in indoor air can
 210 directly be taken up and further metabolised by plants, the VOCs are more extensively removed
 211 by microorganism mostly present in the rhizosphere around the roots of the plants (Pacheco-
 212 Torgal et al. 2015). However, the removal of carbon dioxide, sulfur dioxide, nitrogen dioxide
 213 and ozone appear to be partially or solely plant facilitated (Fikiey et al. 1981; Pacheco-Torgal
 214 et al. 2015; Oh et al. 2011; Torpy et al. 2014a). These VICs are generally taken up by the plant
 215 stomates (gaseous compound exchange pores) during daylight hours (Pacheco-Torgal et al.
 216 2015).

217 **Table 2: Overview of biological system studies for indoor air purification.**

System Design	Airflow (passive/active)	Summary of Results	References
Potted Plant	Passive	VOC removal capacity has successfully been tested for about 200 plant species in about 50 studies. The VOCs most investigated were BTEX and formaldehyde, but some studies included acrylonitrile, trichloroethylene, methanol, ethylhexanol, octane and α -pinene. Typically, 10-20% TVOC removal has been recorded within one hour for a plant in a 10 L gastight glass jar.	(Irga et al. 2018; Wolverton 1997; Wood et al. 2006 ;Yoneyama et al. 2002; Liu et al. 2007; Yang et al. 2009; Irga et al. 2013 ; Soreanu et al. 2013)

	Passive	Ultrafine particle (PM) reduction is illustrated for nearly all plant species tested. The plant foliage density as well as tree architecture seems most relevant enabling a small (11%), yet statistically significant hydrophobic and hydrophilic PM reduction in homes.	(Stapleton and Ruiz-Rudolph 2018; Weerakkody et al. 2017)
	Passive	VIC removal capacity has been proven for CO ₂ , SO _x , NO _x and O ₃ , although NO _x removal may negatively affect plant health.	(Fikiey et al. 1981; Pacheco-Torgal et al. 2015)
	Active	Enhances the flow of pollutants to the root zone compared to passive systems, increasing the VOC and PM removal capacity. Among them around 50% removal of PM _{2.5} and PM ₁₀ .	(Lohr and Pearson-Mims 1996; Liu et al. 2007; Wang and Zhang 2011; Irga et al. 2017a; Treesubuntorn and Thiravetyan 2018)
Plant-assisted Biotrickling Filter	Active	VOC removal (10 – 75% in a single-pass configuration) proven for BTEX, methylethylketone, formaldehyde, acetone, octane, α -pinene, decane, ethylacetate and ethylhexanol.	(Darlington and Dixon 1999; Darlington et al. 2001; Llewellyn et al. 2002; Llewellyn and Dixon 2011; Mikkonen et al. 2018; Irga et al. 2019)
Biofilter	Active	The removal of multiple typical indoor air pollutants such as toluene and formaldehyde was shown to be higher than 90% in a single-pass configuration. A mixture of 71 VOCs was tested in a biofilter inoculated with yeasts with most compounds removed.	(Ondarts et al. 2012; Prenafeta-Boldú et al. 2019)
Biotrickling Filter	Active	Formaldehyde and BTEX effectively reduced (respectively 100% and 65-93%) in a single-pass configuration.	(Lu et al. 2010)
Membrane Bioreactor	Active	Experiments performed with both microporous and dense-phase membranes provided a proof of concept for different VOCs and odorous compounds.	Van Ras 2005; Lebrero et al. 2013)
Capillary Bioreactor	Active	High removal rates (13 or 17 times greater than those tested biotrickling filters) of methanol and toluene were obtained. Mass transfer capacity was studied with methane as model compound showing removal > 100 g m ⁻³ capillary channel h ⁻¹ .	(López De León et al. 2019; Rocha-Rios et al. 2013)
Photo-Bioreactor	Active	CO ₂ reduction up to 95% was proven alongside with the significant removal of VOCs, NO _x and NH ₃ .	(Soreanu and Dumont 2020)

219 **Biofilters, Biotrickling Filters and Bioscrubbers** - While plants assist to support and sustain
220 the active microbial community in plant-based systems, microorganisms are cultivated
221 differently in conventional biological air treatment techniques, traditionally classified as
222 biofilters, bioscrubbers, and biotrickling filters. Biofilters are systems where both the
223 microorganisms and the water phase are immobilized on the biofilter media. Bioscrubbers are
224 traditionally referred to as two separate reactor compartments with recirculating process water
225 between them. In the first reactor unit, contaminants are absorbed from the pollutant air into
226 the liquid through mass transfer. In the second unit, the dissolved contaminants are biodegraded
227 by microbes suspended in an aqueous culture broth (suspended growth biomass). Biotrickling
228 filters are typically defined as single-stage bioreactor units with a packing supporting the
229 growth of a biofilm through which a liquid containing nutrients is trickled. Thus, the absorption
230 of contaminants from the polluted air to the process liquid and their subsequent biodegradation
231 in biotrickling filters take place in one process unit. The microbes are mostly immobilized on
232 the packing (fixed-film biomass). These conventional gas treatment systems have been
233 developed and applied for many industrial applications since the late 1950s. They are
234 recognised as reliable and economical control techniques for treating gaseous stream with
235 relatively low concentrations of contaminants (Kennes and Veiga 2013; Soreanu and Dumont
236 2020; van Groenestijn and Kraakman 2005; Estrada et al. 2011), but more research is needed
237 at the even lower concentrations common in indoor spaces.

238 **Membrane Bioreactors** – These bioreactors for gas treatment use membranes that separates
239 the contaminant-laden gas stream and the process liquid containing the microorganisms and
240 nutrients required for pollutant degradation. A biofilm of microorganisms develops on the
241 membrane at the liquid side and the air pollutants diffuse from the gas stream through the
242 membrane to the biofilm. Biological based air treatment systems using membranes are of
243 interest for indoor air purification because of its multiple advantages compared to their

244 biological counterparts. The first advantage is that the biofilm does not interfere with the gas
245 stream and that the biomass optimal environmental conditions can be controlled in terms of
246 water activity and nutrient supply. Secondly, this configuration eliminates the risk of
247 unintentionally drying out of the biofilm and prevents moisture and microorganisms being
248 released from the air purification system with certain membrane types (e.g. thin dense-phase
249 dimethylsiloxane membranes). Finally, hydrophobic indoor air pollutants with high membrane
250 permeability may be treated more effectively as further discussed by Kumar et al. (2008) and
251 Studer and Von Rohn (2008).

252 **Capillary Bioreactors** – Capillary reactors are small channels where capillary forces become
253 dominant relative to gravity forces. The small channels (capillary channels) can be used to
254 create a train of alternating liquid and air bubbles flowing co-currently. The optimal flow
255 pattern is called segmented flow or Taylor flow in a specific gas to liquid ratio and alternating
256 frequency. The flow pattern creates internal liquid circulation that enhances the mass transfer
257 between the gas and the liquid phase. This plug flow contains no macromixing or axial
258 dispersion while providing local internal mixing with minimum energy due to the low pressure
259 drop over the channels (Kreutzer et al. 2005; Rocha-Rios et al. 2013). Capillary reactors can
260 combine enhanced mass transfer at relatively low pressure drop, both important factors usually
261 influencing the cost of air treatment systems. Laminar gas flow conditions are typically
262 observed in biological gas treatment system and therefore highly dependent on diffusion rate
263 of molecules (rather than advection), which is especially slow in liquids. Improving advection
264 (e.g. by intense mixing) will increase the mass transfer especially through the liquid phase. The
265 flow characteristics in capillary channels has been researched in the framework of chemical
266 reaction optimisation (Kreutzer et al. 2005; Shao et al. 2010) and recently also studied for
267 biological gas treatment (Rocha-Rios et al. 2013; López De León et al. 2019).

