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Biological treatment of indoor air for VOC removal: Potential and challenges

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ABSTRACT

There is nowadays no single fully satisfactory method for VOC removal from indoor air due to the difficulties linked to the very low concentration ($\mu g \ m_s^{-3}$ range), diversity, and variability at which VOCs are typically found in the indoor environment. Although biological methods have shown a certain potential for this purpose, the specific characteristic of indoor air and the indoor air environment brings numerous challenges. In particular, new methods must be developed to inoculate, express, and maintain a suitable and diverse catabolic ability under conditions of trace substrate concentration which might not sustain microbial growth. In addition, the biological treatment of indoor air must be able to purify large amounts of air in confined environments with minimal nuisances and release of microorganisms. This requires technical innovations, the development of specific testing protocols and a deep understanding of microbial activities and the mechanisms of substrate uptake at trace concentrations.

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1. Introduction

Indoor air contamination is a complex problem involving particles (such as dust and smoke), biological agents (molds, spores), radon,

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asbestos, and gaseous contaminants such as CO, CO₂, NO_x, SO_x, 49 aldehydes and VOCs (Volatile Organic Compounds) (Table 1). The 50 latter are strongly suspected to cause many Indoor Air Quality (IAQ) 51 associated health problems and "sick-building" symptoms (Wallace, 52 2001; Jones, 1999; Wieslander et al., 1997; Yu and Crump, 1998).

Singularly, despite the abundance of evidence linking the exposure 54 to VOCs in indoor air with various health effects, only few reports 55 evaluating the existing abatement technologies are currently 56

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Table 1Typical air pollutants in indoor air (US EPA)

Group	Definition/example	Origin	Toxic effects
Particles	Very small liquid or solid substances in suspension in the air: mists, dust, pollen, cigarette smoke, viruses,	Outdoor air, combustion, carpets, human activity, decaying building	Irritation to eyes and/or respiratory tissues, allergies, cancer, indirect effect through biological production of toxins.
	bacteria, molds		
Gaseous	CO, CO ₂ , NO ₃₄ formaldehyde, VOCs	Combustion, human activity, building	Irritation to eyes and/or respiratory tissues, allergies, cancer,
pollutants		materials, furniture, cleaning products, mold	effects on the respiratory liver, immune, reproductive and/or
		development etc.	nervous system
Radon and	Radioactive gases	Rock, soil, groundwater, natural gas, mineral	Lung cancer
its progeny		building materials	

available. Several studies have demonstrated the potential of biological methods to remove indoor VOCs (Wolverton et al., 1984; Wolverton et al., 1989; Darlington et al., 2000; Darlington et al., 2001; Chen et al., 2005; Orwell et al., 2006; Wood et al., 2006). However, there is still a lack of solid and relevant data available to understand the true removal mechanisms in these systems and the apparent mismatch between experimental observations and theoretical results from transfer-based models on air biological treatment, as demonstrated in this review. This paper presents a critical review on the potential of biological technologies for indoor air purification with the dual objective of providing a state of the art of the relevant literature and a roadmap for future research. For the latter purpose, a scaledown approach was used in order to understand the consequences of indoor biological treatment on microbial growth and process design. Then, we identified current knowledge gaps hindering the proper understanding and development of biological processes for indoor air VOC removal. In the following, the term "biological purifier" is used to describe any device including a biological component (botanical or microbial) used for VOC removal; "botanical purifier" was used to specifically describe devices using plants and their associated microorganisms. Classical systems were named according to the conventional air treatment nomenclature (Revah and Morgan-Sagastume, 2005).

2. Indoor air quality

Because Americans spend nearly 90% of their time indoors and nearly 25% of US residents are affected by poor IAQ either at the workplace or at home, the US Environmental Protection Agency (EPA) ranks poor IAQ among its largest national environmental threats. Its counterpart, the European Environmental Agency (EEA) has pointed out IAQ as one of the priority concerns in children's health and similar issues are faced worldwide (Zhang and Smith, 2003; Observatory on Indoor Air Quality, 2006, Zumairi et al., 2006). In fact, some buildings contain such high levels of contaminants that they are qualified as "sick", because exposure to them results in multiple sickness symptoms (e.g. headache, fatigue, skin and eye irritations, or respiratory illness) commonly described as the "sick-building syndrome" (SBS) (Burge, 2004).

Data on IAQ-health related effects is still lacking and sometimes contradictory. For instance, Pilotto et al. (1997) found a strong link between exposure to peak NO₂ concentration and respiratory infections in children aged 6–10 years whereas other authors failed to establish this association (Brunekreef et al., 1990; Samet et al., 1993). Nevertheless, it has been suggested that indoor air pollution causes between 65,000 and 150,000 deaths per year in the US, which is comparable to outdoors pollution induced mortality (Lomborj, 2002). IAQ also impacts work productivity as for instance Wargocki et al. (1999) showed subjects exposed to a typical indoor pollution source (plastic carpet) typed 6.5% less than control subjects. Likewise, empirical studies have shown that the use of ventilation rates lower than 25 L s⁻¹ per person in commercial and institutional buildings was correlated to an increase in the number of short-term sick leaves

(Sundell, 2004). It has therefore been estimated that 40–200 billions 108 USD could be annually saved or gained by improving IAQ in the USA 109 only (in 1996 USD; Fisk, 2000). This problem is already driving an 110 important IAQ market that reached \$5.6 billion in 2003 in the USA 111 where it was expected to rise up to \$9.4 billion by 2008 (Market 112 report; indoor air quality, 2004).

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3. VOCs and indoor air quality

Interestingly, there is no clear or unanimous definition of what is a 115 VOC: The US EPA defines VOCs as substances with vapor pressure 116 greater than 0.1 mm Hg, the Australian National Pollutant Inventory as 117 any chemical based on carbon chains or rings with a vapor pressure 118 greater than 2 mm Hg at 25 °C, and the EU as chemicals with a vapor 119 pressure greater than 0.074 mm Hg at 20 °C. Chemicals such as CO, 120 CO₂, CH₄, and sometimes aldehydes, are often excluded. In addition, 121 sub-classifications such as Very Volatile Organic Compounds (VVOCs) 122 or Semi Volatile Organic Compounds (SVOCs) have been used in the 123 context of IAQ (Crump, 2001; Ayoko, 2004).

Several organizations such as the World Health Organization 125 (WHO), the US EPA, or the OQAI (French Indoor Air Quality 126 Observatory), have established lists of priority indoor air pollutants 127 (WHO, 2000; Johnston et al., 2002; Mosqueron and Nedellec, 2002, 128 OQAI) based on the ubiquity, concentration and toxic effect of the 129 substances involved. These lists are relatively similar and system- 130 atically include aldehydes, aromatics, and halogenates as well as 131 biocides. Differences are due to the type of pollution taken into 132 account (only chemicals for the EPA, no mixtures such as tobacco 133 smoke for the OQAI) and the geographic specificities of indoor air 134 pollution. Indeed, variations in the building materials, cleaning 135 products, or type of ventilation used generate differences in the 136 indoor air pollution (Sakai et al., 2004). These priority lists will most 137 likely evolve upon new analytical and toxicological findings more 138 relevant to IAQ such as the health effects of chronic exposure to 139 multiple pollutants at low concentration (Mosqueron and Nedellec, 140 2002). This lack of relevant knowledge probably explains why there 141 are only few guidelines for VOC indoor air concentration currently 142 available (WHO, 2000; Canada, 1987).

Hundreds of VOCs can be simultaneously found in indoor air. These 144 compounds exhibit very large variations in concentration as well as 145 physical, chemical, and biological properties. Furthermore, the 146 composition of the mixture greatly varies in time as the concentration 147 of VOCs released from coating and furniture generally decreases in 148 time whereas the release of certain substances depends on punctual 149 human activities or even human breathing (Ekberg, 1994; Phillips, 150 1997; Miekisch et al., 2004). Primary emissions of VOCs constitute the 151 major source in new or renovated dwellings during the first months, 152 while physical and chemical deterioration of buildings material 153 (named secondary emission) later becomes the main mechanisms of 154 VOC release (Wolkoff and Nielsen, 2001; Yu and Crump, 1998). Indoor 155 VOC concentrations depend on the total space volume, the pollutant 156 production and removal rates, the air exchange rate with the outside 157 atmosphere, and the outdoor VOCs concentrations (Salthammer, 158

Table 2 Example of VOCs found in indoor air

t2.2 t2.3	Compound	CAS ^a	Priority list		Indoor concentration (µg m ⁻³)			Recommended values (μg m ⁻³)	IARC ^c Health effects		References	
t2.4			EPA	A OQAI	b WHO	Min	Max	Average				
t2.5 t2.6	Acetaldehyde	75-07-0	×	HP		3.24	119 78.0	18.9 12.0	LFC ^d	2B	Respiratory disorders, irritation of the eyes	Weisel et al. (2005) Mosqueron (2002)
t2.7 t2.8	Benzene	71-43-2	×	HP	×	0.48	364 141	2.90 1.57	0.17 LFC ^d	1	Immunological disorders, leukemia, neurological effect	Weisel et al. (2005) Edwards et al. (2001)
t2.9	Dieldrin	60-57-1	×	₩P			6.00×10 ⁻⁴	7.00×10^{-5}	-	3	Neurological effect, cancer of the liver	Mosqueron and Nedellec (2002)
t2.10	Dichlorvos	62-73-7	×	HP			2.24	0.455	-	2B	Neurological effect, cancer of the liver	Mosqueron and Nedellec (2002)
t2.11 t2.12	Formaldehyde	50-00-0	×	HP	×	11.2	53.8 62.3	20.1 33.0	1000 (1 h) 60 ^e	2A	Respiratory disorders, irritation of the eyes	Weisel et al. (2005) Edwards et al. (2001)
t2.13	Naphthalene	91-20-3	×		×	2.20	90.1		-	2B	·	Mosqueron and Nedellec (2004)
t2.14 t2.15	Tetrachlorethylene	127-18-4	×	₩P	×	0.10	20.9 73.6	0.56 1.38	250 LFC ^d	2A	Neurological effect, renal disorder	Weisel et al. (2005) Mosqueron and Nedellec (2002)
t2.16 t2.17	Toluene	108-88-33	} ×	₩P	×	2.83	122 247	10.1 14.6	260	3	Neurological effect	Weisel et al. (2005) Edwards et al. (2001)
t2.18 t2.19	Trichlorethylene	79-01-6	×	¥ P	×	0.04	7.84 41.8	0.12 0.86	2.3 LFC ^d	2A	Neurological effect, cancer of the testicles	Weisel et al. (2005) Mosqueron and Nedellec (2002)

t2.20 a Chemical Abstract Service.

