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Abstract

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

Keywords

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

Abbreviations: AWT, advanced wastewater treatment; LCA, life cycle assessment; LCI, life cycle inventory; LCIA, life cycle impact assessment; MP, micropollutant; GAC, granular activated carbon

1. Introduction

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

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

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

2. Materials and methods

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

2.1. Goal and scope definition

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

2.1.1. Functional unit

Generally, LCA studies on wastewater treatment systems used a functional unit on assumed per capita loadings such as the population-equivalent (PE) defined in the European directive 91/271 (Corominas et al., 2020). Hence, the functional unit selected for this LCA study was “the AWT of an urban (biologically pre-treated) wastewater effluent generated from 50 000 PE during one year”. Such a treatment capacity can be estimated at $2{,}74.10^6$ $m^3.year^{-1}$ based on the assumption that an average French PE discharges 0,15 m^3 of wastewater daily (Mercoiret, 2010; Risch et al., 2011).

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

2.1.2. Scenarios

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

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

Insert Table 1 here.

2.1.3. System boundaries

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

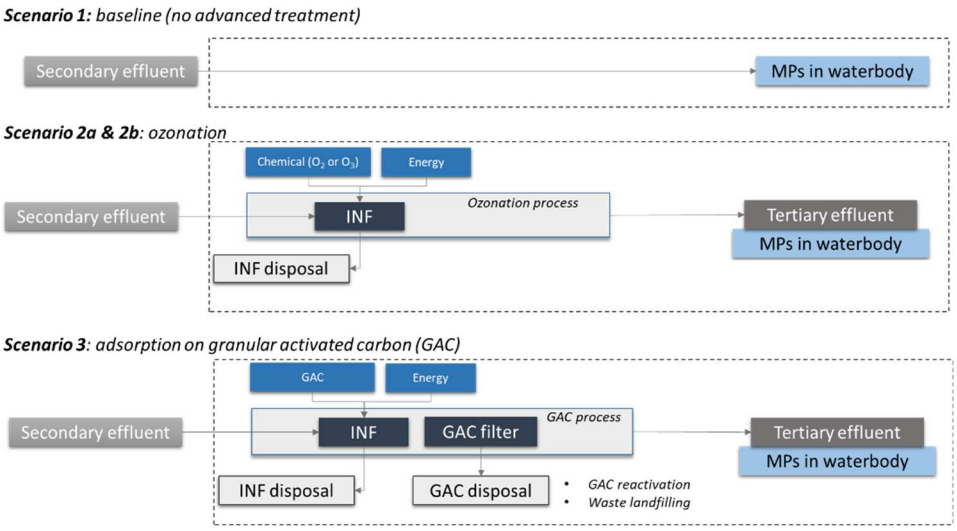


Figure 1. *Flow diagram and system boundaries for the AWT scenarios modelled in this study. System boundaries include the discharge of secondary effluent in a baseline scenario with no advanced treatment (1), discharge of AWT effluent after ozonation treatment with different feed gas (2a, with pure oxygen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

2.2. Inventory analysis

2.2.1. Infrastructure and operational inputs

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

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

The operational energy demand for AWT scenarios were in the same order of magnitude and they represent about 15-20% of the overall energy demand in a WWTP (Arzate et al., 2019). For ozonation this energy demand is strongly dependent on the specific ozone dose, which was set at 6.22 gO_3/m^3 i.e. 0.78 $gO_3/gDOC$ with a dissolved organic carbon load of 8 $gDOC/m^3$ in this study (Choubert et al., 2017b). Hence for ozonation with air, this energy demand is estimated at $1,2.10^{-1}$ kWh/m^3 or $3,2.10^5$ kWh/year with a total treated wastewater volume of $2,74.10^6$ m^3/year based on the average French inhabitant wastewater generation of 0,15 $m^3/(PE.d^{-1})$. This energy demand is contingent to specific parameters such as nitrites and DOC in the secondary effluent and the chosen value for the energy demand in the ozonation step should represent the global consumption of electricity (including ozone destruction, air production, cooling etc.). Replacing the feed-gas with pure oxygen reduces the energy demand of 25% (Choubert et al., 2017b) since the energy demand for air production is then negligible. However, the average pure oxygen demand is estimated at 8.8 gO_2/gO_3 (Annex Table S2) (Larsen et al., 2010; Muñoz et al., 2009; Remy, 2013). In all scenarios the energy was modelled with the French electricity mix reported by ecoinvent v3.6.