268 **Photobioreactors** – Microalgae in photobioreactors fix CO₂ using light energy with an
269 associated release of oxygen. It represents a viable solution for the continuous control of
270 elevated CO₂-concentrations of indoor air in spaces with many occupants such as office
271 buildings or schools. Microalgae can convert CO₂ into biomass through photosynthesis, while
272 also being capable of improving air quality by degrading or bioconverting hydrocarbons as
273 some species exhibit a versatile autotrophic, heterotrophic or mixotrophic metabolism. Both
274 microalgae and botanical plants are based on oxygenic photosynthesis, but microalgae are
275 much more efficient in converting light into biomass due to their small cells and suspended
276 growth in a liquid environment (Brennan and Owende 2010). With over 50,000 species of
277 microalgae identified, the knowledge on microalgal cultivation has notably increased recently,
278 many microalgae studies being carried out with the focus on biofuel production, wastewater
279 treatment and food production. The most used microalgae species are *Spirulina (Arthrospira)*,
280 *Chlamydomonas*, *Scenedesmus* and *Chlorella sp.* because they are known to be robust against
281 contaminants, being easy to grow, nontoxic, edible and of which the first now is commonly
282 used as a dietary supplement. Key environmental conditions for CO₂-fixation by microalgae
283 are light availability and temperature. CO₂-conversion increases with the intensity of light
284 provided until the microalgae culture becomes light saturated, corresponding to light intensities
285 of about 10% of the summer and 20% of the winter light brightness for most microalgae
286 (Richmond and Hu 2013). The optimal temperature commonly ranges from 15 to 30 °C (Zhou
287 et al. 2017). Photo-bioreactors have been studied and tested for different applications including
288 flue gas treatment from coal power plants (Mendoza et al. 2013), combined treatment of
289 exhaust gas and wastewater (Anbalagan et al. 2017), biogas upgrading (Rodero et al. 2019) and
290 fully enclosed spaces such as permanently occupied space stations (Niederwieser 2015;
291 Soreanu and Dumont 2020). The applicability of microalgal systems for indoor air treatment

292 is underdeveloped, as common conditions and the concentrations of indoor air pollutants are
293 far from those of industrial systems.

294
295 ***3.2 Examples of Commercial Bio-based Indoor Air Systems***

296 The market of biological purification systems for improving indoor air quality is rapidly
297 expanding, but only a few commercially available plant-based systems have proven to have a
298 high and long-term removal efficiency for relevant VOCs such as formaldehyde (Torpy et al.
299 2014b). Green walls are not necessarily installed to control the indoor air quality but are an
300 illustration of interior landscaping and are popular in office buildings mostly because they
301 enhance the aesthetics and may help align company brands with dedication to sustainability.
302 Figure 2a shows an example of an aesthetic green wall consisting of moss requiring no extra
303 light or regular watering while adsorbing sound (Ambius 2020).

304 The availability of multiple design concepts using botanical and microbial approaches is clearly
305 promising and deserves to be further investigated. Different system types may have to be
306 developed, ranging from personal mobile air purifiers (Andrea 2020), as shown in Figure 2b,
307 to larger building air purifiers fully integrated with the HVAC system of building, as shown in
308 Figure 2c, where air is circulated through a vertical green wall consisting of plants in a porous
309 rooting material (Nedlaw Living Walls 2020). Maintaining optimal moisture control is critical
310 and automated moistening when a fan pulls air through the plant root zone would be important
311 as incorporated by Phytofilter in their active potted plant air purifier system (Phytofilter 2020).
312 Remote monitoring using sensors and management performing plant care including providing
313 water, light and ventilation may be used to facilitate the management of active green walls in
314 its specific building environment. These basic functions may be combined with a series of
315 critical conditions such as temperature and relative humidity (RH) as well as carbon dioxide
316 and certain VOCs as proposed and tested by Liu and colleagues (2018).

317 Hybrid systems still need to be developed for a more complete treatment of indoor air; that is
318 not only VOCs, but all other pollutants including CO₂, PM, CO and NO_x, so that it can fully
319 contribute to a better indoor air quality.

320

321 <FIGURE 2>

322

323 **3 CHALLENGES FOR BIOLOGICAL INDOOR AIR PURIFICATION**

324 Conventional chemical-physical methods for indoor air purification are, besides the
325 shortcomings mentioned above, also disadvantaged by the diversity and variability in space
326 and time of pollutants in indoor environments. This is an opportunity for biological systems,
327 which can make use of diverse and adaptive microbial communities capable of removing
328 pollutants. These adaptive characteristics have been consistently observed in industrial
329 biofilters successfully treating low concentrations of, often odorous, emissions from different
330 sources with varying concentrations over time (Kennes and Veiga 2013; Soreanu and Dumont
331 2020; van Groenestijn and Kraakman 2005; Estrada et al. 2011). Although microorganisms-
332 based biological abatement has mostly been focussed on the treatment of VOCs and odours,
333 indoor botanical plants have been shown capable of removing also some VOCs (Irga et al.
334 2018), nitrogen dioxide (Coward et al. 1996; Yoneyama et al. 2002), sulfur dioxide (Lee and
335 Kim 1999), as well as ozone (Fikiey et al. 1981) and even particulate matter as demonstrated
336 by Lohr and Pearson-Mims (1996). While biological methods for the abatement of industrial
337 off-gases are reasonably developed, the potential for indoor pollutant abatement is currently
338 emerging (Pacheco-Torgal et al. 2015; Vallero 2011).

339 Biological indoor air purifying systems are considered a ‘green’ technology that can boost the
340 eco-efficiency of smart-buildings and bring extra advantages as to aesthetics and Indoor
341 Environmental Quality (IEQ). This may only be possible when overcoming some of the
342 challenges such as mass transfer limitations, bioavailability of low pollutant concentrations,

343 guaranteed microbial safety, elevated indoor air relative humidity, and incorporate CO₂-
344 removal. In addition, system economic feasibility and reliability are a prerequisite to enable
345 practical implementation and sustain guaranteed performances.

346

347 *3.1 Bioavailability and Mass transfer Limitations*

348 Bioavailability is an expression of the fraction of the pollutant mass present in a compartment
349 that has the potential of being assimilated by the organism (Vallero 2011). Contaminants with
350 high solubility in water such as alcohols and aldehydes are readily removed from the air by
351 biological air filtration, while other pollutants such as long-chain hydrocarbons, with low
352 aqueous solubility, may require an enhancement of the biofiltration performance. In addition,
353 the low concentrations of indoor air contaminants typically cause increased mass transfer
354 limitations and thus a reduced bioavailability for effective removal. One major downside of
355 biological indoor air cleaning systems is the large value of the biological purifier volume to
356 room ratio required due to the high footprint cost of buildings. For instance, an indoor air
357 biological purifier for a room with dimensions 8 m × 5 m × 2.5 m (L × W × H) would require
358 a relatively large volume of about 1100 – 3300 litres, assuming the typical gas residence time
359 of 10 - 30 seconds of industrial applications for a 95% removal efficiency and an room air
360 exchange rate of four per hour, which equals a purifier volume of 1 – 3% of the total room
361 volume (Guieysse et al. 2008). Therefore, it may be challenging for biological based systems
362 to obtain effective indoor air cleaning performances that is reasonably compact in size.

363 Besides overcoming mass transfer limitations, biological indoor air purification systems also
364 need to be able to sustain enough microbial activity under conditions of trace pollutant
365 concentrations. Pollutants are utilized by microbes to obtain energy and synthesize new
366 biomass, but when pollutant concentrations are low such as in indoor air, and due to the
367 variability may decrease even lower, a critical level can be reached below the production of

368 new microbial cells is not good feasible. Attached growth (biofilms) are known to be able to
369 support biomass under changing conditions including carbon and energy source depletion. A
370 typical result of the biomass switching into starvation mode is the removal of cells from a
371 biofilm (dispersal) mediated by the decrease in bacterial adhesiveness and biodegradation of
372 the biofilm matrix (Madigan et al. 2017). Indoor air pollutants may not always be high enough
373 in energy and/or carbon content essential for cell maintenance and growth, and co-metabolism
374 may be required for an effective pollutant removal. As low concentrations are common in the
375 environment, it is relevant to note it is known that microorganisms can develop survival
376 strategies, such as quickly increasing their substrate affinity, changing to metabolic
377 mineralisation or accumulating the limiting substrate when possible (Madigan et al. 2017;
378 Kovárová-Kovar and Egli 1998). The evaluation of pollutant biodegradation at the $\mu\text{g L}^{-1}$ - ng
379 L^{-1} level consistently showed that microorganisms can mineralize pollutants down to pg L^{-1}
380 and that the so-called affinity constant determined at higher concentrations are at trace levels
381 differently (Subba-Rao et al 1982).

