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Review

Tertiary Wastewater Treatment Technologies: A Review of Technical, Economic, and Life Cycle Aspects

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Abstract: The activated sludge process is the most widespread sewage treatment method. It typically consists of a pretreatment step, followed by a primary settling tank, an aerobic degradation process, and, finally, a secondary settling tank. The secondary effluent is then usually chlorinated and discharged to a water body. Tertiary treatment aims at improving the characteristics of the secondary effluent to facilitate its reuse. In this work, through a literature review of the most prominent tertiary treatment methods, a benchmarking of their technical efficiency, economic feasibility, and environmental impact was carried out. The photo-Fenton method proved to be the most technically efficient process, significantly reducing the microbial load and pharmaceutical content (by 4.9 log and 84%, respectively) of the secondary effluent. Chlorination and UV irradiation exhibited the lowest treatment costs (0.004 EUR/m^{-3}) and the lowest global warming potential (0.04 and 0.09 kg CO_2eq. m^{-3}, respectively). After all the data were aggregated, a decision-making tool was constructed in the form of a ternary diagram, which indicates the most appropriate tertiary treatment method according to the weight-per-process aspect (technical, economic, and environmental) selected by the user, with chlorination, UV irradiation, ozonation, microalgae cultivation, and constructed wetlands prevailing in the final results.

Keywords: tertiary wastewater treatment; chlorination; constructed wetlands; microalgae; ozonation; life cycle assessment; technoeconomic

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1. Introduction

Global population growth, as well as modern lifestyles, has resulted in an increase in the amount of produced sewage, thus making wastewater management urgent. According to United Nations estimations, the global population will reach ca. 11 billion by 2100, whereas other researchers estimate that the population will reach a lower peak earlier than 2100 [1]. In any case, human activity is expected to dramatically increase, followed by increased urban wastewater production.

Typically, wastewater can be classified as sewage when derived from domestic activities. Further categorization can divide sewage into blackwater, greywater, and yellow water, with the first corresponding to wastewater contaminated with human waste, the second to sewage derived from non-toilet units (e.g., showers) without being contaminated with human feces or urine, and the latter being polluted by urine collected from sewer channels [2–4]. Apart from domestic and industrial wastewater, hospitals, agricultural activity, and livestock farming can lead to aquatic environmental pollution, including contamination by emerging contaminants (ECs) (e.g., pharmaceuticals), posing a threat to both the environment and humans [5].

The most widespread sewage treatment method involves the activated sludge process. A typical wastewater treatment plant consists of a pretreatment step where grease and large solids are removed from the wastewater, followed by a primary settling tank. After most of the suspended solids are removed, the wastewater is led to the aerobic degradation

process that implements the activated sludge. At this stage, most of the organic content is converted into carbon dioxide and microbial biomass. The effluent of the aerobic process is treated with a secondary settling tank that removes and partially recirculates the produced biomass. These are the steps that are typically included in such a process, with some also implementing a nitrification/denitrification process in the activated sludge process. The effluent of the secondary settling tank (secondary effluent) is then typically chlorinated and discharged into a water body.

Despite the fact that the aforementioned steps are enough to remove most of the organic content from wastewater, residual pollutants such as inorganic nutrients (mostly nitrogen and phosphorus), increased microbial content, and ECs can have a significant impact on the final treated wastewater recipients. Moreover, strict quality criteria need to be met before the treated water can be reused instead of ending up in a water recipient [6]. In this scope, several secondary effluent treatment methods have been developed and implemented, typically called tertiary treatment processes. Among the tertiary treatment methods, chlorination, ultraviolet irradiation, membrane filtration, constructed wetlands, microalgae cultivation, ozonation, and photo-Fenton processes have attracted significant research interest due to their low operational costs, as well as their high efficiency. Some indicative works related to tertiary treatment techniques are demonstrated in the present paper.

An effort was made by the authors to present the most important technical efficacy parameters, namely the ability of each tertiary process to reduce pathogens and remove inorganic nutrients and EC. Furthermore, to put the technical efficacy results into a sustainability perspective, data regarding the treatment costs and global warming potential were also collected. Most of the published literature assesses the technical efficacy of these methods, with a much smaller number of publications examining their economic and life cycle aspects, a fact that unfortunately causes a disparity between the data available for each examined aspect.

2. Tertiary Wastewater Treatment Technologies

2.1. Chlorination

Chlorination is a widely used disinfection method applied in the last stages of sewage treatment. As a strong oxidizer, chlorine reacts with organic compounds, but can also lead to the formation of harmful chlorinated byproducts with negative effects on human health and the environment [7]. Several studies have reported the potency of chlorination in disinfection. In the work of Decol et al. [8], a 2.5-log reduction in *E. coli* was achieved, whereas Francy et al. [9] reported a 0.7- to 2.6-log reduction in a number of microbiological indicators such as *E. coli* (highest log reduction) and viruses. Chlorination does not affect the nutrient content of secondary effluent (in terms of N and P), making this method unsuitable when the treated wastewater is led to recipients prone to eutrophication. Pharmaceutical compounds, on the other hand, are prone to oxidation from chlorine, with Li and Zhang [10] testing the effects of the chlorination of compounds, i.e., sulfamethoxazole, ciprofloxacin, norfloxacin, tetracycline, trimethoprim, and erythromycin, reporting a reduction of 43 to 73%. Regarding the economic aspect of chlorination, it is considered one of the cheapest tertiary methods, with a treatment cost of 0.0003 to 0.006 EUR/m^3 [11–13]. Finally, due to its minimum energy needs, chlorination leads to very low greenhouse gas (GHG) emissions, with Walsh and Mellor [14] reporting GHG as low as 0.004 kg CO_2 eq. m^{-3}, whereas Pasqualino et al. [15] reported a slightly higher value of 0.07 kg CO_2 eq. m^{-3}.

2.2. Ultraviolet Irradiation

Ultraviolet technology, alone or combined with other degradation processes, has been extensively investigated as a tertiary treatment process. Zhang et al. examined the UV process coupled with H_2O_2 for secondary effluent treatment, achieving an ARG reduction equal to 3.48 log at pH 3.0 and 2.32 log at pH 7.0, whereas the cost was estimated at 0.296 USD/m^3 [16]. The coupling of UV with H_2O_2 technology was studied in the

removal of microcystin-RR by Qiao et al., with the reduction percentage of this pharmaceutical being 94.83% [17]. The combination of UV and the ozonation process by Chin and Bérubé in the treatment of surface water resulted in 50%, 80%, and 70% reductions in TOC, trihalomethane formation, and haloacetic acid formation, respectively [18].

