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1 Comparative Life Cycle Assessment of two advanced treatment steps for wastewater micropollutants: how to
2 determine whole-system environmental benefits?

3 Eva Risch^{1,2}, Louis Jaumaux^{1,2}, Camille Maesele^{1,2}, Jean-Marc Choubert³

4 ¹ ITAP, Univ Montpellier, INRAE, Institut Agro, Montpellier, France

5 ² ELSA, Research group for environmental life cycle and sustainability assessment, Montpellier, France

6 ³INRAE REVERSAAL, 69625 Villeurbanne Cedex – France

7

8 E-mail contact: eva.risch@inrae.fr

9 **Abstract**

10 Advanced wastewater treatment (AWT) technologies are now considered to target urban micropollutants
11 (MPs) before discharge into receiving water bodies and to comply with specific criteria for reuse. Extra
12 energy and/or resources are necessary to achieve the elimination of MPs. Using the Life Cycle Assessment
13 framework, this study assesses net environmental efficiencies for two AWTs (i) ozone systems (air-fed and
14 pure oxygen-fed) and (ii) granular activated carbon filter. Sixty-five MPs with proven removal efficiency
15 values and toxicity and/or ecotoxicity potentials were included in this study building on results from recent
16 research. Consolidated Life Cycle Inventories with data quality and uncertainty characterization were
17 produced with an emphasis on operational inputs. Results show that the direct water quality benefits
18 obtained from AWT are outweighed by greater increases in indirect impacts from energy and resource
19 demands. Future research should include water quality aspects not currently captured in life cycle impact
20 assessment, such as endocrine disruption and whole-effluent toxicity, in order to assess the complete policy
21 implications of MP removal strategies.

22

23 **Keywords**

24 Life Cycle Analysis; micropollutants; advanced wastewater treatment; removal efficiencies;
25 environmental impacts; granular activated carbon; ozone treatment

26

27 **Abbreviations:** AWT, advanced wastewater treatment; LCA, life cycle assessment; LCI, life cycle inventory;

28 LCIA, life cycle impact assessment; MP, micropollutant; GAC, granular activated carbon

29 **1. Introduction**

30 Several organic and inorganic micropollutants (MPs) are insufficiently eliminated in conventional
31 wastewater treatment plants (WWTPs) as they are designed to treat biodegradable carbon, nitrogen and
32 phosphorus, and they eliminate non-polar and large organic molecules (Zepon Tarpani and Azapagic, 2018)
33 or metals with affinity with organic matter (Choubert et al., 2011b). Therefore, discharges of MPs in aquatic
34 environments can lead to potential negative effects on living organisms. A large array of MPs found in very
35 low concentrations in treated wastewater make advanced wastewater treatment (AWT) processes technically
36 challenging. Intensive usage of treatment chemicals and energy in AWT processes have been associated
37 with increased life cycle toxicity and other environmental impacts (Dolar et al., 2012; Igos et al., 2012; Papa
38 et al., 2015, 2013; Pasqualino et al., 2011; Rahman et al., 2018). In order to find AWT technologies suited to
39 meeting targets on MPs, it is important to understand environmental benefits achieved by MP removal (e.g.,
40 reduced toxicity) as well as unintended environmental impacts due to additional chemical, energy, and
41 materials requirements. Life cycle assessment (LCA) has been used extensively to characterize and quantify
42 the net environmental impacts of wastewater treatment plants, and to compare treatment options (Byrne et
43 al., 2017; Corominas et al., 2020). Nevertheless, few papers consider MPs due to the lack of toxicity and/or
44 ecotoxicity potentials (Arzate et al., 2019; Igos et al., 2021, 2012; Rahman et al., 2018). However, the effects
45 of MPs in the environment are yet to be fully understood (Eggen et al., 2014) and therefore affected by high
46 uncertainty and low coverage of impact pathways. Furthermore, the extensive identification and
47 quantification of all the MPs in wastewater is a complicated task both technically and economically. For this
48 reason, often just a few target compounds are reported in monitoring studies and potential impacts are
49 calculated with these incomplete data (Aemig et al., 2021; Alfonsín et al., 2014; Rahman et al., 2018). For
50 example, transformation products which arise from oxidative treatments and are not included in the LCA may
51 increase ecotoxicity of the treated effluent (Vogna et al., 2004).

52 Because the scope of LCA includes both direct emissions from WWTPs as well as indirect emissions
53 from producing and transporting all chemicals, energy, and infrastructure required for treatment, LCA can be
54 applied to study both environmental benefits and costs associated with AWT to meet stringent regulations.

55 The aim of this work was to determine the net environmental efficiencies for AWTs like oxidation by
56 ozone produced with pure oxygen or air, or adsorption by activated carbon.

57 **2. Materials and methods**

58 The steps leading to the environmental assessment of two types of wastewater management systems
59 using the life cycle assessment (LCA) method are presented in this section. Thus, we will follow the
60 recommended scheme of a standardized LCA (ISO, 2006a, 2006b): (i) goal and scope definition (Section
61 2.1), (ii) inventory analysis and uncertainty characterization (Sections 2.2 and 2.3), (iii) impact assessment
62 (Section 2.4), (iv) interpretation of results and discussion of the most salient issues when considering MPs in
63 LCA (Section 3).

64 **2.1. Goal and scope definition**

65 The objectives of this study were to assess and compare life cycle environmental impacts of two
66 promising AWT processes targeting wastewater MPs (e.g. ozonation and adsorption on activated carbon),
67 identifying their main environmental hotspots. Additionally, the ecotoxicity and toxicity impacts of MPs in the
68 AWT effluent were estimated and compared to the equivalent impact of the effluent from conventional
69 WWTPs without AWT. The scope of the study covers AWT effluent discharges and by-products generated
70 during the AWT as well as the infrastructures of treatment units, and operational inputs (e.g. required
71 electricity, chemicals or adsorption media).

72 **2.1.1. Functional unit**

73 Generally, LCA studies on wastewater treatment systems used a functional unit on assumed per capita
74 loadings such as the population-equivalent (PE) defined in the European directive 91/271 (Corominas et al.,
75 2020). Hence, the functional unit selected for this LCA study was “the AWT of an urban (biologically pre-
76 treated) wastewater effluent generated from 50 000 PE during one year”. Such a treatment capacity can be
77 estimated at $2,74 \cdot 10^6 \text{ m}^3 \cdot \text{year}^{-1}$ based on the assumption that an average French PE discharges $0,15 \text{ m}^3$ of
78 wastewater daily (Mercoiret, 2010; Risch et al., 2011).

79 The rationale for this choice of functional unit was to ensure good representativeness with a LCA based
80 on an existing full-scale French WWTP using biofiltration (secondary treatment) followed by ozonation and
81 extensively monitored (Choubert et al., 2017b). In the following, different AWT processes were compared
82 using this common functional unit of comparison since these systems were all designed for wastewater
83 treatment.

84 **2.1.2. Scenarios**

85 This study considered two technologically proven AWT processes for the removal of organic MPs with
86 both proven removal efficiencies for use on urban wastewater. First, ozone treatment is currently the most
87 used oxidation process for organic MPs removal in drinking water production and is now recently considered
88 for the advanced treatment of wastewater effluents (Bertanza et al., 2011; Bourgin et al., 2018; Guillossou et
89 al., 2020; Margot et al., 2013; Schindler Wildhaber et al., 2015). Second, adsorption onto granular activated
90 carbon (GAC) has been proven to be a viable process to remove organic MPs (Benstoem et al., 2017;
91 Boehler et al., 2012; Mailler et al., 2016; Sbardella et al., 2018). Four scenarios were built to meet the
92 objectives of this LCA study on AWTs which included a reference scenario “baseline” without any AWT, two
93 ozone systems with different feed gas (a. with pure oxygen, b. with air) and adsorption on granular activated
94 carbon (Table 1). Previous steps of treatment consist of a primary settling and different stages of secondary
95 treatment by submerged biofilters operated to remove suspended solids, biodegradable carbon and
96 ammonium.