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1997). Typical air exchange rates in rooms without mechanical ventilation systems range from 0.1 to 0.4 h⁻¹, as stated above. Indoor VOCs concentrations are generally higher than outdoor concentrations because VOCs can be released from human activities and a wide variety of materials such as floorings, linoleum, carpets, paints, surface coatings, furniture etc (Yu and Crump, 1998). Salthammer (1997) for instance showed furniture coatings could release 150 VOCs (mainly aliphatic and aromatic aldehydes, aromatic hydrocarbons, ketones, esters and glycols) at Total VOC (TVOC) concentrations up to 1288 µg m⁻³ in test chamber studies and TVOCs emission rates as high as 22,280 µg m⁻² h⁻¹ have been recorded from vinyl/pvc flooring (Yu and Crump, 1998). Molds and bacteria can contribute significantly to the presence of particles (spores) of VOCs in indoor pollution (Schleibinger et al., 2004). Microbial development in buildings can be found in places where humidity accumulates including defective heating and air conditioning systems, garbage disposal, bathrooms, water leaks etc. and has been shown to provoke toxic and allergenic responses. Thus, although the individual concentrations of each contaminants are generally low (µg m⁻³), several hundreds contaminants can be found simultaneously, resulting in significant TVOC levels. Kostiainen (1995) found that individual concentrations of selected pollutants were 5–1000 times higher in 38 Finish sick-houses (defined as houses in which people experienced symptoms associated with SBS) than their mean concentrations in 50 normal houses used as reference, with over 200 VOCs being simultaneously detected in 26 houses. The same study also reported a maximal TVOC concentration of 9538 μg m $^{-3}$ in one sick house compared to the mean concentration of 121 μg m $^{-3}$ recorded in the normal houses. Likewise, Brown and Crump (1996) recorded TVOC concentrations up to 11,401 μg m⁻³ in UK homes and Daisey et al. (1994) reported indoor TVOC concentrations of 230–700 μg m $^{-3}$ (geometric mean of 510 μg m $^{-3}$) in 12 Californian office buildings. It is not easy to correlate TVOC with health effects because this generic parameter does not reflect the individual differences in toxicities found among indoor air VOCs; however, experience of eye, nose or mouth irritation has been reported at 5000-25,000 μg TVOC m⁻³ (Andersson et al., 1997).

Although indoor VOCs such as benzene or some polycyclic 195 aromatic hydrocarbons are recognized as human carcinogens, a direct 196 association between VOCs exposure and SBS symptoms or cancer has 197 not been fully established at typical indoor air concentrations 198 (Wallace, 2001). Several studies however correlated exposure to low 199 concentrations of these pollutants with increased risks of cancer or 200 eye and airways irritations (Table 2) (Vaughan et al., 1986, Wallace, 201 1991, Wolkoff and Nielsen, 2001). Symptoms such as headache, 202 drowsiness, fatigue and confusion have been recorded in subjects 203 exposed to 22 VOCs at 25 μg m $^{-3}$ (Hudnell et al., 1992). Likewise, 204 exposure to 1000 µg m⁻³ of formaldehyde causes coughing and eye 205 irritation. In addition, many harmless VOCs can react with oxidants 206 such as ozone, producing highly reactive compounds that can be more 207 harmful than their precursors, some of which are sensory irritants 208 (Sundell, 2004; Wolkoff et al., 1997; Wolkoff and Nielsen, 2001), 209 Finally, most of the reported concentrations based on stationary 210 measurement might lead to an underestimation of the real exposure 211 dose of the subjects evaluated in epidemiological studies because 212 concentrations in the breathing zone could be 2 to 4 times higher than 213 those recorded with traditional methods (Rodes et al., 1991; Wallace, 214 1991; Wolkoff and Nielsen, 2001).

4. Indoor air treatment

Existing solutions to poor indoor air quality include combination of 217 actions such as removing the pollutant sources; increasing ventilation 218 rates and improving air distribution; and cleaning the indoor air (US 219 EPA). Although certain furniture or appliance-manufacturers are 220 already phasing out the use of formaldehyde, removing the pollutant 221 sources is only possible when these are known and control is 222 technically or economically feasible, which is seldom the case. New 223 substances are constantly detected and classified as hazardous and 224 many sources can release compounds for years. In addition, there 225 are fears that many air pollutants are yet to be discovered (Otake 226 et al., 2001; Carlsson et al., 2000; Muir and Howard, 2006) and pre-227 ventive approaches might therefore be needed to ensure indoor air 228

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b HP: Highly Priority, VP: Very Priority.

^c International Agency for Research on Cancer (IARC) classification: Group 1: agent carcinogenic to humans, group 2A: agent probably carcinogenic to humans, group 3: agent not classifiable as to humans, group 4: agent not carcinogenic to humans, ns: non-study.

d NIOSH recommended for the carcinogenic Lowest Feasible Concentration.

e Canada (1987).

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Table 3
Current and emerging indoor air treatment methods: principle, examples and limitations

t3.2	Current and emerging indoor air treatment methods: principle, examples and limitation							
t3.3	Method	Principle						
t3.5	Current methods Filtration	Air is passed through a fibrous material (often coated with a viscous substance), which is efficient for particle removal but not gases. Filters are compact and commonly used but their efficiency decreases as they become saturated (fouling). Microorganisms can also develop in filters and particles reemission might occur.						
t3.6	Electrostatic precipitator with ionization	An electric field is generated to trap charged particles. Electrostatic precipitators are often combined with ion generators that charge particles. Remove efficiently particles, are, compact, commonly used but can generate hazardous charged particles.						
t3.7	Adsorption	Air pollutants are adsorbed onto activated carbon or zeolites, often as filtration post-treatment. The adsorbent might be too specific and might saturate fast because the pollutant are not destructed. There is therefore a potential risk of pollutant reemission.						
t3.8	Ozonation	Ozone is generated to oxidize pollutants. Only remove some fumes and certain gaseous pollutants and might generate unhealthy ozone and degradation products. Ozone-based purifiers are not recommended by the American Lung Association.						
t3.9	Photolysis	High energy ultra violet radiation oxidizes air pollutants and kills pathogens. It can however only remove some fumes and some gaseous pollutants and might release toxic photoproducts. Accidental exposure to UV light is harmful and UV irradiation is energy consuming.						
t3.10	Photocatalysis	High energy ultra violet radiation is used in combination with a photocatalyst (TiO_2) to generate highly reactive hydroxyl radicals that can oxidize most pollutants and kill pathogens. This energy-intensive method is increasingly popular and suitable for a broad range of organic pollutants.						
t3.11	Emerging methods							
t3.13	Membrane separation	Pollutants are passed through a membrane into another fluid by affinity separation. This method is normally recommended for highly loaded streams and has yet to be proven at low VOC levels. If the separated VOC are not reused, membrane filtration must be completed with a destruction step.						
t3.14	Enzymatic oxidation	Air pollutants are transferred into an aqueous phase where they are degraded by suitable enzymes Little information is however available concerning the efficiency of the commercial system (Air and Water Solution Inc., USA, http://www.srebiotech.com/) and new enzymes must be supplied periodically.						
t3.15	Botanical purification	Air is passed though a planted soil or directly on the plants. The contaminants are then degraded by microorganisms and/or plants, the precise mechanisms being unclear. Although the efficiency of botanical purification has not been fully proven, a number of devices have been patented and several commercial products are available.						
t3.16		US patent 6,676,091 for instance discloses a device where air is forced directly through a vertical (or slightly inclined) porous material serving as support for hydroponic plants, the plant's main purpose being to support the activity of pollutant degrading microorganisms in their rizosphere. This device is rather large in regards to other technology but can be use for interior design purposes.						
t3.17	Biofilters and biotrickling filter	Air is passed through a packed bed of a solid support colonized by attached microorganisms that biodegrade the VOCs. In one configuration, air was purified through lava rocks covered with a geotextile cloth supporting mosses (Darlington et al., 2001).						

contaminants are maintained below satisfactory levels at all times. Natural aeration is the easiest alternative but it is often not possible because of outdoor weather, external pollution conditions (Ekberg, 1994; Daisey et al., 1994) or issues of security, safety in high buildings,

climate control, or noise. Periodical air refreshing is often not efficient 233 because many indoor air pollutants are constantly released. Hence, 234 forced ventilation is still one the most common methods used for air 235 treatment (Wargocki et al., 2002). The improvement of indoor air 236 quality and energy savings are encouraged in the EU and by 237 movements such as the "Green Building" (US Green Building Council), 238 which means that forced ventilation should be reduced at the same 239 time as IAQ should be improved. In a worst case scenario (no heat 240 recovery from ventilation) the energy requirements for heating a 241 commercial office of 100 m² and 2.5 m of height at 23 °C ventilated 242 with outdoor air (4 °C) at an air exchange rate of 3 room volumes per 243 hour is approx. 3420 kWh/month (or about 340 USD, based on 244 residential electricity prices in 2006). Consequently, there are few 245 alternatives left than purifying the air inside the building.

Current methods for air purification include combinations of air 247 filtration, ionization, activated carbon adsorption, ozonation, and 248 photocatalysis (Table 3). These processes can be integrated into the 249 central ventilation system (in duct) or used in portable air purifiers (or 250 air cleaner) designed for limited spaces. Efficient strategies for particle 251 removal are now well established and include combinations of 252 filtration and electrostatic precipitation. The situation is however 253 very different for VOC removal. For instance, in a study conducted to 254 compare several commercial air purifiers, Shaugnessy et al. (1994) 255 concluded that, although high efficiency particles air filters (HEPA 256 filters) and electrostatic precipitators were highly efficient for particle 257 removal, none of the techniques tested (HEPA filtration, electrostatic 258 precipitation, ionization, ozonation, activated carbon adsorption) 259 could significantly remove formaldehyde. A similar study was recently 260 conducted to compare 15 air cleaners with a mixture of 16 261 representative VOCs (Chen et al., 2005). The technologies evaluated 262 included sorption filtration, ultraviolet-photocatalytic oxidation (UV- 263 PCO), ozone oxidation, air ionization and a botanical purifier 264 prototype (where contaminated air was blown through the rizosphere 265 of plants and contaminants were in principle removed by soil 266 microorganisms, the plants or their enzymes through various 267 mechanisms). The results were:

- 1. Among the 7 air cleaners using activated carbon in combination 269 with HEPA filter and/or ionizer, the best single pass removal 270 efficiencies achieved for formaldehyde, toluene and dodecane were 271 4, 32 and 39%, respectively (these contaminants were selected 272 for being representative of soluble, semi-soluble and unpolar 273 substances).
- 2. The commercial UV-PCO purifier did not effectively remove any of 275 the tested VOCs, although a "properly designed" device was 276 effective for certain VOCs (no data presented).
- 3. None of the ozone generator systems significantly removed any of 278 the VOCs tested; some even released ozone to concentrations much 279 higher than the safety limit set by the OSHA (Occupational Safety 280 and Health Administration).
- 4. The botanical purifier was able to remove around 20% of 282 formaldehyde (single pass) but no toluene and only 4% of 283 dodecane.

From this data, the authors concluded that only the biological 285 system significantly removed very volatile organic compounds, such 286 as formaldehyde, in contrast to the adsorption processes that 287 generally only satisfactorily removed the poorly soluble contaminants. 288 The overall elimination capacity (g VOC removed d_{-}^{-1}) of the botanical 289 purifier for formaldehyde was however lower than the best activated 290 carbon based device because the biological purifier could not be 291 operated at high air flows. An air exchange rate of 3 room volumes per 292 hour is generally recommended for indoor air treatment, which 293 means that very large amounts of air must be treated into relatively 294 small units (for suitable use in the indoor space without visual or noise 295 nuisances). This might be difficult to achieve in botanical purifiers 296 where air is ventilated into the soil through the roots.

