



Life Cycle Assessment of sulfate radical based-AOPs for wastewater disinfection

S. Guerra-Rodríguez, S. Cuesta, J. Pérez, E. Rodríguez, J. Rodríguez-Chueca *

Department of Industrial Chemical & Environmental Engineering, Escuela Técnica Superior de Ingenieros Industriales, Universidad Politécnica de Madrid, Madrid, Spain

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ABSTRACT

Water stress is a global issue that is increasingly worsening due to rising water demand and uneven rainfall distribution caused by climate change. Moreover, the quality of water bodies is deteriorating, necessitating the exploration of alternative water sources, such as wastewater reclamation and reuse. However, ensuring the absence of pathogens is crucial in mitigating potential health risks. Advanced oxidation processes (AOPs) that utilize sulfate radicals (SR-AOPs) have been proposed as alternatives to conventional disinfection treatments. The SR-AOPs have the advantage of producing less or no disinfection by-products, but their environmental sustainability must be considered. This study aims to evaluate the life cycle impacts of four SR-AOPs that utilize peroxymonosulfate (PMS) for treatment: PMS/UV-A; PMS/H₂O₂/UV-A at 1:1 and 1:3 ratios; and PMS/O₃. Furthermore, the study assesses the impact of the pilot plant where the treatments are carried out. Results show that PMS/H₂O₂/UV-A at a 1:3 ratio had the highest environmental impact in nine of the sixteen categories studied (Product Environmental Footprint method), followed by PMS/UV-A. The PMS/O₃ treatment scenario had the lowest environmental impact and achieved a 5-log removal of *Enterococcus faecalis* in the shortest treatment time. Notably, electricity consumption emerged as the most critical factor in assessing the environmental sustainability of the treatments, accounting for between 65 and 90% of the total impact across all scenarios. Additionally, fossil resource depletion and climate change emerged as the most relevant impact categories across all scenarios. In this way, LCA becomes a key tool when making decisions in the selection of treatment scenarios, not only considering the treatment efficiency.

1. Introduction

Due to the global water scarcity currently being experienced, the reuse of reclaimed wastewater is postulated as a viable alternative to reduce water stress in certain parts of the world. However, there are several concerns that prevent this from becoming a reality [1], such as the presence of contaminants of emerging concern (CECs) and microbiological agents (including antibiotic resistance genes, ARGs) at the outlet of conventional wastewater treatment plants [2–5]. European Regulation (EU) 2020/741 establishes minimum requirements for water reuse, including the control of pathogenic microorganisms, and monitoring of CECs and ARGs in certain situations, with the aim of promoting water reuse in all member states. This new legislation has been in effect since June 2020, but will only be applicable as of 26 June 2023. Therefore, to achieve safe water reuse, it is essential to develop treatments capable of effectively removing these contaminants and complying with the current legislation.

In this context, research on the application of advanced oxidation processes (AOPs) for wastewater reclamation has undergone significant development in recent years. These processes are based on the generation of highly reactive radical species capable of degrading organic pollutants while inactivating microorganisms present in the water. Although processes based on the generation of hydroxyl radicals are the most widespread, treatments based on the generation of sulfate radicals (SR-AOPs) have also demonstrated the ability to degrade pollutants at laboratory and pilot scales, and to a lesser extent, at the industrial scale [6]. Peroxymonosulfate (HSO₅⁻, PMS) is the main precursor for the generation of sulfate radicals. Its activation can occur through multiple routes, with the most studied the use of UV radiation, heat, or homogeneous and heterogeneous catalysts. Recently research has investigated the efficiency of processes combining PMS with other oxidants, such as H₂O₂ or O₃ [7–12]. Table S1 in Supplementary Material shows the main reactions associated with SR-AOPs involving UV radiation or the use of other oxidants such as H₂O₂ or O₃. The advantages of this type of

* Corresponding author.

E-mail address: jorge.rodriguez.chueca@upm.es (J. Rodríguez-Chueca).

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multiple oxidation process are that it allows for process intensification, increasing its efficiency, and reducing the required concentration of PMS, which is typically a limitation due to its high associated economic cost.

Most of the SR-AOPs described in the literature focus on laboratory-scale studies, where the main conclusions are derived from degradation values of different target pollutants. In rare cases, pilot-scale or full-scale studies are presented, which also include economic analysis. However, economic costs are not the sole factor to be considered. When developing processes to address a specific environmental issue, it is crucial to be mindful of the use of resources and energy during treatment, which can result in additional environmental burdens [13]. Therefore, it is necessary to assess the potential environmental impact associated with the process, ensuring that the proposed solution does not create greater environmental burdens than the initial problem being addressed. A valuable tool for this purpose is Life Cycle Assessment (LCA), as it enables the quantification of impacts throughout all stages of a product, service, or process [14].

