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1 Process Technology and Sustainability Assessment of 2 Wastewater Treatment

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15 Abstract

16 Removal of heavy metals in wastewater treatment is crucial to protect the environment, wildlife,
17 and human health. Various techniques have been developed focusing on heavy metal ions,
18 pharmaceuticals and other contaminants' removal from different wastewater sources. The main
19 methods include adsorption, filtration, ion exchange, electrochemical, reverse osmosis,
20 precipitation, floatation flotation/coagulation/flocculation and photocatalytic-based treatments.
21 This paper comprehensively and critically reviews and discusses common technologies used
22 for wastewater process treatment, applications, and their sustainability assessment. The
23 sustainability profile depends majorly on the exact approach followed for each technology,
24 including its energy consumption, type of radiation (where appropriate), auxiliary materials
25 used (e.g. catalysts, adsorbents), and further specific experimental process settings. Thus, while
26 sustainability inevitably provides a multi-faceted answer, the review finally aims for
27 sustainability benchmarking of all technologies, by compressing the manifold outcomes
28 towards a compact information set, such as a table and radar plot.

29 **Keywords:** wastewater treatment, heavy metal removal, pharmaceutical; wastewater process
30 technology, sustainability assessment

34 **1. Introduction**

35 Toxic compounds and heavy metals in wastewater have been found increasingly in the
36 industrial revolutions and human daily activities [1]. It is reported that heavy industries such as
37 textile, paper manufacturing, batteries, mining, plating and electroplating, petrochemicals, etc.
38 are the main sources causing water pollution [2]. The advert effects of contaminated wastewater
39 are enormous since it threatens human health and destroys the balance of the Earth's ecosystem
40 [3]. Especially, the non-biodegradation and carcinogenic generation of heavy metals might
41 cause critical health issues to live organisms, for example, cancer in human beings [4].

42 Traditionally, membrane filtration, reverse osmosis, adsorption, photocatalytic reduction
43 coagulation, flocculation, flotation, ion exchange, chemical precipitation, electrochemical
44 reduction, etc. are popularly used for heavy metal ions removal as shown in Figure 1 [5].
45 Although those technologies were reported to be effective for wastewater treatment, their
46 disadvantages in terms of environmental impacts and huge sludge generation during treatment,
47 causing the additional cost of processing and transportation, cannot be ignored [6].



48
49 **Figure 1** *Different treatment approaches for the removal of heavy metal ions in wastewater (Reprinted*
50 *from [5] with permission of Elsevier)*

51
52 The development of these wastewater treatment processes is largely driven by rising
53 environmental awareness together with more stringent regulatory requirements. They were set
54 in the background of a multitude of large-scale industrial processes, with different kinds of
55 wastewaters necessitating a variety of processing options to meet their cost, environmental and
56 product-quality needs in the diverse origin.

57 Recent publications regarding sustainability focusing on wastewater treatment have revealed
 58 the challenges to remove all kinds of pollutants that are presented in wastewater from traces to
 59 grams per litre. Most of them include applications of Life Cycle Assessments (LCA) as a way
 60 for benchmarking performances of methodologies, pointing out advantages and disadvantages
 61 in each case, with the environmental impacts.

62 In this study, several wastewater treatment technologies will be assessed to determine their
 63 sustainability from the data of the literature review. Our calculations are mainly based on the
 64 reported metrics and general conclusions obtained from the previous publications. The selection
 65 criterion of the literature sources was based on the homogenisation of the functional unit to the
 66 extent, possible in terms of scale. As shown in Table 1 the scope of studies was urban
 67 wastewater, wastewater treatment (including industrial), and water desalination, giving all of
 68 them a *cradle-to-gate* perspective. Ecoinvent database is commonly used for all three software
 69 namely SimaPro, Gabi, and OpenLCA. Furthermore, CML and ReCiPe-2016 were popularly
 70 applied for most of the LCA studies relevant to wastewater treatment.

71 **Table 1** Summary of references and modeling assumptions for assessing the impact of
 72 wastewater treatment.

<i>Technology</i>	<i>Approach</i>	<i>Functional unit</i>	<i>Scope</i>		<i>Software</i>	<i>Methodology</i>	<i>Database</i>	<i>Refs.</i>
<i>Membrane filtration</i>	Monetization	1 m ³ water	Agriculture irrigation	Cradle-to-gate	OpenLCA 1.10.2	ReCiPe-2016	Ecoinvent	[7]
<i>Coagulation</i>	Effluent minimization	1 m ³ water	Urban wastewater	Cradle-to-gate	GaBi Professional 8.6	ReCiPe-2016	Ecoinvent	[8]
<i>Reverse osmosis</i>	Processes benchmarking	1 m ³ water	Water desalination	Cradle-to-gate	SimaPro™ v.8	CML	Ecoinvent 3.1	[9]
<i>Ion exchange</i>	Water boiler treatment	10 ³ m ³ WW	Water desalination	Cradle-to-gate	SimaPro™ v.7.3	CML	Ecoinvent	[10]
<i>Catalytic reduction</i>	Water purification	1 m ³ water	Urban wastewater	Cradle-to-gate	SimaPro™ v.6.0	CML	Ecoinvent	[11]
<i>Electrochemical reduction</i>	COD removal	1 kg of HCOOH	Wastewater treatment	Cradle-to-gate	SimaPro 8.2.3.0	Dynamic model	Ecoinvent 3.0	[12]
<i>Adsorption</i>	Water purification	1 m ³ water	Urban wastewater	Cradle-to-gate	SimaPro™ v.6.0	CML	Ecoinvent	[11]

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 74
 75

76 **2. Process Design and Sustainability of Wastewater Treatment Technologies**

77 In this section, we will provide information on the popular technologies and their process
78 designs that are currently applied worldwide for wastewater treatment, targeting toxic and
79 heavy metal removals. The pros and cons as well as their practical applications will also be
80 mentioned based on the updated literature reviews. Finally, the sustainability of each
81 technology will be evaluated and compared using data from previous papers.

82 **2.1. Membrane Filtration**

83 **2.1.1. Process technology**

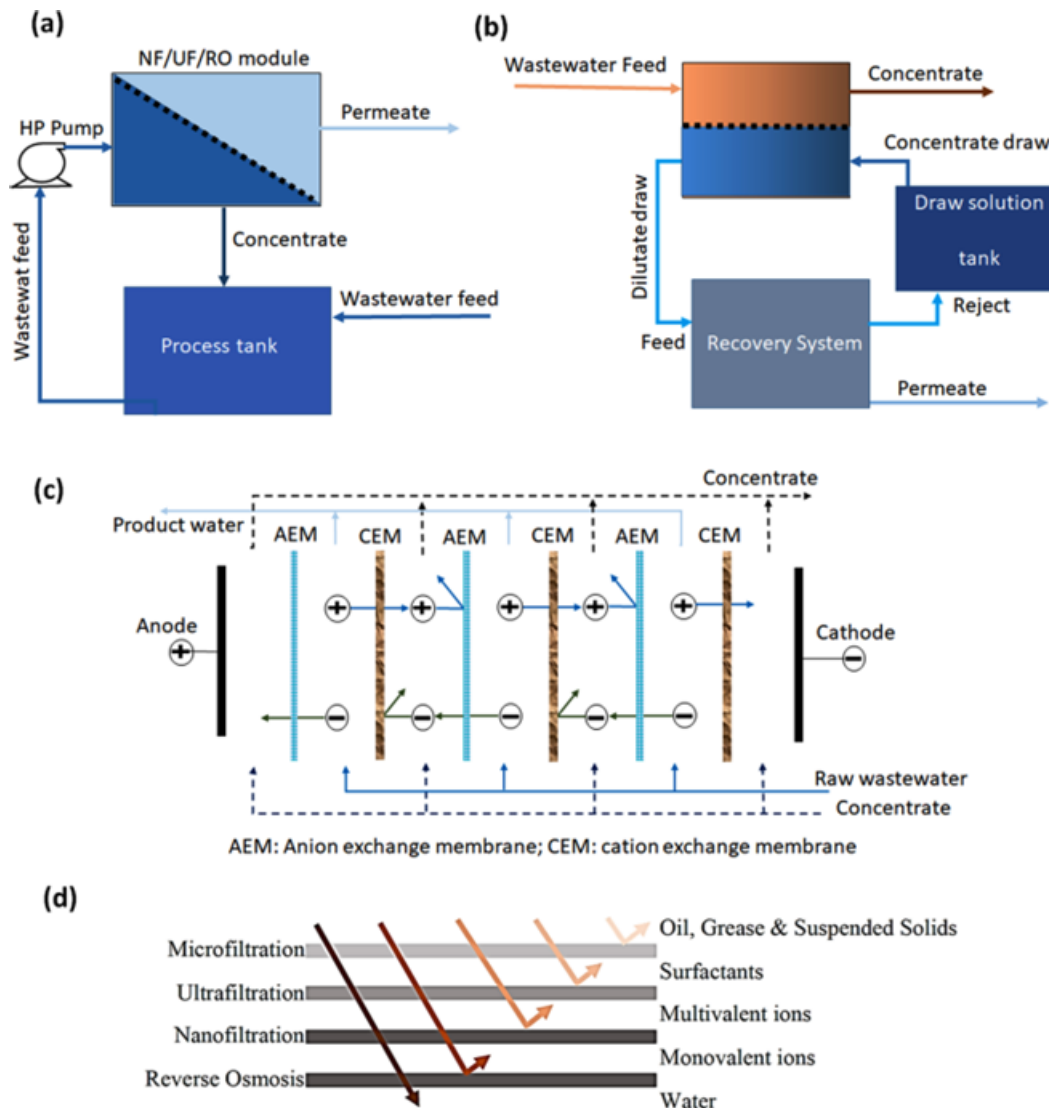
84 Direct membrane filtration (DMF) uses membranes, and typically needs to add driving forces
85 to its core process (direct filtration) to achieve full performance. The various driving forces are
86 pressure, osmosis, thermal, and electrical. DMF receives more attention due to the decreased
87 membrane price and the improved membrane performance. DMF uses membrane separation to
88 remove the organics/nutrients from wastewater, which gives high permeate quality at a high
89 water recovery ratio. DMF processes are compact and require only a small footprint which may
90 have advantages of simplified pre-processing, e.g., not requiring an additional activated sludge
91 process, and reducing energy consumption. The co-production of organics and nutrients at high
92 concentrations can be utilised to generate renewable energy (H_2 , CH_4) or can be converted to
93 fertilisers. In this way, DMF has the potential for carbon neutrality if nutrient recovery is added
94 to the wastewater treatment. As for any membrane process, membrane fouling is a serious threat
95 to industrial application. This holds especially in the presence of organic contents by adding
96 biological degradation, as a further step. This emphasises the role of pretreatment for DMF.

97 DMF can generate high-quality reclaimed water from wastewater. This superior permeate water
98 quality can be used for undrinkable waters such as gardening, reclaimed, irritated, or bathroom
99 flushing waters, etc. Figure 2 shows the concept of membrane-based treatments for toxic
100 compounds and metal ions removal from both waste and salty water.