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Table 4Biodegradability of typical indoor VOCs

Substance	Biodegradability ^a	Henry's law constants		Biological treatment			
		H ^b (atm m ³ mol ⁻¹)	References	Inlet concentration ^c (mg m ⁻³)	Removal efficiency (%)	Biological treatment ^d	References
Acetaldehyde (Ethanal; CH ₃ CHO)	3	5.88 10 ⁻⁵	Zhou and Mopper (1990)	18.1-180.1 ^e	40-80	В	Mohd Adly et al. (2001)
		7.69 10 ⁻⁵ 5.88 10 ⁻⁵	Sander (1999) US EPA (1982)				
Benzene (C ₆ H ₆)	2	6.25 10 ⁻³	Staudinger and Roberts (1996)	1.6 ^e	9–77	В	Ergas et al. (1992)
		5.55 10 ⁻³	US EPA (1982)	0.32-1.28 ^e	50 to 60	BF	Wolverton et al. (1989)
		$4.76 \ 10^{-3}$	Sander (1999)	0.048-0.48 ^e	20	PW	Darlington (2004)
Formaldehyde (Methanal; HCHO)	3	3.33 10 ⁻⁷	Sander (1999)	0.12-0.49 ^e	50 to 60	BF	Wolverton et al. (1989)
•		3.23 10 ⁻⁷	Zhou and Mopper (1990)	0.018-0.18 ^e	90	BF	Darlington (2004)
		3.13 10 ⁻⁷	Staudinger and Roberts (1996)				
Naphthalene (C ₁₀ H ₈)	1	4.76 10 ⁻⁴	Sander (1999)	0.494 ^e	75	TPPB	MacLeod and Daugulis (2003)
		$4.76 \ 10^{-4}$	US EPA (1982)				` ,
Tetrachlorethylene (Tetrachloroethene; C ₂ Cl ₄)	1	2.78 10 ⁻²	US EPA (1982)	0.678 ^e	12–49	В	Ergas et al. (1992)
, , , , , , , , , , , , , , , , , , , ,		1.69 10 ⁻²	Staudinger and Roberts (1996)	0.36-4.80 ^e	0–8	BTr	Torres et al. (1996)
		$1.56 \ 10^{-2}$	Sander (1999)				
Toluene (Methylbenzene; C ₆ H ₅ CH ₃)	2	6.67 10 ⁻³	US EPA (1982)	1.88 ^e	14–78	В	Ergas et al. (1992)
3, 3,		6.67 10 ⁻³	Staudinger and Roberts (1996)	753.5	50	MS	Ergas (1999)
			(1555)	0.226-0.301 ^e		BF	Darlington (2001)
				0.057-0.57 ^e		BF	Darlington (2004)
Trichlorethylene (Trichloroethene; C ₂ HCl ₃)	1	$9.09 \ 10^{-3}$	Sander (1999)	107.44	30	MS	Parvatiyar et al. (1996)
(Themorocanche, egiteis)		1.12 10-2	US EPA (1982)	0.081-0.81 ^e	0	BF	Darlington (2004)
		1.00 10 ⁻²	Staudinger and Roberts (1996)		50 to 60	BF	Wolverton et al. (1989)
			(1000)	0.01-0.04 ^e	0-24	BTr	Torres et al. (1996)

- ^a 1=low biodegradability, 2=moderate biodegradability, 3=good biodegradability (Shareefdeen and Singh, 2005; Devinny et al., 1999).
- b Under standard conditions.

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- ^c Concentrations close to the average concentration observed in indoor air.
- d B = Biofiltration; MS = Membrane Separation; BF = Botanical Filter; TPPB = Two-Phase Partitioning Bioreactor; BTr = Biotrickling Filter.
- ^e In mixture with other compounds.

Membrane mediated VOCs removal in indoors air has been also recently evaluated. Formaldehyde and benzene were significantly removed at concentrations of 11,500 and 6500 μ g m $^{-3}$, respectively using a zeolite membrane operated at permeation fluxes of 0.01 and 0.0056 g m $^{-2}$ L^{-1} , respectively (Aguado et al., 2004). Further studies must however demonstrate pollutant removal at concentrations relevant to indoors environments (approx. one order of magnitude lower than those tested) and provide solutions to destroy the VOCs following separation.

5. Biological treatment of indoor air

There is little data available on the biological removal of VOCs from indoor air and all the studies hitherto conducted have, to the best of our knowledge, focused on botanical purifiers. In a pioneer study supported by the NASA, Wolverton and co-authors demonstrated the potential of plants (and their rizosphere) to remove indoor VOCs in sealed chamber. In their earliest study (Wolverton et al., 1984), the authors found that several plants could remove formaldehyde at $19,000-46,000~\mu g~m^{-3}$ to levels lower than $2500~\mu g~m^{-3}$ (detection limit) in 24~h, Similar studies were conducted with benzene and trichloroethylene at more relevant concentrations of $325-2190~\mu g~m^{-3}$ (Wolverton et al., 1989). It was then found that the 8 plants tested could remove benzene by 47-90% in 24~h compared to 5-10% in the control tests, and that the rizosphere zone was the most effective area for removal. Orwell et al. (2004) later investigated the potential of indoor plants for removing benzene in sealed chamber (0.216 m³) and

found that microorganisms of the plant rizosphere were mainly 322 responsible for benzene removal (40–80 mg m $^{-3}$ d $^{-1}$). These results 323 were obtained at high initial benzene concentrations (81,000- 324 163,000 $\mu g m^{-3}$) and benzene removal rate increased linearly with 325 the dose concentration, suggesting the system might be inefficient 326 under typical indoor air conditions. However, the same team more 327 recently demonstrated that plants significantly reduced toluene and 328 xylene at indoor air concentrations of 768–887 μg m⁻³ (Orwell et al., 329 2006) and even the TVOC concentration in office buildings during 330 field testing under real conditions (Wood et al., 2006). Unfortunately, 331 the divergences in toluene removal reported in the studies of Chen 332 et al. (2005) and Orwell et al. (2006) cannot be explained, especially as 333 the prototype used in the earlier study was not fully described. Many 334 parameters such as the interfacial areas, the moisture content, and the 335 type (hydrophobicity) of the biomass used can influence pollutant 336 removal in biological purifiers. There is therefore a need for a more 337 coordinated research in the area. Various botanical purifiers have also 338 been patented (i.e. US5407470, US5277877) but such devices have not 339 reached a broad market and no data on pollutant removal under 340 relevant conditions is available. Research on the development of a 341 commercial biological purifier has been carried out at the University of 342 Guelph, Canada (Darlington et al., 2000; Air Quality Solution Ltd). In 343 one configuration, air was purified through lava rocks covered with a 344 geotextile cloth supporting mosses (Darlington et al., 2001). This 345 device was operated at relevant influent levels equal or lower than 346 $300 \,\mu g \, m^{-3}$ and displayed a purification efficiency of 30% at the lowest 347

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air flow treated. Water was also added to the filter to compensate for water losses through evaporation (approx. $20~L~d_{\sim}^{-1}$ in $120~m^2$ and $640~m^3$ room). In a second configuration, disclosed in US patent 6,676,091 from the same author, air is forced directly through a vertical (or slightly inclined) porous material serving as support for hydroponic plants which its main purpose is to support the activity of pollutant degrading microorganisms in the rizosphere.

From the studies herein presented, it appears that the role of plants in botanical purifier is often suspected to support a microbial activity that is responsible for pollutant removal. Direct pollutant accumulation or degradation by plants is however known to occur during phytoremediation of contaminated soils (Newman and Reynolds, 2004) and the ability of plant leaves to directly take up and remove pollutant during air treatment is still debated (Wolverton et al., 1984; Schmitz et al., 2000; Schäffner et al., 2002). A recent study has suggested that bacteria growing on plant leaves could also contribute to VOC biodegradation (Sandhu et al., 2007). More generally, there is growing evidence of the complexity, and importance of interactions between plants and bacteria (Dudler and Eberl, 2006) and research in this area is of utmost importance for IAQ. There is a lack of peer-reviewed data available in the literature and an urgent need to improve our understanding of the fundamental mechanisms of VOC uptake or release by plants and their microbial hosts (Kesselmeier and Staudt, 1999). The following discussion will therefore focus on the more established microbial degradation mechanisms.

5.1. Influence of type of VOCs

The biological treatment of organic compounds is based upon the capability of microorganisms to use these molecules as sources of carbon, nutrients and/or energy or to degrade them cometabolically using unspecific enzymes. The intrinsic biodegradability of an organic compound depends on many factors such as its hydrophobicity to the microbial population, the most soluble being generally the most biodegradable, or its toxicity. Toxicity effects, which sometimes limit the biological treatment of industrial air, are likely not a problem at the concentrations found in indoor air and this will not be discussed further in this review. Many VOCs are rather small molecules that are moderately soluble and in fact, are biodegradable (Table 4) although certain xenobiotic compounds, such as chlorinated compounds (i.e. tetrachloroethylene), may be recalcitrant. Synergetic or negative interacting effects within pollutant mixture should also be taken into consideration (Yu et al., 2001). Given the high number of VOC simultaneous found in indoor air, and the huge variations in structures and properties, a biological process suitable for indoor air treatment should rely on diverse, versatile and adaptive microbial communities to ensure all pollutants are removed. This can be achieved in fixed biofilmbased reactors where high microbial diversity and cell proximity favour cellular exchanges (Molin and Tolker-Nielsen, 2003; Singh et al., 2006), acclimation (long cell residence time) and synergetic effects under various growth conditions by the establishment of substrate concentration gradients thorough the biofilm (Beveridge et al., 1997; Marshall, 1994). Completing or combining biodegradation with a physicochemical post-treatment is also possible to ensure the complete removal of all pollutants. Finally, great variations in total and individual pollutant concentrations leading, for instance, to long periods of time when a given compound is not found in the indoor air could lead to permanent or momentary losses in catabolic ability. Such effects need to be further studied and possibly prevented as discussed below.

5.2. Influence of low concentrations on biomass productivity and transfer rates

During the biodegradation process, the concentration of an organic pollutant in the micro-environment where the microorganisms are found has a profound impact on microbial activity and ultimately on the pollutant removal rate. At reasonably high substrate concentrations, the

organic pollutant can be metabolized and used to synthesize more 410 biomass in a process that self-regenerates the biocatalyst. When the 411 concentration is decreased further, a critical level is reached below 412 which new cells are no longer produced. Nevertheless, the contaminant 413 can still be biodegraded if a significant active biomass is available and if 414 the gene(s) responsible for the production of the enzyme(s) required for 415 the pollutant degradation is (are) still expressed (Kovárová-Kovar and 416 Egli, 1998). Gene expression depends, among other factors, on the 417 concentration of the enzyme substrate which can be the pollutant or 418 other molecules. In this context, it is crucial to compare the low 419 concentrations at which indoor VOCs are typically found with known 420 threshold for microbial growth and biodegradation.