As mentioned before, AOPs (Advanced Oxidation Processes) present multiple advantages from the perspective of their effectiveness in removing certain contaminants. However, they can also have certain disadvantages that can translate into associated environmental impacts. For example, the vast majority of AOPs require the use of chemical reactants [15], such as chemical oxidants or catalysts based on the presence of metals, to promote the generation of oxidizing radicals. The use of these chemical substances can pose a post-treatment problem, especially when used in high concentrations [16–18]. On the other hand, many AOPs are photo-assisted, meaning that to increase the efficiency of the process, the use of radiation (of any type depending on the type of treatment) [15] is required. This translates into a significant use of electrical energy, which therefore represents a potential environmental impact [19,20]. Other factors such as the use of temperature in reactions can also pose a potential environmental impact due to the need for an external energy input. Therefore, process optimization is crucial in order to minimize the associated impacts, considering, in any case, the combination of reactants that allows for the highest performance with the lowest necessary concentration, as well as the use of the energy source with the least impact to carry out photo-assisted processes, such as solar radiation.

Unfortunately, there is a general lack of literature regarding the life cycle impacts of advanced oxidation processes. Although some research has conducted LCA of different AOPs [21–23], even comparing them to other advanced treatments such as ozonation or nanofiltration [24,25], only two publications addressing the environmental impacts of SR-AOPs have been found at the time of writing [26,27]. In both cases, the authors found that the production of oxidizing agents accounted for the majority of the environmental footprint of the treatments corresponds to the production of oxidising agents, followed by electricity consumption. However, further studies are necessary to analyse a wide range of scenarios in order to gain a comprehensive understanding of the environmental consequences of implementing these processes at a real scale. To the best of our knowledge, this is the first time that a comparative study of the quantified environmental impacts through LCA of 4 different SR-AOPs used for the disinfection of wastewater samples at a pilot plant level is reported. It should be noted that this is a scaling work from the laboratory scale to the pilot scale, and where subsequently, the LCA along with the results of the target contaminant removal efficiency, serves for decision-making on which treatment is the most sustainable from an environmental standpoint.

In previous work conducted by this research group, various SR-AOPs have been optimised at a pilot scale for the inactivation of *Enterococcus faecalis* [28]. Hence, the primary objective of this study is to conduct a comparative LCA to ascertain which of the optimized treatments is the most environmentally. Furthermore, an LCA of the pilot plant has also been conducted. Additionally, a sensitivity analysis has been performed to assess the impact of the energy mix utilized for electricity generation

on the treatment's outcomes.

2. Materials and methods

2.1. Pilot plant

This work utilized a pilot plant model Photobench LED275-0.8/LED365-16/450-16a, which was developed by APRIA Systems. Figure S1 in Supplementary Material shows front and back images of the pilot plant. The system is equipped with a single feed pump (CM 1–3; Grundfos) that continuously circulates water from a 50 L feed tank (Etatron) through various treatment modules. It has a nominal flow rate of $1,700 \text{ L}\cdot\text{h}^{-1}$ and a maximum operating pressure of 16 bar. To enhance treatment efficiency and protect sensitive components of the system, a safety filter (NW25, Cintropur) is installed downstream of the feed pump to prevent the passage of solids larger than $25 \mu\text{m}$.

The key components of the plant are two parallel ring photoreactors, each comprising two concentric tubes. The inner tube houses the LED lamps, while the outer tube carries the water to be treated. The photoreactor utilized in the evaluated scenarios is composed of 20 UV-A LEDs lamps that emit radiation at a wavelength of 365–370 nm, with irradiance of $1,200 \text{ mW}$, and 20 Vis LEDs lamps arranged alternately in 4 rows. The theoretical maximum radiation emitted by the UV-A and Vis lamps is $1,973 \text{ W}\cdot\text{m}^{-2}$, and $2,969 \text{ W}\cdot\text{m}^{-2}$, respectively, but the total irradiated power can be adjusted in each test. The photoreactors are constructed with borosilicate glass and protected by a black polylactic acid housing, incorporating an air-cooling system and a temperature probe to preserve the LED's irradiation potential and lifespan. The maximum recommended operating flow rate in both reactors is $650 \text{ L}\cdot\text{h}^{-1}$, with a maximum pressure of 1 bar.