A reduction of 100% for both diclofenac and bezafibrate was achieved when coupling UV irradiation with H_2O_2, as reported by De la Cruz et al., whereas the combination of three technologies UV/Fe^{2+}/H_2O_2 resulted in a 100% elimination of diclofenac [19]. Moreover, Guo et al. examined the UV disinfection process over ARB and ARGs present in municipal sewage [20]. The reduction in bacteria resistant to erythromycin and tetracycline was found to be equal to 1.4 and 1.1 log, respectively.

2.3. Membrane Filtration

Membrane filtration methods include a diverse group of processes, with the most common ones being pressure-driven membranes. During pressure-driven membrane filtration, a pressure difference is imposed on the two sides of a semi-permeable membrane, with the kinds of solutes permeating the membrane, further defining the membrane types. Membranes with a pore size on a scale of 1 μm (microfiltration, MF) typically reject suspended solids. Ultrafiltration (UF) has a smaller pore diameter and can reject larger dissolved molecules. Membranes with a pore size on a scale of 1 nm fall within the nanofiltration (NF) group and can reject smaller dissolved molecules (typically up to 200 Da) and divalent ions. Finally, reverse osmosis (RO) has no pores and separation occurs through the different diffusion rates of the solutes in the polymer of the membrane. RO membranes can even reject monovalent ions [21].

An integrated pilot unit combining UF, RO, and electrooxidation to manage municipal sewage was introduced by Urtiaga et al. [22]. All the target compounds, namely naproxen, ofloxacin, furosemide, bezafibrate, and fenofibric acid, were rejected with a percentage higher than 99%. Cheng et al. studied the efficiency of an anaerobic MF system in the removal of some antibiotic resistance bacteria (ARB) and their associated ARGs found in municipal sewage and the microbial load reduction was equal to 2–3 log units [23]. The elimination of ARB and ARGs present in secondary wastewater effluent by employing a TiO_2-modified polyvinylidene fluoride (PVDF) UF membrane was investigated by Ren et al. [24]. ARGs were removed at a rate of 98%.

Dolar et al. examined performance as it concerned the degradation of selected veterinary Phs present in pharmaceutical sewage using a laboratory and pilot scale RO/NF membrane treatment process [25]. The removal of TOC and COD was 70.8% and 35.4%, respectively, whereas the degradation percentage of the selected Phs ranged from 94% to 100% for the NF and RO membranes, respectively. Furthermore, Ho et al. examined palm oil mill effluent treatment by implementing graphene oxide (GO)/multi-walled carbon nanotube (MWCNTs) conductive membranes [26].

Treatment costs using membrane filtration can range from 0.4 to 1 EUR/m^3 [27] and the produced emissions can range from 0.2 to 2.3 kg CO_2 eq. m^{-3}, with the total emissions depending on the number of membrane steps and the required transmembrane pressure [28,29].

2.4. Constructed Wetlands

Constructed wetlands (CW) have been used since the 1950s as a waste management option [30]. The premise of this wastewater treatment method is based on the naturally occurring processes involving vegetation, soil, and microorganisms in a controlled environment [30].

Breitholtz et al. [31] achieved a BOD reduction of 40%, whereas reductions in 92 pharmaceuticals (Phs) ranged between 42 and 52%. The treatment of wastewater derived from residential areas was investigated over horizontal flow constructed wetlands (HFCW), vertical flow constructed wetlands (VFCW), and biofilters by Adrados et al. [32]. According to their study, the TN reduction ranged from 21 to 85%. Younger and Henderson [33] reported

41%, 59%, and 66% reductions in BOD, P-PO_4, and N-NH_4, respectively, employing an innovative full-scale mine water/sewage cotreatment CW for polluted mine waters. The yield of a vertical up-flow CW for swine wastewater was studied by Huang et al. [34] with remarkable results. Reductions in COD, TN, N-NH_4, and TP were 92.2%, 92.7%, 94.4%, and 97.8%, respectively, whereas the degradation of Phs ranged between 98.3% and 99.9%. The removal of various ECs (77.2%) and antibiotic resistance genes (ARGs) derived from landfill leachate was investigated by Yi et al. [35] by employing a full-scale horizontal subsurface flow CW. An integrated surface flow CW was employed over a 10-year period for the removal of ARGs found in domestic wastewater, as reported by Fang et al. [36]. The COD, BOD, TN, TP, and N-NH_4 degradations were 70.8%, 75.2%, 60.2%, 55.6%, and 61.3%, respectively. Chen et al. examined the ARG (85.8%) and antibiotic eliminations from wastewater derived from residential areas using mesocosm-scale horizontal subsurface flow CWs [37]. VFCWs were applied for the removal of ciprofloxacin HCl, oxytetracycline HCl, and sulfamethazine from swine sewage achieving elimination rates of 85%, 95%, and 73% for each of the aforementioned antibiotics, as found by Liu et al. [38]. The elimination of ARGs from municipal wastewater was studied by Nõlvak et al. [39] by employing horizontal subsurface flow CWs, achieving a 92% and 25% removal of BOD and TN, respectively. Chen et al. studied the effect on the degradation of antibiotics and ARGs of domestic sewage by employing six mesocosm-scale CWs [40]. The removal rates of COD, TOC, TN, and N-NH_3 were 80.2%, 80.3%, 54.7%, and 44%, respectively, whereas the total removal of all detected antibiotics was 98.6%. A 62% reduction in the rate of tet genes and a 90% average total removal rate of oxytetracycline and difloxacin antibiotics from swine sewage were achieved by applying a VFCW by Huang et al. [41].