Granular activated carbon (GAC) filter media requires fresh activated carbon and can be regenerated with spent media. The amount of fresh GAC used in a filter bed during its 5 years of operational lifetime was estimated with 20% of losses during a regeneration cycle. In this study, the GAC filter media underwent regeneration every 6 months before final disposal after 5 years (Rahman et al., 2018).

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

Insert Table 2 here.

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

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

Measured concentrations of MPs in secondary effluent may range widely from 1,7 $ng.L^{-1}$ and 29,7 $\mu g.L^{-1}$ (Bruchet et al., 2015; Choubert et al., 2011a, 2017b; Coquery et al., 2011; Li et al., 2019; Martin Ruel et al., 2012; Miège et al., 2008). The arithmetic means of the reported concentrations are used in the present 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; Guillossou 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,

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

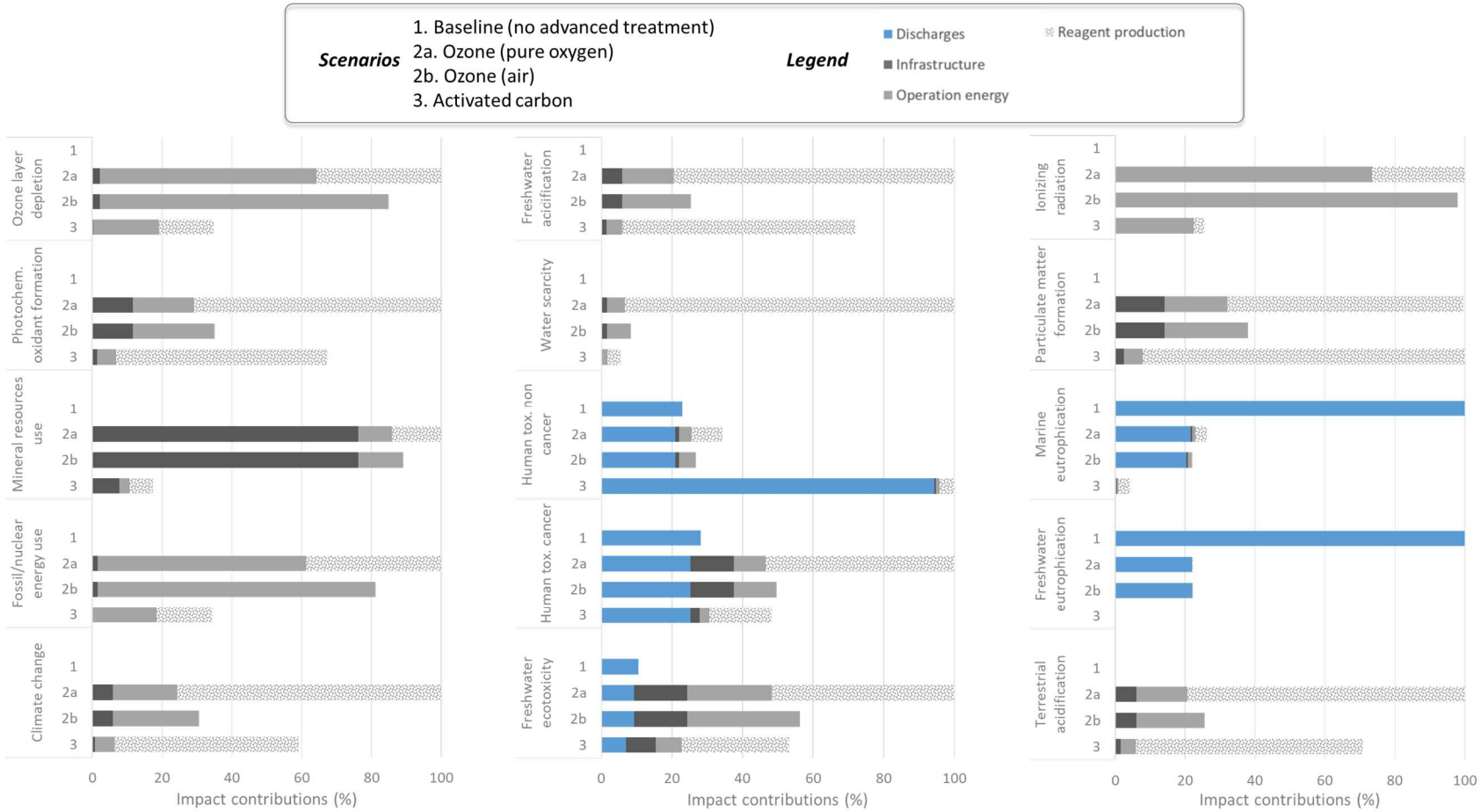


Figure 2. Contribution analysis on the fifteen midpoint indicators of Impact World+ method (Bulle et al., 2019) for the four AWT scenarios which include baseline (1), ozone (2a and 2b) and activated carbon (3). All scenarios are compared using the same functional unit, being the advanced wastewater treatment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

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

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

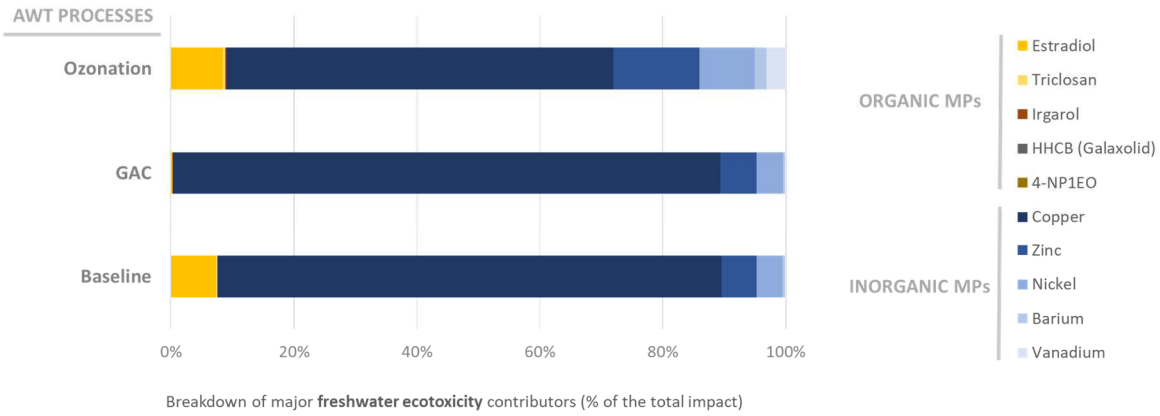


Figure 3. Top ten contributors on freshwater ecotoxicity for AWT discharges, expressed in % of the total impact