Toluene indoor air concentrations of 0.58_{-17} µg m $^{-3}$ have been 422 reported in Californian office buildings (Daisey et al., 1994). Assuming 423 toluene must first transfer into an aqueous phase before being 424 biodegraded, the maximum aqueous toluene concentration (C_{aq} *) at 425 which microorganisms will be exposed to can be calculated from the 426 Henry's law constant (H) coefficient:

$$C_{\text{aq}}^* = \frac{P_i}{H_i} \tag{1}$$

Where P_i is the partial pressure of the target contaminant in the 430gas phase and H_i its Henry constant. For toluene ($H=6.67\ 10^{-3}$ atm m³ 431 mol^{-1} ; Table 4), this will result in a C_{aq} * of 2–60 ng L^{-1} under normal 432 conditions of temperature and pressure. If toluene is removed by 90%, 433 microorganisms would actually be exposed to concentrations of 0.2-434 6 ng L^{-1} (under continuous treatment under a steady state). At such 435 concentration, toluene can be reasonably considered as the limiting 436 substrate if it is the only carbon source available. By comparison, the 437 threshold growth concentration of bacteria from drinking-water 438 biofilm has been estimated to about 0.1 µg L⁻¹ (van der Kooij et al., 439 1995) which is in the same range of reported toluene mineralization at 440 aqueous concentrations of 0.9 μ g L^{-1} with active bacteria (Roch and 441 Alexander, 1997). Hence, from the data currently available, it seems 442 unlikely that indoor air VOC can support growth. However, micro- 443 organisms might still be capable to use certain VOCs as energy source 444 for maintenance or cometabolically biodegrade them by using another 445 substrate for growth.

The specific biomass growth rate (μ, h^{-1}) at sub-inhibitory 447 substrate concentration can often be expressed as:

$$\mu = \frac{\mu_{\text{max}} S}{K_{\text{S}} + S} - k_{\text{d}} \tag{2}$$

Where $\mu_{\rm max}$ (h $_{\sim}^{-1}$) is the maximum specific growth rate, $K_{\rm S}$ is the 451 saturation constant (mg L $_{\sim}^{-1}$), $k_{\rm d}$ is the endogenous decay coefficient 452 (h $_{\sim}^{-1}$) and S is the limiting substrate concentration (mg L $_{\sim}^{-1}$). The 453 substrate uptake volumetric rate is equivalent to:

$$r_{\rm su} = \frac{\mu X}{Y} + mX \tag{3}$$

where X is the biomass concentration, Y is the true biomass yield (g 456 biomass g substrate $^{-1}$), and m is the maintenance coefficient. This 457 equation shows that substrate consumption for maintenance can 458 occur even under no growth conditions ($\mu_{\text{max}}S/(K_S+S) < k_d$), although 459 cell decay would cause a continuous decrease in catabolic activity.

Based on the toluene biodegradation kinetics reported by 461 Alagappan and Cowan (2003) in *Pseudomonas putida* F1 cultures 462 (μ_{max} =0.37 h $_{-}^{-1}$ and K_{s} =0.44 mg L $_{-}^{-1}$), the specific cell production rate 463 under indoor conditions should range from 5×10^{-5} —1.7 × 10^{-6} h $_{-}^{-1}$. 464 These values are far below the cellular maintenance rates reported in 465 the literature for aromatic compounds (0.019 h $_{-}^{-1}$ for ethylbenzene 466 and 0.016 h $_{-}^{-1}$ for benzene), and the death cells coefficients for *P*. 467 *putida* F1 during the degradation of toluene (0.06 h $_{-}^{-1}$; Alagappan and 468 Cowan, 2003). Thus, in this particular situation, neither would 469 pollutant supply meet maintenance requirements nor would the 470 specific growth rate meet the cellular decay rate.

Although these kinetics parameters were obtained at pollutant concentrations higher than those found in indoor air, this simple calculation shows that indoor air biological treatment will likely require the development of specific methods to provide and maintain a suitable catabolic activity. First, due to the complexity and variability of indoor air, an inoculum that possesses the suitable catabolic ability might be difficult to obtain. These microorganisms would also likely need to be pre-cultivated at higher VOC concentration to obtain a significant cell number in a relative short time, which might impair their ability to take up substrates at trace levels (microorganisms can loose selective traits when the corresponding selection pressure is released). Attached growth should therefore be recommended for inoculum preparation to combine, through the establishment of substrate concentration gradients inside the biofilm, microbial growth at the liquid-biofilm interface with continuous selection of microorganisms acclimated to low-substrate concentration inside the biofilm (Beveridge et al., 1997). Second, maintaining catabolic activity (and not only cell mass or cellular activity) could be challenging as microorganisms can loose their ability to biodegrade certain substrates when deprived from them during long periods of time. Biofilm systems could ensure constant performance under fluctuating operating conditions by allowing substrate accumulation at the biofilm interface until degradation becomes possible (Singh et al., 2006). As mentioned above, attached microorganisms also form diverse and dynamic communities (cell communication and cellular exchanges such as horizontal gene transfer being favoured by the cell proximity) that can respond more quickly to changes, which in return favours functional redundancy (Rittmann and McCarty, 2001). Biofilms are in fact ubiquitous and prevail under diluted environment (Beveridge et al., 1997). Finally, even under conditions when suitable degradation-enzymes are expressed, microbial activity must be capable to reduce the contaminant at concentration low enough to permit significant mass transfer. Roch and Alexander (1997) showed toluene mineralization at $0.9 \,\mu\mathrm{g}\,\mathrm{L}_{2}^{-1}$ but the pollutant still remained at 79 ng L⁻¹ after 8 days of incubation. Similar findings were reported by Pahm and Alexander (1993) when studying the biodegradation of pnitrophenol at trace concentration although addition of a secondary carbon source was capable to trigger pollutant removal at concentrations of 1 μ g L⁻¹. However, the feasibility of removing estrogens at 100 ng L_{\perp}^{-1} to below 2.58 ng L_{\perp}^{-1} (detection limit) with pure laccase

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from *T. versicolor* was recently demonstrated (Auriol et al., 2007), 512 showing biological systems should be able to perform at indoor air 513 concentrations.

Clearly, the development of biological methods for indoor air 515 filtration faces several challenges and requires more research on the 516 microbial mechanisms of acclimation, survival, substrate recognition, 517 accumulation and uptake at trace concentration. Most published 518 biodegradation studies have been conducted under irrelevant condi- 519 tions (single substrate, high substrate concentration, suspended 520 growth) using isolates cultivated in the laboratory. Current kinetics 521 data might therefore underestimate the capacity of natural strains to 522 take up trace pollutants (Subba-Rao et al., 1982; Pahm and Alexander, 523 1993). Low concentrations are common in the environment and 524 certain microorganisms have developed original survival strategies 525 under such conditions by for instance accumulating limiting substrate 526 before starting to growth (Singh et al., 2006). New models to correlate 527 growth with substrate concentration are therefore needed at trace 528 concentration, as suggested by Butterfield et al. (2002) in a study on 529 drinking-water biofilm formation under carbon-limited conditions 530 $(<2 \text{ mg L}^{-1}).$

The simultaneous presence of many contaminants in indoor air 532 might sustain microbial growth or, at least, induce pollutant miner- 533 alization, as suggested by the experience of Pahm and Alexander 534 (1993) described above. Furthermore, under starving conditions, 535 certain microorganisms are capable to quickly increase their affinity 536 for the limiting substrates or acclimate to simultaneously mineralize a 537 high number of carbon sources (Kovárová-Kovar and Egli, 1998, Tros 538 et al., 1996). Particles and other air macropollutants (carbon dioxide, 539 carbon monoxide, ammonia, etc) or plants might also provide enough 540 energy and carbon substrates for growth, as evidenced by the 541 microbial colonization of indoor surfaces and air handling systems or 542 the microbial activity recorded in the rizosphere of botanical purifier. 543 In addition, certain microorganisms are able to grow both hetero- 544 trophically and autotrophically (Larimer et al., 2003) or on myriads of 545 different organic compounds (Chain et al., 2006). Such metabolic 546 versatility would give obvious advantages under conditions where 547 numerous potential carbon and energy sources are simultaneously 548 found at very low concentrations and would greatly enhance the 549 treatment of indoor air. The question is therefore not if microbial 550 growth would occur, but if it will cause VOC reduction. For instance, 551

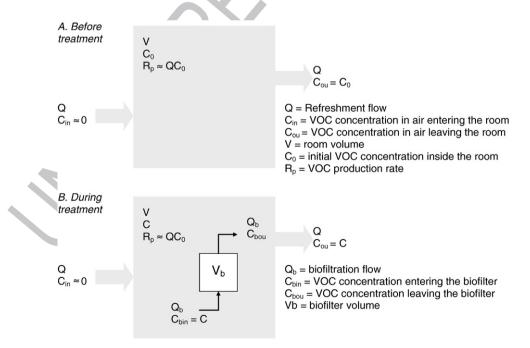


Fig. 1. Schematic presentation of a simple steady state mass balance analysis of indoor air treatment.

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from field studies on the efficiency of plants to purify air from office buildings, Wood et al. (2006) suggested that a TVOC concentration of 100 ppb was sufficient to induce a biological response that could reduce the TVOC concentration up to 75%. In addition, during laboratory studies on VOC removal by potted plants in sealed chamber, Orwell et al. (2006) observed the presence of toluene accelerated the removal of x-xylene, although the reciprocal was not true.

Several authors have also challenged the mass transfer and microbial uptake theories use to predict the effect of substrate concentration in biological purifiers. Active transfer by enzymatic transformation has for instance been reported and mechanisms of direct uptake at the air_cell interface have been suggested. For instance, Miller and Allen (2005) reported that direct pollutant diffusion through the aqueous layer surrounding the biofilm could not explain the surprisingly high performances of biological systems treating the highly hydrophobic alpha-pinene. Likewise, it has been suggested that the aerial mycelia of fungi, which are in direct contact with the gas phase, might promote the direct uptake of VOC from the gas phase. This uptake is faster than if a flat biofilm of bacteria directly contacts the gas phase because of a high gas—mycelium interfacial area of the fungal mat and the highly hydrophobic nature of the fungal cell wall (Arriaga and Revah, 2005; Kennes and Veiga, 2004; Van Groenestiin and Kraakman, 2005, Vergara et al., 2006).

5.3. Impact of purification efficiency on design

A simple steady state mass balance analysis can be used to design a device for indoor air purification (Fig. 1) based on the following equation:

$$V\frac{\mathrm{d}C}{\mathrm{d}t} = QC_{\mathrm{in}} - QC_{\mathrm{out}} + R_{\mathrm{p}} \tag{4}$$

where V is the room volume (m³); C the TVOC concentration in the room (µg m¬³); C is the refreshment flow rate (m³ h¬¹) through the room, C_{in} is the TVOC concentration of the air entering the room (µg m¬³), C_{out} is the TVOC concentration of the air leaving the room (µg m¬³), and the C_p is the TVOC production rate inside the room (µg h¬¹).