Ledón et al. found a 90% reduction in the BOD rate from domestic wastewater by employing horizontal subsurface flow (HSSF) CW with the HSSF pretreatment cost being equal to 1903 USD/p.e, whereas the GHG emissions ranged between 3.8 and 4.7 kg $CO_{2\text{-eq}}$/kg for BOD_5 [42]. The coupling of microbial fuel cell (MFCs) technology with a conventional HSSF CW was investigated for municipal wastewater treatment by Corbella et al., achieving a remarkable 85% BOD reduction, where the cost of a conventional CW was estimated at 430 EUR/p.e [43]. Winery sewage treatment employing various scenarios of CW operations was examined by Flores et al., with the GHG emissions derived from the LCA being 1.3 kg $CO_{2\text{-eq}}/m^3$ [44]. Significant variations in costs were observed via life cycle costing (LCC) analysis of two different small-scale WWTPs coupled with CW technology for the treatment of wastewater produced by a student residential building and its coffee shop located in Brazil [45]. The cost of the scenario implementing a mobile CW (2.42×10^4 kg $CO_{2\text{-eq}}$) was found to be higher than that of the scenario using a decentralized VFCW (1.03×10^4 kg $CO_{2\text{-eq}}$) for wastewater treatment by Lakho et al. [46]. Pan et al. investigated the treatment of wastewater produced by the residential area of Changzhou in China using a vertical subsurface flow CW system, achieving 96% and 83% reductions in BOD and N-NH_4, respectively, whereas the total GHG emissions were estimated at 38.83 kg $CO_{2\text{-eq}}$/d [47]. A subsurface flow CW system was implemented for blackwater and greywater treatment coming from a rural area in Southern Brazil and showed an impressive COD, BOD, TKN, N—NH_3, and total P reduction, as described by Lutterbeck et al. [48]. The LCA revealed GHG emissions of 1.33×10^3 kg $CO_{2\ eq}$. Finally, Garfí et al. examined the performance of a combined VFCW and HFCW system for the treatment of sewage produced by small rural areas [49]. The BOD reduction was 89%, the capital cost was 210.36 EUR/p.e., the operational and maintenance costs were 0.4 EUR/m^3, and the GHG emissions were ca. 990 g $CO_2/m^3{}_{water}$.

2.5. Microalgae

WWTPs based on microalgae have gained significant attention since they combine environmentally friendly tertiary treatment technology with enhanced biomass production [50]. Two different microalgae-based WWTPs were compared considering their performance in agro-industrial sewage treatment, as reported by Magalhães and co-workers [50]. Both

proposed WWTPs were examined for wastewater treatment, as well as microalgae biomass production. The first system was a bubble column photobioreactor (PBR), whereas the second was a high-rate pond (HRP). The COD reduction of the former was 54.3%, whereas the reduction percentage of the latter was 47.7%. The removal rates of N-NH_4 and P were found to be complete in the case of the PBR and were 59.5% and 100%, respectively, in the case of the HPR, highlighting the superior performance of the PBR. Silambarasan et al. studied the performance of coupling microalgae (*Scenedesmus* sp. and *Chlorella* sp.) with lipid augmentation in order to remove nutrients from domestic sewage [51]. The reduction percentages of COD, TOC, TN, N-NH_4^+, N-NO_3^-, and PO_4^{3-} were estimated at 83%, 86%, 94%, 98%, 96%, and 95%, respectively.

Marangon et al. compared the environmental impact of two different scenarios for domestic sewage treatment [52]. According to the first scenario, a high-rate algal pond (HRAP) treatment system resulted in 0.1 kg CO_{2eq} GHG emissions, whereas following the second scenario based on a hybrid reactor formed by an HRAP and a BR, the emissions corresponded to 0.19 kg CO_{2eq}. Li and co-workers investigated the treatment of municipal sewage by a non-separated nutrient resource derived from municipal wastewater and integrated in order to facilitate biofuel production from microalgae [53]. The GHG emitted from the proposed process were 20,881 kg CO_{2eq}/y.

2.6. Ozonation

Ozonation was used for wastewater treatment purposes in 1906 in Paris, France [54]. In ozonation processes, ozone reacts with organic contaminants and degrades them but can form intermediary toxic products, and its low water solubility leads to low process efficiency [54]. This process has been examined in the literature as a tertiary treatment method, producing great results for the microbial load and the pharmaceuticals removed, but did not significantly affect the nutrient content of the secondary effluent. More specifically, Lamba and Ahammad [55], reported a 4 log reduction in *coliforms*, whereas Nasuhoglu et al. [56], Shi et al. [57], and Maniakova et al. [58], reported a 2.2- to 5.3-log reduction in *coliforms*, *E. coli*, *Salmonella*, and *Enterococcus*. Regarding pharmaceutical removal, Liu et al. [59] found that ozonation was capable of removing a range of pharmaceuticals by 5 to 80%. Antoniou et al. [60] also reported a removal of 70 to 100% for carbamazepine, naproxen, beclomethasone, and memantine. Regarding the treatment costs, 0.03 EUR/m^3 has been reported in the literature [61], with a global warming potential of 0.025 to 0.3 kg CO_2 eq. m^{-3}.

2.7. Photo-Fenton

Among the various advanced oxidation processes (AOPs), the Fenton oxidation process has gained significant attention, mostly because of its low operational costs [62]. The photo-Fenton process was employed in order to be studied for the removal of sulfamethazine, resulting in complete degradation of this antibiotic, whereas the TOC reduction was 56%, as reported by Pérez-Moya et al. [62]. A 100% elimination of amoxicillin and an 81% TOC removal were obtained by implementing the photo-Fenton process as presented by Trovó et al. [63]. The degradation of 22 micropollutants present in municipal wastewater was investigated by employing a photo-Fenton (UV254/H_2O_2/Fe) process, as shown by De la Cruz et al. [19]. The average percentage concerning the removal of all 22 pollutants was 80%.

The solar photo-Fenton process was quite effective for ARB and ARG removal from urban sewage, as mentioned by Giannakis et al. [64]. Various Phs, namely ofloxacin, carbamazepine, flumequine, ibuprofen, and sulfamethoxazole, were found to be almost completely removed from municipal sewage when coupling NF and solar photo-Fenton technology, as reported by Miralles-Cuevas et al. [65]. Quite interesting is the work of Elmolla and Chaudhuri, who combined the photo-Fenton process with a sequencing batch reactor (SBR) to study performance concerning the treatment of antibiotic wastewater [66]. They achieved an 89% reduction in soluble COD.

Reductions in the Phs present in wastewater produced by a Spanish pharmaceutical industrial unit were investigated by considering the environmental impact of employing an LCA over heterogeneous and homogeneous Fenton processes, as reported by Rodríguez and co-workers [67]. The heterogeneous Fenton process exhibited lower GHG emissions of 0.04 kg CO_{2eq}. An LCA study was performed by Pesqueira et al. of various solar-based treatments including solar circumneutral photo-Fenton (SPF) [68]. The GHG emissions of the latter were estimated at 0.331 kg CO_{2eq}. The emitted GHG of 554 kg CO_{2eq}/1000 m^3 as it concerns the operation of a solar photo-Fenton process at acidic pH for municipal wastewater treatment were found to be lower compared to systems operating at neutral pH, as presented by Gallego-Schmid and co-workers [69].