382

383 ***3.1.1 Fundamentals of Gas-Liquid Mass Transfer in Bioreactors***

384 Two processes are occurring simultaneously in biological air purification systems. The first
385 step is the transfer of the gas pollutant from the air to the biofilm or aqueous cultivation broth,
386 after which the second step, biological degradation of the pollutants, can occur. These
387 processes of mass transfer and biological degradation occur almost instantaneously, typically
388 rate limited by either the mass transfer or the bio-kinetics. Since biological gas treatment
389 techniques are typically most practical at relatively low concentrations, at least partial mass
390 transfer limitation can be expected (Kennes and Veiga 2013; Kraakman et al. 2011). This
391 limitation is further triggered by the hydrophobic nature of a fraction of indoor air pollutants
392 (CO , NO_2 , aromatic and aliphatic hydrocarbons, cycloalkanes, halocarbons, terpenes, etc.),

393 which hinders their mass transport from the air emission to the micro-organisms which are
394 often surrounded by a moist (hydrophilic) environment.
395 Mass transfer of gas pollutant or oxygen can be limiting and may occur at different locations
396 of the reactor such as near the liquid/biomass interface, inside the biofilm, or near the outlet of
397 the reactor due to the low residual concentrations in the gas phase. Mass transfer can be
398 described with the theory of Lewis and Whitman (Lewis and Whitman 1924). This two-film
399 model uses two phases (e.g. air and water) that have not reached equilibrium as to Henry's law.
400 Such an equilibrium only exists at the direct air-water interface, while the target pollutant
401 transfers to or from this interface with a specific rate. This transfer rate is determined by the
402 pollutant properties and the air and water properties and is defined with mass transfer rate
403 coefficients. The overall mass transfer coefficient (k_{overall}) can be simplified to a mass transfer
404 rate coefficient of different parts of the system, as shown in Eq. (1).

$$405 \quad 1/k_{\text{overall}} = 1/k_G + 1/k_L + 1/k_B \quad (1)$$

406 Where k_G , k_L and k_B are the mass transfer rate coefficients for respectively the gas phase, the
407 liquid phase and the biofilm. Mass transfer coefficients are not only determined by physical–
408 chemical properties of the pollutant and the air and liquid media, but also the reactor type and
409 its operating conditions (Kim and Deshusses 2008; Dorado et al. 2009; Kraakman et al. 2011).
410 Mass transfer limitation have been observed in the gas phase under laminar gas flow conditions
411 and low pollutant concentrations (van Ras et al. 2005) as well as between the liquid phase and
412 biofilm under turbulent conditions and high pollutant concentrations (Estrada et al. 2014).
413 However, under the most common conditions the mass transfer resistance in the gas and the
414 biofilm can be expected to be negligible. Therefore, the overall mass transfer rate per reactor
415 volume R ($\text{g m}^{-3} \text{s}^{-1}$) from the gas to the liquid phase may be described by Eq. (2) (Koch 1990).

$$416 \quad R = k_L a (C_G / H - C_L) = (D_{AL} / \delta_{\text{film}}) a (C_G / H - C_L) \quad (2)$$

417 where D_{AL} is the gaseous pollutant diffusivity in the liquid ($m^2 s^{-1}$), H the Henry coefficient
418 (dimensionless) and δ_{film} the liquid film thickness (m). C_G the gas pollutant concentration (g
419 m^{-3}) and C_L the liquid phases pollutant concentrations. The term k_{La} (s^{-1}) is a volumetric
420 coefficient that determine the mass transfer rate by factors independent of the concentration,
421 where k_L is the liquid phase mass transfer coefficient ($m s^{-1}$) and “a” is the specific gas-liquid
422 interfacial area ($m^2 m^{-3}$).

423 Pollutant mass transfer takes place through both diffusion and advection. Diffusion is the
424 random Brownian motion of individual pollutants in a medium, while advection is the larger-
425 scale motion of the medium containing the pollutants. The mass transfer enhancement in gas–
426 liquid pollutant exchange reactors usually requires an intensification in power consumption
427 through boosted mixing or increased turbulence. Unlike turbulent systems (e.g. aerated stirred
428 tanks) where the suspension of air-liquid-cells can be assumed a well-mixed system with mass
429 transfer expected to be comparable throughout the reactor, in laminar contactors (e.g. biofilters
430 for air treatment) heterogeneities caused by pollutant gradients as well as irregular moisture
431 content and biomass concentrations may create limitations for optimum mass transfer.
432 Fundamental processes like mass transfer in the heterogenic biological air purification systems
433 are more difficult to describe mathematically, which is desired to fully understand the rate-
434 limiting steps in a system and eliminating a design of the bioreactor that is primarily based on
435 empirical experience (Popat and Deshusses 2010).

436

437 ***3.2 Microbial Safety Challenges***

438 The potential formation of microbial air pollutants (harmful bioaerosols) may be considered a
439 potential drawback of biological air purification systems. Bioaerosols refer to both living and
440 non-living components, for example pollen, dust, spores, mites, allergens, viruses, bacteria and
441 fungi. Bioaerosols contaminate the air via pets, mould, insects, sick occupants, (de)humidifiers

442 or improper air filters and may be attached to dust particles. Potted plants may also contribute
443 to indoor bioaerosols (Soreanu and Dumont 2020). Air contaminated with bioaerosols may
444 cause allergic reactions and infectious diseases. In addition, indoor environments can generate
445 conditions that can amplify certain microbial species including pathogens and may include
446 aerosols from plumbing, wetted surfaces or damp indoor environments. Conditions supporting
447 the accumulation of stagnant water is for example known to allow the proliferation of
448 *Legionella*. *Legionella* growth requires, besides stagnant water, a carbon source and a
449 temperature around 38°C (Grimes 1991). However, although the proliferation and transmission
450 of *Legionella* is limited under typical indoor space temperatures (Burchett et al. 2007),
451 increased temperatures may occur as a result of lighting systems or integrated water
452 recirculation pumps. Hence, an indoor air purifying system needs to eliminate the risks of
453 nuisance due to the release of harmful bioaerosols.

454 Normally fungal amplification in indoor spaces such as bathrooms is primarily caused by
455 elevated room humidity levels above about 80 % RH (Adan and Samson 2011). Biological
456 systems require a high water-activity near the active zones to sustain microbial activity, which
457 holds the risk of increasing the RH of the air treated in indoor rooms, where RH of 30- 60% is
458 common for comfort. In addition, too many plants in one room potentially may increase the
459 RH beyond the comfort zone and entails an associated risk due to mould development
460 (Darlington et al. 2001; Llewellyn et al. 2008).

461 In addition, spores can be produced which may involve reproductive cells of plants, fungi or
462 algae as part of their life cycle as well as bacterial cells adapted in adverse conditions for
463 dispersal and/or survival. Spores are hardy and typically inactive and may require more
464 aggressive conditions to control or contain compared to active microbial cells.

465 Besides the potential for the proliferation of microbial species and spores, the amplification of
466 microbial products can also be a source of diseases and may involve volatiles or other microbial

467 metabolites including toxics. For example, many indoor fungi produce metabolites that can
468 induce respiratory or allergic diseases upon exposure. Several hundred of mycotoxins have
469 shown to be potentially harmful with respect to food contamination (Alshannaq and Yu, 2017),
470 while the effects of inhalation of indoor-related mycotoxins are far less clear at this moment
471 (Korkalainen et al., 2017). These microbial toxins are typically odourless and can be released
472 even from dead microbial material.