Furthermore, the plant is equipped with an ozone generation unit that can be utilized for an ozonisation stage prior to the photochemical treatment or as the primary treatment. The ozonisation system comprises an ozone generation unit and a Venturi tube for in-line dosing of the generated ozone. The ozone generator (GHBZO3-E, ZonoSistem) has the capability to produce an adjustable ozone dose, with a maximum flow rate of $2.5 \text{ g}\cdot\text{h}^{-1}$. Additionally, the plant incorporates an in-line air extractor with a Kosner activated carbon filter (model KE-100) positioned above the feed tank to control the gases generated during the process. The filter allows for a maximum air flow of $198 \text{ m}^3\cdot\text{h}^{-1}$ when operating at 2,200 rpm.

Moreover, the installation includes a temperature control system that enables for temperature-controlled conditions. This system comprises a heat exchanger, a thermostatically controlled bath, and an external temperature probe. The plant is equipped with a Programmable Logic Controller (PLC) that monitors, controls, and regulates the temperature and power consumption of the LED lamps. Additionally, it features an online controller consisting of a pH and temperature probe, along with an analytical transmitter that displays the data obtained by the probe (both from Mettler-Toledo).

2.2. Description of the scenarios evaluated

A previous study conducted by researchers at the UPM has enabled the optimization of various SR-AOPs and the pre-selection of four different water treatment scenarios based on their operational outcomes. Detailed information regarding the treatment optimization, which serves as the foundation for this comparative LCA, can be found in the study published by Guerra-Rodríguez et al. [28]. The concentration of *E. faecalis* over time in the optimized treatments can be referenced in Table S2 in the supplementary material. These four treatments are discontinuous and utilize a volume of 35 L of simulated wastewater. The physicochemical characteristics of the simulated wastewater can be consulted in Table S3 in the Supplementary Material. The primary objective of these treatments is to eliminate *Enterococcus faecalis* down to its limit of detection (LOD), which is $10 \text{ CFU}\cdot\text{mL}^{-1}$. PMS

(Sigma-Aldrich) is used as the oxidizing agent and is activated by UV-A radiation (365 nm). In some scenarios, the influence of H₂O₂ (Panreac) at 30% v/v and ozone as additional reagents in the activation of PMS is investigated. Table 1 presents the specific conditions for each treatment, with durations ranging from 20 min (for the O₃/PMS system) to 120 min (required in the PMS/H₂O₂/UV-A (1:3) system) and PMS concentrations ranging from 0.5 to 1.5 mM.

2.3. Life cycle assessment

The manuscript follows all four phases of a LCA in accordance with ISO 14040:2006 and ISO 14044:2006 [29,30]. These phases include defining the goal/scope, conducting a life cycle inventory (LCI), performing impact assessment, and interpretation. The subsequent subsections will provide further details on each of these stages.

2.3.1. Goal/scope, functional unit and system boundaries

The main objective of the LCA is to assess the potential environmental impact of the photochemical pilot plant and compare the potential effects of the proposed AOP in the four described scenarios described. This approach enables the determination of the optimal treatment scenario based on environmental criteria.

The functional unit (FU) serves as the basis for calculation and on which the material and energy balances are performed. Two FU have been established, one for each LCA conducted. The first FU corresponds to a volume of treated water with a specific amount of contaminant removed, specifically up to the limit of detection of *Enterococcus faecalis*. Therefore, the established FU is: "Removal of 10⁵ CFU/mL (colony-forming units/mL) of *Enterococcus faecalis* in 35 L of treated water effluent". The second FU pertains to the LCA of the entire pilot plant, with the FU defined as "a photochemical treatment pilot plant with UV-A LED technology".

Fig. 1 illustrates the system boundaries and the scope of the LCA, demonstrating a division into two stages. The initial stage encompasses the phases from raw material extraction to the pilot plant's arrival at its final location in Madrid. It represents a cradle-to-gate LCA of the pilot plant. The subsequent stage focuses on the comparative LCA of the selected treatment scenarios.

The pilot plant, equipped with photochemical technology using LED, is situated in the research laboratories of the Escuela Técnica Superior de Ingenieros Industriales at Universidad Politécnica de Madrid in Spain. The manufacturing process of the plant was by APRIA System, a company located in Cantabria, Spain. However, it should be noted that the components used were not all sourced from the same geographical location but were acquired through third-party suppliers.

The time frame for this study spans from September 2020 to September 2021, encompassing the construction phase of the pilot plant until the last completion of the final treatments in the study.