A semi-industrial solar photo-Fenton reactor was investigated by Foteinis et al. concerning the environmental sustainability of the proposed process over real wastewater effluent derived from a pharmaceutical laboratory [70]. The TOC reduction was found to be 79%, whereas the GHG emissions obtained from the LCA corresponded to 2.71 kg CO_{2eq} m^{-3}. Photo-Fenton processes in compound parabolic concentrator-type solar reactors have attracted significant interest recently, which is mostly attributed to their enhanced performance over the degradation of recalcitrant pollutants [71]. As mentioned in the analysis performed by Belalcázar-Saldarriaga and co-workers of the above-mentioned process employed for the degradation of acid orange 52 dye (AO52), the obtained COD and TOC reduction percentages were 55% and 35%, respectively, and the GHG emitted from the system operation were 0.762 kg CO_{2eq}/m^3 wastewater [71].

3. Benchmarking of Tertiary Wastewater Treatment Technologies

The data collected from the literature regarding the aforementioned methods are presented in Appendix A and summarized in Table 1. As can be observed in Table 1, the different tertiary treatment methods exhibited different strengths and weaknesses in the examined aspects. The physicochemical and advanced oxidation methods had a significant impact on the microbial load of the secondary effluent, whereas the biological methods did not exhibit microbial load reductions (or this was not reported at all in the examined literature). On the other hand, the biological processes were the only ones with significant nutrient removal through their assimilation with the produced biomass. Regarding the presence of pharmaceuticals, all the examined tertiary methods showed significant reductions. Regarding the economic aspect, the simpler (and more widespread) methods, such as chlorination, UN irradiation, and ozonation, had the lowest treatment costs. Finally, the GWP of the examined methods seems to follow a similar trend to their energy consumption, with the energy-intensive processes, i.e., membrane filtration, exhibiting the highest GHG emissions.

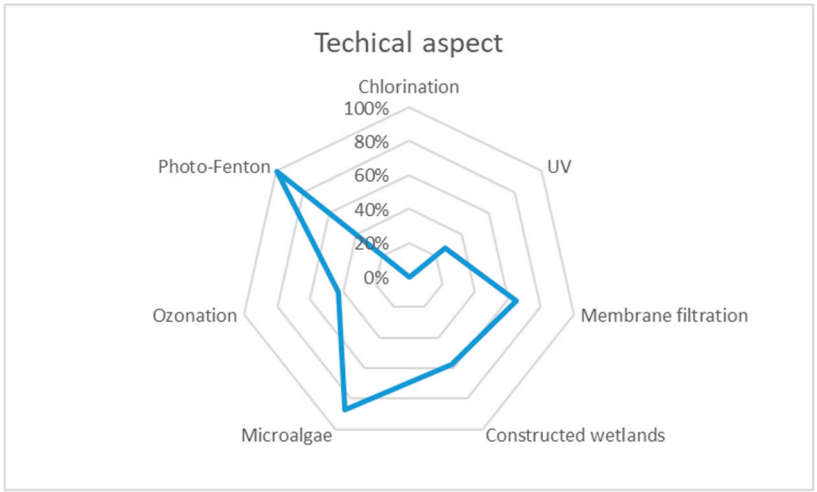
Table 1. Average technical economic and life cycle performances of the examined tertiary treatment methods.

Category	Treatment Method	Technical			Economic (Cost EUR/m^3)	Life Cycle (kg CO_2 eq. m^{-3})
		Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)		
Physicochemical	Chlorination	2.14	0	42	0.004	0.040
	UV	2.92	0	53	0.004	0.086
	Membrane filtration	3.50	6	70	0.614	0.754
Biological	Constructed wetlands	0.87	53	57	0.784	0.511
	Microalgae	0.00	77	73	0.291	0.468
Advanced oxidation	Ozonation	3.18	0	63	0.030	0.219
	Photo-Fenton	4.93	0	84	0.405	0.549

In order to facilitate a comparison of the different tertiary treatment methods investigated, a scoring methodology was implemented, with a score given for the three aspects (technical, economic, and life cycle) for each treatment method. In this way, the performance of each tertiary treatment method could be put into perspective and easily compared with that of the other examined processes. The scoring system ranged from 0% (worst score) to 100% (best score), with different normalization steps being needed for each aspect. Here, it should be noted that despite making the results of the literature review more comprehensible, the necessary assumptions for the scoring step, such as using data for the removal of specific pathogens or pharmaceuticals for each tertiary treatment method, increased the uncertainty. For the highest accuracy, the reader should refer to the raw data extracted from the literature.

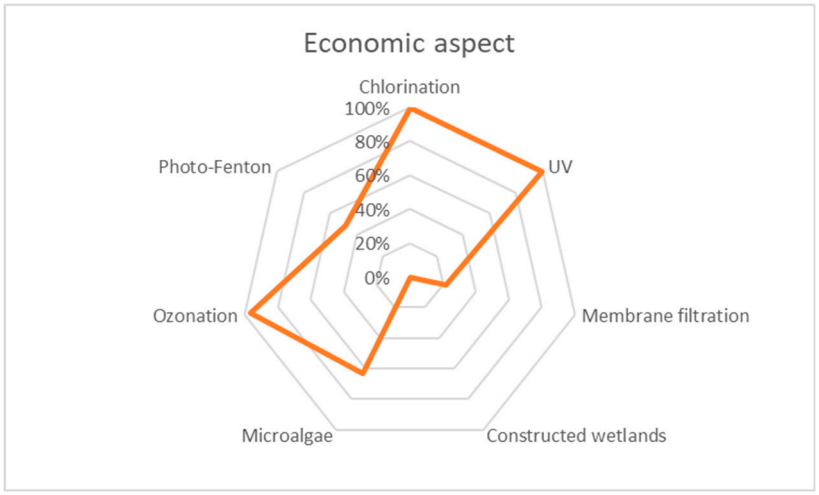
Regarding the technical aspect, the three subcategories of microbial load, nutrient, and pharmaceutical reductions were considered to contribute equally to the average technical aspect score. After normalizing the microbial reduction data by dividing by the highest value, an average reduction was calculated for each method using the microbial, nutrient, and pharmaceutical reductions. Finally, in order to have the technical aspect scores ranging from zero to one hundred, the average reduction values were normalized a second time by subtracting the lowest average reduction value and dividing by the highest average reduction value.