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

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

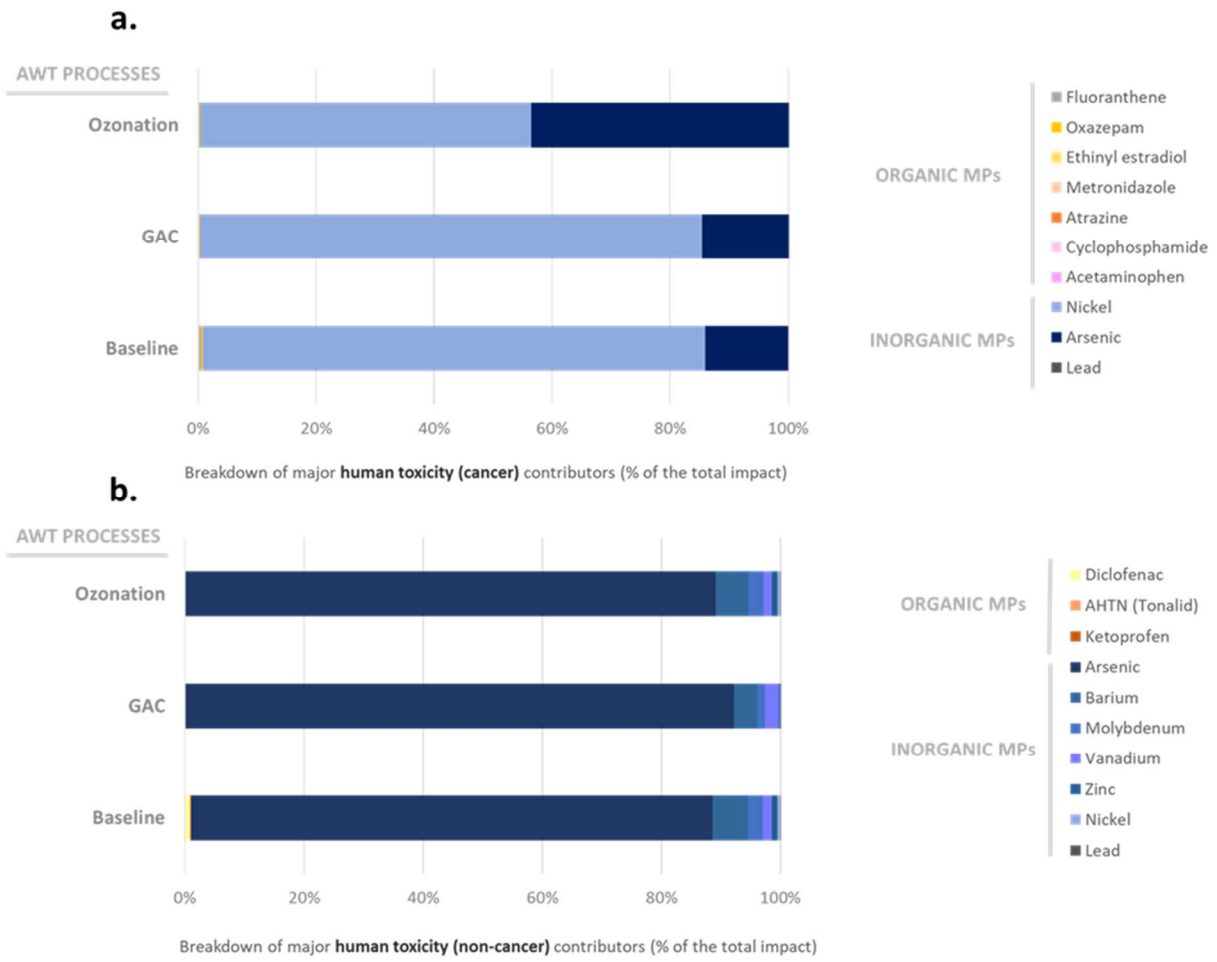


Figure 4. Top ten contributors on human toxicity (a. cancer and b. non-cancer) for AWT discharges, expressed in % of the total impact.