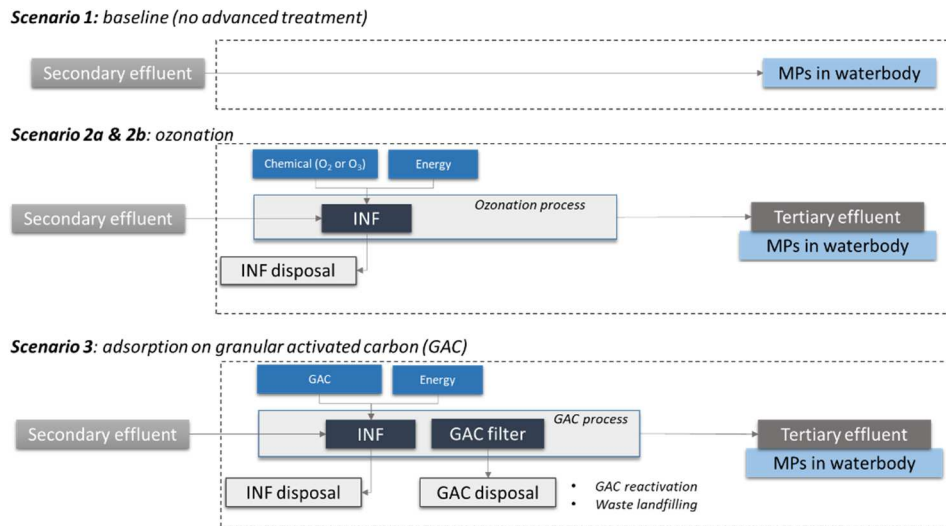
97 *Table 1. AWT scenarios modelled to meet the objectives of this LCA*

98 *Insert Table 1 here.*

99

100 **2.1.3. System boundaries**

101 The system boundaries included the materials for the construction of AWT infrastructures, as well as their
102 final disposal, and the resources required for their operation. Outlet pipes for discharge of treated water from
103 the WWTP to the receiving environment were assumed invariant in all scenarios therefore not modelled in
104 the scenario comparisons (by the ceteris paribus condition). The schematic diagrams of four wastewater
105 treatment scenarios with different AWT processes are shown in Figure 1.



106

107 **Figure 1.** Flow diagram and system boundaries for the AWT scenarios modelled in this study. System
 108 boundaries include the discharge of secondary effluent in a baseline scenario with no advanced treatment
 109 (1), discharge of AWT effluent after ozonation treatment with different feed gas (2a, with pure oxygen and
 110 2b, with air); and discharge of AWT effluent after adsorption on GAC (3) as well as the end-of life of the GAC
 111 filter, material demands for the infrastructure construction and end-of-life.

112 **2.2. Inventory analysis**

113 **2.2.1. Infrastructure and operational inputs**

114 Infrastructure requirements for AWTs were modelled after a reference plant with a treatment capacity of
 115 100 000 PE (Larsen et al., 2010). Then infrastructure demands were scaled down to the 50 000 PE capacity
 116 in the chosen functional unit (Annex Table S1), using a power law function to account for the non-linear
 117 relationships between the equipment capacities and the respective material demands (Arzate et al., 2019;
 118 Gallego-Schmid and Tarpani, 2019).

119 A detailed source analysis was first performed on each data source for energy and chemical requirements
 120 since these parameters showed a great variability across literature. The results from the source analysis
 121 were used to filter the data to select representative values (Annex Table S2). Then life cycle inventories
 122 (LCIs) were built for each scenarios using a horizontal weighted-averaging approach (Henriksson et al.,
 123 2013) which accounted for inherent uncertainties and spread, thereby quantifying data uncertainty.
 124 Inventories of operational requirements are summarized in Table 2 with respect to infrastructure
 125 (construction and disposal), energy, chemicals and emissions. Background data was sourced from the
 126 Ecoinvent v3.6 database (Ecoinvent, 2019) using French (FR)-specific processes where possible and
 127 modelled in the SimaPro 8.4 LCA software (Pré Consultants, 2017).

128 The operational energy demand for AWT scenarios were in the same order of magnitude and they represent
129 about 15-20% of the overall energy demand in a WWTP (Arzate et al., 2019). For ozonation this energy
130 demand is strongly dependent on the specific ozone dose, which was set at 6.22 gO₃/m³ i.e. 0.78 gO₃/gDOC
131 with a dissolved organic carbon load of 8 gDOC/m³ in this study (Choubert et al., 2017b). Hence for
132 ozonation with air, this energy demand is estimated at 1,2.10⁻¹ kWh/m³ or 3,2.10⁵ kWh/year with a total
133 treated wastewater volume of 2,74.10⁶ m³/year based on the average French inhabitant wastewater
134 generation of 0,15 m³/(PE.d⁻¹). This energy demand is contingent to specific parameters such as nitrites and
135 DOC in the secondary effluent and the chosen value for the energy demand in the ozonation step should
136 represent the global consumption of electricity (including ozone destruction, air production, cooling etc.).
137 Replacing the feed-gas with pure oxygen reduces the energy demand of 25% (Choubert et al., 2017b) since
138 the energy demand for air production is then negligible. However, the average pure oxygen demand is
139 estimated at 8.8 gO₂/gO₃ (Annex Table S2) (Larsen et al., 2010; Muñoz et al., 2009; Remy, 2013). In all
140 scenarios the energy was modelled with the French electricity mix reported by ecoinvent v3.6.
141 Granular activated carbon (GAC) filter media requires fresh activated carbon and can be regenerated with
142 spent media. The amount of fresh GAC used in a filter bed during its 5 years of operational lifetime was
143 estimated with 20% of losses during a regeneration cycle. In this study, the GAC filter media underwent
144 regeneration every 6 months before final disposal after 5 years (Rahman et al., 2018).

145 *Table 2. Inventory data to treat secondary effluent from 50 000 PE during one year through the AWT processes under study*
146 *Insert Table 2 here.*

147 **2.2.2. MP concentrations and loads in secondary effluent, and removal efficiencies in AWT**

148 Among the broad range of MPs routinely found in wastewater (Pistocchi et al., 2019; Ternes et al., 2004;
149 Verlicchi et al., 2012), 65 MPs from different chemical groups are selected for the current study. This short-
150 list includes 30 pharmaceuticals, 9 pesticides, 9 inorganics, 8 various industrial substances, 5 hormones, 3
151 fragrances and 1 polycyclic aromatic hydrocarbon (Annex Table S3a). These short-listed MPs were chosen
152 considering (i) available monitoring data in secondary effluents and AWT effluents (i.e. with quantified
153 removal efficiencies) and, (ii) available toxicity and/or ecotoxicity potentials in life cycle impact assessment
154 (LCIA) methods.

155 Measured concentrations of MPs in secondary effluent may range widely from 1,7 ng.L⁻¹ and 29,7
156 µg.L⁻¹ (Bruchet et al., 2015; Choubert et al., 2011a, 2017b; Coquery et al., 2011; Li et al., 2019; Martin Ruel
157 et al., 2012; Miège et al., 2008). The arithmetic means of the reported concentrations are used in the present
158 study as influent MP loads feeding to the AWT removal processes (Annex Table S3b). Removal efficiencies

159 achieved for ozonation and GAC processes were quantified using recent research (Bourgin et al., 2018;
160 Choubert et al., 2014, 2017b; Guillosoou et al., 2019, 2020; Li et al., 2019; Mailler et al., 2015; Martin Ruel et
161 al., 2012; Rahman et al., 2018). A particular attention was given to the selection of reliable removal
162 efficiencies in AWT effluents given the low MP concentrations and sampling measurement uncertainties to
163 ensure unbiased MP analyses (Choubert et al., 2017a). MP loads in secondary effluent and average removal
164 efficiencies for the two AWT processes are listed in Table 3.