473 ***3.2.1 The Risk of Microbial Emissions***

474 Relatively dry conditions in a biological air treatment system stimulating the growth of fungi
475 may increase the risk of fungal spore release (Pasanen et al. 1991), a focus of interest because
476 they may possess multiple hazard potential for human health (allergies, toxicosis, infections).
477 Irga et al. (2017b) assessed an active green wall and concluded that active botanic biofilters are
478 unlikely to release fungi to hazardous levels if the system is maintained properly. Darlington
479 and colleagues did observe increased fungal spores in the indoor air during the first year of
480 operation of a moss biofilter using lava rock a support media but remained within typical
481 reported fungal counts of indoor spaces such as flats containing house plants (100–200 CFU
482 m^{-3}), considered a healthy level (Darlington et al. 2000). This was more recently confirmed by
483 Fleck et al. (2020) who concluded that fungal spores are emitted in concentrations well below
484 WHO safety guidelines from active green walls when operated under well monitored
485 conditions. Some organisms present in buildings may have infectious potential, but some are
486 not highly virulent and rarely cause infection in people with healthy lungs and healthy immune
487 systems. Moreover, a study by Ibanga et al. (2018) on an industrial biofilter treating odorous
488 foul air from a Material Recovery Facility showed that organic biofilters containing woodchip-
489 based media can remove bioaerosols. The average removal of the four clusters of bioaerosols
490 studied, total fungi, total mesophilic bacteria, *Aspergillus fumigatus* and Gram-negative
491 bacteria, accounted for 71%, 68%, 70% and 50%, respectively, regardless of the gas contact

492 time tested in the biofilter. Although the emitted bioaerosols concentrations from the biofilter
493 still exceeded ambient background concentrations, the results confirm earlier studies by Becker
494 and Rabe (1997) that measured a 69% and 89% reduction in *Aspergillus fumigatus* spores from
495 an industrial biofilter treating foul air from an indoor composting facility. Although the risk of
496 elevated spore concentrations in the treated air from biological systems may seem limited (and
497 not much more than background indoor air concentrations), there is however still too little data
498 available in the case of faulty equipment or unforeseen upsets.

499 The risk of elevated concentrations of secondary products like volatiles or other microbial
500 metabolites including toxics in biological air purification systems is currently unclear and
501 would require further study. Endotoxins and mycotoxins may also be more difficult to remove
502 in biological filters. A study evaluating the effectiveness of various types of biofilter media to
503 purify the ventilation air from a chicken hatchery room reveal that endotoxic was only slightly
504 removed in a typical biofilter. The endotoxic removal ranged from 11 to 51%, while dust-
505 particles were moderately (about 82%) removed and gram-negative bacteria highly (about
506 99%) removed (Tymczyna et al., 2007).

507 Human health safety parameters need to be established for indoor air purification applications
508 that allow the comparison of physical-chemical and biological treatment systems. As example
509 could be used the “tolerable” level of 10^{-6} DALYs per person per year which was introduced
510 in the WHO Guidelines for Drinking-Water Quality (2004) by analogy to the established
511 “tolerable lifetime risk” for carcinogenic chemicals of 1 case of cancer per 100,000 exposed
512 people over a 70-year lifetime. The metric DALYs for a disease or health condition are
513 calculated as the sum of the Years of Life Lost (YLL) due to premature mortality in the
514 population and the Years Lost due to Disability (YLD) for incident cases of the health
515 condition.

516 While the focus should always be on reducing the exposure to harmful microbes, there is also
517 an opportunity to encourage introduction of beneficial microbes. Beneficial microbes, so-called
518 environmental probiotics, may provide a protection against opportunistic pathogens or their
519 expansion. Indoor airborne microbes may also be a source and passage of the diversity of
520 human microbes, the so-called the human microbiome, that assists for example in providing
521 nutrients for our cells and benefits the programming of our immune system and thus potentially
522 prevent or reduce the risk of suffering diseases (The National Academies of Science 2017).
523 Nevertheless, these hypothetic connections on human health by plants-associated microbes in
524 biological systems such as green walls remain to be fully explored.

525

526 ***3.3 CO₂ as Indoor Air Pollutant***

527 Because occupants produce CO₂, its concentration in indoor spaces occupied by humans and/or
528 animals is higher than the concentrations outdoors. Concentrations above 1,000 ppm_v are
529 defined as an indoor air pollutant by the American Society of Heating, Refrigerating and Air-
530 Conditioning Engineers (ASHREA 2019) and in most green building certification schemes
531 threshold CO₂ concentrations are considered (Wei et al. 2015). With the growing trend of
532 constructing airtight buildings to provide energy consumption savings, the difference in
533 indoor–outdoor CO₂ concentration increases as the ventilation rate per person decreases (i.e.
534 rate of outdoor air supply to an indoor space). With the current global average outdoor
535 concentration of about 400 ppm_v, CO₂ levels in urban areas can be expected to be higher
536 (Persily 1997) and CO₂ concentrations inside occupied indoor spaces typically vary from
537 outdoor levels up to several thousand ppm_v (Persily et al. 2008). Elevated CO₂ concentrations
538 in office building can be expected especially in the afternoons and in meeting rooms where
539 important decisions are sometimes made.

540 Several studies have shown that human performance is directly influenced by the CO₂
541 concentration. Indeed, a decline in workplace productivity and student academic performance
542 have been shown with elevated CO₂ levels (Satish et al. 2012; Bakó-Biró et al. 2004; Seppänen
543 et al. 2006; Shaughnessy et al. 2006). Satish et al. (2012) showed a moderate but statistically
544 significant adverse effects of 1,000 ppm_v CO₂ in six out of a nine scales of human decision-
545 making performance and a large reduction in seven scales at 2,500 ppm_v when compared to a
546 baseline level of 600 ppm_v. Two previous studies with only 10 participants showed that they
547 performed proofreading significantly more poorly at CO₂ concentrations of 4,000 ppm_v and
548 marginally but significant differences were recorded at 3,000 ppm_v versus 600 ppm_v. The
549 difference in reading performance was observed in the errors found, not in the reading speed.
550 The quality of sleep is also affected by the CO₂ concentration in the sleeping room, alongside
551 the freshness of the sleeping room air perceived and the next day performance (Strøm-Tejsen
552 et al. 2016). In addition, negative symptoms like dry eyes, sore throat, nose congestion (related
553 to the mucous membranes) and drowsiness, short breath, cough and panting (related to the
554 lower respiratory tract) have been associated with elevated CO₂ levels (Erdmann and Apte
555 2004).

556 Although many elevated CO₂ concentrations are the result of insufficient supply of ambient
557 outside air as per current professional standards, even the ventilation rates in the leading
558 ASHRAE standard (ASHREA 2019) can result in CO₂ concentrations higher than 1,000 ppm_v
559 in generously occupied spaces (Satish et al. 2012).

560

561

562 **4 EFFECTIVE BIOLOGICAL INDOOR AIR PURIFICATION SYSTEMS**

563 ***4.1 Overcoming Bioavailability and Mass transfer Limitations***

564 To overcome mass transfer limitations, several strategies may be considered. Enzymatic
565 transformations outside the cell to facilitate mass transfer have been suggested as a microbial
566 mechanism to explain extremely high performances of conventional biofilters treating
567 hydrophobic pollutants (Miller and Allen 2005). In addition, direct pollutant uptake from the
568 air under dry conditions has been shown in fungal-based systems (Arriaga and Revah 2005;
569 Vergara-Fernández et al. 2006; Lebrero et al. 2016). The use of bioactive polymeric coatings
570 that hold a high affinity for the target pollutant has been shown to enhance pollutant uptake
571 from the air emission in packed bed bioreactors, thus avoiding any mass transfer required
572 through the water layer covering conventional biofilms. This biocatalytic approach has been
573 shown with biocatalytic activity being retained for months under growth-limiting conditions
574 (Flickinger et al. 2017; Estrada et al. 2015). Moreover, extracellular metabolites such as
575 biosurfactants can enhance the mass transfer of gas pollutants (Albino and Nambi 2009) or the
576 addition of chemical surfactants as shown by Song et al. (2012).