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

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

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

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

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

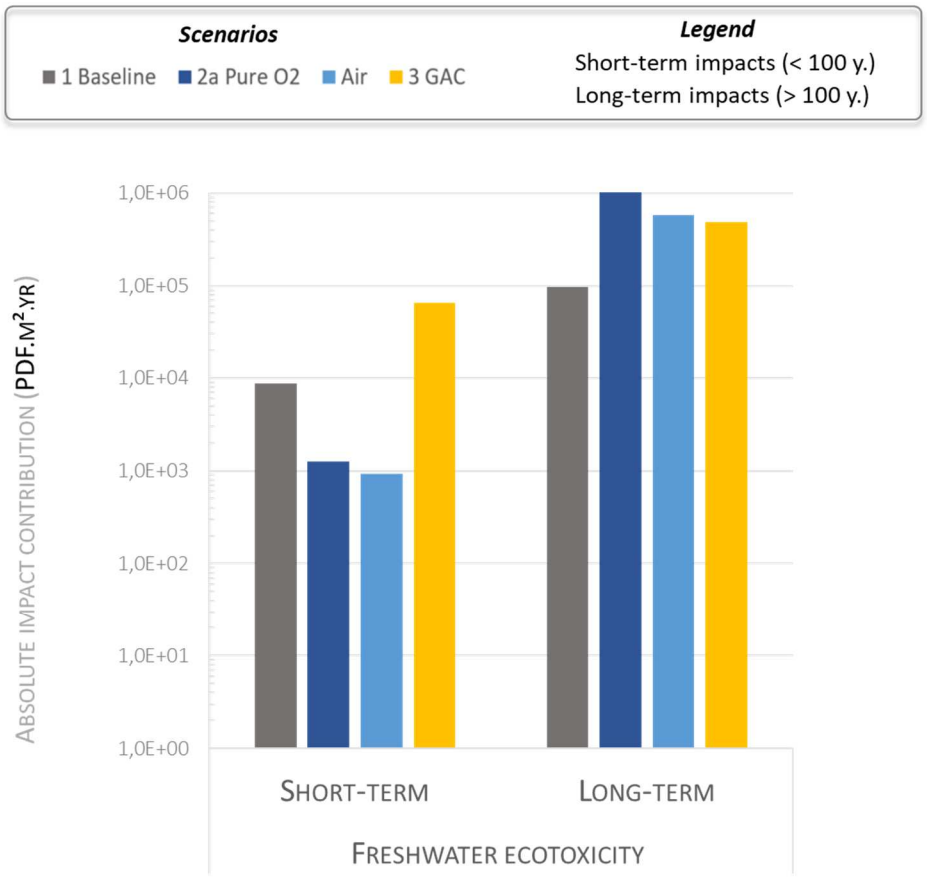


Figure 5. Temporal horizon distinction on freshwater ecotoxicity indicator (mid-to-endpoint level, IW+ method). Short-term impacts occur after emission until 100 years, while long-term impacts occur beyond 100 years. Impact results are expressed on a logarithmic scale to inform on the order of magnitude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

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

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

3.3. Comparative analysis of AWTs

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

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

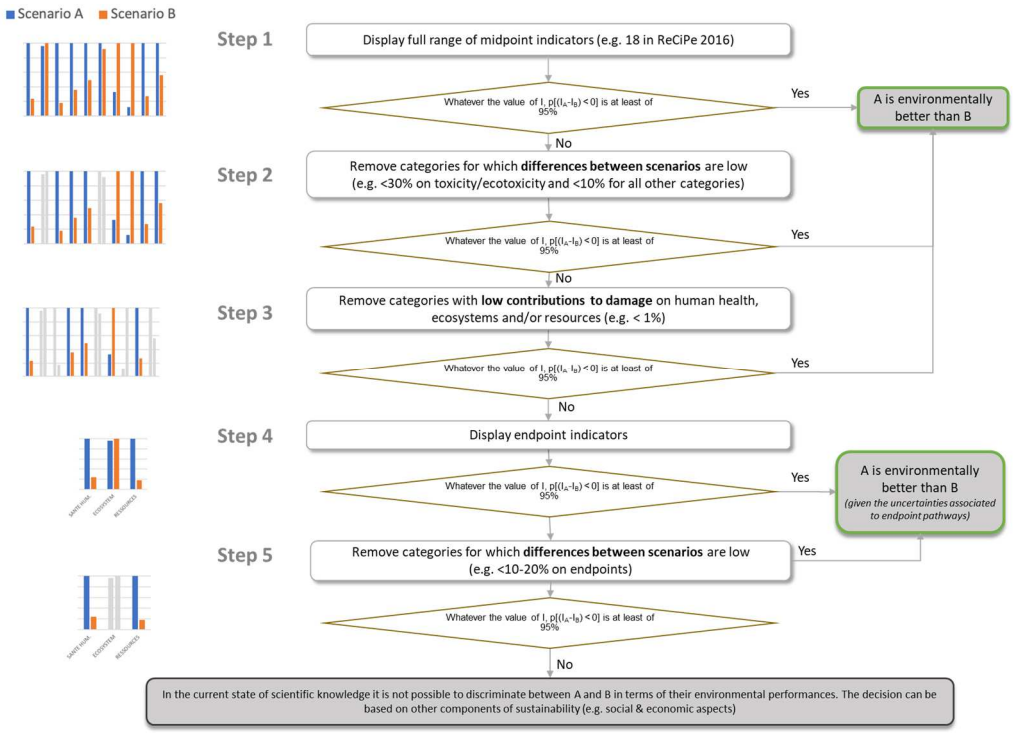


Figure 6. Decision-support procedure for the analysis of LCIA results leading to a choice backed on biophysical criteria among several scenarios (adapted from Guérin-Schneider et al, 2018)