At steady state under completely mixed conditions within the control volume analyzed dC/dt=0 and $C=C_{\rm out}$. Assuming that $C_{\rm in}$ is negligible compared to $C_{\rm out}$ (indoor air concentrations are usually higher than outdoor), Eq. (5) gives:

$$R_{\rm p} = QC_0 \tag{5}$$

where $C_0 = C_{\text{out}}$ represents the initial pollutant concentration in the room, before air is being cleaned. When an air treatment unit (i.e. a biological purifier) is started in the room the mass balance analysis becomes:

$$V\frac{dC}{dt} = QC_{\rm in} - QC_{\rm out} + R_{\rm p} - R_{\rm b}$$
 (6)

Where, for a biological purifier, R_b is the TVOC biological removal rate (ug h^{-1}).

If we assume that $R_{\rm p}$ is constant (which is unlikely as the rate of VOC evaporation from coatings depends on the concentration of the VOC in the room and the concentration in the material) and $C_{\rm in} \approx 0$, the rate of TVOC biologically removed under steady state conditions can be expressed as:

$$R_{b} = Q(C_{0} - C) \tag{7}$$

If we now consider the biological purifier, R_b can also be expressed as:

$$R_{b} = Q_{b} \left(C_{b_{in}} - C_{b_{out}} \right) \tag{8}$$

Where $C_{b_{in}}$ and $C_{b_{out}}$ are the TVOC concentration in the air entering 608 and leaving the biological purifier, respectively; and Q_b is the air flow 609 treated by the biological filter.

By definition, the single pass efficiency (%) of the biological purifier 611 is expressed as:

$$\eta = \frac{C_{b_{\text{in}}} - C_{b_{\text{out}}}}{C_{b_{\text{in}}}} \tag{9}$$

Based on the assumption of completely mixed conditions in the 615 room $C_{\rm b_m}$ = C, then:

$$R_{\rm b} = \eta C Q_{\rm b} \tag{10}$$

Combining Eqs. (8) and (11) gives the following expression for Q_b : 619

$$Q_{b} = \frac{C_{0} - C}{\eta C} Q \tag{11}$$

The required biological purifier air flow is therefore a function of 622 the single pass biofiltration efficiency, the initial and required TVOC 623 concentrations, and the refreshment rate. It can also be expressed as a 624 function of the ratio between initial and final concentrations in the 625 room ($A = C_0/C$), which is another expression of the removal efficiency 626 according to:

$$Q_{b} = \frac{A-1}{\eta}Q \tag{12}$$

The overall performance of the purification device can then be 630 expressed as its effectiveness (*R*, %) according to:

$$R = \frac{\zeta_1 - \zeta_0}{\zeta_0} \tag{13}$$

Based on this equation, a simulation was made to determine Q_b 634 under varying R and η values at C_0 =500 μ g m⁻³ (Fig. 2) as typical 635 TVOC values in sick houses range from 100 to 1000 µg m⁻³. The 636 refreshment rate was assumed to 0.35 h⁻¹ in a room of 100 m³ 637 $(Q=35 \text{ m}^3 \text{ h}^{-1})$ as described by Wolkoff et al. (1991). This simulation 638 shows that a purification flow (Q_b) of approx. 400 m³ h⁻¹ is required to 639 achieve a satisfactory overall purification effectiveness of 90% with 640 η =80%. This is rather logical since the removal efficiency of any 641 system is intrinsically limited by the low concentration of the 642 pollutants. These values are in accordance with the recommended 643 air cleaner true effectiveness of 80% for particle removal, which is 644 equivalent to providing 4-5 room volumes of clean air per hour 645 (Shaughnessy and Sextro, 2006). Hence, it is not only the single pass 646 purification efficiency of the biofiltration device but the overall 647 purification capacity (ηQ_b) that is important, explaining why the 648 concept of Clean Air Delivery Rate (CADR = ηQ_b , the amount of purified 649 air delivered per unit or time) was introduced to evaluate and 650 compare the various devices proposed for air removal (Shaughnessy 651 and Sextro, 2006). Interestingly, at equivalent CADR, purification 652 devices with high single pass efficiencies should be preferred because 653 of their lower energy requirement (lower required flow rate).

The biological purifier can be designed using another engineering 655 approach based on the gas residence time (t_{res}) in the purifier and the 656 purifier refreshment capacity (α):

$$t_{\rm res} = \frac{V_{\rm b}}{Q_{\rm b}} \tag{14}$$

where $V_{\rm b}$ is the volume of the biological purifier.

$$\alpha = \frac{\eta \cdot Q_b}{V} = \frac{\text{CADR}}{V} \tag{15}$$

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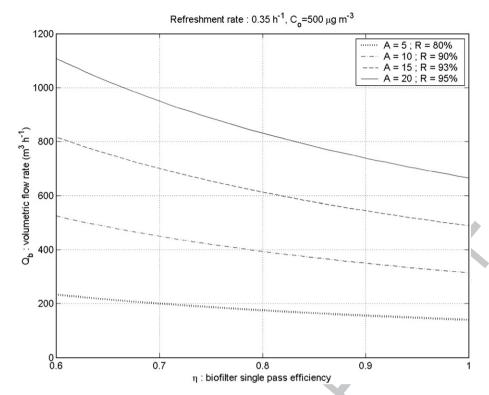


Fig. 2. Simulated performance of the effect of the biological purifier single pass efficiency (η, %) on the required biofiltration volumetric flow rate (Q_b, m³ h⁻¹) to achieve an overall effectiveness R = 80 - 95% (with $R = (C_0 - C_0 - C_0)/C_0$ where C and $C_0 = 500 \,\mu g \, m^{-3}$ are the steady state TVOCs concentrations before and during the biological purifier operation, respectively) in a 100 m³ room with a refreshment rate of 0.35 h⁻¹. The air cleaner effectiveness can also be expressed as the ratio $A = C/C_0$.

Thus, the biological purifier volume required can be expressed as: $\frac{(17)}{n}V_{\rm b} = \frac{\alpha \cdot t_{\rm res} \cdot V}{n}$

Assuming a typical gas residence time (industrial applications) of 30 s, a purifier refreshment capacity of 4 h_{\perp}^{-1} , and a 80% single pass efficiency, the biological purifier volume required for a 100 m³ room would be 4.2 m³, which is a prohibitive volume in indoor environment. Even at such a short residence time as 2 s, the volume of the bioreactor needed in our simulation would be 278 L. By comparison, the biological purifier developed by Darlington et al. (2001) was tested at surface loading rates of 90–720 m³ m $^{-2}$ h $^{-1}$, which is within the range of operation of industrial biotrickling filter and bioscrubbers (100-1000 m³ m⁻² h⁻¹; Van Groenestijn and Hesselink, 1993). The 1920 L (9.6 m²) bioscrubber (not including plants and aquarium used for water circulation) was also operated in a 640 m³ room at influent toluene, ethylbenzene and xylene concentrations of 0-300 $\mu g m^{-3}$. Single pass efficiencies of approx. 10–30% were achieved depending on the flow (the higher the flow, the lower the single pass efficiency) and temperature, which accounted for a CADR of 720 m³ h₋⁻¹ equivalent to a refreshment rate of 1.1 h_{\perp}^{-1} . Although it is difficult to draw conclusions from this single study, the data provided seems consistent with our simulation. It clearly shows that the engineering of compact biological purifiers with high effectiveness will be an important challenge.

The model described above can also be used to simulate the dynamic changes in pollutant concentration in a room where air purification is started by using Eq. (4) expressed as:

$$V\frac{\mathrm{d}C}{\mathrm{d}t} = QC_0 - QC - \eta Q_b C \tag{16}$$

Which can then be integrated as:

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$$C_{t} = C_{0} \exp \left(-Bt\right) + \frac{A}{B} (1 - exp\left(-Bt\right)) \tag{17}$$

$$B = \frac{Q + \eta Q_b}{V} = \frac{Q + \text{CADR}}{V} \tag{18}$$

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$$A = \frac{QC_0}{V} \tag{19}$$

In Eq. (17), $C_0 \exp(-Bt)$ represents the combined VOC removal from 699 refreshment and biofiltration and $\frac{A}{B}(1-exp(-Bt))$ represents the 700 production within the room. Using the example described above 701 (V=100 m³, Q=35 m³ h^{-1} , C_0 =500 μ g m $^{-3}$), Fig. 3 shows that at the 702 range of purification flows required for such application, steady states 703 conditions are achieved rather quickly (1–2 h). Similar models are 704 used to estimate the single pass efficiency of purification devices in 705 sealed chamber test where pollutant are introduced at a certain 706 amount but where there is no production (Chen et al., 2005). Thus, 707 Wolverton et al. (1989) reported a decreased in benzene concentration 708 from 765 to 78 μ g m $_{\wedge}^{-3}$ in 24 h in a sealed chamber containing a plant, 709 which resulted in a *B* coefficient in Eq. (18) of 0.1 h_{Δ}^{-1} . The *B* coefficient 710 is composed of the pollutant leakage rate from the system (Q/V) and 711 the pollutant removal in the air purifier (CADR/V=purifier refresh-712 ment capacity). The same author conducted a leak experiment which 713 allowed calculating the leak contribution to approx. 0.01 h_{\blacktriangle}^{-1} . Hence, 714 the botanical purifier used in this study generated an amount of 715 purified air equivalent to 0.09 room volume per hour (CADR of 716 purified air equivalent to 0.09 from volume per flour (CADK of 716 0.075 $\mathrm{m}^3 \, \mathrm{h}^{-1}$) and would not significantly improve IAQ under realistic 717 conditions. Low refreshment rates of 0.02–0.3 h^{-1} were also achieved 718 by Orwell et al. (2006) in sealed-chambers containing potted plants 719 and initially supplied with 768–886 $\mathrm{\mu g} \, \mathrm{m}^{-3}$ of m-xylene or toluene, 720 based on VOC exponential removal rate constants of 0.52–7.44 d^{-1} . 721 Likewise, Chen et al. (2006) achieved the highest CADR of 8.3 $\mathrm{m}^3 \, \mathrm{h}^{-1}$ 722 Q4 (refreshment rate of 0.15 h^{-1}) with the botanical purifier compared to 723

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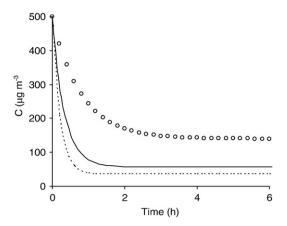


Fig. 3. Simulated changes in TVOC concentration (C, μ g m $^{-3}$) in a 100 m 3 room where air purification is started at t=0 at a flow of 500 (dashed line), 300 (plain line) or 100 (circles) m 3 h_-^{-1} in a biological purifier with 90% single pass efficiency. The refreshment rate in the room is 0.35 h_-^{-1} and the initial TVOC concentration is 500 μ g m $^{-3}$.

values above 200 m³ h⁻¹ with other portable devices. Despite this, a significant TVOC removal was recorded when using potted plants during field testing in office (Wood et al., 2006) and even if such results should be reproduced under better controlled conditions, they might indicate that our current evaluation models are inadequate.