165 **Table 3. Selected MPs with average concentrations, estimated mass loadings in secondary treated effluents of WWTPs**
166 **entering AWT and associated removal efficiencies**
167

168 *Insert Table 3 here.*

169 In general both ozone and GAC demonstrate removal efficiencies greater than 75% on fragrances,
170 industrial substances and hormones. Ozone treatment targets better pharmaceuticals, while GAC shows
171 overall greater removal efficiencies on pesticides. As a rule, inorganics are not affected by ozone (Ruel et al.,
172 2011) while GAC may partially adsorb (e.g. copper and chromium) or release (e.g. arsenic and vanadium) in
173 treated effluent. Indeed, industrial-scale GAC filters use carbon extracted from coal which contains traces of
174 metallic elements potentially leached from the GAC filter during the AWT (Choubert et al., 2014).

175 **2.3. Uncertainty characterization**

176 In order to ensure transparency and credibility of the LCA results obtained in this study, it is critical
177 that the uncertainty sources are quantified and their effects on results are communicated (Gavankar et al.,
178 2015). Furthermore, the treatment of uncertainty will help interpret differences in the studied systems in the
179 comparative LCA (e.g. to determine whether the preference for one system can be questionable due to
180 result uncertainties) as shown in Igos et al. (2018). Uncertainty ranges for unit processes taken from the
181 Ecoinvent database (Ecoinvent, 2019; Wernet et al., 2016) were defined by the “data pedigree” algorithm
182 available in SimaPro (Pré Consultants, 2019) as proposed by Weidema and Wesnaes (1996). This algorithm
183 relates the datum uncertainty to its source characteristics – i.e. reliability of the source, representativeness of
184 the sample, currency of the period, geographical correlation, technological correlation and sample size. The
185 uncertainty ranges associated to infrastructure construction and AWT operation (e.g. amounts of building
186 materials, energy and ancillary chemicals used, MP discharges etc) are described in Table 4.

187 **Table 4. Data quality and uncertainty estimation for two life-cycle stages: construction and use (including operation,**
188 **maintenance and AWT effluent discharges)**
189

190 *Insert Table 4 here.*

191 Based on uncertainty data in the LCI, uncertainty propagation in the modelled systems was estimated
192 using a random sampling method i.e. the available Monte-Carlo routine in SimaPro with 1500 runs to yield
193 mean and standard deviation values estimated with 95% confidence intervals (Annex Table S7b). Finally, the
194 influences of uncertainty in two key parameters were studied using sensitivity analyses (See section 3.3).

195 **2.4. Life cycle impact assessment**

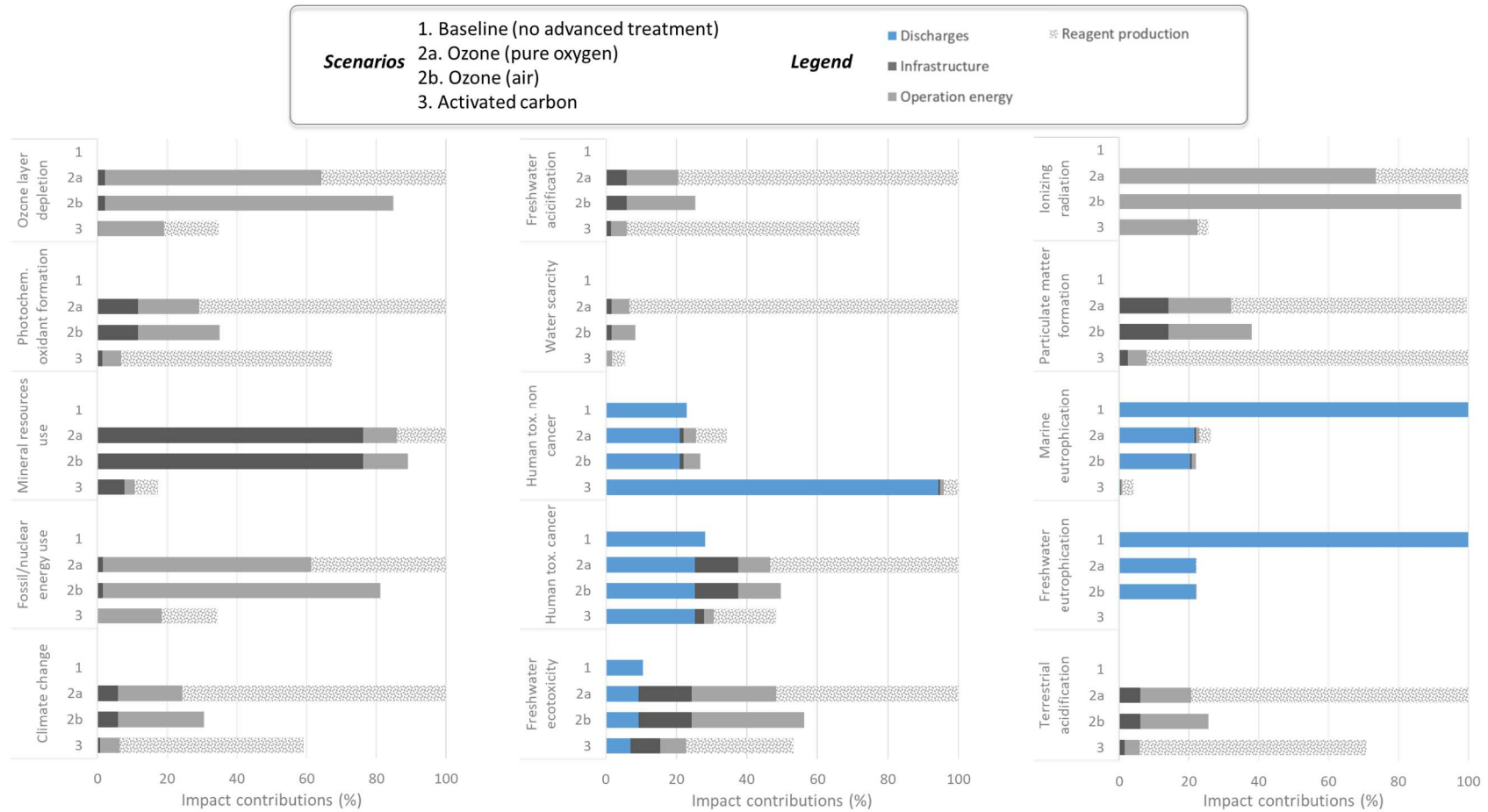
196 The Impact World+ method (Bulle et al., 2019) was selected for its robust assessment of human toxicity
197 and freshwater ecotoxicity potentials based on the consensual midpoint-level USEtox v2 model (Rosenbaum
198 et al., 2008). The library of the USEtox v2 model in SimaPro was complemented with 31 new
199 characterization factors (Alfonsín et al., 2014; Maillard et al., 2019) corresponding to the MP shortlist used in
200 this study (Annex Table S3a and Table S4). Detailed characterization factors are given after Maillard et al.
201 (2019) for 107 substances including organic MPs and pesticides in freshwater ecotoxicity (Table S8a and
202 S8b) and human toxicity (Table S9a and S9b). In Impact World+ at the damage level, some indicators
203 including long-term effects (climate change, marine acidification, toxicity cancer, toxicity non-cancer, and
204 freshwater, terrestrial, and marine ecotoxicity) are subdivided into separate mid-to-endpoint indicators for
205 short-term and long-term in order to differentiate impacts occurring in the short-term and long-term (after 100
206 years). Hence, added MPs were also characterized at the damage level (mid-to-endpoint) for the short-term
207 toxicity and ecotoxicity indicators (namely freshwater ecotoxicity and human toxicity cancer and non-cancer)
208 using conversion coefficients (Bulle et al., 2019; Huijbregts et al., 2005). However on long-term toxicity and
209 ecotoxicity indicators, characterization factors for organic MPs discharged in water were estimated to be nil
210 under the hypothese that organic MPs undergo complete degradation within 100 years after discharge.