577 New reactor designs can also provide enhanced mass transfer with minimum power
578 consumption, with the examples of hollow-fibre membrane bioreactors, capillary bioreactors
579 or two-liquid phase partitioning bioreactors. Hollow-fibre membrane bioreactors use
580 membranes that separates the contaminant-laden gas stream from the process liquid containing
581 the microorganisms. Hydrophobic indoor air pollutants with high membrane permeability may
582 be treated more effectively as the hydrophilic barrier (water) can be avoided. Capillary reactors
583 are small channels where capillary forces become dominant relative to gravity forces. These
584 small channels can be used to create a train of alternating liquid and air bubbles flowing co-
585 currently. This flow patron can create internal liquid circulation that enhances the mass transfer
586 between the gas and the liquid phase. Two-liquid phase partitioning bioreactors (TPPBs) are
587 employed with a non-aqueous liquid phase that is water-immiscible and non-volatile (e.g.
588 silicon oil), that exhibits a high affinity for the target gas pollutants and have been developed

589 specifically to overcome limitations of mass transfer (Muñoz et al. 2012). Direct uptake of the
590 hydrophobic target pollutant from the silicon oil by micro-organisms at the oil-water interface
591 is assumed the mechanism for improved performance. A non-aqueous liquid phase may also
592 provide buffer capacity against starvation periods or pollutant surges by providing a VOC
593 reservoir, which in addition may result in enhanced process stability and system reliability
594 (Darracq et al. 2009).

595 Besides overcoming mass transfer limitations, biological indoor air purification systems also
596 need to be able to sustain enough microbial activity under conditions of trace pollutant
597 concentrations. Indoor air pollutants may not always be high enough in energy and/or carbon
598 content essential for cell maintenance and growth, and co-metabolism may be required for an
599 effective pollutant removal. The bioactive zones of plants (i.e. soil, plants roots and plant
600 leaves) can provide extra energy and carbon substrates to sustain overall microbial eco-system
601 stability, while hydroponic substrates may be used as secondary substrate in plant-based
602 systems for biological indoor air purification systems (Guieysse et al. 2008; Soreanu et al.
603 2013).

604

605 ***4.2 Bio-based Hybrid System:***

606 ***4.2.1. Capillary Reactor Combined with a Botanical Green wall***

607 As mentioned before, hybrid systems combining several technologies may be more suitable for
608 effective indoor air purification. A capillary reactor could be combined with an existing vertical
609 green wall so that the process liquid from the capillary reactor is fed to the root zone of the
610 plant in the green wall (Figure 3). The advantage of such a combination is that the benefits of
611 the plant-based green wall such as aesthetics and support of microbial activity (through root
612 exudates produced by photosynthesis that enhances mutual benefit between plant and
613 microbes) are combined with the high mass transfer capacities of a capillary reactor.

614 This hybrid system could be obtained using existing vertical green walls that use a hydroponic
615 substrate with a recirculation water flow so that one liquid stream is combined in the two
616 systems. It is estimated by the authors that the size, materials and labour involved in installing
617 a green wall will be significantly greater compared to adding a capillary reactor. The additional
618 costs would be limited to the capillary reactor that might require an additional liquid flow
619 regulating valve and a separate air ventilation device to control the liquid and air flow through
620 the capillary reactor. This hybrid system could advance existing (mainly aesthetic) green walls
621 and foster HVAC energy savings.

622

623 <FIGURE 3>

624

625 *4.2.2 UV Photolysis-Based Systems*

626 Combining a biological purifier with ultraviolet (UV) photolysis for the removal of bioaerosols
627 may be a simple solution to polish the biologically treated air under all conditions. UV
628 photolysis-based removal of bioaerosols has been proven effective at UV light wavelengths of
629 200 to 280 nm. UV disinfection has been widely used in hospitals and health care facilities,
630 drinking water industry, food industry, wastewater treatment industry and pharmaceutical
631 industry as a rather environmentally friendly technology (Chuaybamroong et al. 2010; Hu et
632 al. 2020).

633 Viruses are most susceptible to UV radiation, while bacteria and fungal spores are more
634 resistant to UV inactivation (ASHRAE 2009). Biofilters post-treatment to control spores and
635 bioaerosols has been investigated by Valdez-Castillo et al. (2019), who reported 70%
636 inactivation efficiency of bioaerosol in a photocatalytic post-treatment with an active catalyst
637 ZnO/Perlite, while Saucedo-Lucero et al. (2014) observed a spore deactivation efficiency of
638 98% from a fungal biofilter with photolytic and photocatalytic post-treatment processes.

639 Alternatively, the use of silver-based nanomaterials known for their antimicrobial properties
640 might be useful as coating or filter fabric in a post-treatment air purification device after a
641 biological purifier (Deshmukha et al., 2018).

642 ***4.2.3. Membrane Bioreactors Combined with a Botanical Green Wall***

643 Bacterial emission from an innovative biofiltration system was studied by Lu et al. (2012),
644 where the active bacteria were immobilised (embedded in calcium alginate gel). Although the
645 air velocity was much lower (0.01 m s^{-1}) than can be expected in building air conditioning
646 systems, an effective formaldehyde biodegradation (69 – 94%) was obtained with no bacterial
647 release detected. Alternatively, dense-phase membranes may be used in biological indoor air
648 purification systems, which have been investigated for space applications and showed the
649 potential for an effective pollutant removal, while eliminating the release of bioaerosols and
650 moisture in the air (van Ras et al. 2005; Soreanu et al. 2013; Kumar et al. 2008; Llewellyn et
651 al. 2008; Kraakman et al. 2007). The use of small membranes through the root zone of botanical
652 plants in a so-called botanical-membrane hybrid biofilter was proposed by Llewellyn et al.
653 (2008). Similarly, a membrane bioreactor using dense-phase membranes could be combined
654 with an existing vertical green wall that uses a hydroponic substrate so that the process liquid
655 from the membrane bioreactor could be fed to the root zone of the plant in the green wall
656 (Figure 4). In such a combination, the benefits of the plant-based green wall such as aesthetics
657 and support for microbial activity are combined with a limited release of bioaerosols and
658 moisture to the ambient air.

659

660 <FIGURE 4>

661

662 ***4.2.4 CO₂ Abatement Using Botanic Plants and Microalgae***

663 The use of biology to remove indoor CO₂ offers opportunities that have received limited
664 research attention, while indoor plants have proven to be able to reduce CO₂ in indoor
665 environments. Oh et al. (2011) and Pennisi and van Iersel (2012) studied the capacity of several
666 indoor plant species to reduce CO₂ and concluded that an impractical large quantity of indoor
667 plants would be needed due the limited photosynthetic rate to significantly reduce the indoor
668 CO₂ concentration. Plant-based green wall technology increases the efficiency of capturing
669 CO₂ compared to passive potted plants, because plant density and thus photosynthetic rate
670 increases, along with the ease to increase the light intensity (Torpy et al. 2014b).