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

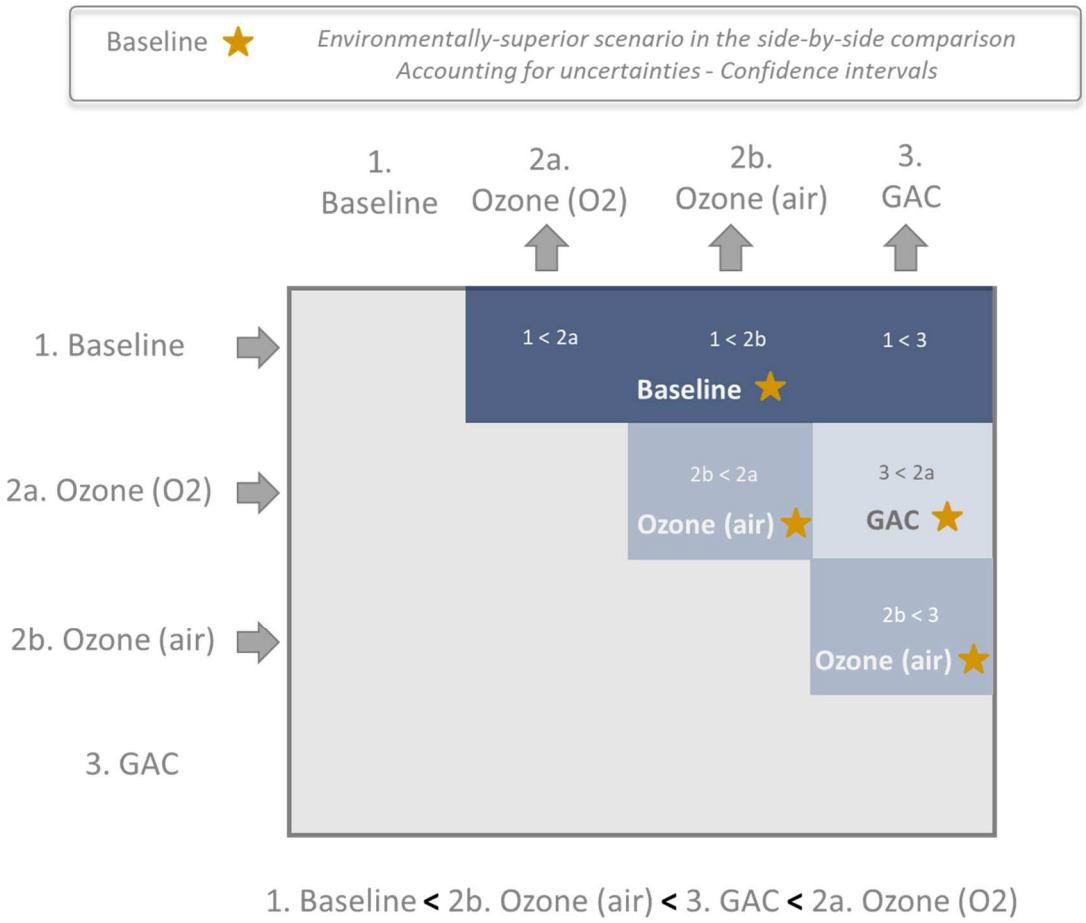
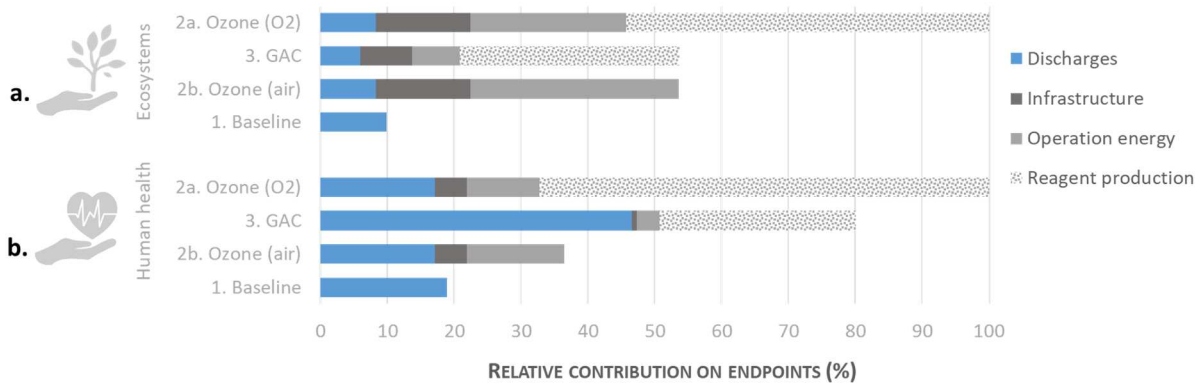


Figure 7. Pairwise comparisons of scenarios using the simplified decision-support procedure (Guérin-Schneider et al. 2018)