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Figure 2 Membrane technologies to remove pollutants from waste and salty water: a) reverse osmosis; b) forward osmosis; c) electrodialysis and d) the separation capabilities of different membranes against different pollutants (Reprinted from [2] with permission of Springer-Nature)

109 DMF is the most simple method and is among the best ones as being able to be operated as a
110 direct pressure-driven membrane process [13]. Here, the membrane removes particles,
111 organics, nutrients, and pathogens from wastewater by the principle of physical separation. This
112 has the advantage of not necessitating biological processes, resulting in an overall simplified
113 process. DMF can add osmosis (FO) membrane filtration, allowing the passing of water through
114 a semi-permeable FO membrane [14]. The input solution is usually kept at relatively low
115 osmotic pressure as compared to drawing the solution, at high osmotic pressure, which
116 generates the differences in osmotic pressure. FO has the advantage in terms of energy savings
117 since it can effectively remove both organic and inorganic matters without the need for high

118 pressure. This reduces costs on both energy consumption and capital equipment (e.g. pumps).
119 As a crucial advantage, the relatively low formation and compaction of cake layers in FO result
120 in a less fouling potential in comparison to other membrane techniques.

121 In membrane distillation (MD), physical membrane separation is combined with thermal
122 distillation [15]. The key performance parameter is to establish a high vapor pressure difference,
123 which is achieved by setting a high-temperature difference between the feed and permeate
124 solutions. Another key performance parameter is material-based and relates to the membrane
125 characteristics. Influential is here the choice of materials and their chemical, mechanical, and
126 thermal resistance, pore size, hydrophobicity, and surface roughness. MD is favoured, when
127 industrial wastewater is available at high temperature, which means utilising this energy of the
128 wastewater stream and reducing the overall energy demand [16]. In this way, waste streams of
129 low-grade thermal energy sources can be purified such as that origin from waste heat, solar
130 energy, and geothermal heat. In addition, municipal wastewater can be treated with high
131 performance, ensuring high-quality permeate for reclamation and highly concentrated retentate
132 for resource recovery. However, severe fouling might be a problem for MD due to the
133 significant differences in operating temperatures.

134 Electrodialysis (ED) is another upgrade of direct membrane filtration and leverages ion
135 exchange through membranes to transport ions using electric potential. ED technology works
136 best with industrial wastewater having the same content of contaminants, such as comprising
137 brine wastewater and seawater [17]. Consequently, ED is applied for desalination to produce
138 clean water. In practice, ED is preferred since it offers a simple technology that can overcome
139 the limitations of RO in terms of water recovery, modularity, process control, membrane
140 durability, and operating costs, etc. ED in conjunction with adequate post-treatment favors co-
141 valorisation by enabling selective separation of nutrients, toxic compounds, heavy metal ions,
142 and organic and inorganic materials.

143 **2.1.2. Applications**

144 DMF systems were first utilised in the 1990s by Butler and MacCormick [18]. Since then,
145 positive pressure DMF systems have been employed for the treatment of different types of
146 wastewater including greywater, municipal wastewater, and industrial wastewater. In all of
147 these processes, the main role of the membrane was to remove particles, nutrients, and
148 pathogens (to some parts) from the bulk of the wastewater stream. DMF can generate water
149 quality for domestic and industrial non-potable water such as planting, fertilisation of the soil,

150 toilet flushing, cooling water, and water for stripping columns. Recently, DMF is used for
151 resource recovery as well. For example, Li et al. [19] experimented with a lab-scale UF with
152 the size of 0.0062 μm at a constant pressure of 0.12 bar to treat greywater sourced from the
153 ecological settlement. They successfully treated TOC (from 161 to 28.6 mg/L), while reporting
154 a reduction in the treatment flux from 10 to 6 LMH within two weeks of continuous operation.
155 The same trend was also observed by Jin et al. [20] in which a lab-scale MF (size: 1 μm) was
156 utilised to treat municipal wastewater with a COD concentration of 200-500 mg/L. The
157 permeate after the treatment had < 30 mg/L COD showing successful treatment of wastewater.
158 However, the flux decreased from 20 to 10 LMH within 350 hours of continuous operation.
159 Overall, DMFs are plausible for industrial and greywater treatment, the treatment flux reflected
160 a transient behaviour for all cases and decreased with time spanning.

161 **2.1.3. Sustainability assessment**

162 Concerning this technology, Canaj et al. [7] probed the high dependence of local conditions on
163 environmental measurements, discriminating between local and global LCA impact categories.
164 Global warming potential (GWP), fossil fuel, and toxicities were considered global effects. For
165 example, the release of CO_2 into the atmosphere (GWP increase) is a global phenomenon,
166 changing the temperature of the planet. Differently, other categories such as water consumption
167 and marine eutrophication were considered local effects, as the raise of nutrient-induced
168 phytoplankton productivity occurs on a local scale and there is no spread to the wider
169 environment. In this study, up to 18 environmental impacts on wastewater treatment were
170 calculated using membrane filtration, including an associated costs assessment. The latter
171 showed how positive local environmental aspects can be turned into a financial gain, which can
172 also help to mitigate the negative global environmental effects. In terms of technology, it was
173 found that wastewater membrane technology is beneficial for the local environment, but harms
174 the greater environment. The negative impact of electricity is the main cause of the latter. Teow
175 et al. [21] also concurred with the dominance of electricity consumption (41.6 MJ/6 m^3 water
176 in the best scenario) in the overall environmental impact using membrane filtration and reverse
177 osmosis (described below). Categories such as terrestrial acidification, eutrophication, and
178 human toxicity are also highly influenced by energy consumption, due to the combustion of
179 fossil fuels, and the subsequent emissions of carbon dioxide (CO_2), sulphur dioxide (SO_2),
180 nitrogen oxides, and volatile mercury. Electricity consumption accounts for 73% of GWP, 80%
181 of terrestrial acidification, 51% of eutrophication, and 43% of human toxicity. These results are
182 consistent with the ones reported by Canaj et al. [7] regarding membrane filtration, showing

183 positive impact on ecotoxicity (0.63 kg 1,4-dichlorobenzene equivalent (1,4-DCB-eq)/m³ ww),
184 terrestrial acidification (0.02 kg SO₂-eq/m³ ww), freshwater eutrophication (0.001 kg PO₄-
185 eq/m³ ww) and marine ecotoxicity (0.01 kg 1,4-DCB-eq/m³ ww). This finally and importantly
186 scores in the global warming (0.41 kg CO₂-eq/m³ ww) as compared with other methodologies.

187 **2.2. Reverse Osmosis**

188 **2.2.1. Process technology**

189 Reverse Osmosis (RO) is an industrially applied unit operation used extensively for wastewater
190 treatment. It is a membrane process, which works under the pressure (150 to 600 psig) [22].
191 The membrane material is either a thin film composite or a cellulose acetate membrane. RO
192 recovery and salt removal rates are not complete, yet on a typical level of 70-90% and 90-99%,
193 respectively. RO can be coupled to ion exchange to increase its efficiency [23].

194 On the positive side, RO membranes often have higher rejection rates for different
195 contaminants; thus, are widely applicable. They also facilitate the process of simplicity, as RO
196 can directly treat sewage into high-quality water without the need of adding chemical,
197 biological, and sorption processes [24]. On the negative side, fouling has to be mentioned. As
198 far as many membrane processes, the long-duration stability and operability depend largely on
199 the prevention of fouling, mineral scaling, and chemical degradation. RO Membranes are
200 solution-based, with permeants dissolving in the materials and then diffusing throughout the
201 membrane. As a consequence, they rely on a diffusion-controlled process, and mass transfer of
202 permeants takes place through a dense membrane. RO membranes are typical of hydrophilic
203 nature to allow them for processing water as a preferred solvent, thus not adding unnecessary
204 flow resistance [25]. Membrane processes, when properly working, can substantially reduce
205 costs; mainly by reducing operational costs in terms of materials and energy. They demand,
206 however, high initial capital costs, and also require high-cost pretreatment requirements [25].
207 On the environmental side, RO has concentrated disposal problems.

208 Membranes are often assembled in a spiral wound module of a tube format, which then can be
209 numbered up [26]. This needs the presence of separation spacer mesh in the feed channel which
210 is known to trap fouling particles and reduce flow throughput. Alternative RO design and
211 operations have been developed over the years to overcome the mentioned disadvantages. For
212 example, modified "open channel" modules with a limited scaling and fouling potential [27].
213 Advantages of RO separation are the absence of a need for a phase change, e.g. unlike
214 distillation, that is requiring only low-energy consumption. RO systems are also compact and

215 have ease of operation which allows also to take unskilled personnel into operation. The
216 inherent modular design of membrane processes facilitates their scale-up to a certain scale
217 (when the numbering-up becomes too costly). RO systems do not require regular maintenance.
218 Yet, there are also disadvantages of RO processes. The built-up of permeate flow requires high
219 pressure to exceed the osmotic pressure of the feed [28]. The RO membranes are susceptible to
220 concentration polarization and fouling, which requires pretreatment of the feed solution;
221 meaning to make them cleaner [29]. The membranes are high-performance materials, and as
222 such mechanically not very robust. Malfunction in the operation procedures can lead to their
223 deterioration. This also means a need for extensive cleaning procedures [28]. RO is also not a
224 fast procedure since it takes relatively long for purification. To the sophisticated equipment,
225 initial capital costs are high, and there has to be belief in the regaining of the investment over
226 the years of operation.

227 **2.2.2. Applications**

228 Chemical removal from industrial wastewater is one of the industrial and key examples of using
229 RO systems for treating chemically poisoned cooling water to preserve the environment. For
230 example, boron contents are one of the challenging chemical compounds to be removed from
231 the water. It is been demonstrated that a single-stage RO unit is not sufficient for reducing boron
232 from water to meet the threshold, defined in world standards and guidelines for water treatment
233 [30]. Recently, Hilal et al. [31] conducted a study to compare the feasibility of RO and its
234 techno-economic analysis for producing freshwater using single, or double pass RO for
235 seawater treatment containing $\sim 5 \text{ mg L}^{-1}$ of boron. They reported that for a double pass RO
236 system, the cost of treatment can be $0.55 \text{ \$ m}^{-3}$ for a permeating quality of $< 0.4 \text{ mg L}^{-1}$, while a
237 single pass RO represented $0.38 \text{ \$ m}^{-3}$. Similarly, Alabduljalil et al. [32] reported that the
238 production cost can be intensified to $1.43 \text{ \$ m}^{-3}$ when RO is utilised for deboronation of Persian
239 Gulf seawater containing 5.01 mg L^{-1} of boron. Overall, RO systems have been experimentally
240 and industrially proven for chemical removal from water.

241 **2.2.3. Sustainability assessment**

242 Reverse osmosis was assessed and compared in terms of sustainability indicators of different
243 wastewater treatment methods by Zhou et al. [33]. As concluded, the environmental assessment
244 varied when using the same functional unit to the extension of the life cycle inventory which
245 characterisation models of material flows were used. In another study, Pazouki et al. [9]
246 reported the same scale (1 m^3 wastewater) for a comparative desalination configurations study.