5.4. Humidification and biohazards

Since biological purifiers are typically saturated with water and since indoor air treatment requires high flows, indoor biological purification might increase the moisture content in the room or building where it is used. This beneficial effect when indoor air is too dry (moisture contents of 30-60% are generally recommended for comfort) could also trigger to the excessive growth of fungi with negative impact on IAQ (Schleibinger et al., 2004), although these effects are still uncertain (Robbins et al., 2000; Pasanen, 2001). Darlington et al. (2000) for instance reported that the use of an indoor biological purifier significantly increased the concentrations of total suspended spores, although these values were similar to concentrations found in flats containing house plants, and still remained within healthy levels (100–200 CFU m⁻³). In addition, none of the 17 fungal species identified was known pathogenic. Likewise, Ottengraf and Konings (1991) reported that the concentration of microbial germs (mainly bacteria) in the outlet of full scale industrial biofilters was within the range of typical indoor air concentration, and only slightly higher than typical outdoor air concentrations, which was more recently confirmed by Zilli et al. (2005). There is however too little data available and the potential release of microorganisms from indoor biological purifiers (especially in the case of faulty equipment or accidents) should be better studied and prevented if necessary.

5.5. Esthetic, noise, purification perception

Besides being efficient for pollutant removal, indoor air purifiers must be esthetic (unless it is integrated into the ventilation system) and silent, which is rather challenging considering the flow required and the volumetric constrains. As mentioned above, indoor air pollution involves many types of pollutants (particulates, inorganic etc.), which concentrations must all be reduced below a certain "perception" level for users to feel the improvement of air quality. Especially, the effects of macropollutants such as CO₂ and H₂O should-also be considered.

5.6. Evaluating performance

Setting_up realistic purification goals is difficult as there are only few guidelines on indoor VOC concentration. Evaluating performance

is no less challenging due to the analytical difficulty to detect 764 numerous substances at very low concentrations, the lack of knowl- 765 edge about contaminants and their effects, and because of the number 766 of associated effects of air purification (i.e. moisture, temperature, 767 physiological impacts on user). Ultimately, blind testing in sick- 768 buildings that could correlate pollutant removal with customer/user 769 satisfaction (by survey, measurement of productivity, etc) and health 770 improvements would be necessary. Unfortunately, such methods 771 cannot be used during phases of design and optimization for 772 economical reasons and because of the need for well-defined and 773 reproducible testing conditions, replicate and control, Hence, the 774 efficiency of air purifiers is generally evaluated by either direct 775 measuring of the single pass efficiency or by using test-chambers. In 776 the first case, an artificially contaminated air stream is passed at a 777 certain flow through the purification device and the concentrations of 778 the target contaminants are measured at the inlet and outlet of the 779 purifier (Howard-Reed et al., 2002). This method is rather simple and 780 the influence of parameters such as the effluent composition or the 781 treatment flow can easily be tested under continuous inlet air 782 composition. Test-chamber assays are however often preferred, 783 perhaps because they offer more flexibility and better simulate the 784 indoor environment. Here, pollutants are injected in a hermetical 785 chamber equipped with the air purifier as well as various sampling, 786 analysis, and air conditioning devices and their concentrations are 787 measured over time. The purifier efficiency can then be evaluated 788 using the basic model described above. Depending on the size and 789 complexity of the chamber, the pollutant can be replaced by pollutingmaterial and treatment efficiency can be evaluated, for instance, in 791 terms of work productivity by monitoring the activity of human test- 792 workers operating inside the chamber (Wargocki et al., 1999). The use 793 of artificially contaminated air also brings its own challenges as it is 794 very difficult to determine a universal "model indoor air" due to the 795 diversity and variability of pollutants concerned (Ekberg, 1994; Yu and 796 Crump, 1998; Otson et al., 1994). However, recent progresses have 797 been made in that direction (Ondarts et al., 2007).

VOC analysis from indoor air normally requires large air samples 799 that are passed through a solid or liquid absorbent that serves to 800 concentrate the contaminants, followed by further extraction/separa- 801 tion and analysis (Crump, 2001). However, many pollutants are not 802 known or cannot be detected at indoor air concentration levels 803 (Ondarts et al., 2007) and the costs of monitoring all known 804 compounds would be prohibitive. In such cases, the TVOC (for details 805 on protocols see Crump, 2001) can be measured although this 806 parameter can be exclusive and does not take into consideration the 807 different intrinsic toxicities of each compounds and interaction effects 808 between the pollutants (Wolkoff, 2003; Wolkoff and Nielsen, 2001; 809 Molhave, 2003).

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6. Designing biological purifiers

Common biological processes for VOCs abatement include bio- 812 scrubbers, biotrickling filter, and biofilters (Iranpour et al., 2005; 813 Burgess et al., 2001; Delhoménie and Heitz, 2005; Revah and Morgan-814 Sagastume, 2005). In bioscrubbers, the air is washed with an aqueous 815 phase into which the pollutants transfer, and the aqueous phase is 816 transferred into a bioreactor where the pollutants are biodegraded. In 817 biotrickling filters, microorganisms are grown on an inert material 818 (plastics resins, ceramics etc). An aqueous solution containing the 819 nutrients required for microbial growth is continuously distributed 820 and recirculated at the top of the reactor and percolates by gravity, 821 thus covering the biofilm with an aqueous layer. Contaminated air is 822 introduced as co-or counter current and the contaminants diffuse into 823 the aqueous phase where they are biodegraded. The purpose of the 824 packing material is to facilitate the gas and liquid flows and enhance gas/825 liquid contact, to offer a surface for microbial growth, and to resist 826 crushing and compaction. In biofilters, air is passed through a moist 827 porous material which supports microbial growth. Water remains within the packing material and is added intermittently to maintain humidity and microbial viability. The packing material is generally a natural material (peat, compost, wood shavings, etc.) which is biodegradable and provides nutrients to the microorganisms although intensive research has been done to use synthetic materials (Jin et al., 2006).

Because they provide large interfacial areas for exchange, biofilters are typically recommended for low-substrate concentrations and poorly soluble substances. On the other hand, biotrickling filters and bioscrubbers allow higher surface volumetric loading rates and are more suitable for conditions of fast transfer (high pollutant concentration) or when pH needs to be controlled. As indoor air biofiltration implies low transfer and high volumetric flow, it is not clear which of the configurations described above will be the best. However, biofilmbased technologies offer many advantages in regards of the microbial properties required and the utilization of a biodegradable (i.e. compost) or bioactive (i.e. soil, plants roots, plant leaves) support for growth might allow microbial activity at low-substrate concentration. It is therefore not surprising that botanical purifiers, which are based on configurations taking advantage of air exchange through the plant root-soil or plant-foliage areas, have been more extensively studied for indoor air purification so far.

Emerging technologies could provide a more suitable platform for indoor air biological purification. Biological indoor air treatment can potentially release dust, microorganisms, and water. These problems can be simultaneously solved by using membrane bioreactors which physically disconnect the sorption step (air—water exchange) from the biodegradation step. The use of membrane bioreactors for VOC removal is nowadays only established at high pollutant loads (Ergas et al., 1999) but recent studies have demonstrated that such systems could be efficient at indoor air concentration levels (Llewellyn and Dixon, 2006, Ramis et al., 2007). The use of membrane could allow more compact designs for indoor air treatment. This is clearly a very promising technology which should be further investigated.

An additional common limitation to all biological air treatment processes is the need to transfer contaminants into an aqueous phase prior to their biodegradation, which is especially problematic in the case of hydrophobic pollutants such as hexane. The addition of a hydrophobic organic phase into the bioreactors (two liquid phase partitioning bioreactors) could significantly enhance the transfer of the pollutants to the microorganisms and thereby, their removal (Muñoz et al., 2007). Other possibilities include the addition of activated carbon or other adsorbents in combination with the biological system. Such approaches should be investigated in the case of indoor air treatment as they could also concentrate the contaminants to levels suitable for growth.

7. Conclusions

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Poor indoor air quality is a worldwide problem with tremendous human health and economical consequences. Although technologies for particle removal are rather well established, there are nowadays no satisfactory methods for VOC control because removing indoor VOC sources or increasing ventilation rates is often not feasible or economical. There is a therefore a need for designing specific air purifying devices to clean and circulate the air inside affected buildings. Among the technology potentially suitable for this purpose are biological systems replying upon the ability of plants and/or organisms to detoxify organic compounds. However, a critical review of the existing literature in regards to biological and engineering constrains reveals numerous problems that must be solved before biologically-based air purifiers can be designed and implemented.

First, our current knowledge on microbial kinetics and the thresholds for substrate uptake, consumption and gene expression raise serious doubt concerning the feasibility of microbial degradation of VOCs at indoor air concentrations. Yet, we also know biological systems (in a broad sense) respond to indoor VOCs because of the linkages between 892 VOC occurrence and SBS. In addition, there is experimental evidence 893 that VOCs can be biologically removed at indoor concentration even if 894 the precise mechanisms are unknown. This apparent contradiction is 895 perhaps explained by the fact that our current knowledge was derived 896 from studies conducted under conditions (single strains with single 897 substrate at high concentration) irrelevant to the indoor air environment 898 (diverse communities exposed to multiple substrates at low concentra- 899 tions and direct pollutant uptake). Clearly, there is a need for 900 fundamental research under indoor relevant conditions. This would 901 not only help to design and optimize indoor air biological purifiers but 902 also to solve growing environmental issues linked to trace contaminants 903 in water resources. One of the most interesting areas of research is 904 perhaps the study of heterotroph-phototroph relationships such as 905 those observed in plants. This could explain why plants "appear" to 906 remove VOC at trace concentration when this hardly benefit them (from 907 a detoxification point of view) and why microorganisms could degrade 908 indoor VOCs and survive starvation.

Second, the design of biological air purifier requires the develop- 910 ment of new technologies for highly efficient pollutant transfer (from 911 air to the biological catalyst) in order to allow high volumetric 912 treatment flows while maintaining high treatment efficiencies. 913 Current biological purifiers have shown some potential but are all 914 limited by their low treatment capacity. Solutions could be found 915 among technologies (i.e. membrane bioreactors) that liberate from the 916 need to transfer VOC to an aqueous phase. This opens interesting 917 possibilities for cross-disciplinary research initiatives.

Finally, as IAQ is linked to the presence of pollutants other than VOC 919 and as biological methods might always be limited in the cases of poorly 920 soluble or recalcitrant substances, there is a need to develop combined 921 physicochemical-biological methods. This is especially necessary to 922 eliminate potential nuisances from the biological purifier itself.