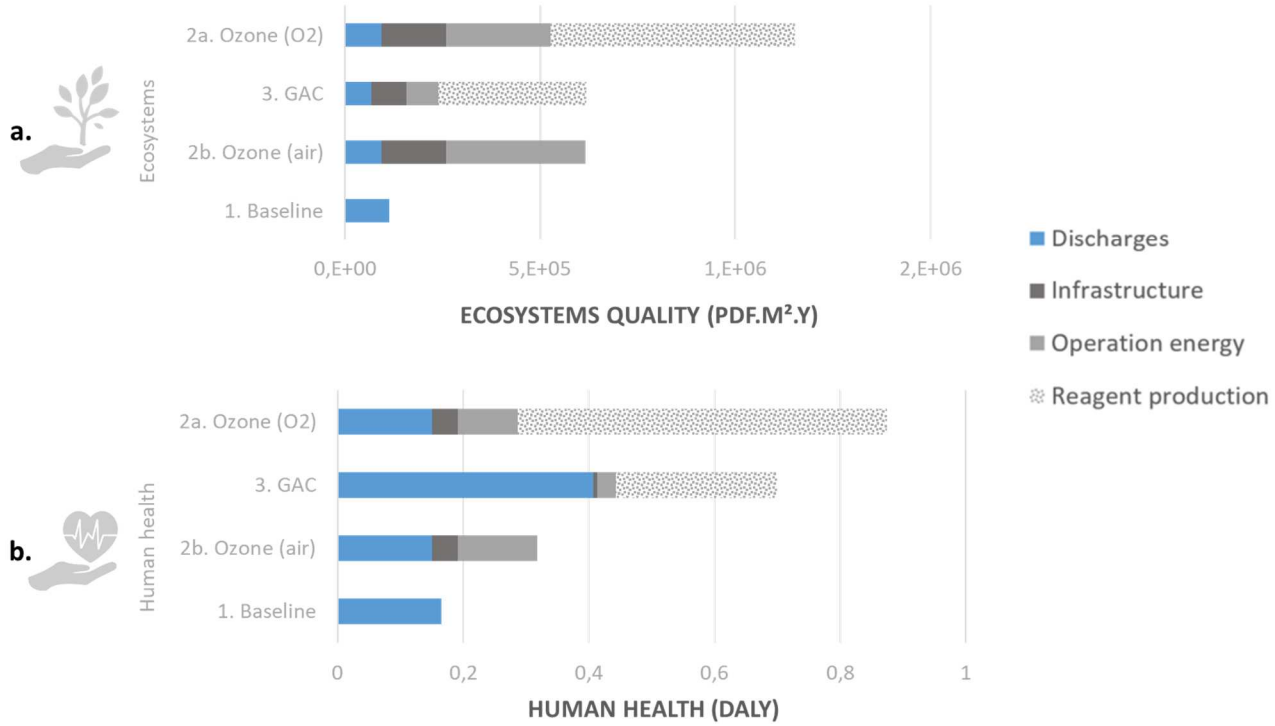
Indeed results show that use of ozone produced with air and GAC are equivalent on ecosystems quality with less impacts than oxygen-fed ozone. Also, ozone produced with air appears as a better AWT choice compared with GAC or oxygen-fed ozone in terms of human health endpoints for the specific operating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



318

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

321



322

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

325

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

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

Under the modelling assumptions of this study oxygen-fed ozone did not show net environmental benefits relative to air-fed ozone due to a significant increase in impacts from the extra energy demand, based on a weighted mean value across reviewed studies around 19 kWh/kgO_3. However, using detailed measurements on a full scale French WWTP with ozone treating 30 000 PE, the ozonation step (comprising of air production, ozone generation, ozone destruction, ventilation and cooling) had a global energy demand of 32 kWh/kgO3 (See Annex Table S2). A sensitivity analysis on this energy demand for ozone scenarios showed that the latter value yielded impact increases of about 13% and 18% on human health and ecosystems endpoints (See Annex Figure S2), which underlines the significance of this choice of parameter value in the LCA in agreement with Igos et al. (2021).

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

4. Conclusions

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

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

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

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598

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

Scenario	*Advanced wastewater treatment (AWT) process*
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	
Building materials, kg (1)	*m*	*2SD*	*m*	*2SD*	*m*	*2SD*
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 efficie

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
Industrial substances (various)	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
	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
Pharmaceuticals	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
	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; Guillossou 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)

(1) CV= coefficient of variation. Uncertainty information is provided with mean average values and standard deviation.

(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.

(3) Estimated uncertainty (SD) based on expert judgement