211 **3. Results and discussion**

212 **3.1. Environmental hotspots within AWT systems (midpoint level)**

213 Contribution analyses for the baseline scenario and the three AWT scenarios are shown on 15
214 midpoint indicators (Figure 4). These results indicate that ozone with pure oxygen (scenario 2a) yields the
215 greatest impacts on 10/15 indicators due to feed-gas production and storage, with the exclusion of two
216 indicators with non-significant differences with another scenario (e.g. less than 10% difference). As
217 expected, reduction of MPs in the effluent (discharges, in blue) after AWT leads to reduced direct
218 environmental and health impacts. However, these reductions can hardly be seen as overall life cycle
219 impacts are dominated for most impact categories by indirect emissions from upstream production of energy,

220 chemicals, and (to a lesser extent) infrastructure materials. For example, AWTs reduce MPs concentration in
221 the effluent locally as shown on midpoint toxicity impacts (i.e. freshwater ecotoxicity and human toxicity from
222 the USEtox v2 model) yet their implementation lead to net increases in overall life cycle impacts.



223

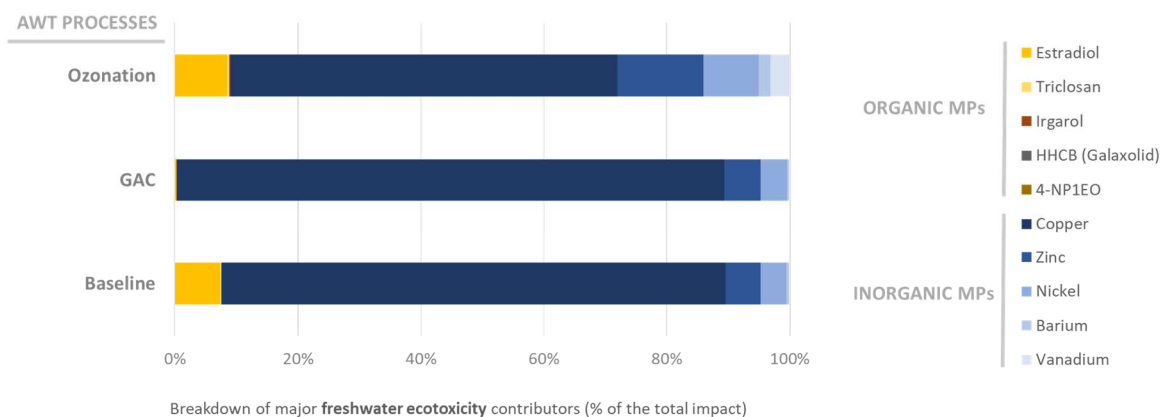
224 **Figure 2. Contribution analysis on the fifteen midpoint indicators of Impact World+ method (Bulle et al., 2019) for the four AWT scenarios which include baseline (1), ozone (2a and 2b) and**
 225 **activated carbon (3). All scenarios are compared using the same functional unit, being the advanced wastewater treatment from 50 000 PE during one year. Relative contributions (in %) are**
 226 **indicated with the scenario having the greatest impact at 100%.**

227

228 These findings are consistent with previous LCA studies (Arzate et al., 2019; Igos et al., 2012;
 229 Rahman et al., 2018) and this can be explained by low MP concentrations and low values of LCIA impact
 230 factors for discharged MPs. However, the effects of these substances in the environment are yet to be fully
 231 understood (Eggen et al., 2014) and therefore affected by high uncertainty and low coverage of impact
 232 pathways. Furthermore, the extensive identification and quantification of all the MPs in wastewater is a
 233 complicated task both technically and economically. For this reason, often just a few target compounds are
 234 reported in monitoring studies and potential impacts are calculated with these incomplete data (Aemig et al.,
 235 2021; Alfonsín et al., 2014; Rahman et al., 2018). For example, transformation products which arise from
 236 oxidative treatments and are not included in the LCA may increase ecotoxicity of the treated effluent (Vogna
 237 et al., 2004).

238 The breakdown of the freshwater ecotoxicity impact contributions demonstrate that copper and zinc
 239 are by far the greatest contributors (90%) in baseline and AWT effluents, with organic MPs accounting for
 240 less than 10% of the total impact (Annex Table 5a). These results on freshwater ecotoxicity impact also
 241 underline that five organic MPs ranked among the top ten contributors, estradiol, triclosan, cybutryne
 242 (irgarol), HHCB (Galaxolid), and 4-NP1EO which have significant ecotoxicity potentials (Figure 3).

243



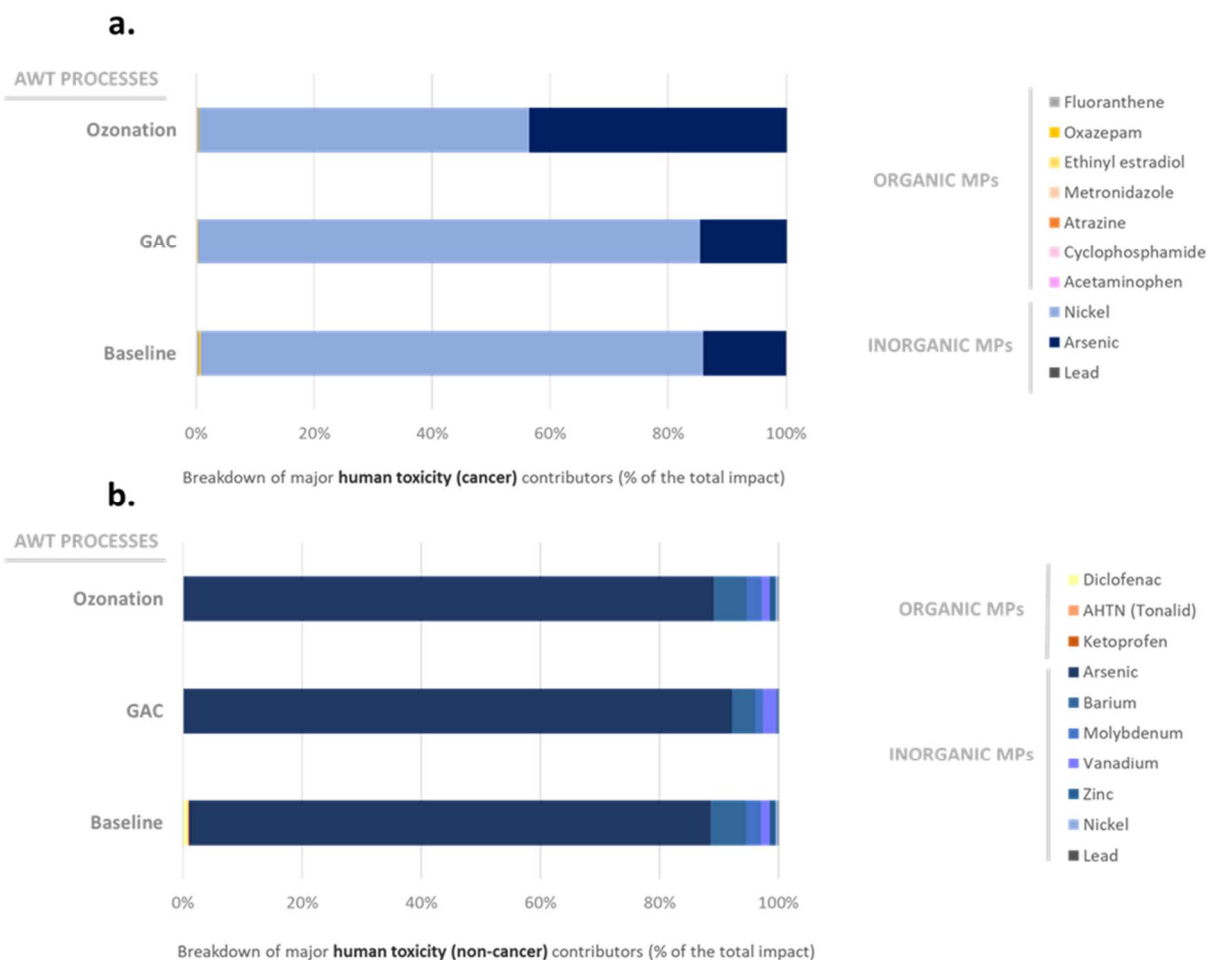
244

245 **Figure 3. Top ten contributors on freshwater ecotoxicity for AWT discharges, expressed in % of the total impact**

246 Similarly, the breakdown of human toxicity impacts (respectively, cancer and non-cancer) proves once
 247 again that nickel and arsenic contribute almost single-handedly to the total impacts. Seven organic MPs rank
 248 among the top ten contributors on the human toxicity (cancer) impact: fluoranthene, oxazepam, ethinyl
 249 estradiol, metronidazole, atrazine, cyclophosphamide and acetaminophen. While on the human toxicity (non-
 250 cancer), diclofenac, AHTN (tonalid) and ketoprofen are among the leading organic MPs (Annex Table S5b).
 251 Five novel organic MPs (HHCB, 4-NP1EO, AHTN and ketoprofen) that were added in the USEtox v2 model

252 appeared among the first ten contributors on these impacts, which highlights the importance of
 253 complementing existing (eco)toxicity models to refine further the environmental assessment of AWT systems
 254 (Figure 4).