Because the economic and life cycle aspects had a negative impact on the evaluation of the processes, the values used for scoring were the comparison of the methods and the worst results for each criterion. For example, constructed wetlands had the highest treatment cost amongst the processes examined, whereas chlorination and UV had the lowest. The value used for scoring the economic aspect of microalgae was $0.784 - 0.291 = 0.493$ EUR/m^3, which was then normalized by dividing by the difference between the highest value and the minimum value among the processes. This way, the worst-performing method had a score of zero and the best-performing method had a score of one hundred after normalization. The three obtained scores for each aspect examined are presented in Figure 1.

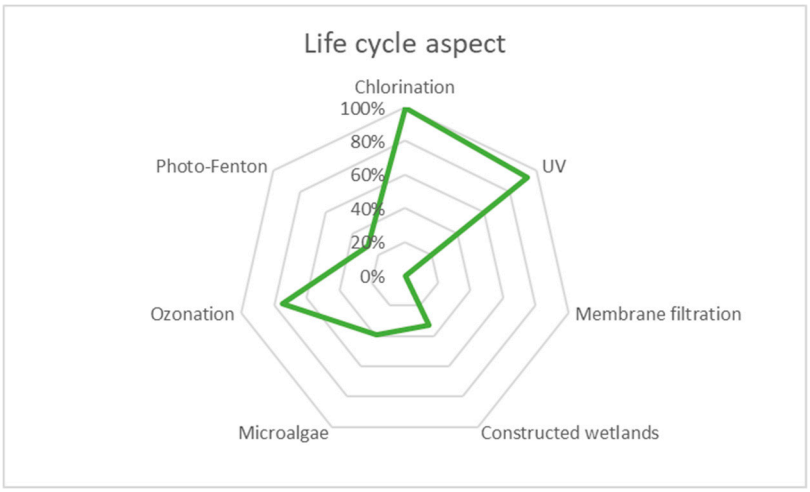


(**a**)

Figure 1. *Cont.*



(**b**)



(**c**)

Figure 1. Normalized scores for the three aspects examined for each tertiary treatment method; (**a**) technical aspect, (**b**) economic aspect, (**c**) life cycle aspect.

Because of the complex nature of choosing the most appropriate tertiary treatment method, since each method had certain strengths and weaknesses, the weight given to each of the three examined aspects was decided. In order to facilitate this process, a ternary diagram was constructed, with its three axes corresponding to the different weights to be given to each aspect (Figure 2). Each point in Figure 2 denotes the method with the highest weighted score for the three corresponding weight values. The ternary diagram was constructed by calculating the overall score of all the examined methods at each point and indicating the tertiary method with the highest overall score. The overall score for each method at each point was calculated using the three weights for the three examined aspects (corresponding to the coordinates of the points on the diagram) and the score for each aspect (as depicted in Figure 1). For example, for the points 0.2, 0.6, 0.2 (technical, economic, and life cycle) in the ternary plot, the overall score of all the methods was calculated using the formula 0.2 × Technical aspect score + 0.6 × Economic aspect score + 0.2 × Life cycle aspect score. The resulting scores for all the methods at this point in the plot were compared and the UV method was proven to have the highest overall score. This process was repeated for all the points in Figure 2 in increments of 0.025, calculating a total of 860 sets of overall scores. In this way, the best-performing (in terms of overall score) methods were determined and are depicted in Figure 2. The fact that not all the examined tertiary treatment methods appear in the final plot corresponds to the fact that the missing

methods did not exhibit the highest overall score amongst the examined methods at any point in the plot. The final plot can be used to indicate the tertiary method with the highest overall score for a set of weights determined by the user, corresponding to the importance of the technical, economic, and life cycle aspects of the application under investigation.

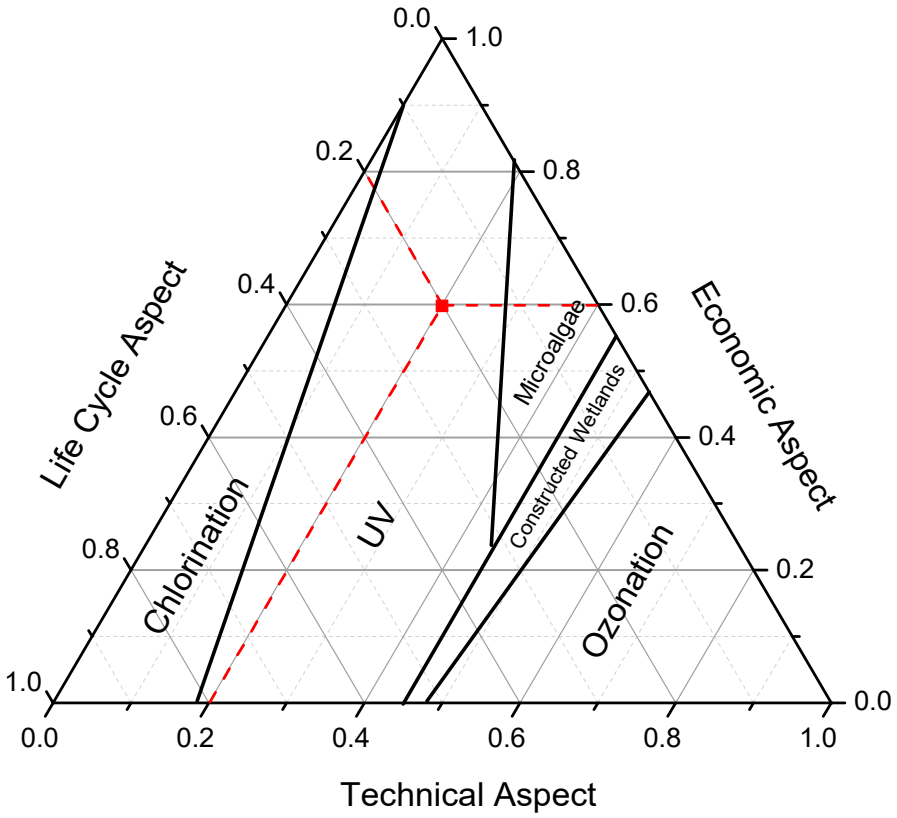


Figure 2. Ternary diagram for the evaluation of tertiary treatment methods, with an example of 0.2, 0.6, and 0.2 weights for the technical, economic, and life cycle aspects, respectively.