2.3.2. Data quality assessment

A detailed inventory has been compiled, utilizing various data sources including process diagrams, analytical data, construction drawings, equipment datasheets, and direct information obtained from vendors and manufacturers. It is important to note that there are practical limitations in collecting input data, particularly when equipment consist of multiple parts with varying masses and materials. Consequently, Table 2 categorizes the collected data based on quality, ranging

Table 1

Description of the scenarios evaluated by LCA. [H₂O₂] = 1.5 mM.

Scenario	Treatment	Operation time (min)
1	PMS (1.5 mM)/UV-A	90
2	PMS (0.5 mM)/O ₃	20
3	PMS (1.5 mM)/H ₂ O ₂ /UV-A	60
4	PMS (0.5 mM)/H ₂ O ₂ /UV-A	120

from highest (A) to lowest (D).

2.3.3. Emission inventory and impact assessment method

SimaPro [31] is one of the most widely used and comprehensive software tools available on the market to conduct LCA. This tool encompasses a diverse range of databases for the life cycle inventory (LCI) phase. The most used one is EcoInvent [32], with information on energy supplies, energy production, transportation, and waste disposal services, all with associated environmental performance.

The characterization of impact categories was conducted according to the Product Environmental Footprint (PEF), a methodology proposed by the European Commission as a common way of measuring environmental performance [33]. All impact categories included in PEF methodology are evaluated: acidification terrestrial and freshwater (AP), cancer human health effects (HTP-C), climate change (GWP), ecotoxicity freshwater (FETP), eutrophication freshwater (F-EP), eutrophication marine (M-EP), eutrophication terrestrial (T-EP), ionising radiation (IRP), land use (LU), non-cancer human health effects (HTP-NC), ozone depletion (ODP), photochemical ozone formation (POCP), use of fossil resources (F-RD), resource use mineral and metals (M-RD), respiratory inorganics (PMF), water scarcity (W-RD).

3. Results and discussion

3.1. Life cycle inventory

Following the standardized LCA procedure, two distinct LCA were conducted: the first to compare the different scenarios, and the second to determine the impacts associated with the pilot plant. The first life cycle inventory (LCI), related to the different scenarios, is shown in Table 3. It includes the quantities of reagents used in the treatments and calculates the energy consumption required to achieve the level of treatment described by the FU. Scenario 2 is the one in which the least number of reagents is used and, moreover, the one with the lowest energy consumption. This is because, although the ozonator consumes electrical energy for ozone generation, the overall consumption is lower than in the other scenarios evaluated. Taking this into account, it is reasonable to expect that this is the scenario with the lowest associated impact. However, it is not so easy to make a prediction for the rest of the scenarios, since it will be decisive to know whether the reagents used, or the electricity consumed is the factors that has the greatest impact on the LCA.

As can be observed, this inventory only includes the inputs to the system, without considering the emissions or the consumption of resources derived from the extraction of raw materials, product synthesis, energy generation, etc. However, when the available information is entered into the software, a much more comprehensive inventory is generated, consisting of a total of 1,894 records for each of the proposed scenarios. This complete inventory has not been included in this work due to its length, but it contains, among other things, atmospheric emissions and emissions to the aquatic environment. In the case of electrical energy, the generation mix in Spain in the year 2020 has been considered, based on data from REE. This mix has been specifically constructed since the data from Ecoinvent was older.

All the data used to create this inventory are either experimental (duration of the experiments and doses of reagents used) or provided by the manufacturers (electrical consumption of the different equipment). Therefore, the quality of this information has been determined as class A in all cases, which is why it has not been included in the table.

On the other hand, Table 4 presents the LCI of the pilot plant. In contrast to the previous case, the quality of these data varies, resulting in a lower level of accuracy in the evaluation below. Despite the willingness of the manufacturing company to provide data, in some instances, the obtained information has been insufficient, necessitating estimations. This is evident in the case of the aluminium frame, where the weight has been calculated based on the length of the profiles (measured