671 Microalgae in photobioreactors devoted to indoor air treatment can reduce CO₂ levels more
672 effectively than higher plants as photosynthetic rates are higher due to their small cell and
673 suspended growth in a liquid medium. Heterotrophic–phototrophic symbiotic relationships
674 such as those observed in plant root systems offer opportunities for a more complete indoor air
675 purification that includes CO₂ removal. Combined algae-bacterial biotreatment systems or
676 mixotrophic algae can offer opportunities for indoor air purification comparable to those found
677 in wastewater treatment (Anbalagan et al. 2017). Up till now, microalgal cultivation has been
678 mainly focussed on green energy and food/fine chemicals production and did not fully explore
679 the potential for the purification of air pollutants. Soreanu and co-workers showed that CO₂
680 can be removed up to 95% at inlet CO₂ concentrations of about 400 ppm_v with a sparged
681 photobioreactor, alongside with the removal of VOCs, NO_x and NH₃ (Soreanu and Dumont
682 2020). Microalgae are capable of biotransforming VOCs, able to use NO_x and indirectly
683 assimilate SO_x (Giordano and Wang 2017). The development of microalgae photobioreactors
684 for indoor air purification may have to explore different reactor configurations, including
685 combinations of traditional tubular algae bioreactors with air lift reactors, membrane
686 bioreactors, capillary reactors or biotrickling filters and deserves further research to explore
687 their full potential in the context of indoor air purification. At this point it should be stressed

688 that the combination of attractive photobioreactors and illumination designs, together with the
689 aesthetic of microalgae culture, represents a competitive advantage of this technology in indoor
690 applications. The microalgae reactor could be combined with an existing vertical green wall
691 that uses a hydroponic substrate so that the process liquid from the microalgae bioreactor could
692 be feed the root zone of the plant in the green wall as illustrated in Figure 5. It is estimated by
693 the authors that the size, materials and labour involved in installing a green wall will be
694 significantly greater compared to adding a photo bioreactor. The additional costs would be
695 limited to the photo bioreactor, additional liquid stream piping and valving and a separate air
696 ventilation device to control the air flow through the photo bioreactor. This hybrid system could
697 advance existing (mainly aesthetic) green walls and could provide CO₂ removal more
698 effectively. The additional light requirements as well as the potential release of excess moisture
699 into the indoor environment brought about by the intensive air sparging typically applied in
700 photo-bioreactors should be considered (Cervera and Gomez, 2015).

701

702 <FIGURE 5>

703

704

705 **5 BENEFITS OF BIOLOGICAL INDOOR AIR PURIFICATION**

706 *5.1 Building Energy Costs Savings*

707 Building ventilation desires the intake of fresh outdoor air, which will need to be heated or
708 cooled in order to meet indoor requirements for temperature and humidity. Ventilation of
709 buildings with ‘fresh’ ambient air only or ventilation in combination with air cleaning methods
710 can be used to maintain or improve IAQ. However, to obtain heating and cooling energy cost
711 savings a reduced intake of outdoor air is desired with buildings better sealed against the
712 outside climate conditions. A reduction in outdoor air intake without compromising IAQ can

713 only be obtained when low levels of all relevant pollutants can be maintained, that is for all
714 relevant VOCs, PM and VICs, including CO₂. Hybrid systems such as advanced botanical-
715 microbial or combined biological-physical systems may be able to provide a more complete
716 treatment of indoor air providing an opportunity for building energy cost savings without
717 compromising the IAQ.

718 The performance of air treatment systems is usually evaluated by the removal efficiency of
719 pollutants of a single pass through the system (the difference in concentration between inlet
720 and outlet air assuming no significant change in pressure and temperature in the treated air) at
721 a specific loading rate (the amount of pollutant removed over time, typically expressed per
722 system volume). Rather than the single pass purification efficiency, the overall purification
723 capacity per volume of indoor space is more important for indoor air purification systems. The
724 clean air delivery rate (CADR) is the reduction in outdoor air intake that can be obtained with
725 an indoor air purifying system, while maintaining low levels of pollutants in the room
726 (Shaughnessy and Sextro 2006). Moreover, the CADR can be translated in total energy
727 expenditure to evaluate potential costs savings for HVAC. The concept of CADR was
728 introduced to evaluate various indoor air purification devices (Shaughnessy and Sextro 2006),
729 where CADR is defined as the volume of purified air delivered per unit of time providing a
730 specific air purifier refreshment capacity α (h⁻¹) for an indoor room:

$$731 \quad \alpha = \text{CADR} / V = \eta \cdot Q / V \quad (3)$$

732 Where V is the volume of the indoor room (m³), η is the single-pass removal efficiency of the
733 purifier (%) and Q is the airflow treated by the purifier (m³ h⁻¹).

734 Baseline fresh air rate for non-process conditioned spaces of three room-volumes per hour is
735 typically recommended but is dependent of the type of room and its occupancy (AESHRAE
736 2019). Calculating the operating effectiveness (CADR/kW) makes it possible to compare

737 ventilation with or without air purifiers and may need consideration of the air mixing
738 performance of an air purifier as discussed further by Noh and Yook (2016).
739 Rodgers et al. (2013) installed an active plant-based biotrickling filter into a newly built
740 residential house and proved significant savings in total energy expenditure for HVAC in the
741 summer, when compared to the air-conditioning system alone. The biological system was
742 evaluated on VOC and CO₂ reduction and general climate conditions in the room such as
743 temperature and RH. Green walls are open enough to easily move the air through the system
744 and barely any increase in energy is required when incorporated into an existing HVAC system
745 with most of the energy required to provide enough light to the plant (Soreanu et al., 2013).
746 Active green walls integrated in the HVAC system can significantly reduce the intake of fresh
747 outdoor air and have been claimed to save energy up to 60% typically used by conventional
748 HVAC systems (Nedlaw Living Walls 2020).

749

750 **5.2 Green Building Certifications**

751 Biological indoor air purifiers can also provide credits towards Indoor Environmental Quality
752 (IEQ) for green building certification schemes that stimulates to realise sustainable buildings
753 that are healthy, energy-efficient and environmentally friendly. IEQ has a large impact on our
754 typical modern life and requires high indoor air quality to prevent health effects such as dry
755 eyes, headache, tiredness, allergies, respiratory infection and sick building syndrome (SBS)
756 (Burge, 2004). While building professionals and building owners may recognise the
757 importance of IAQ, they often do not appreciate how routine design and construction decisions
758 can ultimately result in IAQ problems. Sustainable building creates physical structures and uses
759 processes that are environmentally responsible and resource-efficient and take into account the
760 full lifecycle of a building. Green building certification was introduced in Europe and the
761 United States in the early 1990s, including the BRE Environmental Assessment Method

762 certification (BREEAM; in the United Kingdom), Leadership in Energy and Environmental
763 Design certification (LEED; in the United States) and Haute Qualite Environnementale
764 certification (HQE; in France). Many other green building schemes have been generated since
765 that have been adapted to national environmental and economic conditions (Wei et al. 2015).

766

767 ***5.3 Improvement of Indoor Comfort and Overall Well-being***

768 In addition to building energy cost savings and improving IEQ, biological indoor air purifying
769 systems can contribute to occupants' mental health in indoor spaces and may directly influence
770 human performance and productivity. IEQ is typically quantified by indoor air pollutant
771 concentrations (e.g. CO₂ concentration) and indoor climate conditions (i.e. temperature,
772 relative humidity and air movement), while occupants' comfort and overall well-being may be
773 quantified in terms of a physical sensation, a persons' mental state or both at the same time.
774 Physical health has been typical quantified the physiological reactions of blood pressure and
775 perspiration rates. Mental health can be estimated by psychological responses (e.g. verbal scale
776 vote of occupant what he/she considers a feeling of comfort). Kim et al. (2020) showed a
777 statistically significant negative relationship between the indoor climate and CO₂
778 concentrations and occupants' mental health, which was determined by blood pressure and
779 psychological responses. In this context, green plants in indoor spaces without specifically
780 being designed to clean indoor air have already been proven to provide valuable improvements
781 on indoor comfort and well-being, resulting in environments that are healthier and aesthetical
782 more pleasant to work and live in. Plant may help evaporate moisture lowering the temperature,
783 produce oxygen through photosynthesis or may help reduce sound levels as an acoustic
784 absorption system. Vegetation has also shown to affect emotions of consumers (Tifferet and
785 Vilnai-Yavetz 2017). Vegetation brings elements of nature inside a building that may provide
786 spaces that could create an aesthetical pleasant environment and potentially reduce stress.

787 Plant-based systems may improve worker productivity and creativity as well as comfort or
788 perception of their indoor space quality creating a more desirable place to work (Moya et al.
789 2018). Maybe the further development and benefits of (biological) indoor air purifying systems
790 should be focussed on the overall human well-being and productivity, as it may be easier to
791 motivate people and companies than air-pollutant-related chronic health benefits that occur
792 decades in the future (Siegel 2019).