4. Conclusions

Many options for tertiary treatment have been examined in the literature, with each treatment process exhibiting different positive and negative points. The first step of the analysis presented herein was the aggregation of data regarding the technical, economic, and environmental impact of each process. This step had the fewest assumptions, as the data were reported as extracted from the literature, but a further aggregation of the results could facilitate a comparison of the tertiary treatment methods examined. As a next step, the data were normalized (assuming the equivalent significance of microbial load, nutrient, and pharmaceutical reductions) and scored. Despite the fact that this process facilitated a comparison of the examined methods, some applications may have attributed higher importance to one technical aspect, for example, when the water recipient was prone to eutrophication or if the sewage derived from a hospital with a high pharmaceutical concentration. The final step was the construction of the ternary diagram illustrating the tertiary treatment method with the highest weighted score for each weight set chosen by the user. This tool can be used to quickly illustrate the strengths and weaknesses of the examined methods and also for decision making in a typical tertiary treatment process, but as previously stated, when specific challenges need to be overcome, directly examining the data aggregated from the literature would be more suitable. UV, ozonation, and chlorination dominated the optimal tertiary treatment methods according to Figure 2, followed by microalgae cultivation and constructed wetlands. The most important advantage of UV and chlorination appears to be their lower treatment costs and low GHG emissions, whereas ozonation exhibited increased efficiency in microbial load and pharmaceutical reductions

with moderate treatment costs. The increased treatment costs and GHG emissions of membrane filtration and photo-Fenton did not allow for their assessment as optimal tertiary methods under any set of weights, as seen in Figure 2. It should be noted that these results were dependent on the implemented methodology, and the high efficiency of both the membrane filtration and photo-Fenton methods in the removal of pathogens and pharmaceuticals can be of high value in cases where increased treatment efficiency is required. Through the analysis presented herein, the complementarity of different tertiary treatment methods is illustrated, facilitating their combination in multi-step processes for the removal of all contaminants (i.e., combining biological and physicochemical or advanced oxidation methods).

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Conflicts of Interest: The authors declare no conflict of interest.

Abbreviations

AO52	acid orange 52
AOPs	advance oxidation processes
ARB	anaerobic resistant bacteria
ARG	antibiotic resistance genes
BOD	biochemical oxygen demand
BR	biofilm reactor
COD	chemical oxygen demand
CW	constructed wetlands
EC	emerging contaminants
GHG	greenhouse gases
GO	graphene oxide
HFCW	horizontal flow constructed wetlands
HRAP	high-rate algal pond
HRP	high-rate pond
HSSF	horizontal subsurface flow
LCC	life cycle costing
MF	microfiltration
MFCs	microbial fuel cells
MWCNTs	multi-walled carbon nanotubes
NF	nanofiltration
PBR	photobioreactor
PVDF	polyvinylidene fluoride
RO	reverse osmosis
SBR	sequencing batch reactor
SPF	solar photo-Fenton
TOC	total organic carbon
UF	ultrafiltration
UV	ultraviolet
VFCW	vertical flow constructed wetlands

Appendix A

Table A1. Literature data used for scoring the chlorination method.

Process Information	Technical			Economic (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
Sulfamethoxazole 220 ng/L			73			[10]
Ciprofloxacin 153 ng/L			66			[10]
Norfloxacin 92 ng/L			50			[10]
Tetracycline 86 ng/L			39			[10]
Trimethoprim 155 ng/L			65			[10]
Erythromycin 273 ng/L			43			[10]
Diclofenac 40 µg/L			97			[72]
Ibuprofen 40 µg/L			0			[72]
Clofibric acid 40 µg/L			5			[72]
Naproxen 40 µg/L			11			[72]
Gemfibrozil 40 µg/L			45			[72]
Mefenamic acid 40 µg/L			12			[72]
E. coli 3.7 Log CFU/100 mL	2.5					[8]
E. coli 4.34 Log CFU/100 mL	2.57					[9]
Enterococci 3.46 Log CFU/100 mL	1.18					[9]
Fecal coliforms 4.57 Log CFU/100 mL	2.34					[9]
F-specific coliphage 2.33 Log CFU/100 mL	0.71					[9]
Somatic coliphage 3.92 Log CFU/100 mL	1.68					[9]
Adenovirus 0.97 Log CFU/100 mL	0.81					[9]
Norovirus 0.74 Log CFU/100 mL	0.74					[9]
Coliforms 4 Log CFU/100 mL	4					[55]
Antib. Resist. Genes 6 Log	1.97					[13]
E. coli 7 Log CFU/mL	5					[57]
				0.0003		[11]
				0.005		[12]
				0.006		[13]
					0.046	[73]
					0.007	[15]
					0.004	[14]

Table A2. Literature data used for scoring the UV method.

Process Information	Technical			Economic (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
E. coli 5 × 10^5 CFU/100 mL	4					[74]
E. coli 7.7 Log × 10 CFU/L	3.82					[9]
Enterococci 8.56 Log × 10 CFU/L	3.38					[9]
Fecal coliforms 8.26 Log × 10 CFU/L	3.89					[9]
F-specific coliphage 6.4 Log × 10 CFU/L	1.17					[9]
Somatic coliphage 7.36 Log × 10 CFU/L	2.98					[9]
Adenovirus 2.73 Log gc/L	0.24					[9]
Coliforms 5 Log CFU/mL	4					[55]
Antib. Resist. Genes 6 Log	1					[13]
Antib. Resist. Genes 5 Log copies/L	2.5					[20]
E. coli 2 × 10^7 CFU/mL	5.1					[57]
Sulfamethoxazole 250 ng/L			100			[75]
Trimethoprim 90 ng/L			100			[75]
Erythromycin 200 ng/L			100			[75]
Acetaminophen 0.1 mM, caffeine 0.12 mM, antipyrine 0.05 mM, doxycycline 0.03 mM, ketorolac 0.05 mM			100			[76]