247 Considering the metric calculations shown in Table 2, reverse osmosis has a positive impact on
248 key indicators such as global warming, scoring in $0.07 \text{ kgCO}_2\text{-eq/m}^3$ wastewater, with 0.008
249 kg1,4-DCB-eq/m^3 for human toxicity, and $0.0002 \text{ kgSO}_2\text{-eq/m}^3$ for terrestrial acidification. Yet,
250 negative environmental impact is created through fossil abiotic depletion (2.59 MJ/m^3), as also
251 mentioned above regarding its correlation with energy consumption.

252 Ras et al. [10] investigated the environmental footprint of boiler feedwater desalination, and
253 used an unusually large scale of life cycle inventory (1000 m^3 as a functional unit), which means
254 that the results taken are supposed to be closer to a real life application. While increasing the
255 evaluation scale by three orders of magnitude as compared to Pazouki et al. [9], which used 1
256 m^3 as a functional unit, the impact on global warming, terrestrial acidification, and human
257 toxicity increased by four orders of magnitude. This possibly shows that LCA assessments
258 taken at too small might not be representative of real life processes. Yet Ras et al. also found a
259 positive result within their large scale evaluation, which is that marine ecotoxicity decreased by
260 a factor of 10.

261 **2.3. Ion exchange**

262 **2.3.1. Process technology**

263 The concept of this technology is based on the exchange of ions between two electrolytes or an
264 electrolyte solution and a complex. The applications of ion exchange are seen in process
265 purification, separation, or contamination removals from aqueous and heavy metal
266 contaminated solutions [34]. Typically, soil humus, zeolites, and ion exchange resins are
267 popularly employed based on cost-effectiveness [35]. Ion exchangers can also be categorised
268 as cation and anion exchangers following their electrochemical properties in which cation
269 exchangers will exchange positively charged ions while anion ones will exchange negatively
270 charged ions. Ion exchange, together with absorption and adsorption, can also be classified as
271 a form of sorption.

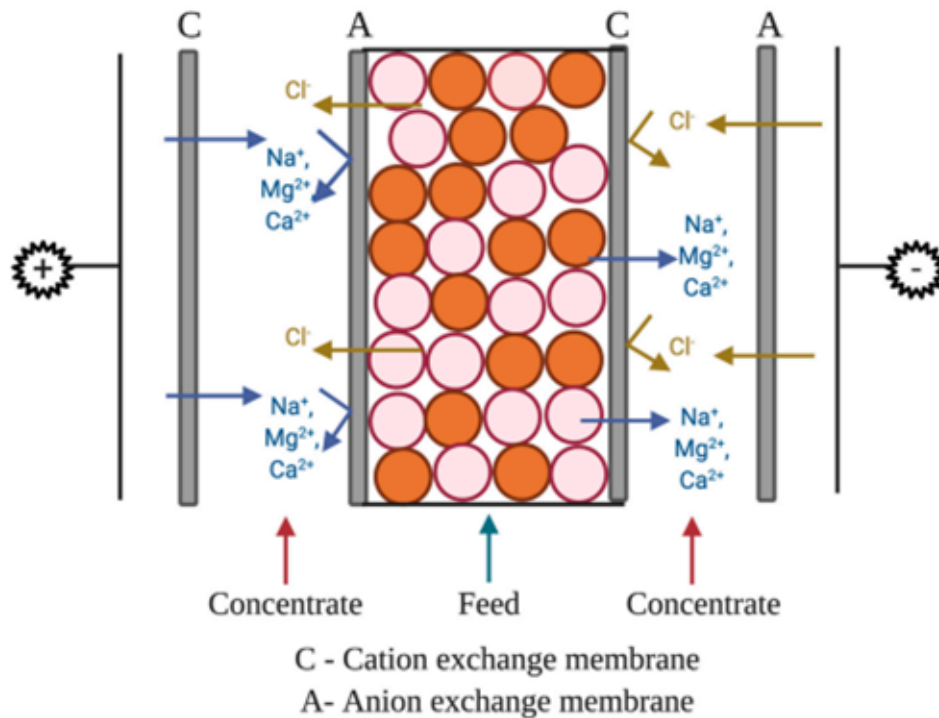
272 Ion exchange resins are among the most popular technologies and are usually applied in an ion
273 exchange system on a cyclic basis [36, 37]. To this end, the contaminated water is allowed to
274 pass through the resin basis until it is saturated. Normally, the water leaving the resin layers
275 will have a higher concentration of heavy metal ions than expected. The positive is that resin
276 can be then reactivated by backwashing to remove the accumulated solids, flushing the
277 unwanted ions from the resin by applying a concentrated solution of replacement resin [38].

278 Thus, the process is usually limited by the generation of backwash which requires further
279 treatment.

280

281 In practice, to increase the lifetime of the ion exchange resin unit, soluble organics should be
282 first removed to avoid the unit being overloaded before the demineralised process happens [39].

283 Figure 3 shows the water softening process using ion exchange resins.



284

285 **Figure 3** Illustration of electrodesion (ion exchange membranes) process for water
286 treatment. Reprinted from [40] with permission of MDPI

287 2.3.2. Applications

288 Ion exchange processes are widely used in industrial applications owing to their advantages
289 in terms of cost and energy saving [2, 41, 42]. The applications of ion exchange processes vary
290 from water treatment for power engineering, metal finishing, food and beverage industries, to
291 pharmaceutical production, and heavy industry wastewater treatment [43-45]. Especially, ion
292 exchange has been considered one of the most reliable methods to purify water that can be
293 utilised for high-purity required processes such as power generation, nuclear, and electronic
294 industries [45, 46].

295 Ion exchange processes are also popularly used for households since it provides a simple
296 solution for water purification to upgrade tap water to drinkable water. They are mainly
297 embedded in tap water filters to enhance the production of soft water through the removal of
298 both cation and anion found in the supply water [47, 48]. In wastewater treatment, inorganic or
299 polymeric ion exchangers are preferred due to cost-effectiveness and their ability to remove
300 heavy metals from highly polluted water [49, 50].

301 **2.3.3. Sustainability assessment**

302 Following a recent study conducted by Ras et al. [10], ion exchange technology was assessed
303 at a 10,000 m³ ww processing scale. Despite having several advantages in comparison to reverse
304 osmosis, marine ecotoxicity is very high, reaching 37 kg1,4-DCB-eq/1000 m³ ww which is
305 approximately 22 times higher than reverse osmosis. Similarly, the human toxicity impact,
306 takes a high score of 51 kg1,4-DCB-eq/1000 m³, being a factor of 2.2 higher than for reverse
307 osmosis. Yet, ion exchange technology provides benefits for some other environmental impacts
308 over reverse osmosis. For example, it scores better by a factor of 1.10, 1.49 and 2.22
309 respectively, in the LCA categories freshwater eutrophication (0.3 kgPO₄-eq/10³ m³), elements
310 of abiotic depletion (3.9 kgSb-eq/10³ m³), and global warming (534 kg CO₂-eq/10³ m³
311 wastewater). It is noteworthy to mention that the environmental footprint is caused by
312 fundamental elements of the two technologies, under investigation here; for example, the ion
313 exchange material determines largely the footprint of the whole technology concerning toxicity,
314 recycling, and biodegradability.

315 In this context, Choe et al. [51] conducted an LCA study on the influence of material
316 regeneration to mediate the environmental backpack of key materials of a process. For example,
317 brine material loss carries more than 80% of the total environmental impacts of the process,
318 which regeneration can improve. The negative impact is particularly strong on human toxicity,
319 ecotoxicity, and eutrophication above 90%, which is in line with the results provided by Ras et
320 al. [10]. Regeneration could decrease more than 80 % for all of the three impacts, thereby
321 reducing the former environmental harm below 20 %. This fact also produced a 25 % reduction
322 in GWP impact. The most significant reduction was ozone depletion with an impact reduction
323 higher than 95 %. This study probes the potential of this technology once the regeneration step
324 is solved.

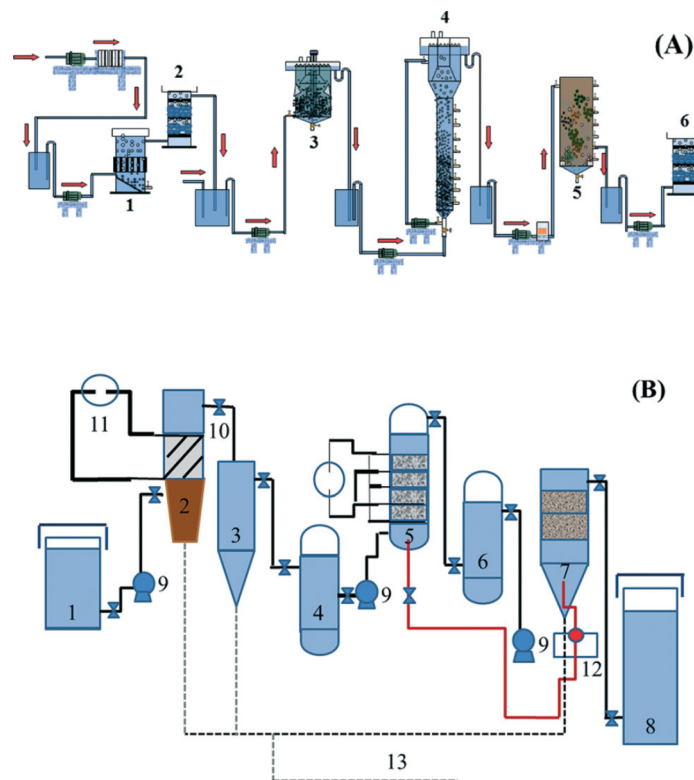
325 **2.4. Electrochemical Reduction**

326 **2.4.1. Process technology**

327 Electrochemical technology is one of the most effective technologies to remove heavy metals
328 and organic matters [52]. The fundamental of electrochemical is based on advanced oxidation
329 processes (AOP) and can be classified into subcategories such as electrochemical reduction,
330 electro-dialysis, electro-coagulation/flotation, and anodic oxidation [52].

331 The advantages of electrochemical technology are remarkable since it can be easily integrated
332 into other technologies to improve both removal capacities and energy efficiencies [53]. This
333 technology also offers a low operating cost solution, fewer chemicals used, and relatively high
334 purity of metallic and organic pollutant removal. Significantly, both heavy metals and organic
335 compounds can be separated and removed from wastewater at the same time through the
336 application of electrochemical technology [54].

337 In wastewater treatment, electrochemical technologies are usually applied at the pretreatment
338 stage to improve the biodegradability of the pollutants or as an advanced methodology to further
339 remove COD or decolourisation the water to satisfy the environmental standards. Figure 4
340 shows combined processes developed by Feng et al. [55] in which electrochemical technology
341 is applied as a finishing (A) or a screening process (B).