793

794 **6. CONCLUSIONS**

795 The indoor concentration of air pollutions is almost always higher than the outdoor
796 concentration of air pollutions because outdoor-sourced contaminated air enters indoor
797 occupied spaces and combines with indoor-sourced pollutants. The threats posed by a long-
798 term exposure to poor indoor air quality have been acknowledged in recent years as buildings
799 are progressively sealed due to the increasingly stricter safety guidelines and against the outside
800 environment to obtain heating and cooling energy cost savings. Many buildings now rely
801 entirely on mechanical ventilation to recirculate indoor air with a minimized outdoor air intake,
802 leading to the accumulation of indoor pollutants. Currently there is not a single technology that
803 can efficiently provide a complete and satisfactory purification of indoor air. Biological
804 systems for improving indoor air quality are promising, but challenges need to be considered
805 to properly address the bioavailability of low pollutant concentrations, control indoor air
806 relative humidity, guarantee microbial safety, and incorporate CO₂-removal. Indoor air
807 treatment supported by hybrid technologies such as advanced botanical-microbial or combined
808 biological-physical systems could foster HVAC energy savings and green building certification
809 schemes, advance existing ecstatic green walls, and enhance indoor comfort and overall well-
810 being. Overall, there is an urgent need for fundamental research under relevant indoor
811 conditions to reveal the full potential for biological indoor air purification.

812

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819

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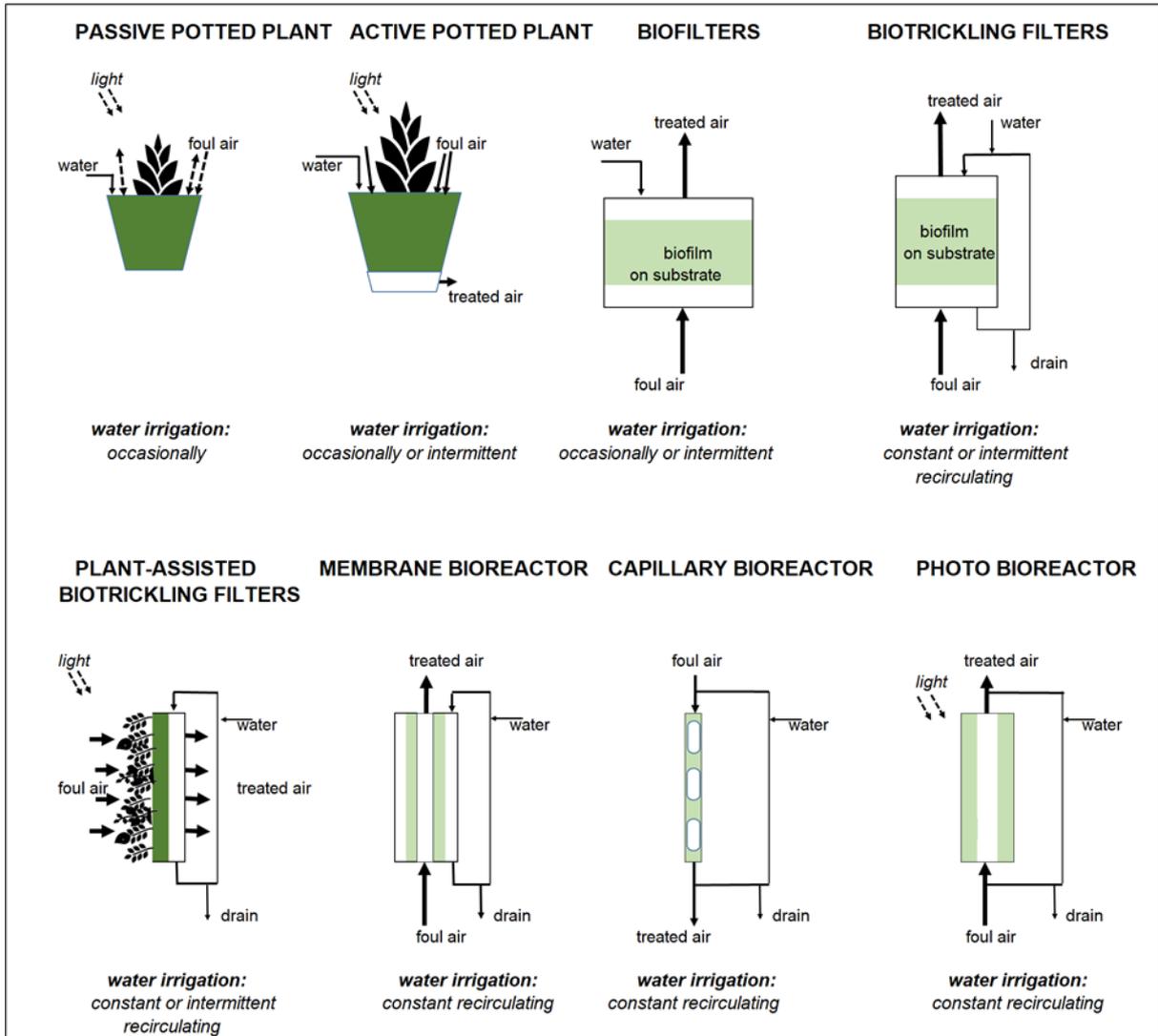
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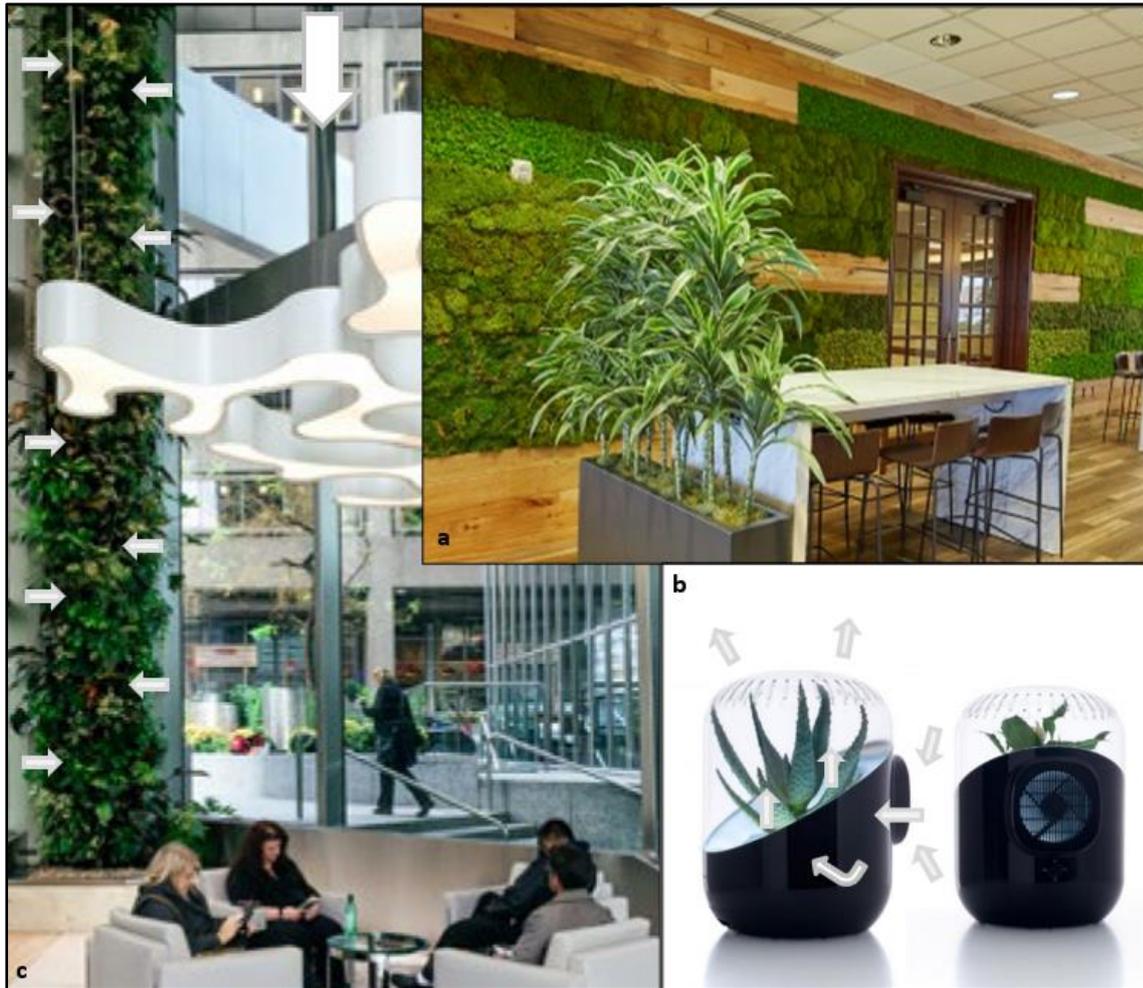
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1211

1212 **Figure 1:** Process schematics of typical biological air treatment system design configurations.