255



256

257 **Figure 4. Top ten contributors on human toxicity (a. cancer and b. non-cancer) for AWT discharges, expressed in % of the total**
 258 **impact.**

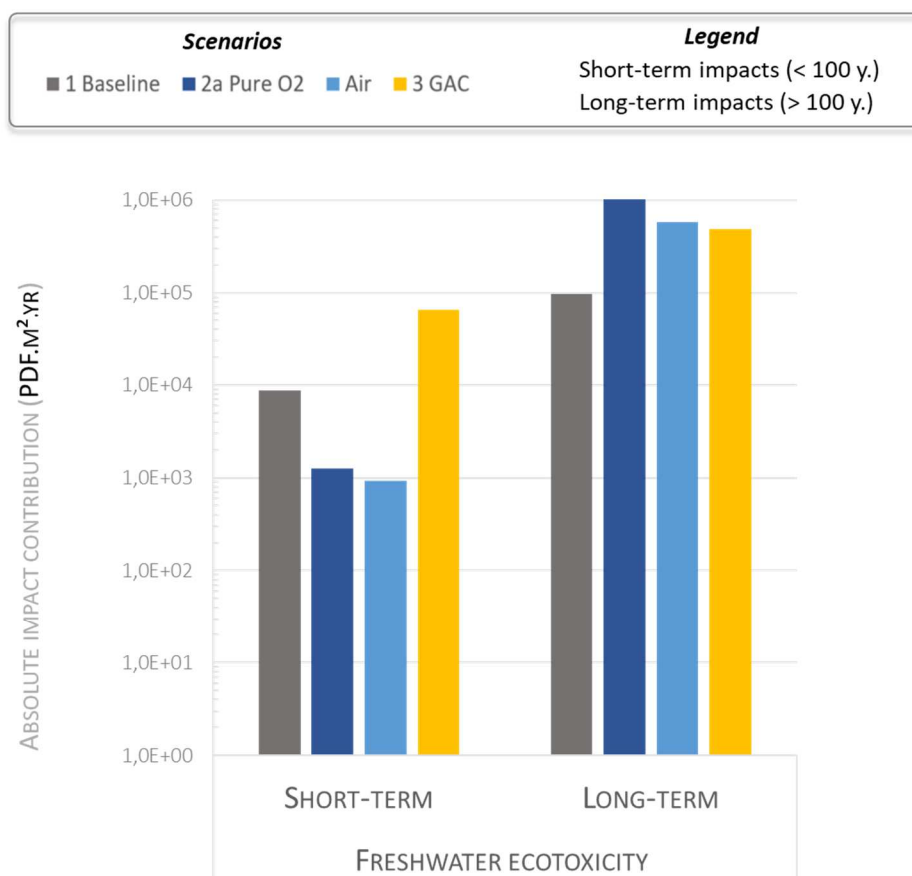
259 On the whole, impacts of effluent discharges are generally reduced after GAC treatment except on
 260 human toxicity non cancer, with arsenic release (Choubert et al., 2014) and on freshwater eutrophication due
 261 to phosphates based on a pilot-scale study (Mailler et al., 2015). However, these removal efficiencies should
 262 benefit from consolidated measurements to provide greater accuracy in scenario comparisons.

263 3.2. Long term vs short term impacts (mid-to-endpoint)

264 A distinction between long term and short term seems necessary to further analyse toxicity-related
 265 impacts with a strong dominance of inorganic MPs over organic MPs. Indeed, current impact assessment

266 methods are not adequately mechanistic or capable of combining the potential impacts of organic
267 compounds with those of metals (Pradinaud et al., 2019). Due to this limitation in LCIA methods, metals tend
268 to significantly impact on toxicity and ecotoxicity indicators compared to degradable organic MPs due to their
269 persistence in the environment in different dissolved or particulate forms (Aemig et al., 2021; Brudler et al.,
270 2019; Lorenzo-Toja et al., 2016; Risch et al., 2018). With the temporal horizon distinction at the mid-to-
271 endpoint level in the Impact World+ method (Bulle et al., 2019), the impacts of metals and persistent organic
272 pollutants can be differentiated into short-term (first 100 years) and longer-term impacts (beyond 100 years).

273 Results for the ozone scenarios effectively highlight significant organic MP removal and reduced
274 negative impacts of about an order of magnitude on the short-term horizon for aquatic ecosystems
275 (freshwater ecotoxicity short-term indicator) compared to baseline scenario as shown in logarithmic scale in
276 Figure 5. However, as shown in Figure 4 and more detailed in Annex Table S7, AWT effluent discharges
277 contributed modestly (respectively 10% and 7% for ozone scenarios and GAC) to the overall freshwater
278 ecotoxicity indicator, behind indirect resource demands (respectively 90% and 93%). Hence, the net
279 increases in freshwater ecotoxicity long-term indicators for AWT scenarios with metals emissions originate
280 mostly from background processes.



281

282 *Figure 5. Temporal horizon distinction on freshwater ecotoxicity indicator (mid-to-endpoint level, IW+ method). Short-term*
 283 *impacts occur after emission until 100 years, while long-term impacts occur beyond 100 years. Impact results are expressed*
 284 *on a logarithmic scale to inform on the order of magnitude differences between short and long-term impacts. Detailed*
 285 *contributions on the short-term freshwater ecotoxicity indicator are given in Supplementary information (Table S6).*

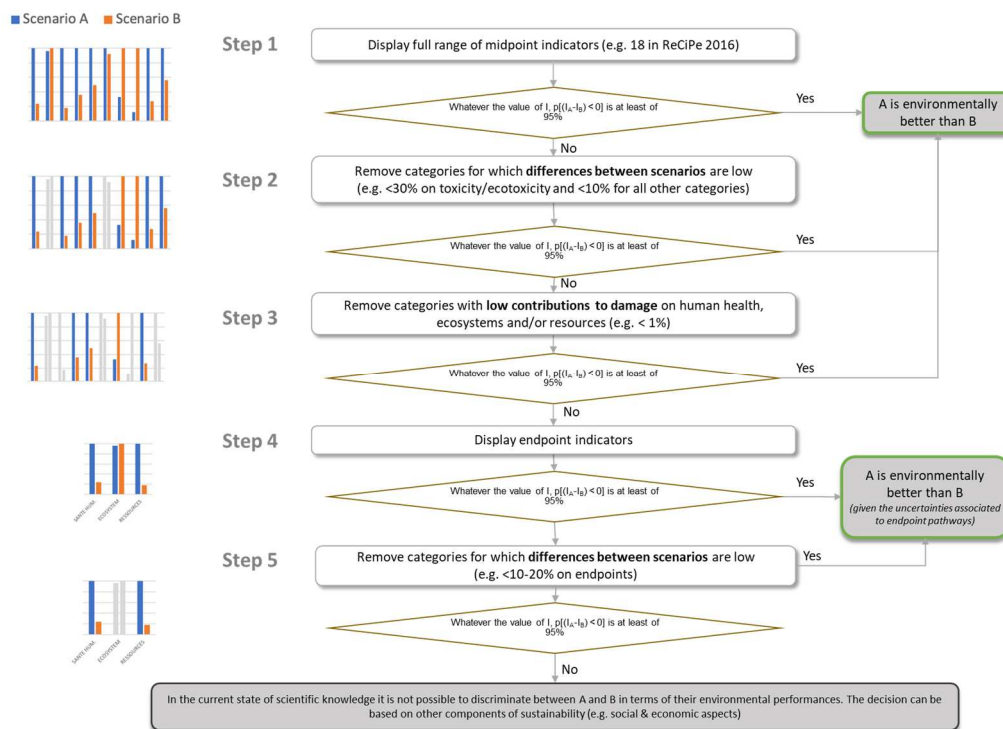
286 Conversely, benefits of organic MP removal are not evidenced on (human) health-related impacts
 287 (See Annex Figure S1). As demonstrated in Aemig et al. (2021), the total impact of organic MPs released in
 288 the aquatic environment through WWTP effluents on human health was low due to (i) no direct exposure
 289 routes for organic MPs (e.g. to capture potential endocrine disruption effects), (ii) the buffer role of the
 290 environment where the MP loads are diluted, and (iii) the low MP loads after AWT (Simazaki et al., 2015).
 291 Unsurprisingly, metals are leading the long-term freshwater ecotoxicity impacts, with copper, zinc and nickel.