8. Uncited references

US EPA 925 US Green Building Council 926

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References 936

Aguado S, Polo AC, Bernal MP, Coronas J, Santamaria J. Removal of pollutants from 937 indoor air using zeolite membranes. J. Membr. Sci. 2004;240:159-66. Alagappan G, Cowan R. Substrate inhibition kinetics for toluene and benzene degrading 939 pure cultures and a method for collection and analysis of respirometric data for 940 strongly inhibited cultures. Biotechnol. Bioeng. 2003;83:798-810. Andersson K, Bakke JV, Bjørseth O, Bornehag CG, Clausen G Hongslo JK, et al. TVOC and 942 health in non-industrial indoor environments. Indoor Air 1997;7:78-91. 943 944 Air Quality Solution Ltd. www.naturaire.com Arriaga S, Revah S. Improving hexane removal by enhancing fungal development in a 945 microbial consortium biofilter. Biotechnol. Bioeng. 2005;90:107-15. 946 Auriol M, Filali-Meknassi Y, Tyagi RD, Adams CD. Laccase-catalyzed conversion of natural 947 and synthetic hormones from a municipal waster. Water Res. 2007;41:3281-8. Avoko GA, Volatile organic compounds in indoor environments, In: Hutzinger O, editor, 949 The handbook of environmental chemistry. Springer-Veriag; 2004, p. 1-35 950 Beveridge TJ, Makin SA, Kadurugamuwa JL, Li Z. Interaction between biofilms and the 951 environment, FEMS Microbiol, Rev. 1997;20:291-303. 952

Brown VM, Crump DR. Volatile organic compounds. In: Berry et al RW, editor. Indoor air 953 establishment. Watford, Herts: Indoor Environment Ltd; 1996. 954
Brunekreef B, Houthuijs D, Dijkstra L, Boleij JS. Indoor nitrogen dioxide exposure and 955 children's pulmonary function. I. Air Waste Manage. Assoc. 1990:40:1252–6. 956

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1027

1028

1029

1030

1031

1034

1035

1036

- Burge PS, Sick building syndrome, Occup, Environ, Med. 2004;61:185-90.
- Burgess JE, Parsons SA, Stuetz RM. Developments in odour control and waste gas treatment biotechnology: a review, Biotechnol, Adv. 2001:19:35-63.
 - Butterfield PW Camper AK Ellis BD Jones WI. Chlorination of model drinking water biofilm: implications for growth and organic carbon removal. Water Res. 2002:36:4391-405.
 - Canada MdAeS Directives d'exposition concernant la qualité de l'air des résidences Rapport du Comité consultatif fédéral-provincial de l'hygiène du milieu et du travail. Canada: Ottawa, Ministre des Approvisionnement et Services: 1987. p. 1-31
 - Carlsson H, Nilsson U, Ostman C. Video display units: an emission source of the contact allergenic flame retardant triphenyl phosphate in the indoor environment. Environ. Sci. Technol. 2000:34:3885-9.
 - Chain PSG, Denef VJ, Konstantinidis KT, Vergez LM, Agullo L, et al. Burkholderia xenovorans LB400 harbors a multi-replicon, 9.73-Mbp genome shaped for versatility, PNAS 2006:103:15280-7
 - Chen W, Zhang JS, Zhang Z. Performance of air cleaners for removing multiple volatile organic compounds in indoor air. ASHRAE Trans. 2005:111:1101-14.
 - Crump D. Strategies and protocols for indoor air monitoring of pollutants. Indoor Built. Environ 2001:10:125-31
 - Daisey JM, Hodgon AT, Fisk WJ, Mendell MJ, Ten Brinke J. Volatile organic compounds in twelve Californian office buildings: classes, concentrations and sources. Atmos. Environ, 1994:28:3557-62.
 - Darlington A, Chan M, Malloch D, Pilger C, Dixon MA. The biofiltration of indoor air: implications for air quality. Indoor Air 2000;10:39-46.
 - Darlington AB, Dat JF, Dixon MA. The biofiltration of indoor air: air flux and temperature influences the removal of toluene, ethylbenzene, and xylene. Environ. Sci. Technol. 2001:35:240-6
 - Darlington AB. An integrated indoor air biofiltration system for municipal infrastructure. University of Guelph; 2004. p. 1-50.
 - Delhoménie MC, Heitz M. Biofiltration of air: a review. Crit. Rev. Biotechnol. 2005:25:53-72.
 - Devinny JS, Deshusses MA, Webster TS. Biofiltration for air pollution control. Boca Raton: Lewis Publishers; 1999.
 - Dudler R, Eberl L. Interactions between bacteria and eukaryotes via small molecules. Curr. Opin. Biotech. 2006;17:268-73.
 - Edwards RD, Jurvelin J, Koistinen K, Saarela K, Jantunen M. VOC identification from personal and residential indoor, outdoor and workspace microenvironment samples in EXPOLIS-Helsinki. Finland Atmos. Environ. 2001;35:4829-41.
 - Ekberg LA. Volatile organic compounds in office buildings. Atmos. Environ. 1994;28:3571-5
 - Ergas SJ, Schroeger ED, Chang DPY. Biodegradation technology for volatile organic compound removal from airstream. Final report. Contract No. A032-127. California Environmental Protection Agency, Air Resources Board, Research Division; 1992. May.
 - Ergas SJ, Shumway L, Fitch MW, Neemann JJ. Membrane process for biological treatment of contaminated gas streams. Biotechnol. Bioeng. 1999;63:431-41.
 - Fisk WJ. Health and productivity gains from better indoor environments and their relationship with building energy efficiency. Annu. Rev. Energy Environ.
- Howard-Reed C, Nabinger SJ, Emmerich SJ. Measurement and simulation of the indoor air quality impact of gaseous air cleaners in a test house. 9th International Conference on Indoor Air Quality and Climate. Proceedings, vol. 2. . Indoor Air; 2002.
- Hudnell HK, Otto DA, House DE, Mølhave L. Exposure of humans to a volatile organic mixture II. Sensory. Arch. Environ. Health 1992;47:31-8.
- Iranpour R, Cox HHJ, Deshusses MA, Schroeder ED. Literature review of air pollution control biofilters and biotrickling filters for odor and volatile compound removal. Environ. Prog. 2005;24:254-67.
- Jin Y, Veiga MC, Kennes C. Development of a novel monolith-bioreactor for the treatment of VOC-polluted air. Environ. Technol. 2006;27:1271-7.
- Johnston PK, Hadwen G, McCarthy J, Girman JR. A screening-level ranking of toxic
- chemicals at levels typically found in indoor air. US-EPA 2002:930-6. Jones AP. Indoor air quality and health. Atmos. Environ. 1999;33:4535-64.
- 1020 Kennes C, Veiga MC. Fungal biocatalysts in the biofiltration of VOC-polluted air. J. 1021 Biotechnol. 2004;113:305-19. 1022 Kesselmeier J, Staudt M. Biogenic volatile organic compounds (VOC): an overview on
- 1023 emission, physiology and ecology. J. Atmos. Chem. 1999;33:23-88. 1024 Kostiainen R. Volatile organic compounds in the indoor air of normal and sick houses. 1025
- Atmos. Environ. 1995;29:693-702. 1026
 - Kovárová-Kovar K, Egli T. Growth kinetics of suspended microbial cells: from singlesubstrate-controlled growth to mixed-substrate kinetics. Microbiol. Mol. Biol. Rev. 1998:62:646-66.
 - Larimer FW, Chain P, Hauser L, Lamerdin J, Malfatti S, Do L, et al. Complete genome sequence of the metabolically versatile photosynthetic bacterium Rhodopseudomonas palustris. Nat. Biotechnol. 2003;22:55-61.
- 1032 Llewellyn D. Dixon MA. Botanical-membrane hybrid for the biofiltration of indoor air. Proceedings of the USC-TRG Conference on Biofiltration for Air Pollution Control, 1033 Long Beach, California; 2006. p. 5-84.
 - Lomborj B. The skeptical environmentalist. Cambridge: Cambridge University Press; 2002, p. 182.
- MacLeod CT, Daugulis AJ. Biodegradation of polycyclic aromatic hydrocarbons in a two-1037 1038 phase partitioning bioreactor in the presence of a bioavailable solvent. Appl. Microbiol, Biotechnol, 2003;62:291-6. 1039
- 1040 Market report: indoor air quality, Filtration Industry Analyst, 2004, p.14.
- 1041 Marshall KC. Microbial adhesion in biotechnological processes. Curr. Opin. Biotechnol. 1042 1994:5:296-301.

- Miekisch W. Schubert IK. Noeldge-Schomburg GFE. Diagnostic potential of breath 1043 analysis – focus on volatile organic compounds. Clin. Chim. Acta 2004;347:25–39. 1044
- Miller MJ, Allen DG. Modeling transport and degradation of hydrophobic pollutants in 1045 biofilters biofilms. Chem. Eng. J. 2005;113:197-204. 1046
- Mohd Al, Hiroyasu M, Yoshimasa Y, Kimitoshi F, Katsumi N. Removal of mixtures of 1047 acetaldehyde and propionaldehyde from waste gas in packed column with 1048 immobilized activated sludge gel beads. Biochem. Eng. J. 2001;8:9-18. 1049
- Molhave L. Organic compounds as indicators of air pollution. Indoor Air 2003;13:12-9. 1050 Molin S. Tolker-Nielsen T. Gene transfer occurs with enhanced efficiency in biofilms and 1051 induces enhanced stabilisation of the biofilm structure. Curr. Opin. Biotechnol. 1052 2003:14:255-61 1053
- Mosqueron L, Nedellec V. Hiérarchisation sanitaire des paramètres mesurés dans les 1054 bâtiments par l'observatoire de la qualité de l'air intérieur. Observatoire de la 1055 Qualité de l'Air Intérieur: 2002, p. 2-98. 1056
- Mosqueron L, Nedellec V. Revue des enquêtes sur la qualité de l'air intérieur dans les 1057 logements en Europe et aux Etats-Unis. Observatoire de la qualité de l'air intérieur; 1058 2004. p. 1-55. 1059
- Muir DCG, Howard PH. Are there other persistent organic pollutants? Environ. Sci. 1060 Technol. 2006:40:7157-66. 1061
- Muñoz R, Villaverde S, Guieysse B, Revah S. Two phase partitioning bioreactors for the 1062 treatment of volatile organic compounds. Biotechnol. Adv. 2007;25:410-22. 1063