342

343 **Figure 4** *Flow diagram of combined electrochemical processes: (A) two phase anaerobic*
344 *(CSTR + EGSB)–aerobic (SBR)–electrocatalytic oxidation; (B) coagulation–electrocatalytic*
345 *oxidization–biological contact oxidation combined process (Reprinted from [55] with*
346 *permission of Royal Society of Chemistry)*

347 **2.4.2. Applications**

348 The electrochemical reduction can be classified into electrodeposition, cathodic
349 electrochemical dechlorination, and cathodic electrochemical denitrification [55].

350 Electrodeposition is widely applied in metallurgical and electroplating industries, printed circuit
351 boards, and battery manufacturing. Its ability to remove heavy metal ions or recover precious
352 metals is highly appreciated. The very first industrial application of electrodeposition was
353 reported back in mid-17th when it was applied to recover copper from cupriferous mine waters
354 [56]. Nowadays, electrodeposition is still operated in the mining industry to recover gold-rich
355 alloys through the application of a filter-press-type electrochemical flow reactor with highly
356 polished vitreous carbon (VC) and titanium (Ti) flat cathodes [57].

357 Cathodic dechlorination is a special electrochemical process designed for the removal of
358 chlorinated organic compounds (COCs) found in wastewaters related to herbicides, fungicides,
359 and pesticides industries. The advantages of this method are highlighted through its ability to
360 be effectively applied for the removal of a wide range of COCs such as chlorinated volatile
361 organic compounds (VOCs) [58, 59], polychlorinated hydrocarbons [60-62], and
362 polychlorophenols [63, 64]. The electrochemical dechlorination of aromatic compounds, such
363 as chlorobenzenes, was also reported for both academic and industrial applications.

364 Cathodic electrochemical denitrification is mainly applied for the removal of nitrate and nitrite
365 ions found in the ground waters [65-67]. The applications of precious alloys such as coinage
366 (copper, silver and gold) and transition-metal electrodes (platinum, palladium, rhodium,
367 ruthenium, iridium) have been reported for the treatment of solutions containing up to 0.1 M
368 nitrate ions in acid solutions [68-70]. It was also reported that the characterisation of the
369 electrode surface plays a vital role in the efficiency of the process, making it a very good topic
370 for electrochemical studies [71, 72].

371 **2.4.3. Sustainability assessment**

372 A study by Shemfe et al. [12] showed good environmental impacts regarding freshwater
373 eutrophication with 2.10^{-5} kgPO₄-eq/kg of HCOOH, human toxicity with 7.10^{-4} kg1,4-DCB-

374 eq/kg of HCOOH, terrestrial acidification with 0.002 kgSO₂-eq/kg of HCOOH, and ozone
375 depletion with 10⁻⁸ kgCFC11-eq/kg of HCOOH; see also Table 2. However, the global warming
376 potential scored low, with 0.17 kgCO₂-eq/kg of HCOOH. This disadvantage can be settled
377 through environmental benefits in freshwater ecotoxicity and fossil abiotic depletion with 6.78
378 kgCO₂-eq/kg of HCOOH (the highest in Table 2) and 2.79 MJ/kg HCOOH respectively.

379 Sustainability metrics can also refer to other functional units as secondary compounds or useful
380 by-products obtained in wastewater treatment. This would support reusing or recycling
381 compounds extracted from waste in a circular economy fashion. Indeed, this seems to be a
382 missed opportunity in wastewater research, considered by only a very few papers. One of those
383 exemptions is research from Shemfe et al. [12], which used 1 kg of formic acid (HCOOH) as
384 the LCA functional unit for a bio-electrochemical reduction-mediated water purification study.
385 HCOOH was a by-product recycled from the wastewater purification process. The assessment
386 included a dynamic LCA simulation and a techno-economic assessment for decreasing the
387 oxygen demand, and supporting the production of HCOOH. The costs for production of
388 HCOOH were in the range of €0.015 – 0.005 g⁻¹, which has to be seen because of the low
389 productivity of 0.094–0.26 kg y⁻¹. The study showed good environmental impacts regarding
390 freshwater eutrophication with 2.10⁻⁵ kgPO₄-eq/kg of HCOOH, human toxicity with 7.10⁻⁴
391 kg1,4-DCB-eq/kg of HCOOH, terrestrial acidification with 0.002 kgSO₂-eq/kg of HCOOH,
392 and ozone depletion with 10⁻⁸ kgCFC11-eq/kg of HCOOH; see also Table 2. To put those
393 results into a wider perspective, the environmental results suggest a large impact for the
394 recycling of HCOOH, in view that it is a product of global high demand.

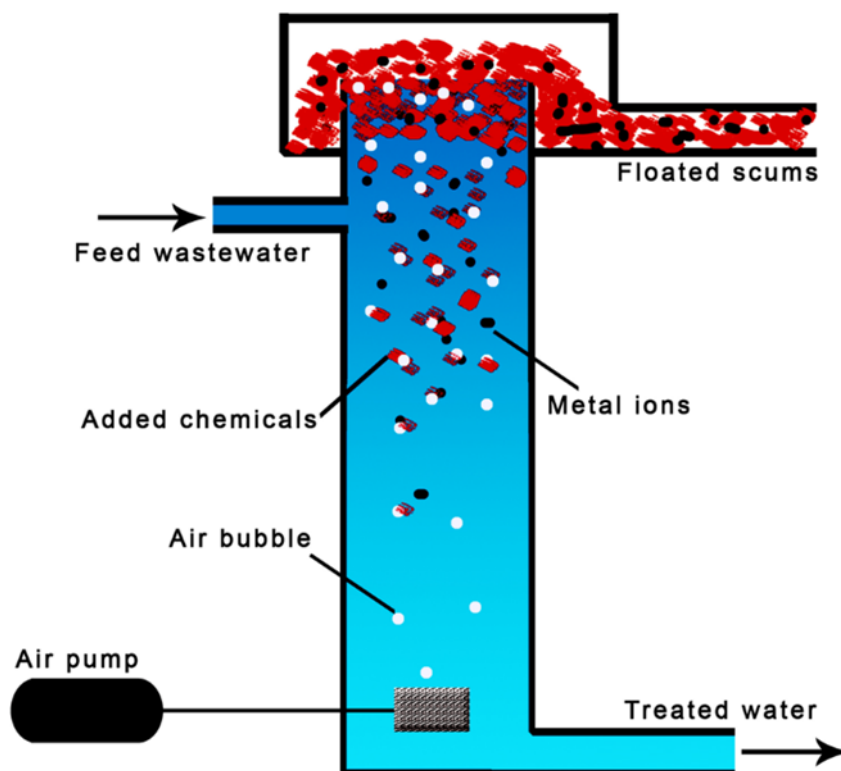
395 As an alternative valuable by-product, the use of bio-electrochemical reactors allows for the
396 secondary synthesis of methanol via the by-products CO₂ and H₂. Setting 1t methanol as a
397 functional unit, Streeck et al. [73] developed a microbial electrolysis cell using renewable
398 energy. This new technology's environmental impacts were compared with fossil-based
399 methanol production, and showed less global warming potential and consumption of fossil fuels
400 by a factor of 2.1^{-2.8} and 0.7^{-0.9}, respectively. A drawback of the bio-electrochemical process is
401 the increased material demand for metals and synthetic materials per unit of product, thus
402 making a negative contribution to the metal depletion potential by a factor of 0.3-0.6. Other
403 environmental impacts such as terrestrial acidification and freshwater eutrophication are also
404 worsened by a factor of 0.1-1.1 and 0.6-0.9, respectively.

405 **2.5. Coagulation and flocculation**

406 **2.5.1. Process technology**

407 The coagulation–flocculation process is among the widely used treatment processes for
408 industrial wastewaters [74]. The process occurs in two steps; strongly agitated mixing of the
409 dispersed coagulant within wastewater is followed by more soft agitation to initiate flocculation
410 by agglomeration of small particles into well-defined flocs. Those flocs are allowed to settle to
411 a sludge that is removed. The treated wastewater (supernatant) is then treated subsequently or
412 discharged. The key is to generate the gas bubbles in kinds of micro, medium, or macro-
413 bubbles. The coagulation–flocculation mechanism can be based on either ion, precipitate, or
414 sportive flotation [75]. A typical arrangement of a flotation system comprises the following
415 process equipment: a conditioning/feed tank with a mechanical mixer, a peristaltic pump, liquid
416 rota-meters, flotation column, a foam collection tank, an air compressor, washing trap, an
417 airflow meter, a porous diaphragm, a mercury U-tube manometer and effluent tank, a pH meter,
418 and a dosing pump.

419 In-line mixers are typically used for the first highly agitated mixing step, which has to be fast
420 [76]. For example, in-line mechanical mixers with variable speed impellers, and in-line jet
421 mixers are used. The establishment of velocity gradients is essential to the flocculation
422 performance and is achieved, i.e by baffling chambers; granular media beds; diffused air; spiral
423 flow chambers; reciprocating blades, and rotating blades. Figure 5 illustrates a schematic of the
424 flotation treatment process.



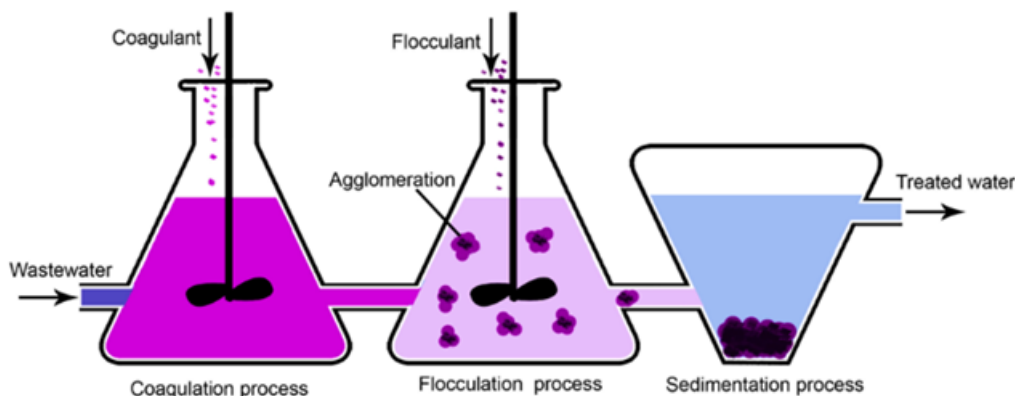
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426 **Figure 5** Schematic of the flotation treatment process (Reprinted from [2] with permission of
427 Springer-Nature)

428 More innovative process equipment has been described in the literature. One example is an in-
429 line mixing helical reactor for the flocculation and solid-liquid separation, termed flocs
430 generator reactor (FGR) and the flocculation-flotation reactor (FF) with integrated zigzag or
431 static mixers, which enables the generation of very light flocs (with entrained and entrapped
432 air) [77].