292 Detailed impact contributions on short-term freshwater ecotoxicity (Annex Table S6) reveal the most
 293 potent ecotoxic MPs in baseline and AWT effluents which are hormones (estradiol), pesticides (triclosan),
 294 industrial substances (alkylphenols group) and a fragrance (HHCB).

295 3.3. Comparative analysis of AWTs

296 In this section a comparative LCA is performed between three AWT systems and the baseline
 297 scenario using the chosen functional unit. Given the four studied scenarios, in total six pair-wise

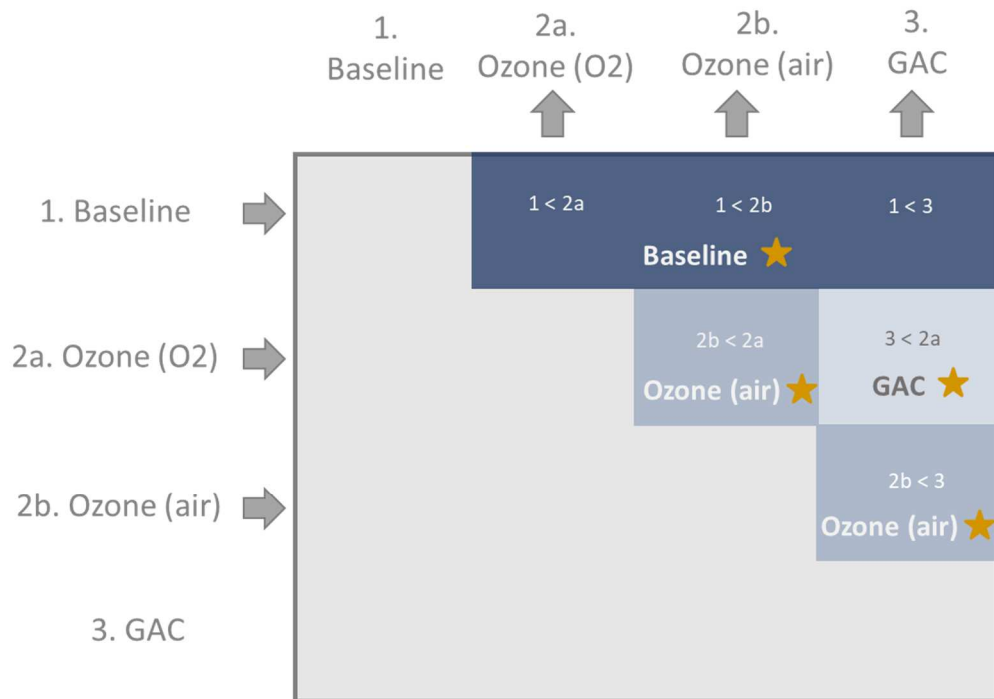
298 comparisons were performed to rank the compared systems (Igos et al., 2018). However, as noted by
 299 Guérin-Schneider et al. (2018) the interpretation of results for different scenarios on the full range of 18
 300 impact categories defined in Impact World+ is often complex due to cognitive obstacles. Hence, a decision
 301 tree (Guérin-Schneider et al., 2018) in Figure 6 helped structure the decision-making by proposing explicit
 302 simplification modalities to select relevant impact categories (Annex Table S11).
 303



304
 305 **Figure 6. Decision-support procedure for the analysis of LCIA results leading to a choice backed on biophysical criteria**
 306 **among several scenarios (adapted from Guérin-Schneider et al, 2018)**

307 Overall, the simplified interpretation of results leads to the following ranking of scenarios in order of
 308 increasing environmental impacts: 1. Baseline < 2b. Ozone (air) < 3. GAC < 2a. Ozone (O2).

Baseline ★ *Environmentally-superior scenario in the side-by-side comparison
Accounting for uncertainties - Confidence intervals*



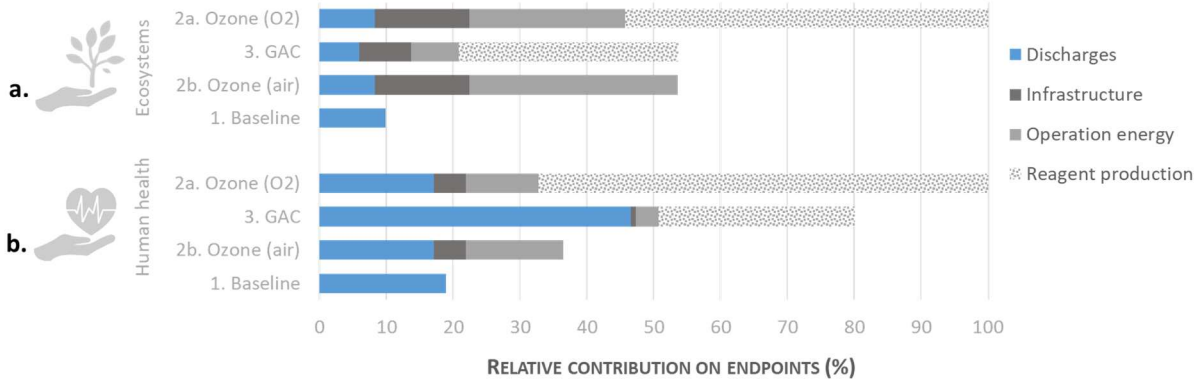
1. Baseline < 2b. Ozone (air) < 3. GAC < 2a. Ozone (O2)

309

310 **Figure 7. Pairwise comparisons of scenarios using the simplified decision-support procedure (Guérin-Schneider et al. 2018)**

311 Indeed results show that use of ozone produced with air and GAC are equivalent on ecosystems
 312 quality with less impacts than oxygen-fed ozone. Also, ozone produced with air appears as a better AWT
 313 choice compared with GAC or oxygen-fed ozone in terms of human health endpoints for the specific
 314 operating conditions in this study (Figure 8 and Figure 9) accounting for inventory data quality and spread
 315 (on the 65 MPs monitored in AWT discharges (See Annex Table S2). However, the baseline scenario (no
 316 AWT) demonstrate clear increases in net environmental impacts for all AWT scenarios at this endpoint level.