1213



1214

1215 **Figure 2:** Examples of commercially available plant-based systems for indoor environments:

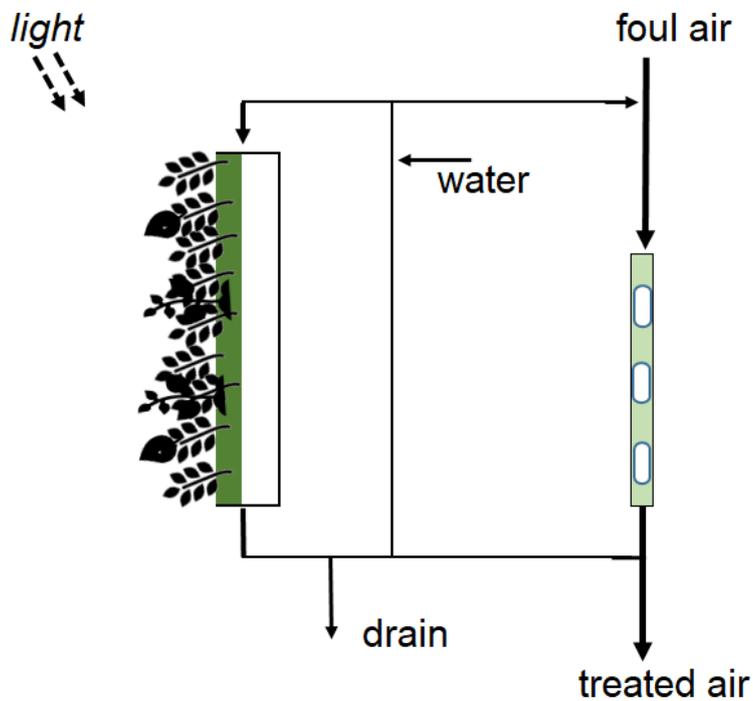
1216 (a) an aesthetic passive moss wall (courtesy of Ambius), (b) an active potted plant (courtesy of

1217 Andrea) and (c) a vertical active green wall integrated with the building HVAC system

1218 (courtesy of Nedlaw Living Walls).

1219

HYBRID: CAPILLARY ASSISTED GREEN WALL



water irrigation:
constant or intermittent constant recirculating
recirculating

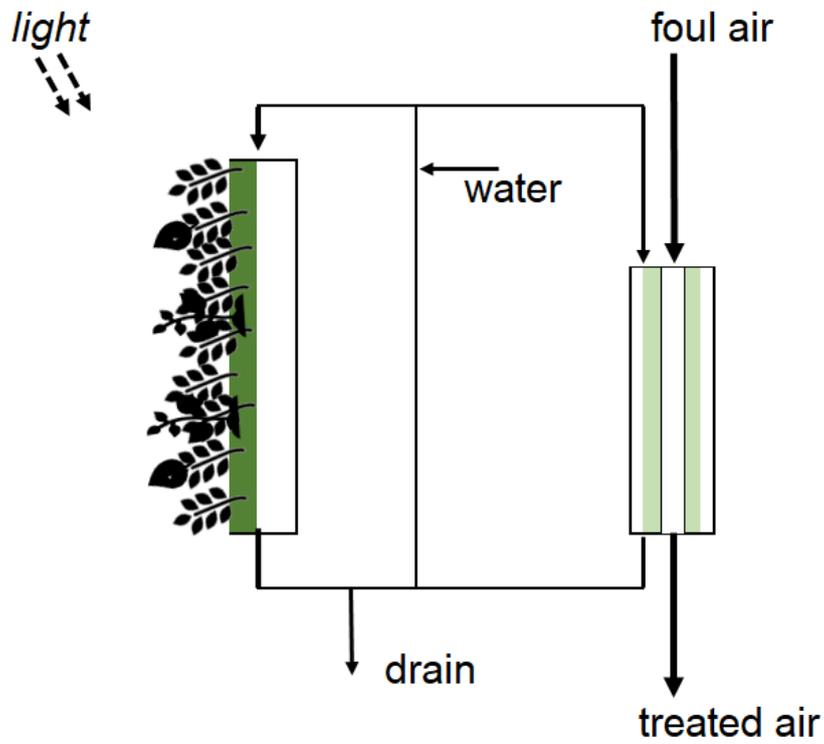
1220

1221 **Figure 3:** A schematic of a vertical active green wall combined with a capillary bioreactor to

1222 enhance mass transfer, improving the overall indoor air purification capacity.

1223

HYBRID: MEMBRANE ASSISTED GREEN WALL



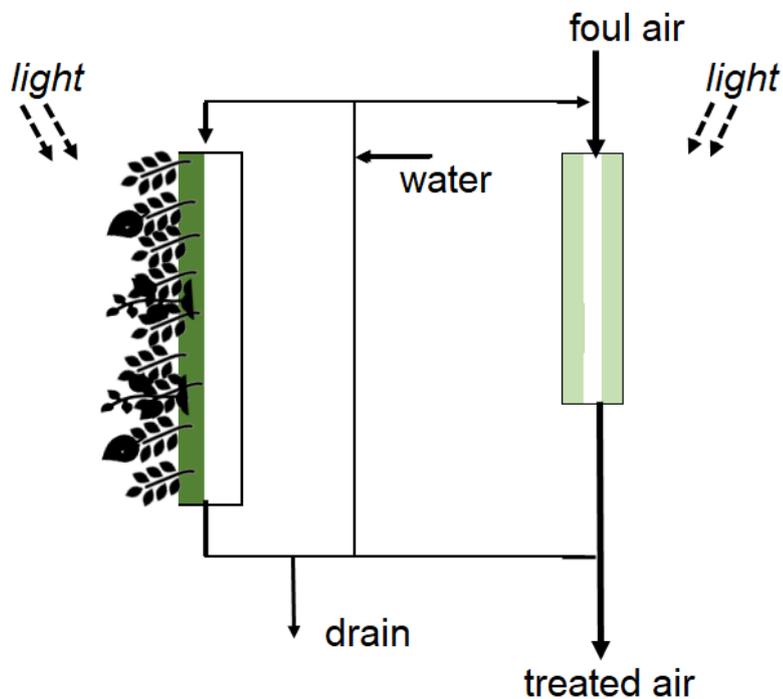
water irrigation:
constant or intermittent *constant recirculating*
recirculating

1224

1225 **Figure 4:** A schematic of a vertical active green wall combined with a dense-phase membrane
1226 bioreactor to enhance air purification while eliminating the release of bioaerosols and moisture.

1227

HYBRID: MICRO-ALGAE ASSISTED GREEN WALL



water irrigation:
constant or intermittent constant recirculating
recirculating

1228

1229 **Figure 5:** A schematic of a photo-bioreactor containing microalgae combined with a vertical

1230 active green wall to enhance CO₂ abatement of botanical-plant green walls.

1231