433 Flocculation

434 Flocculation is conventionally performed as precipitation–aggregation (coagulation/-
435 flocculation) settling as hydroxides or insoluble salts [78]. Flocculation can even be used for
436 co-valorisation, e.g. to selective recover critical minerals such as gold, palladium, silver.
437 Conventional processes are electro-flotation, dispersed (induced) air flotation, and dissolved air
438 (pressure) flotation (DAF). Emerging technologies give priority to stronger agitation, and are
439 nozzle flotation (NF), column flotation, centrifugal flotation (CF), and jet flotation. Figure 6
440 illustrates the fundamental of a coagulation-flocculation process.



441
442 **Figure 6.** An illustrative schematic of the coagulation-flocculation treatment process
443 (Reprinted from [2] with permission of Springer-Nature)

444 The coagulation–flocculation process has proven to be both simple in-process nature and
445 reactor design, energy-efficient, and effective in the treatment performance [79]. The main
446 process issues are decreasing flocculation times, achieving compact equipment size (small
447 footprint), and high efficiency. Coagulation and flocculation processes have also limitations
448 and challenges. This concerns the toxicity and health hazard posed by inorganic coagulants,
449 formation of a large amount of toxic sludge, ineffectiveness in removing heavy metals and other

450 contaminants, increase in effluent color, inefficient pollutant removal using natural coagulants
451 [2]. On top of that, the scaling up of coagulation–flocculation processes is complex [80].

452

453 **2.5.2. Applications**

454 Due to its high flexibility to various contaminants and wastewater types, coagulation and
455 flocculation have become popular and used widespread; for the treatment of dye/textile,
456 agricultural, food, pulp and paper, tannery, landfill leachate, and other industrial wastewaters
457 [81].

458 One of the most popular applications of coagulation and flocculation is to treat wastewater
459 generated through the textile industries. The utilisation of natural coagulants such as chitosan
460 or creature shells has been highlighted owing to their abundance, biodegradability, nontoxic
461 nature, and unique physicochemical properties because of the presence of primary amino
462 groups and high nitrogen content [78, 82].

463 Regarding the food industry, the coagulation-flocculation process was applied to treat molasses,
464 a byproduct obtained through sugar production, causing the dark color of sugar and generating
465 significant organic loadings to the wastewater. Liang et al. [83] reported the treatment of two
466 melanoidins-based molasses effluent, with different COD concentrations, employing ferric
467 chloride and alum. Results showed that higher melanoidin removal was obtained in the case of
468 ferric chloride since it possesses a higher affinity to the reaction sites of melanoidins than alum.

469 Additionally, coagulation and flocculation are also useful for the treatment of wastewater from
470 winery and brewery industries [84-86], pulp and paper industries [87-89], and leather industries
471 [90-92].

472 **2.5.3. Sustainability assessment**

473 Coagulation and flocculation technologies are close technologies since flocculation usually
474 follows coagulation to produce larger settleable flocs. Their way of connection determines the
475 entire process configuration, which can impact the energy expenses to the disadvantage of the
476 impact categories in a sustainability assessment. In this context, Rahmberg et al. [8] described
477 the precipitation of phosphorous (P) derivatives for wastewater P-removal using different
478 configurations which differ in time and method of precipitation. In terms of environmental
479 metrics, pre-precipitation turned out to be the best option. The calculated impacts, referred to

480 as 1m³ treated wastewater, resulting in this best scenario is 0.3 kg CO₂-eq/m³ wastewater for
481 global warming, 10⁻² kg PO₄-eq and 4.10⁻⁴ kg SO₂-eq in freshwater eutrophication and
482 terrestrial acidification, respectively. As noted in Table 2, 1.78 MJ and 7.10⁻⁸ kg Sb-eq are the
483 contributions regarding fossil abiotic and elements abiotic depletion, respectively, scoring
484 lowest among all impact categories as compared with other technologies applied to wastewater
485 treatment.

486 The sustainability of flocculation/coagulation can be improved by the use of natural coagulants.
487 This approach was taken by Ferreira et al. [93] by using a tandem of coagulation and
488 flocculation process sections followed by gravitational sedimentation of wastewater.
489 Environmentally friendly, non-toxic tannin-based coagulants were used together with algae
490 culture cells, which gave additional benefits by biomass harvesting. In the best scenario, the
491 best environmental performance was achieved for global warming (with 3.58 kgCO₂-eq/79.78
492 g of harvested biomass) and human toxicity (0.11 kg 1,4-DCB-eq/79.78 g of harvested
493 biomass). Adding to that, good environmental profiles resulted for terrestrial, freshwater, and
494 marine ecotoxicities, due to using the algae culture and not synthetic additives. Within all steps
495 in the whole process, cultivation and solids removal were the highest factors for the
496 environmental metrics was cultivation and solids removal, which in turn means the best process
497 solution was minimal for those stages.

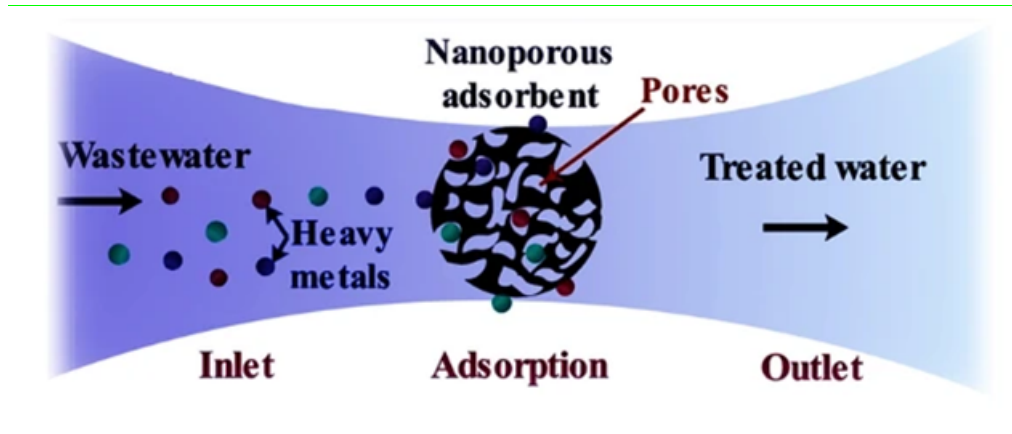
498 **2.6. Adsorption**

499 **2.6.1. Process technology**

500 Adsorption technology is the use of adsorbents to remove toxic compounds or heavy metals
501 from the wastewater based on the physicochemical properties [94]. Normally, the pollutants
502 will be adsorbed onto the surface of the adsorbent which has been modified or integrated with
503 a functional group. The efficiency of the process typically depends on the operating parameters
504 such as temperature, residence time, amount of adsorbent, level of the pollutants and pH value,
505 etc [94].

506 Adsorption is widely applied in industries since it offers a low cost and an effective method to
507 treat highly polluted water [95-97]. The technology is relatively simple while adsorbents can
508 be easily regenerated and recycled for further uses. Recently, nanoporous adsorbents have been
509 developed and effectively applied to remove metal ions from industrial wastewater as shown in
510 Figure 7 [97-99]. The fundamental of this application is to develop a highly porous nano-scale

511 material with a large surface area to absorb the metal ions which can be selectively designed
512 to target a specific metal. Significantly, the nanomaterials can be regenerated using a desorbing
513 agent that is not harmful to the environment.



514

515 **Figure 7** Schematics of an adsorption process (Reprinted from [2] with permission of
516 Springer-Nature)

517 In general, energies are required for the adsorption although it depends on the mechanism of
518 the process. Usually, physical adsorption requires fewer energies than chemical adsorption
519 since it does not require the establishment of bonding between the interface and the absorbed
520 species. Moreover, physical adsorption also happens quicker although this process is reversible.
521 Nevertheless, chemical adsorption tends to be more stable thanks to the strong forces (bonding)
522 between the adsorbate and the adsorbent established through the adsorption process [100].

523 The Gibbs free energy required for both physical and chemical adsorption processes can be
524 calculated by Equation 1 [101].

525
$$dG_{\text{ads}} = dH_{\text{ads}} - TdS_{\text{ads}} \quad (\text{Eq. 1})$$

526 where dG_{ads} , dH_{ads} , dS_{ads} represent the adsorption Gibbs free energy, the adsorption enthalpy
527 and the adsorption entropy, respectively, while T is the temperature of the system.

528 2.6.2. Applications

529 Wastewater treatment by adsorption fills gaps in the practical application of other wastewater
530 technologies and is a major answer to the environmental threat caused by heavy metals to
531 human health. This holds especially for emerging pollutants in the marine ecosystem, where

532 conventional wastewater treatment approaches fail. The commonality of the adsorption
533 technique arises from its ability towards flexible design, operation, low installation expense,
534 high performance and cost-effectiveness. Adsorption is mainly governed by material
535 inventions, and activated carbon is the most prominent adsorbent used; which includes biochars
536 as well as nano and hybrid adsorbents. However, separation and cost issues ask for the testing
537 of novel materials for wastewater adsorption treatment. The quest is for a large surface area,
538 great mechanical strength, and high chemical inertness.

539 In recent years, applications of adsorption were reported for the treatment of synthetic solutions.
540 The processes are effectively applied to remove a single adsorbate [102-104] while only a few
541 reports mentioned their applications for multi adsorbates removal [105, 106]. Magnetic
542 adsorbents have also been developed to enhance the phase separation after adsorption, allowing
543 the separation of adsorbents from the liquid by a simple application of a magnetic field [107-
544 110]

545 **2.6.3. Sustainability assessment**

546 The sustainability impacts of adsorption technology in wastewater treatment are highly
547 influenced by the choice of the adsorbent, *e.g.* activated carbon. This determines the
548 environmental profile and is highest for ozone depletion, fossil abiotic depletion, and global
549 warming. Muñoz et al. [11] quantified those to $7 \cdot 10^{-7}$ kg CFC11-eq, 155 MJ and 11 kg CO₂-eq,
550 respectively, which rank among the best 25% of the technologies (see Table 2). The same study
551 provides a high value for terrestrial acidification (0.12 kg SO₂-eq), freshwater ecotoxicity and
552 human non-carcinogenic toxicity (0.5 kg 1,4-DCB-eq in both cases), while listing a milder
553 impact in freshwater eutrophication (0.022 kg PO₄-eq). In particular, the toxicity of the
554 adsorbents has to be considered, which is central to human health. For example, the toxicity of
555 granular activated carbon, one of the most used adsorbents, was investigated using different
556 analysis methodologies, termed Environmental Design of Industrial Products (EDIP) [111],
557 Environmental Product Development Strategies (EPS) [112], and Ecoindicator 99 [113].
558 Regarding EDIP, the impacts concerning water chronic ecotoxicity ($7.8E+3$ m³) and human air
559 toxicity ($4.9E+6$ m³) scored highest, while human soil toxicity (34 m³) and soil chronic
560 ecotoxicity (107 m³) were the lowest. Regarding EPS, activated carbon is most critical for life
561 expectancy (1.9E-05 person.year), as well as nuisance (6.2E-04 person.year). In Ecoindicator
562 99, the reduction in life expectancy and diminished quality of life are considered. They are
563 expressed as DALY (Disability-Adjusted Life Year), being the sum of the life expectancy at

564 the time of death (YLL) and the number of years disabled (YLD). Applying these premises,
 565 respiratory inorganics were found to be the most adverse category (2.0E-05 DALY) while
 566 respiratory organics scored lowest (7.7E-09 DALY). The value of carcinogens (1.9E-06
 567 DALY) is high as well.