Table A2. *Cont.*

Process Information	Technical			Economic (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
Atrazine diuron, alachlor, pentachlorophenol 1 mg/L			72			[76]
Boldenone 6.57 µM			98			[76]
BPA 60 µM			22			[76]
Butylparaben 8×10^{-5} M			97			[76]
Carbamazepine 3 µM			52			[76]
Chlorfenvinphos			91			[76]
Ciprofloxacin			99			[76]
Chloromycetin 10 mg/L			80			[76]
Clofibric acid 10 mg/L			98			[76]
Cyclophosphamide 10 µg/L			28			[76]
Cytarabine 10 mg/L			10			[76]
Diatrizoate 50 µM			97			[76]
Diclofenac 20 mg/L			74			[76]
Diphenhydramine 5 µM			26			[76]
Doxycycline 5×10^{-5} M			27			[76]
E1 20 mg/L			69			[76]
E2 20 mg/L			59			[76]
EE2			37			[76]
Hydrochlorothiazide 1 µM			59			[76]
Ibuprofen 10^{-4} M			74			[76]
Iopromide			53			[76]
Iohexol 3 µM			12			[76]
Irinotecan 10 µg/L			18			[76]
Isoproturon 1 mg/L			12			[76]
Ketoprofen 50 µM			99			[76]
Mefenamic acid 5.5 Log M			56			[76]
Melatonin 20 mg/L			32			[76]
Metoprolol 5×10^{-4} M			69			[76]
Metronidazole 6 µM			55			[76]
Naproxen 3 µM			65			[76]
NDMA 1 mM			100			[76]
Norfloxacin 5×10^{-5} M			55			[76]
Oxtetracycline			93			[76]
Phenazone 5 µM			96			[76]
Phenytion 5 µM			88			[76]
Primidone 50 µM			9			[76]
Propranolol 100 mg/L			61			[76]
Sulfadimethoxine 3.2 mM			99			[76]
Sulfamethoxazole 10 mg/L			83			[76]
Tamoxifen 10 µg/L			43			[76]
TCE 8.14×10^{-3} mol/L			95			[76]
Tibetene 0.03 mM			87			[76]
Bezafibrate 112 ng/L			0			[19]
Metformin 1736 ng/L			27			[19]
Carbamazepine 333 ng/L			48			[19]
Gabapentin 1508 ng/L			0			[19]
Diclofenac 925 ng/L			96			[19]
Ketoprofen 40 ng/L			97			[19]
Naproxen 372 ng/L			70			[19]
Primidone 65 ng/L			3			[19]
Atenolol 320 ng/L			0			[19]
Metoprolol 255 ng/L			0			[19]
Ciprofloxacin 72 ng/L			56			[19]
Clarithromycin 187 ng/L			10			[19]
Sulfamethoxazole 355 ng/L			3			[19]
Trimethoprim 31 ng/L			0			[19]
Iohexol 4313 ng/L			16			[19]
Iomeprol 5806 ng/L			0			[19]
Benzotriazole 6736 ng/L			18			[19]
Atrazin 25 ng/L			58			[19]
Isoproturon 4 ng/L			0			[19]
Mecoprop 365 ng/L			0			[19]
Terbutryn 23 ng/L			39			[19]
				0.00001		[11]

Table A2. *Cont.*

Process Information	Technical			Economic (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
				0.00644		[13]
				0.0063		[13]
					0.013	[73]
					0.026	[15]
					0.22	[77]

Table A3. Literature data used for scoring the membrane filtration method.

Process Information	Technical Aspect			Economic Aspect (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
UF enterococcus 1.87 × 10^5 CFU/100 mL	5					[6]
UF other coliforms 5.05 × 10^4 CFU/100 mL	2					[6]
UF N 3.62 mg/L		10				[6]
UF P 1.86 mg/L		9				[6]
UF K 16.15 mg/L		0				[6]
NF-90 2 µg/L			73			[78]
NF-200 neutral PhACs 65 µg/L			70			[78]
NF-200 ionic PhACs 65 µg/L			94			[78]
NF-90 neutral 65 µg/L			97			[78]
NF-90 ionic 65 µg/L			99			[78]
NF-90 65 µg/L			73			[78]
UF 2000 0.5 mg/L			70			[78]
UF-NF90 750 µg/L			50			[78]
NF 270 RO 2 µg/L			95			[78]
NF 150-RO 100 ng/L			95			[78]
NF 200 100 ng/L			80			[78]
UF 8000-NF 600 10 ng/L			60			[78]
UF 8000 10 ng/L			30			[78]
NF 90-RO 10 mg/L			99			[78]
NF 200 21 ng/L			100			[78]
NF 270 10 mg L			60			[78]
NF270 800 µg/L			58			[78]
NF90 750 µg/L			97			[78]
RO 0.55 mg/L			100			[78]
NF90 10 mg/L			90			[78]
NF270 10 mg/L			61			[78]
NF90 0.5 mg/L			98			[78]
NF270 0.5 mg/L			71			[78]
RO 0.5 mg/L			89			[78]
NF90 5400 µg/L			77			[78]
NF270 5400 µg/L			58			[78]
RO 5400 µg/L			93			[78]
UF-Atenolol 778 ng/L			0			[22]
UF-Bezafibrate 208 ng/L			21			[22]
UF-Caffeine 17,725 ng/L			0			[22]
UF-Fenofibric acid 139 ng/L			0			[22]
UF-Furosemide 1302 ng/L			17			[22]
UF-Gemfibrozil 18,504 ng/L			71			[22]
UF-Hydrochlorothiazide 16,628 ng/L			90			[22]
UF-Ibuprofen 2514 ng/L			1			[22]
UF-4-AAA 7364 ng/L			0			[22]
UF-Naproxen 2672 ng/L			12			[22]
UF-Nicotine 10,954 ng/L			63			[22]
UF-Ofloxacin 94 ng/L			0			[22]
RO-Atenolol 1044 ng/L			100			[22]
RO-Bezafibrate 164 ng/L			100			[22]
RO-Caffeine 6288 ng/L			99			[22]
RO-Fenofibric acid 194 ng/L			100			[22]

Table A3. *Cont.*

Process Information	Technical Aspect			Economic Aspect (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
RO-Furosemide 811 ng/L			100			[22]
RO-Gemfibrozil 1035 ng/L			99			[22]
RO-Hydrochlorothiazide 239 ng/L			95			[22]
RO-Ibuprofen 574 ng/L			97			[22]
RO-4-AAA 4472 ng/L			99			[22]
RO-Naproxen 2583 ng/L			98			[22]
RO-Nicotine 75 ng/L			76			[22]
RO-Ofloxacin 87 ng/L			95			[22]
Including RO				0.46		[27]
FO-NF				0.96		[27]
UF-RO				0.4		[27]
UF				0.45		[27]
UF-RO				0.8		[27]
UF-RO					2.32	[29]
NF					0.2	[28]
UF					0.25	[77]
UF					0.40	[73]
MF-RO					0.89	[73]
UF-RO					0.91	[73]

Table A4. Literature data used for scoring the constructed wetlands method.