317



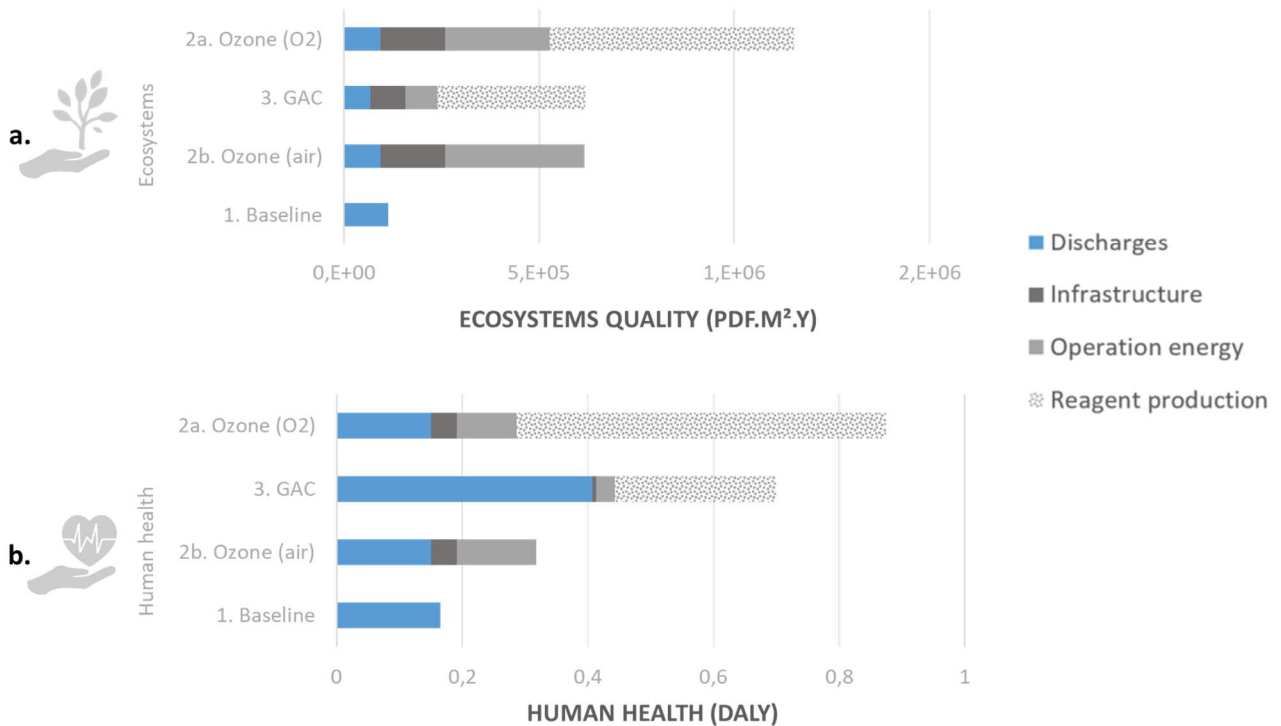
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Figure 8. Comparison of AWT scenarios at endpoint level (IW+ method) with detailed contribution breakdown in relative values (%) on the damage indicators: a. Ecosystems quality and b. Human health.

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Figure 9. Comparison of AWT scenarios at endpoint level (IW+ method) with detailed contribution breakdown in absolute values on the damage indicators: a. Ecosystems quality (PDF.m².yr) and b. Human health (DALY).

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Uncertainties around impact results were estimated (Annex Table S7) with mean values and standard deviations with 95% confidence intervals using the Monte Carlo randomization on process parameters. Sensitivity analyses on selected parameters such as energy source and inputs used in the process which showed significant impact contributions at the endpoint level are important for wastewater systems in general (Igos et al., 2021; Rebello et al., 2020).

331 Considering an European electricity mix (Wernet et al., 2016) instead of the French electricity mix with
332 an important nuclear energy share, for the ozone scenarios (2a and 2b) greatly alters the results of the
333 comparative LCA with now non-significant differences between air-fed ozone generators and oxygen-fed
334 ozone generators (Annex Table S10). This result underlines the importance of specifying the geographical
335 context of this study.

336 Under the modelling assumptions of this study oxygen-fed ozone did not show net environmental
337 benefits relative to air-fed ozone due to a significant increase in impacts from the extra energy demand,
338 based on a weighted mean value across reviewed studies around 19 kWh/kgO₃. However, using detailed
339 measurements on a full scale French WWTP with ozone treating 30 000 PE, the ozonation step (comprising
340 of air production, ozone generation, ozone destruction, ventilation and cooling) had a global energy demand
341 of 32 kWh/kgO₃ (See Annex Table S2). A sensitivity analysis on this energy demand for ozone scenarios
342 showed that the latter value yielded impact increases of about 13% and 18% on human health and
343 ecosystems endpoints (See Annex Figure S2), which underlines the significance of this choice of parameter
344 value in the LCA in agreement with Igos et al. (2021).

345 Regarding inputs such as reagent production, the GAC scenario could benefit from a further reduction in
346 impacts from activated carbon since its contributions to endpoints were significant despite the hypotheses on
347 GAC reactivation with 20% of GAC lost during regeneration as an alternative of using fresh activated carbon
348 after every breakthrough event (Igos et al., 2021; Rahman et al., 2018; Sbardella, 2019). A sensitivity
349 analysis on the GAC regeneration rates shows that using 10% regeneration losses (compared to 20%)
350 yielded moderate reductions in impacts at endpoint level of 5% on Human health and 8% on ecosystems
351 quality (See Annex Figure S3).

352 **4. Conclusions**

353 The objective of this study was to carry out an environmental impact assessment of AWTs (e.g. ozone
354 treatment and granular activated carbon adsorption) taking into account removal efficiencies on 65 MPs
355 among different chemical groups. Major results are highlighted in the following, as well as recommendations
356 and challenges on the way to conduct such a LCA:

- 357 • Efficiencies of AWT systems were determined in this LCA study with 31 new toxicity and ecotoxicity
358 potentials for MPs in the USEtox v2 model library. Metals (e.g. copper and zinc) contribute
359 significantly to ecotoxicity impacts.

- 360 • Five novel organic MPs (HHCB, 4-NP1EO, AHTN and ketoprofen) that were added in the USEtox v2
361 model library appeared among the first ten contributors on freshwater ecotoxicity and human toxicity
362 impacts.
- 363 • Comparative LCA results reveal multiple environmental trade-offs of AWT when considered on a life
364 cycle basis. The local water quality benefits achieved from the reduction of MPs in effluent are
365 largely outweighed by the indirect toxicity at a regional level resulting from high resource use (e.g.
366 electricity consumption and feed-gas).
- 367 • Hence it is important to develop detailed life cycle inventories and modelling assumptions concerning
368 background processes, with uncertainty quantification on future LCA studies for AWT systems
369 especially with comparative LCAs as some major parameters can alter the comparison outcomes;
- 370 • Results for the ozone scenarios effectively highlight significant organic MP removal and reduced
371 negative impacts of about an order of magnitude on the short-term horizon for aquatic ecosystems.
- 372 • Most potent ecotoxic MPs in short-term freshwater ecotoxicity in all discharges were hormones
373 (estradiol), pesticides (triclosan), industrial substances (alkylphenols group) and a fragrance
374 (HHCB).
- 375 • Air-fed ozone and GAC are better AWT choices compared with oxygen-fed ozone on the
376 ecosystems quality endpoint for the specific operating conditions in this study (using a French
377 electricity mix). On human health, air-fed ozone is by far the best AWT choice, followed by GAC and
378 finally pure oxygen-fed ozone. However these results must be interpreted carefully and provide an
379 understanding of regionally distributed life cycle impacts which have to be complemented with
380 knowledge of local water quality issues;
- 381 • More research on toxicity data and impact assessment models to improve organic/inorganic MP
382 distinction in existing endpoints and to account for additional MP-relevant endpoints (e.g., endocrine
383 disruption, synergistic effects of mixtures) and transformation products would improve the utility and
384 representativeness of the LCA results, while process improvements may increase removal rates
385 while lowering energy and chemical requirements. In light of the current results, however, life cycle
386 environmental impacts should be considered while adopting management and risk mitigation
387 strategies for MPs in combination with local risk assessment approaches.