568 The main factor contributing to the sustainability of adsorption refers to the adsorbent itself,
 569 and in particular its adsorption capacity and speed, together with the recyclability. Therefore,
 570 new types of adsorbents have been investigated with a supposed better sustainability profile
 571 than conventional adsorbents such as activated carbons or aluminas, resins, zeolites, or
 572 molecular sieves. Sukmana et al. [114] introduced non-conventional adsorbents from renewable
 573 resources such as agriculture and other sources of biological by-products. These new adsorbents
 574 are claimed to have low cost, as these can directly be used after minor treatments [115] which
 575 lowers the environmental impacts associated with their production, together with the fact that
 576 waste materials are reused.

577 From a performance viewpoint, those new adsorbents are rich in functional groups, supporting
 578 their adsorptive capacity [116]. Bellahsen et al. [117] used pomegranate peel powder for
 579 ammonium removal, achieving in the best scenario 79 % efficiency (Table 2). Furthermore,
 580 Baby et al. [118] used palm kernel shells to remove wastewater heavy metals, achieving 99 %
 581 efficiency for Pb^{2+} and Cr^{6+} , and 83 % for Zn^{2+} and Cd^{2+} , with adsorption capacities between
 582 43 and 49 mg/g. In the same line, rice husk has shown good performances in nitrate and
 583 phosphate removal with good adsorption capacities as stated in Table 2. Combinations of bio-
 584 based materials with activated carbon can also be found in the literature. In this context,
 585 Zulkania et al. [119] used activated carbon (10 %) and a bio-sorbent produced from palm fibre
 586 wastes (30 %) with phosphoric acid for methylene blue removal, reporting adsorption capacities
 587 in the range of 10 mg/g with an efficiency around 99 % in 90 min.

588 **Table 2** Examples of natural bio-based adsorbents and their efficiencies and capacities
 589 regarding pollutant removal.

Bio-adsorbent	Adsorbate	Efficiency, %	Capacity, mg adsorbate/g adsorbent	Reference
Pomegranate peel powder	Ammonium	79	6.2	Bellahsen et al. [117]
Palm kernel shell	Heavy metals	83 – 99	43 – 49 (60 - 120 min.)	Baby et al. [118]

Triamine-activated rice husk ash	Nitrate		>160 (10 cycles)	Phan et al. [120]
10 % activated carbon + 30 % biosorbent + PO ₄ ³⁻	Methylene blue	99	9.8 (90 min)	Zulkania et al.
Activated rice husk ash	Phosphate	89	(120 min)	Mor et al. [121]
Magnesium chloride modified carbonized rice hull	Ammonium	86 – 90	41 (27 hours)	Thuy et al. [122]
Rice husk	Methylene blue and crystal violet	53 – 98		Quansah et al. [123]

590

591 Yet, there is an economic flip side of these sustainable bio-adsorbents, since Mo et al. [124]
592 reported drawbacks such as the low adsorption speed and the high pH efficiency dependence.

593 Such complex environmental response to the details of the adsorbent materials, and the way
594 they are made, is also found in other life-cycle impact categories; especially when comparing
595 adsorbent materials made from renewable and fossil resources. In principle, one would expect
596 from a renewable resource that the global warming potential is lower for the conventional fossil-
597 based adsorbent. Yet, literature shows antithetic evidence. Joseph et al. [125] found that the
598 CO₂ emissions from fossil sources obtained through biomass incineration resulted in an
599 increase in the global warming potential (GWP) at end-of-life, even considering that energy
600 recovery helped to reduce the negative effect. In this case, the authors assessed bio-based
601 activated carbon production from biomass mixture and biowaste, towards fossil-based
602 conventional equivalents at the end-of-life. 0.3 kg CO₂-eq. and 3.41 kg CO₂-eq. per kg
603 adsorbent were determined, respectively; which means an impressive improvement by a factor
604 of more than 11. The GWP advantage of the renewable route could be increased by optimized
605 processing, which improves the incineration step of biochar production. Yet, literature reports
606 result in conflict about similar bio-based adsorbents with GWP impacts of 11.1 kgCO₂-eq. for
607 olive waste cake [126], 1.15 kgCO₂-eq. for coconut shell [127], or 8.4 to 11.1 kgCO₂-eq., for
608 bituminous coal [128]. Consequently, the type of raw material and the production process
609 highly influence the global warming potential and sustainability, to an extent that may be higher
610 than for fossil-based procedures and adsorbents.

611 Similarly, mixed results are obtained for the energy expenses (cumulative energy demand,
612 CED) by Joseph et al. [125] being in the range of 15 – 30 MJ/kg, around half of the conventional
613 fossil-based procedure (53 MJ/kg). However, adverse results are known as well. The CED can

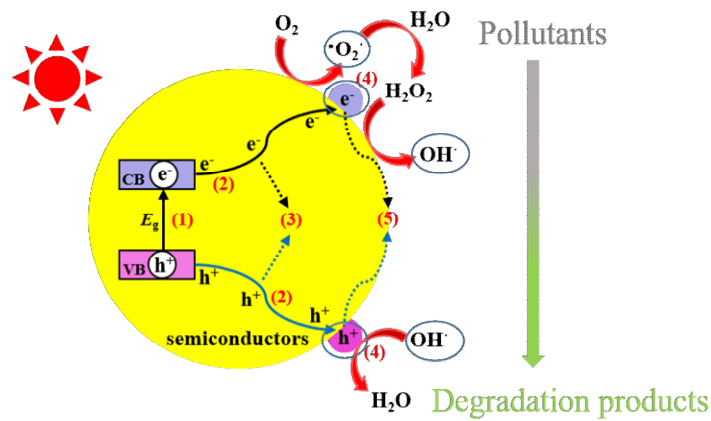
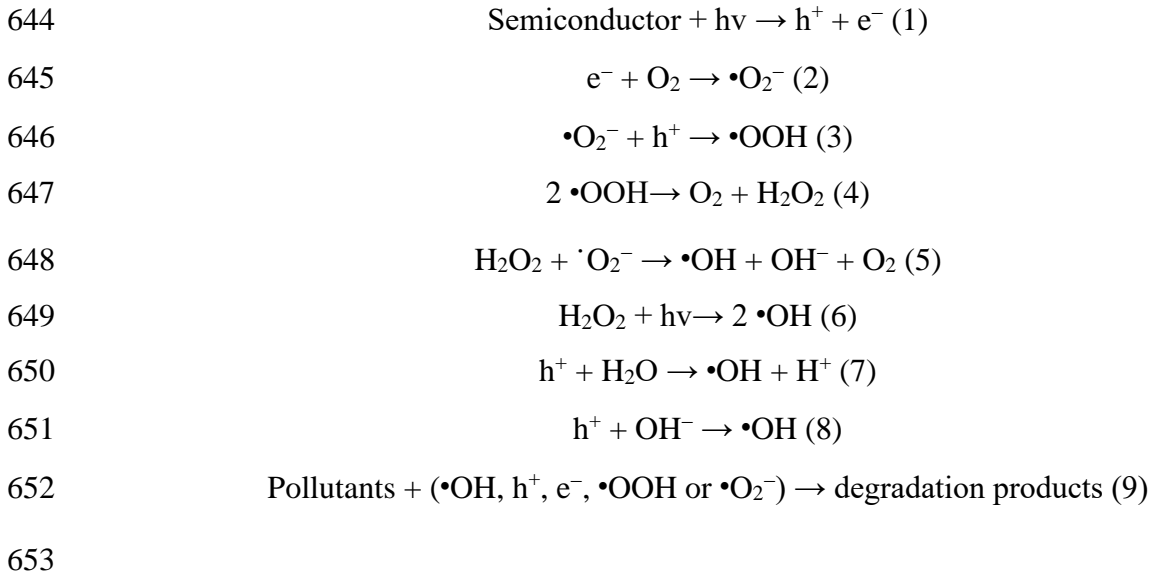
614 be higher for other bio-adsorbents, with Gu et al. [129] quantifying 153 – 241 MJ/kg for wood-
615 chip or coal-based activated carbons.

616 Regarding ecotoxicity, Joseph et al. found 20 to 80 % higher pollutant removal efficiency for
617 conventional carbon production as compared to bio-based adsorbents [125].

618 **2.7. Photocatalytic Reduction**

619 **2.7.1. Process technology**

620 Photocatalytic reduction is effective at degrading recalcitrant organic contaminants and is
621 considered to be an advanced oxidation process (AOP). Photocatalysis is proposed to be
622 combined with other treatment processes, such as biological treatments, to partially reduce total
623 organic carbon, break down macromolecular organic compounds, increase biodegradability,
624 and reduce the toxicity of produced water. It generates reactive oxygen or free radical species
625 as strong oxidants to degrade pollutants of nontoxic molecules. The light used is UV or visible
626 light, and the photocatalytic reaction is facilitated by semiconductors, with TiO₂ being most
627 commonly used. In this way, hydroxyl and superoxide anion radicals are generated, such as the
628 hydroxyl radical, and the superoxide anion radical. In principle, the excitation of electrons (e^-)
629 of the semiconductors transferred from the valence band to the conduction band, generating
630 holes (h^+) behind the valence band. For this reason, the narrower the bandgap is, the more
631 visible-light photons are captured. Another step of photocatalysis is the separation process of
632 photogenerated electrons and holes. Among them, the photogenerated electrons are widely
633 considered a reductant for directly reducing some heavy metals ions. The separated holes may
634 react with a hydroxyl ion (OH^-) or a water molecule to produce hydroxyl radicals ($\bullet\text{OH}$), and
635 also directly participate in the oxidative decomposition due to their strong oxidizability, which
636 is the primary pathway of production of $\bullet\text{OH}$. In addition, the separated electrons can react with
637 dissolved oxygen in the water to produce superoxide radicals ($\bullet\text{O}_2^-$); upon further reaction, the
638 decomposition produces $\bullet\text{OH}$. Before the photocatalytic reactions being occurred, the
639 photocatalytic nanomaterials absorb pollutants in the aqueous solution, increasing the charge
640 mobility and thus enhancing its redox ability. Then, various chemical reactions occur between
641 reactive oxygen species and pollutants, resulting in degradation products. The mechanism of
642 photodegradation is described in Figure 8. The photocatalytic processing is chemically
643 described in this the following equations:



654

655

656 **Figure 8** Photocatalytic degradation in the water treatment process (Reprint from [130] with

657 permission from MDPI)

658 **Table 3** Typical photocatalytic nanomaterials in wastewater treatment processing

659

Name	Target Pollutants	References
ZnO	Methylene blue	[131]
CaO	Methylene blue,	[132]
	Toluidene blue,	
ZnWO ₄	Rhodamine B	[133]
	Rhodamine-B	
	(RhB) and gaseous formaldehyde	
WO ₃	Amoxicillin	[134]