Process Information	Technical Aspect			Economic Aspect (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
16 s rDNA, intI1, and tet genes	1.78					[34]
14 antibiotic resistance genes	0.50					[36]
ARGs 8–9 Log of copies/mL	0.49					[40]
N 23.4 mg/L		5				[31]
P 0.2 mg/L		0				[31]
N 29.3 mg/L		35				[31]
P 0.2 mg/L		50				[31]
N 17.3 mg/L		39				[31]
P 0.2 mg/L		50				[31]
N 1.39 mg/L		66				[33]
P 3 mg/L		46				[33]
N 84.4 mg/L		63				[34]
P 28.2 mg/L		92				[34]
N 35 mg/L		46				[40]
N 72 mg/L		99				[79]
P 11.7 mg/L		97				[79]
65 pharmaceuticals 4.3 µg/L			64			[31]
55 pharmaceuticals 300 ng/L			43			[31]
53 pharmaceuticals			50			[31]
56 pharmaceuticals 190 ng/L			32			[31]
6 pharmaceuticals 7.6–150 µg/L			93			[80]
Antibiotics 300 ng/L			58			[40]
Pharmaceuticals 50–200 ng/L			59			[79]
				1.224		[45]
				0.729		[45]
				0.4		[49]
					0.129	[73]
					0.432	[42]
					0.646	[42]
					0.911	[42]
					0.5	[42]
					0.26	[47]
					0.7	[49]

Table A5. Literature data used for scoring the microalgae.

Process Information	Technical			Economic (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
N 40 mg/L		55				[81]
P 80 mg/L		15				[81]
N 52 mg/L		82				[82]
P 8.5 mg/L		95				[82]
N 18 mg/L		100				[83]
P 1.4 mg/L		84				[83]
N 46 mg/L		94				[51]
P 5.5 mg/L		95				[51]
Metronidazole 5 µM			100			[84]
Florfenicol 46 mg/L			97			[84]
Enrofloxacin 1 mg/L			23			[84]
Tetracycline 100 µg/L			99			[84]
Methyl parathion 20 mg/L			80			[84]
Trimethoprim, Sulfamethoxazole, Triclosan 1.6 ng/L, 360 ng/L, 8 ng/L			44			[84]
7-amino cephalosporanic acid 100 mg/L			70			[84]
Cefradine 100 mg/L			94			[84]
β-estradiol			93			[84]
17 α-estradiol, 17 β-estradiol, Estrone, Estriol 5 µg/L			90			[84]
Sulfathiazole, Sulfapyridine, Sulfamethazine, Sulfamethoxazole, Tetracycline, Oxytetracycline 200 µg/L			47			[84]
Norfloxacin mg/L			37			[84]
				0.42		[49]
				0.162		[85]
					0.6	[49]
					0.336	[86]

Table A6. Literature data used for scoring the ozonation method.

Process Information	Technical			Economic (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
Coliforms 4 Log CFU/100 mL	4					[55]
Coliforms 5 Log MPN/100 mL	2.5					[56]
Coliforms 5 Log MPN/100 mL	2.2					[56]
Coliforms 7 Log MPN/100 mL	4.8					[56]
E. coli 7.3 Log CFU/mL	5.3					[57]
E. coli 4 Log CFU/mL	2.2					[58]
Salmonella 2.9 Log CFU/mL	2.2					[58]
Enterococcus 3 Log CFU/mL	2.2					[58]
Carbamazepine			75			[59]
Alachlor			20			[59]
Bisphenol A			60			[59]
Atrazine			5			[59]
Pentachlorophenol			35			[59]
17-α thinylestradiol			80			[59]
Carbamazepine 1 µg/L			100			[60]
Naproxen 1 µg/L			100			[60]
Beclomethasone 1 µg/L			70			[60]
Memantine 1 µg/L			80			[60]
				0.03		[61]
				0.03		[61]
					0.025	[87]
					0.25	[88]
					0.3	[28]
					0.3	[89]

Table A7. Literature data used for scoring the Photo-Fenton method.

Process Information	Technical			Economic (Cost EUR/m^3)	Life Cycle (kg CO_2 eq./m^3)	Ref.
	Microbial (Log Reduction)	Nutrients (% Reduction)	Pharmaceuticals (% Reduction)			
S. aureus 6 Log CFU/mL	6					[64]
MRSA ATCC 29213 6 Log CFU/mL	6					[64]
E. coli 6 Log CFU/mL	6					[64]
K. pneumoniae 6 Log CFU/mL	6					[64]
MSSA 1112 6 Log CFU/mL	6					[64]
MSSA 1112 RifR 6 Log CFU/mL	6					[64]
MSSA 1112 CipR 6 Log CFU/mL	6					[64]
MSSA 133 6 Log CFU/mL	6					[64]
MSSA 133 CipR 6 Log CFU/mL	6					[64]
MRSA PC1 6 Log CFU/mL	6					[64]
VISA PC# 6 Log CFU/mL	6					[64]
E. coli 4 Log CFU/mL	1					[58]
Salmonella 2.9 Log CFU/mL	2					[58]
Enterococcus 3 Log CFU/mL	0					[58]
Sulfamethazine 50 mg/L			100			[62]
Amoxicillin 50 mg/L			100			[63]
Bezafibrate 112 ng/L			0			[19]
Gemfibrozil 9 ng/L			96			[19]
Metformin 1736 ng/L			63			[19]
Carbamazepine 333 ng/L			94			[19]
Gabapentin 1508 ng/L			77			[19]
Diclofenac 925 ng/L			100			[19]
Ketoprofen 40 ng/L			97			[19]
Naproxen372 ng/L			97			[19]
Primidone 65 ng/L			77			[19]
Atenolol 320 ng/L			87			[19]
Metoprolol 255 ng/L			90			[19]
Ciprofloxacin 72 ng/L			61			[19]
Clarithromycin 187 ng/L			76			[19]
Sulfamethoxazole 355 ng/L			82			[19]
Trimethoprim 31 ng/L			88			[19]
Iohexol 4313 ng/L			94			[19]
Iomeprol 5806 ng/L			87			[19]
Benzotriazole 6736 ng/L			95			[19]
Atrazin 25 ng/L			82			[19]
Isoproturon 4 ng/L			32			[19]
Mecoprop 365 ng/L			93			[19]
Terbutryn 23 ng/L			83			[19]
Ofloxacin 110 µg/L			100			[65]
Carbamazepine130 µg/L			96			[65]
Flumequine 145 µg/L			98			[65]
Ibuprofen 130 µg/L			95			[65]
Sulfamethoxazole 140 µg/L			90			[65]
				0.25		[90]
				0.56		[90]
					0.331	[68]
					0.554	[69]
					0.762	[71]

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