1066 **Q**7

1070

1082

1090

- Newman LA, Reynolds CM. Phytodegradation of organic compounds. Curr. Opin. 1064 Biotechnol. 2004:15:225-30. 1065
- OQAI (Observatoire de la Qualité de l'Air Interieur). http://www.air-interieur.org/
- Ondarts M, Platel V, Hort C, Sochard S. Evaluation of indoor air treatment by two pilot- 1067 scale biofilters packed with compost and compost-based material. Proceedings of 1068 the 15th International Conference on Air Pollution 2007: Modelling, Monitoring 1069 and Management of Air Pollution. Algarve, Portugal; 2007.
- Orwell RL, Wood RL, Tarran WJ, Torpy F, Burchett MD. Removal of benzene by the indoor 1071 plant/substrate microcosm and implications for air quality. Water Air Soil Pollut. 1072 2004:157:193-207.
- Orwell RL, Wood RA, Burchett MA, Tarran J, Torpy F. The potted-plant microcosm 1074 substantially reduces indoor air VOC pollution: II. Laboratory study. Water Air Soil 1075 Pollut. 2006;177:59-80.
- Otake T, Yoshinaga J, Yanagisawa Y. Analysis of organic esters of plasticizer in indoor air 1077 by GC-MS and GC-FPD. Environ. Sci. Technol. 2001;35:3099-102. 1078
- Otson R, Fellin P, Tran Q. VOCs in representative Canadian residences. Atmos. Environ. 1079 1994;28:3563356-9. 1080
- Ottengraf SPP, Konings JHG. Emission of microorganisms from biofilters. Bioprocess 1081 Eng. 1991;7:89-96.
- Pahm MA, Alexander M. Selecting inocula for the biodegradation of organic compounds 1083at low concentration. Microb. Ecol. 1993;25:275-86.
- Parvatiyar MG, Govind R, Bishop DF. Treatment of trichloroethylene (TCE) in a 1085 membrane biofilter. Biotechnol. Bioeng. 1996;50:57-64. 1086
- Pasanen AL. A review: fungal exposure assessment in indoor environments. Indoor Air 10872001;11:87-98.
- Phillips M. Method for the collection and assay of volatile organic compounds in breath. 1089Anal. Biochem. 1997;247:272-8.
- Pilotto LS, Douglas RM, Attewell RG, Wilson SR. Respiratory effects associated with 1091 indoor nitrogen dioxide exposure in children. Int. J. Epidemiol. 1997;26:788-96.
- Ramis A, Hort C, Sochard S, Platel V. Treatment of the confined air of a spacecraft cabin 1093 biotechniques for air pollution control. International Congress for Air Pollution 1094 Control, A Coruna, Spain 3-5 October; 2007. p. 323-73.
- Revah S, Morgan-Sagastume JM. Methods for odor and VOC control. In: Shareefdeen Z, 1096 Singh A, editors. Biotechnology for odour and air pollution. Heidelberg: Springer- 1097 Verlag; 2005. p. 29-64.
- Rittmann BE, McCarty PL. Environmental biotechnology: principles and applications. 1099 Mc-Graw Hill; 2001.
- Robbins CA, Swenson LJ, Nealley ML, Gots RE, Kelman BJ. Health effects of mycotoxins in 1101 indoor air: a critical review. Appl. Occup. Environ. Hyg. 2000;15:773-84.
- Roch F, Alexander M. Inability of bacteria to degrade low concentrations of toluene in 1103 water. Environ. Toxicol. Chem. 1997;16:1377-83. Rodes C, Kamens R, Wiener RW. The significance and characteristics of the personal 1105
- activity cloud on exposure assessment measurements for indoor contaminants. 1106 Indoor Air 1991:1:123-45.
- Sakai K, Norbäck D, Mi Y, Shibata E, Kamijima M, Yamada T, et al. A comparison of indoor 1108 air pollutants in Japan and Sweden: formaldehyde, nitrogen dioxide, and 1109 chlorinated volatile organic compounds. Environ. Res. 2004;94:75-85.
- Salthammer T. Emission of volatile organic compounds from furniture coatings. Indoor 1111 Air 1997:7:189-97
- Samet JM, Lambert WE, Skipper BJ, Cushing AH, Hunt WC, et al. Nitrogen dioxide and 1113 respiratory illnesses in infants. Am. Rev. Respir. Dis. 1993;148:1258-65. 1114
- Sander R. Compilation of Henry's law constants for inorganic and organic species of 1115 potential importance in environmental chemistry; 1999. http://www.mpch-mainz. 1116 mpg.de/~sander/res/henry.html. 1117
- Sandhu A, Halverson LJ, Beattie GA. Bacterial degradation of airborne phenol in the 1118 phyllosphere. Environ. Microbiol. 2007;9:383-92. 1119
- Schäffner A, Messner B, Langebartels C, Sandermann H. Genes and enzymes for in- 1120 planta phytoremediation of air, water and soil, Acta Biotechnol, 2002;22:141-52. 1121
- Schleibinger H. Keller R. Rüden H. Indoor air pollution by microorganisms and their 1122 metabolites. In: Pluschke P, editor. The handbook of environmental chemistry, vol. 1123 4. Berlin Heidelberg: Springer -Verlag; 2004. p. 149–77. Part F. 1124
- Schmitz H, Hilger U, Weinder M. Assimilation and metabolism of formaldehyde by leaves 1125 appear unlikely to be of value for indoor air purification. New Phytol. 2000;147:307-15. 1126
- Shareefdeen Z, Singh A. Biotechnology for odor and air pollution control. Heidelberg: 1127 Springer-Verlag; 2005. 1128

ARTICLE IN PRESS

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1129 Shaughnessy RJ, Levetin E, Blocker J, Sublette K. Effectiveness of portable indoor air 1130 cleaners: sensory testing results. Indoor Air 1994;4:179–88.

Shaughnessy R, Sextro R. What is an effective portable air cleaning device? A review. J.
 Occup. Environ. Hyg. 2006;3:169–81.
 Singh R. Debarati PD. Jain RK. Biofilms: implications in bioremediation. Trends

1133 Singh R, Debarati PD, Jain RK. Biofilms: implications in bioremediation. Trends 1134 Microbiol. 2006;14:389–97.

Staudinger J, Roberts PV. A critical review of Henry's law constants for environmentalapplications. Crit. Rev. Environ. Sci. Technol. 1996;26:205–97.

Subba-Rao RV, Rubin HE, Alexander M. Kinetics and extent of mineralization of organic chemicals at trace levels in freshwater and sewage. Appl. Environ. Microb. 1982;43:1139–50.

Sundell J. On the history of indoor air quality and health. Indoor Air 2004;14:51–8.
 Torres EM, Basrai SS, Kogan V. Evaluation of two biotechnologies controlling POTW air
 emissions. Proceedings of 1996 USG-TRG Conference on Biofiltration. Los Angeles,
 CA: University of Southern California; 1996. p. 182–97.

Tros ME, Bosma TNP, Schraa G, Zehnder AJB. Measurement of minimum substrate concentration (Smin) in a recycling fermentor and its prediction from the kinetic parameters of *Pseudomonas sp.* Strain B13 from batch and chemostat cultures. Appl. Environ. Microb. 1996;62:3655–61.

US EPA. Air and steam stripping of toxic pollutants. Tech. Rep. EPA-68-03-002.
 Cincinnati, OH, USA: Industrial Environmental Research Laboratory; 1982.

Q81150 US EPA. Indoor Air Quality. http://www.epa.gov/iaq/index.html;

van der Kooij D, Vrouwenvelder HS, Veenendaal HR. Kinetic aspects of biofilm
 formation on surfaces exposed to drinking water. Water Sci. Technol. 1995;32:61–5.
 US Green Building Council. www.usgbc.org

1154 Van Groenestijn JW, Hesselink PGM. Biotechniques for air pollution control. 1155 Biodegradation 1993;4:283–301.

Van Groenestijn JW, Kraakman NJR. Recent developments in biological waste gas
 purification in Europe. Chem. Eng. J. 2005;113:85–91.

Vaughan TL, Strader C, Davis S, Daling JR. Formaldehyde and cancers of the pharynx,sinus, and nasal cavity. II. Residential exposures. Int. J. Cancer 1986;38:685–8.

Vergara A, Haaren B, Revah S. Phase partition of gaseous hexane and surface
 hydrophobicity of Fusarium solani when grown in liquid and solid media with
 hexanol and hexane. Biotechnol. Lett. 2006;28:2011–7.

1163 Wallace LA. Personal exposure to 25 volatile organic compounds. EPA's 1987 team study
 1164 in Los Angeles. California. Toxicol. Ind. Health 1991;7:203–8.

Wallace LA. Human exposure to volatile organic pollutants: implications for indoor airstudies. Annu. Rev. Energy Environ. 2001;26:269–301.

Wargocki P, Wyon DP, Baik YK, Clausen G, Fanger PO. Perceived air quality, sick building
 syndrome (SBS) symptoms and productivity in an office with two different
 pollution loads. Indoor Air 1999;9:165–79.

Wargocki P, Sundell J, Bischof W, Brundrett G, Fanger PO, et al. Ventilation and health in 1170 non-industrial indoor environments: report from a European Multidisciplinary 1171 Scientific Consensus Meeting (EUROVEN). Indoor Air 2002;12:113–28. 1172

Weisel C, Zhang J, Turpin BJ, Morandi MT, Colome S, et al. Relationships of indoor, 1173 outdoor, and personal air (RIOPA), Part I. Collection Methods and descriptive 1174 Analyses. Health Effect Institute; 2005. p. 1–144.

WHO. Air quality guidelines for Europe. Second Edition. Copenhagen: World Health 1176 Organisation; 2000.

Wieslander G, Norbäck D, Björnsson E, Janson C, Boman G. Asthma and the indoor 1178 environment: the significance of emission of formaldehyde and volatile organic 1179 compounds from newly painted indoor surfaces. Int. Arch. Occup. Environ. Health 1180 1997;69:115–24.

Wolkoff P, Clausen PA, Nielsen PA, Molhave L. The Danish Twin Apartment Study, Part I: 1182 Formaldehyde a long term VOC measurements. Indoor Air 1991;1:478–90. 1183

Wolkoff P, Clausen PA, Jensen B, Nielsen GD, Wilkins CK. Are we measuring the relevant 1184 indoor pollutants? Indoor Air 1997;7:92–106. 1185
Wolkoff P, Nielsen GD. Organic compounds in indoor air their relevance for perceived 1186

workon P, Neisen GD. Organic Compounds in indoor air their relevance for perceived 1160 indoor air quality? Atmos. Environ. 2001;35:4407–17.

Wolkoff P. Trends in Europe to reduce the indoor air pollution of VOCs. Indoor Air 1188

pollutants from energy-efficient homes. Econ. Bot. 1984;38:224–8. 1191 Wolverton, B.C., Johnson, A., Bounds, K., Interior landscape plants for indoor air 1192

pollution abatement, National Aeronautics and Space Administration, 1989. p.1–22. 1193 Wood RA, Burchett MA, Alquezar R, Orwell RL, Tarran J, Torpy F. The potted-plant 1194

microcosm substantially reduces indoor air VOC pollution: 1. Office field-study. 1195 Water Air Soil Pollut. 2006;175:163-80. 1196 Yu H, Kim BJ, Rittmann BE. A two-step model for the kinetics of BTX degradation and 1197

Yu H, Kim BJ, Rittmann BE. A two-step model for the kinetics of BTX degradation and 1197 intermediate formation by *Pseudomonas putida* F1. Biodegradation 2001;12:465–75. 1198

Yu C, Crump D. A review of the emission of VOCs from polymeric materials used in 1199 buildings. Build. Environ. 1998;33:357–74.

Zhang JJ, Smith KR. Indoor air pollution: a global problem. Br. Med. Bull. 1201 2003;68:209-25.

Zhou X, Mopper K. Apparent partition coefficients of 15 carbonyl compounds between 1203 air and seawater and between air and freshwater: implications for air-exchange. 1204 Environ. Sci. Technol. 1990;24:1864–9.

Zilli M, Camogli G, Nicolella C. Detachment and emission of airborne bacteria in gas-, 1206 phase biofilm reactors. Biotechnol. Bioeng. 2005;91:707–14.

Zumairi MS, Roulet CA, Tham KW, Sekhar SC, Cheong KWD, Wong NH, et al. A 1208 comparative study of VOCs in Singapore and European office building. Build. 1209 Environ. 2006;41:316–29.