ZrO ₂	Methyl orange	[135]
BiTiO ₃	Rhodamine-B (RhB)	[136]
BiOBr	Rhodamine-B (RhB)	[137]
ZnFe ₂ O ₄	Malachite green (Isofar), rhodamine B	[138]

660

661 Toxic metal ions are changed to their metallic element state, metal oxides, or converted their
662 valence to non-toxic or lower toxicity via oxidation or reduction reactions. This kind of
663 processing offers advantages in terms of environmental friendliness, ease of handling, no need
664 to utilise additional chemicals, easy installation, significant efficiency, and no need to involve
665 the formation of sludge [139]. The catalytic activity of TiO₂ originates from its electronic
666 structure and photoelectric characteristics. The bandgap of this material enables its high
667 photocatalytic reaction activity, consisting of the valence band and conduction band. Electron-
668 hole pairs are generated when photocatalytic material is exposed to the light with equal or larger
669 energy than that of the photocatalyst's bandgap. Those hole-electronic pairs can conduct
670 oxidation and reduction. Important process issues to consider are the reuse of photocatalysts
671 and having an estimation for the costs. UV radiation is the most practical and effective
672 irradiation for the photocatalytic wastewater treatment process. The most relevant part of a
673 photocatalytic setup is the light source. UV generation was traditionally done by conventional
674 UV lamps, which are expensive, generate heat, and produce highly toxic waste. It stands to
675 reason that solar energy is the best and most natural radiation source, taken from an economic
676 and environmental point of view. However, the intensity of solar energy depends largely on
677 the site utilized in the world, and some countries have only moderate or low availability. Thus,
678 alternative radiation sources have to be used, even when heading for a solar-analog visible light
679 spectral emission. The use of light-emitting diodes (LEDs) has given unprecedented
680 opportunities to provide visible light in all spectral and dose flexibility, and to UV as well.
681 Consequently, the number of studies on photocatalytic water treatment with alternative visible
682 light and UV sources, such as solar energy and light-emitting diodes (LEDs) are used is
683 increasing, and as a result, there is an emerging number of photocatalysts have been developed.

684 The photocatalytic process might be accelerated by coupling it to a chemical process, turning
685 the initial photo intermediate molecule into the final reactive species for wastewater treatment.
686 This is done with the homogeneous Fenton reaction which creates reactive oxygen species by
687 decomposing H_2O_2 in the presence of iron salts in aqueous media.

688

689 **2.7.2. Applications**

690 Wastewater treatment by photocatalysis destroys a myriad of dyes made by industrial processes,
691 and thus helps the ecosystem by removing a broad toxic product portfolio [140]. The negative
692 environmental impact of those dyes is magnified by the fact that they are soluble in a water
693 environment and resistant to biodegradation. Thus, photocatalytic wastewater treatment can
694 manage to prevent a large ecosystem spread because of their toxicity. Rhodamine B, Methyl
695 Orange, and Methylene Blue are prominent dye examples.

696 Wastewater treatment by photocatalysis is also able to cope with the otherwise chemically
697 persistent hydrocarbon pollutants, comprising alkanes, olefins, and polycyclic aromatic
698 hydrocarbons [141]. In this way, wastewater treatment makes a notable contribution to
699 preserving Earth's marine environment being the prime effluence of hydrocarbon pollutants
700 [142].

701 Photocatalysis also can conveniently decompose phenols in wastewater. This impact is huge,
702 as phenols are platform chemicals, meaning, when coming directly out of refineries, they
703 determine a plethora of downstream chemical products ranging from paints and
704 pharmaceuticals to polymers. Similarly, as given above, the environmental profile of phenols
705 is harmed by their high water solubility, and being so toxic that they have been coined
706 "biological recalcitrant" [143]. The photocatalytic process can generate hydroxyl radicals
707 which attack the electron-rich (reactive) aromatic moiety to form oxidation decomposition
708 products such as hydroquinone, catechol, and p-benzoquinone. Finally, those molecules are just
709 intermediates on the way to small molecules such as acetylene, maleic acid, carbon monoxide,
710 and carbon dioxide. Some phenols such as chlorophenols are considered to be carcinogenic.
711 materials [144].

712 Photocatalytic wastewater applications also include the removal of heavy metal ions such as
713 Cr, Hg, Pb, Cd, Ni, Zn, and Mn ions into low-valence ions or zero-valence metals that have

714 lower harm. They have a mixed environmental profile; while being beneficial to metabolism,
715 they are toxic at high concentrations in drinking water and the food chain without any chance
716 for biodegradation [145]. Major global industries produce toxic heavy metal ions, including
717 metallurgy, mining, nuclear energy, and chemical manufacturing. Their removal adds a major
718 contribution to keeping clean surface and underground water resources. This majorly
719 contributes to human, animal, and crop health, because heavy metal ions destroy cells in living
720 organisms, by complexing into nucleic acids, proteins, and small metabolites.

721 In a similarly efficient way, the technology can treat pharmaceuticals, and their adverse impacts
722 on the ecosystem [146]. Other conventional wastewater treatment methods have limited success
723 with pharmaceuticals so photocatalysis becomes a unique solution here. A prime problematic
724 pollutant tackled by photocatalysis is antibiotics [147]. Similarly, the technology can degrade
725 anti-Inflammatories and lipid regulators.

726 Moving from medicines to food, pesticides are largely used in agriculture with a major impact
727 on water pollution and biological toxicity; which is intensified by their impact on human health
728 and carcinogenic potential [148]. This leads to the presence of viruses in wastewater that cause
729 a variety of gastrointestinal diseases. Their chemical stability and biodegradation resistance
730 make pesticides even more harmful [149, 150], Photocatalytic wastewater technology can cope
731 with those challenges [151-154]. Concerning the inactivation of microorganisms,
732 photocatalysis can provide reactive oxygen species (ROS) to destruct proteins and other
733 macromolecules and finally cause cell death [155, 156].

734 **2.7.3. Sustainability assessment**

735 Energy expenses are typically considered the main environmental hotspot for photocatalytic
736 wastewater technology. Yet, it must be admitted that the main material involved, the advanced
737 photocatalysts, are not given in LCA databases, which makes it difficult or impossible to
738 include those and perform a complete LCA inventory [157]. The use of laboratory data might
739 not be reliable here, as an extrapolation of their sustainability data to a large scale may be
740 erroneous. It might be even doubted, because of the complexity of the synthesis, if an industrial
741 scale can be ever realised for many of the modern advanced photocatalysts. Hence, what is
742 available are a few references regarding cost assessments, which are very specific for the
743 photocatalyst and its application is chosen [157]. Caution is needed to generalise those findings.

744 Coming back to energy expenses and seeing their sustainability from the side of the application,
745 clear thresholds can be drawn. Oxidation processes with $< 1 \text{ kWh/m}^3$ are considered suitable

746 for industrial wastewater applications to generate drinking water. Processes that reach this bar
747 are based on using chemical oxidants and radiation such as O₃, O₃/H₂O₂, O₃/UV, UV/H₂O₂,
748 UV/persulfate, UV/chlorine, and electron beam. More advanced technologies such as those
749 based on photo-Fenton, plasma, and electrolytic-related processes are middle energy consumers
750 (in the range of 1–100 kWh/m³), while UV-based photocatalysis, ultrasound, and microwave-
751 based photocatalysis rank high in energy consumption with >100 kWh/m³ [158]. As an
752 example, Benotti et al. used a UV/TiO₂-photocatalytic reactor for removing 32 pharmaceuticals
753 in wastewater [159]. Under the best conditions, the process consumed 4.24 kW h/m³, with a
754 removal efficiency of higher than 70% for 29 of the 32 compounds, and withdrawing only three
755 compounds to less than 50%.

756 Providing an assessment of other environmental categories, Muñoz et al. [11] investigated
757 photocatalytic reduction, showing a good profile in terrestrial acidification (0.02 kgSO₂-eq).
758 Nevertheless, this technology showed relevant impacts on freshwater and human ecotoxicities,
759 scoring 1.25 and 0.5 kg1,4-DCB-eq respectively. Regarding global warming, photocatalytic
760 reduction scored 5.9 kgCO₂-eq, which is an average compared with other technologies. In
761 addition, the technology ranked in a similar range with regard to freshwater eutrophication
762 (0.019 kgPO₄-eq). Interesting to note is a penalty for energy demand, being 64 MJ, which is
763 much above the average of other technologies. The ozone stratospheric depletion is low (3.10⁻
764 ⁷ kgCFC11-eq).

765 Yet, viewing photocatalytic reduction from its environmental purpose and not holistically, it is
766 highly efficient for contaminant removal; similarly, as adsorption is. The latter has attracted
767 attention due to its impact on eliminating toxicity, carcinogenic capacity and presence in
768 industrial wastewater [160] In this connection, Magdy et al. [161] evaluated the environmental
769 performance of phenol removal using either adsorption in front of different kinds of
770 photocatalysis such as electro-Fenton, solar photo-Fenton, solar photocatalysis by TiO₂, and a
771 tandem TiO₂-mediated photocatalysis and adsorption. The photo-Fenton process resulted to be
772 the most environmentally friendly, and the second most cost-effective, while Electro-Fenton
773 turned out to be the most expensive procedure and least environmentally friendly; especially
774 regarding the global warming potential and depletion of abiotic resources, due to high energy
775 demand and human toxicity. In between ranked adsorption and photocatalytic reduction
776 processes, the latter is highly influenced by the production of TiO₂. Adsorption is more cost-
777 effective with \$0.8 /m³ wastewater than photocatalytic reduction, which is double the cost. On
778 the contrary, adsorption is environmentally more beneficial than photocatalytic reduction,

779 especially for eutrophication, marine aquatic ecotoxicity, photochemical oxidation, and
780 acidification. In terms of Eco-indicator, photocatalytic reduction ranked ahead absorption with
781 0.26 pt, as compared with 0.68 pt scored by adsorption.

782 Pesqueira et al. [162], however, found the opposite result, that the photo-Fenton process had a
783 higher environmental impact than the TiO₂-mediated photocatalysis. As per the investigation
784 of the third process solar photolysis scored 0.025 kgCO₂-eq./kg FU (FU = 1 m³, ReCiPe) for
785 the global warming potential, while the solar TiO₂ and photo-Fenton processes showed more
786 release with 0.11 and 0.85 kgCO₂-eq./kg FU, respectively. Among all impact categories
787 investigated, the solar process scored the lowest, and the differences between TiO₂/photo-
788 Fenton and solar/photo-Fenton were found to be the second-highest. The most prominent
789 differences when comparing with those processes were given regarding the terrestrial
790 ecotoxicity, having gaps of 0.18, 0.6, and 0.7 kg1,4-DCB-eq./kg FU for the solar/TiO₂,
791 TiO₂/photo-Fenton, and solar/photo-Fenton processes, respectively (at absolute scores of 0.081,
792 0.27 and 0.87 kg1,4-DCB-eq./kg FU for solar, TiO₂, photo-Fenton). The photo-Fenton process
793 delivered the highest impacts for all impact categories, with exception of marine eutrophication,
794 land use and mineral resource scarcity. Here, the TiO₂ process gave the highest environmental
795 impact with 9.07E-6 kg N eq., 5.39E-3 m² eq. and 7.74E-3 kg Cu eq., respectively. The three
796 processes (for solar, TiO₂ and photo-Fenton) showed the lowest impact on the stratospheric
797 ozone depletion category (8.59E-9, 4.03E-8 and 5.96E-8 kgCFC-11 eq., respectively).