388

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Table 1. AWT scenarios modelled to meet the objectives of this LCA

<i>Scenario</i>	<i>Advanced wastewater treatment (AWT) process</i>
1	Baseline (no advanced treatment)
2a	Ozone (pure oxygen)
2b	Ozone (air)
3	Granulated activated carbon (GAC)

Table 2. Inventory data to treat secondary effluent from 50 000 PE during one year through the AWT processes under study

Scenario	Ozone (air)		Ozone (pure oxygen)		GAC	
	<i>m</i>	<i>2SD</i>	<i>m</i>	<i>2SD</i>	<i>m</i>	<i>2SD</i>
Building materials, kg (1)						
Concrete	2.6E+04	2.6E+03	2.6E+04	2.6E+03	4.9E+01	4.9E+00
Plastics	1.7E+00	1.7E-01	1.7E+00	1.7E-01	2.1E+01	2.1E+00
Metals	1.8E+03	1.8E+02	1.8E+03	1.8E+02	1.8E+02	1.8E+01
Electricity consumption, kWh (2)	3.2E+05	6.4E+04	2.4E+05	4.8E+04	7.3E+04	1.5E+04
Ancillary chemicals consumption, kg (2)						
Oxygen (pure)			1.5E+05	3.0E+4		
Activated carbon (fresh)					1.1E+04	2.2E+03
Activated carbon (reactivated)					2.8E+04	5.6E+03

Details in Supporting information: (1) Table S1, (2) Table S2

Table 3. Selected MPs with measured concentrations, estimated mass loadings in secondary effluents from WWTPs entering advanced treatment processes and associated removal efficiencies

Group	Compounds	measured concentration ^a in secondary effluent (ng/L)	estimated mass loading in secondary effluent (g/L) with 50000 PE capacity and 150L/(PE.d)	Removal efficiencies ^b of AWT processes (%)		
				ozonation	activated carbon	
Fragrances	ADBI (Celestolid)	1.69E+03	1.27E+01	80.0	95.0	
	AHTN (Tonalid)	7.60E+02	5.70E+00	80.0	95.0	
	HHCB (Galaxolid)	3.06E+03	2.29E+01	80.0	87.0	
Hormones	Estradiol	2.23E+01	1.68E-01	97.5	76.0	
	Estrone	1.59E+01	1.20E-01	93.5	82.0	
	Ethinyl Estradiol	1.67E+00	1.25E-02	92.5	95.1	
	Progesterone	2.24E+01	1.68E-01	0	79.5	
	Testosterone	2.90E+01	2.18E-01	95.0	80.0	
	4-nonylphenol	4.79E+02	3.59E+00	22.4	75.0	
	4-tert-butylphenol	1.42E+02	1.07E+00	50.0	90.0	
	Bisphenol A	4.78E+02	3.58E+00	95.0	74.5	
Industrial substances (various)	Ethylparaben	1.40E+02	1.05E+00	99.0	71.7	
	Methylparaben	5.67E+02	4.25E+00	98.0	73.3	
	NP1EO	4.41E+02	3.31E+00	95.0	80.6	
	NP2EO	3.40E+02	2.55E+00	95.0	67.7	
	Propylparaben	4.38E+02	3.29E+00	80.0	85.0	
Polycyclic Aromatic Hydrocarbons	Fluoranthene	6.82E+01	5.12E-01			
				50.0	42.0	
	Acetaminophen	3.71E+01	2.78E-01	90.4	89.7	
	Alprazolam	3.48E+00	2.61E-02	79.0	0	
	Amitriptyline	2.91E+01	2.19E-01	48.0	46.0	
	Atenolol	1.27E+03	9.56E+00	93.4	95.9	
	Azithromycin	2.30E+02	1.73E+00	84.5	69.0	
	Carbamazepine	7.89E+02	5.92E+00	97.9	80.5	
	Ciprofloxacin	4.78E+02	3.59E+00	71.5	49.0	
	Citalopram	1.70E+02	1.28E+00	75.0	57.5	
	Clarithromycin	5.10E+02	3.82E+00	96.3	92.0	
	Cyclophosphamide	6.75E+00	5.06E-02	84.0	0	
	Diazepam	8.79E+01	6.59E-01	86.5	71.5	
	Diclofenac	8.57E+02	6.43E+00	97.4	78.3	
	Erythromycin	2.22E+02	1.66E+00	89.9	68.4	
	Iohexol	1.52E+04	1.14E+02	38.0	49.8	
	Pharmaceuticals	Ketoprofen	1.11E+03	8.35E+00	73.3	68.5
		Metoprolol	1.17E+02	8.80E-01	92.5	97.0
		Metronidazole	7.88E+02	5.91E+00	72.0	79.0
		Naproxen	8.58E+02	6.43E+00	94.0	49.7
Norfloxacin		3.34E+02	2.51E+00	83.8	87.0	
Ofloxacin		1.56E+02	1.17E+00	91.8	75.8	
Oxazepam		5.29E+02	3.97E+00	84.3	60.5	
Propranolol		1.92E+02	1.44E+00	97.5	95.8	
Roxithromycin		4.30E+01	3.22E-01	84.7	69.0	
Simvastatine		7.18E+02	5.39E+00	70.0	65.0	
Sotalol		1.84E+03	1.38E+01	98.0	90.0	
Sulfamethoxazole		3.86E+02	2.90E+00	96.1	64.9	
Sulfapyridin		4.92E+02	3.69E+00	91.0	82.2	
Tetracycline		2.50E+01	1.88E-01	77.5	54.9	
Trimethoprim		3.39E+02	2.54E+00	93.9	91.6	
Venlafaxine		2.56E+02	1.92E+00	85.5	64.0	
Inorganics		Arsenic	5.68E+02	4.26E+00	7.0	-107
		Barium	6.85E+03	5.13E+01	15.0	-28.0
		Chromium	1.44E+03	1.08E+01	-4.0	92.0
		Copper	5.72E+03	4.29E+01	4.0	84.0
	Lead	5.03E+02	3.77E+00	35.0	97.0	
	Molybdenum	4.20E+03	3.15E+01	8.5	-1.0	
	Nickel	9.83E+03	7.37E+01	10.5	56.0	
	Vanadium	1.34E+03	1.00E+01	15.0	-205.0	
	Zinc	2.97E+04	2.22E+02	9.0	49.0	
	Pesticides	AMPA	2.06E+03	1.55E+01	50.0	6.4
Atrazine		2.34E+01	1.76E-01	42.0	86.8	
Cybutryne (Irgarol)		3.00E+01	2.25E-01	33.3	50.0	
Diuron		2.07E+02	1.55E+00	88.2	93.1	
Imidaclopride ("Neonicotinoid")		6.94E+01	5.21E-01	67.0	89.0	
Isoproturon		4.34E+01	3.25E-01	68.0	87.0	
Simazine		2.18E+01	1.64E-01	50.0	91.0	
Terbutryn		3.80E+01	2.85E-01	85.0	80.0	
Triclosan		9.83E+02	7.37E+00	75.0	56.3	
Classical pollutants		Nitrite (NO2-)	1.35E+05	1.01E+03	78.0	94.2
	Phosphate (PO4-)	8.30E+05	6.23E+03	78.0	-26.3	

^a Sources of MP concentrations: AMPERES project (Martin Ruel et al., 2012; Bruchet et al., 2015), MICROPOLIS-PROCEDES project (Choubert et al., 2017b), Li et al. (2019)

^b Sources of MP removal efficiencies: Li et al., 2019; Guillosoy et al., 2019, 2020; Mailler et al., 2015; Rahman et al., 2018; Choubert et al., 2014, 2017b; Bourgin et al., 2018.

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Table 4. Data quality and uncertainty estimation for two life-cycle stages: construction and use (including operation, maintenance and AWT effluent discharges)

Life cycle stage	Inventory (LCI) component	Distribution type	Uncertainty characterisation (1)	Uncertainty source
Construction stage (infrastructure)	Materials manufacturing (kg)		Ecoinvent (2)	
	Amount of material used (kg)	Normal	CV=5%	see (3)
Use stage (Operation & maintenance, discharges)	Electricity production (kWh)		Ecoinvent (2)	
	Amount of electricity consumption (kWh)	Normal	CV=10%	see (3)
	Ancillary chemicals production (kg)		Ecoinvent (2)	
	Amount of ancillary chemicals used (kg)	Normal	CV=10%	see (3)
	Micropollutants load discharged in effluents (g)	Normal	CV=20%	(Versini et al., 2016)
<p>(1) CV= coefficient of variation. Uncertainty information is provided with mean average values and standard deviation.</p> <p>(2) Uncertainty from the Ecoinvent database when defined, assessment of data quality based on a pedigree matrix (Weidema and Wesnaes, 1996). For our system, up to 70% of the Ecoinvent processes have available uncertainty information.</p> <p>(3) Estimated uncertainty (SD) based on expert judgement</p>				