798 **3. Overview of Environmental Profiles of Common Wastewater Treatment Technologies**

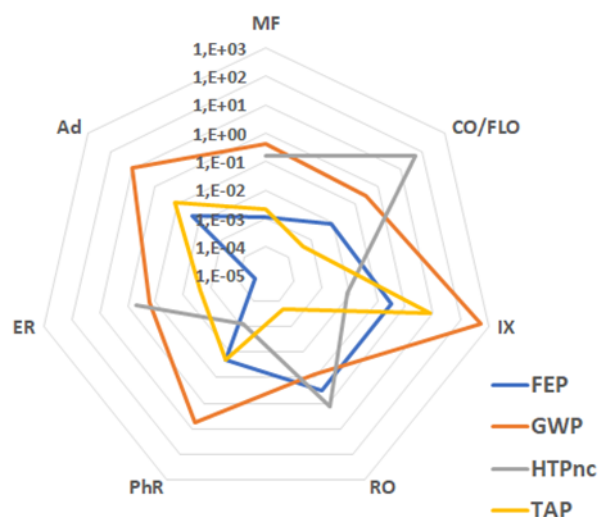
799 The above discussion in Section 2 has been made to embrace as much as possible criteria,
800 methodology, functional units, etc. It turns out that generalised statements judging the
801 sustainability of a whole class of process technology (versus another) can hardly be drawn in
802 the way presented in Section 2. The sustainability findings rather strongly depend on the
803 individual case. Different criteria have been used in all studies (regarding the functional unit,
804 database, assessment methodology, etc. This diversification of software and database supplies,
805 together with using different methodologies (CML, ReCiPe, etc.), limits such comparison.
806 Therefore, benchmarking LCAs inevitably has to face limitations regarding differences in
807 modelling hypotheses as noted in Table 1. A future standardisation would add much to
808 comparability as recently suggested by Müller et al. [163]

809 Yet, a technology selection requires nonetheless solid benchmarking criteria beyond all the
810 understandable complexity. Another way of data representation and discussion may help here.

811 In this sense, Table 4 has assembled the data for process studies compactly discussed in Section
 812 2 to facilitate comparison. This compilation may provide order-of-magnitude statements for
 813 technology-class-related sustainability impact to decipher some principal ecological advantages
 814 and disadvantages of each technology.

815 Table 4 shows that the ion exchange-based technology comprises the highest global warming
 816 potential, followed by adsorption and photochemical reduction while reverse osmosis has the
 817 lowest impact. Regarding freshwater eutrophication, the most advantageous technology is the
 818 electrochemical reduction in this study, whereas both reverse osmosis and ion exchange show
 819 the highest negative impacts. Flotation/coagulation/flocculation technologies are associated
 820 with the highest human toxicity, but in that aspect, photocatalytic reduction can be named a
 821 healthy technology. Finally, terrestrial acidification is significantly high when using the ion
 822 exchange process, while reverse osmosis tends to have a lower environmental impact
 823 concerning this indicator

824 More compactly and effectively than Table 4, Figure 9 shows comparative plots of the different
 825 metrics obtained for different technologies applied to wastewater purification. Accordingly, the
 826 technology which delivers the highest global warming potential impact is ion exchange (IX),
 827 followed by adsorption (Ad) and photochemical reduction (PhR), and the lowest impact is given
 828 for reverse osmosis (RO). Regarding freshwater eutrophication (FEP), the most advantageous
 829 technology is electrochemical reduction, whereas the highest impact is found for reverse
 830 osmosis and ion exchange. **The highest human toxicity potential (HTPnc) is given for**
 831 **coagulation/flocculation technology, while photocatalytic reduction scoring lowest in this**
 832 **category.** Finally, terrestrial acidification (TAP) is especially positive when using ion exchange,
 833 and even lower when using reverse osmosis.



834

835 **Figure 9** Comparative wastewater treatment environmental impacts from different LCA
836 sources

837 **Table 4** Comparative environmental impacts of wastewater treatment using different technologies obtained from different LCA sources

Indicator	Unit	Membrane	Coagulation	Ion	Reverse	Reverse	Photocatalytic	Electrochemical	Adsorption
		Filtration (MF) <small>Error! Bookmark not defined.</small>	Fe & Al salts (CO/FLO) <small>Error! Bookmark not defined.</small>	Exchange (IX) <small>Error! Bookmark not defined.</small>	Osmosis (RO) <small>Error! Bookmark not defined.</small>	Osmosis (RO) <small>Error! Bookmark not defined.</small>	Reduction (PhR) <small>Error! Bookmark not defined.</small>	Reduction (ER) <small>Error! Bookmark not defined.</small>	(Ad) <small>Error! Bookmark not defined.</small>
TWW technology →	Functional unit →	1 m ³ WW	1 m ³ WW	10 ³ m ³ WW	10 ³ m ³ WW	1 m ³ WW	1 m ³ WW	1 kg of HCOOH	1 m ³ WW
Fine particulate matter formation (PMPF)	kg PM2.5-eq	6.77E-04						1.40E-04	
Fossil resource scarcity (FFP)	kg oil-eq	1.20E-01							
Freshwater ecotoxicity (FETP)	kg 1,4-DCB-eq	7.58E-03					1.25E+00	6.78E+00	5.00E-01
Freshwater eutrophication (FEP)	kg PO ₄ -eq	1.14E-03	8.59E-03	3.00E-01	3.30E-01		1.90E-02	2.50E-05	2.25E-02
Global warming (GWP)	kg CO ₂ -eq	4.10E-01	3.00E-01	5.34E+02	6.81E+02	7.45E-02	5.90E+00	1.70E-01	1.10E+01
Human carcinogenic toxicity (HTPc)	kg 1,4-DCB-eq	8.71E-03						7.70E-04	
Human non-carcinogenic toxicity (HTPnc)	kg 1,4-DCB-eq	1.60E-01		5.10E+01	2.30E+01	8.00E-03	1.25E+00	7.70E-04	5.00E-01
Inonising irradiation (IRP)	kBq Co-60-eq	6.68E-02						6.32E+00	
Land use (LU)	m ² a crop-eq	7.00E-03						2.30E-03	
Marine ecotoxicity (METP)	kg 1,4-DCB-eq	1.03E-02		3.70E+01	1.70E+00	1.40E+00			
Marine eutrophication (MEP)	kg N-eq	3.18E-03			0	1			
Mineral resource scarcity (SOP)	kg Cu-eq	1.55E-03							

Human health ozone formation (HOFP)	kg NOx -eq	8.60E-04							
Ecosystem ozone formation (EOFP)	kg NOx -eq	1.99E-03							
Stratospheric ozone depletion (ODP)	kg CFC11- eq	2.04E-07					3.20E-07	1.20E-08	7.50E-07
Terrestrial acidification (TAP)	kg SO ₂ -eq	2.04E-03	4.41E-04	8.20E+00	7.80E+0 0	2.00E-04	1.90E-02	2.30E-03	1.20E-01
Terrestrial ecotoxicity (TETP)	kg 1,4- DCB-eq m ³	6.30E-01				7.30E-03		1.76E+00	
Water consumption (WCP)	consumed	-1.80E-01						2.40E-03	
Fossil abiotic depletion (ADP fossil)	MJ		1.78E+00			2.59E+0 0	6.40E+01	2.79E+00	1.55E+02
Elements abiotic depletion (ADP elements)	kg Sb-eq		6.71E-08	3.90E+00	5.80E+0 0				
Photochemical Oxid. Formation Potential	kg eq C ₂ H ₄						9.00E-04		2.20E-03

839 4. Conclusions

840 Wastewater treatment is run has been used with professional process technology for many
841 decades. Nonetheless, the innovation cycle is high, both on the critical process and materials side.
842 There are numerous fundamental studies, new concepts, and even entire new technologies are
843 introduced. The most relevant innovations on the conceptual side are probably given by modern
844 photocatalysis and nanomaterials science.

845 “Clean Water and Sanitation” is among the 17 United Nations’ Sustainability Goals. Different
846 applications of wastewater treatment with different contaminants to be removed, both on the
847 organic and inorganic side, justify the plethora of technologies and concepts. The technology
848 needs to be conditioned to its purpose. Urban wastewater, for example, has different properties
849 and compositions than industrial wastewater (which may carry pharmaceuticals, resins, plastics,
850 textile, petrochemicals, etc). Adsorption technology, as a second example, has proven to be
851 effective in removing specific contaminants e.g. metals, together with photocatalytic reduction.

852 While some traditional process technologies such as flocculation/coagulation can take recourse
853 to well-established, traditional processing, others reached an industrial scale with ~~still~~ a strong
854 revolution such as adsorption. Still, others are at the most advanced edge, with large promises,
855 yet lacking somewhat of demonstrated scale-up such as photocatalysis when using advanced
856 catalysts. Process engineering literature is underdeveloped as compared to the numerous material
857 and conceptual studies. More process design studies are needed for wastewater technologies,
858 which are on the rising trajectory. This finally needs to move on towards scale-up, moving from
859 the laboratories to the industrial site.

860 As a consequence of the lack of process in the studies for some of the most promising concepts,
861 it is not simple to make a sustainability assessment. Many studies, if at all available, characterise
862 a laboratory approach, without proven application near a real case (rather being a simplified
863 model study). Their scope of validity could be increased if a more anticipatory (‘ex-ante’) life-
864 cycle assessment would be used, extending and broadening the actual results with hot-spot and
865 scenario analysis. That might give more generalised answers.

866 As of now, this review shows adverse results in literature in the sense that none of the wastewater
867 technologies is intrinsically good or bad in its environmental profile. It depends often largely on
868 the setting of one asset such as using clean energy (e.g. by sunlight) or having (catalyst,
869 adsorbent, flocculating, etc.) materials of a green ecological backpack. It predicts that the focused
870 research with one or two key priorities might give notably better sustainability outcomes. Yet,

871 also within the complexity of all the sustainability assessments, which are confusing, at first sight,
872 we could draw some relevant conclusions when staying with one class process technology.

873 Finally, in Section 3, we aimed at the most difficult exercise which is benchmarking the classes
874 of process technologies with each other; which demands a normalised and transparent procedure.
875 Besides all limitations discussed, we could derive some conclusions and recommendations
876 concerning the generic sustainability impact for each of the classes of process technologies
877 presented.

878 **Acknowledgments**

879 The authors acknowledge support from the ERC Synergy Grant Surface-CONfined fast
880 modulated Plasma for process and Energy intensification (SCOPE) from the European
881 Commission with Grant No. 810182.

882

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