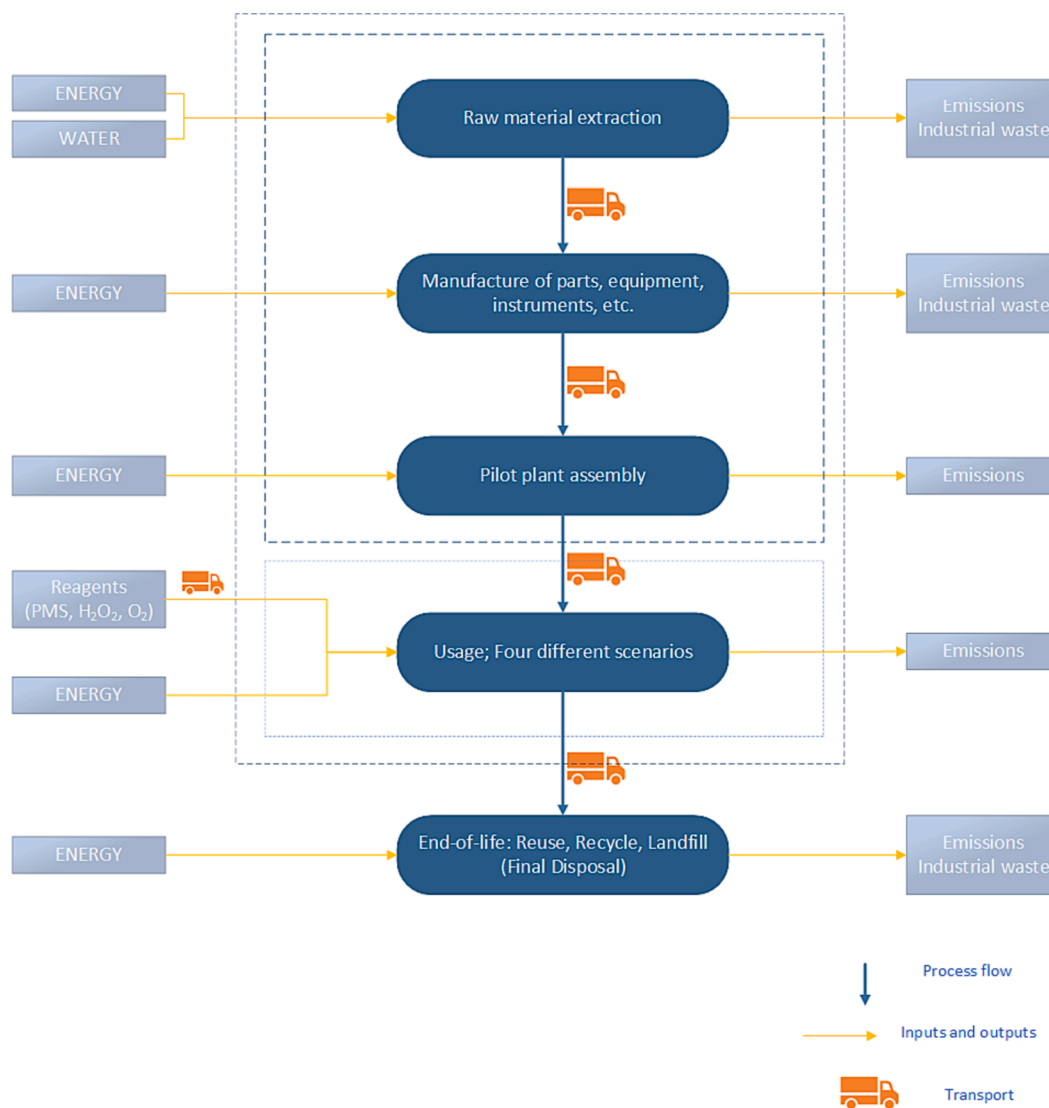


Fig. 1. System boundaries and LCA scope.

Table 2
Quality levels for data classification.

Data quality	Origin of the data
A	Data from official entities, manufacturers, suppliers, or experts
B	Data of similar assets in terms of material and mass
C	Data obtained from approximate volumes and densities
D	Data obtained by means of proximate proportions of each component over total weight

Table 3
LCI: reagents and energy used in the different scenarios.

Scenario	Reagents (kg)		Energy, electricity (kWh)			
	PMS	H ₂ O ₂	Pump	UVA light	Ozonator	Total
1	$1.61 \cdot 10^{-2}$	0	0.68	$1.44 \cdot 10^{-2}$	0	0.69
2	$5.39 \cdot 10^{-3}$	0	0.15	0	0.2	0.35
3	$1.61 \cdot 10^{-2}$	$1.79 \cdot 10^{-3}$	0.45	$9.60 \cdot 10^{-3}$	0	0.46
4	$5.39 \cdot 10^{-3}$	$1.79 \cdot 10^{-3}$	0.90	$1.92 \cdot 10^{-2}$	0	0.92

on-site) and their density. Additionally, the table includes the manufacturing process selected from the EcoInvent database for each of the components. This information is crucial to determining, for instance, the energy required during the molding of the plastic elements or the extrusion of the aluminium profiles.

Once again, upon entering this information into the calculation software, an expanded LCI was obtained, comprising nearly 2,000 entries. This comprehensive LCI encompasses the inputs and outputs of the system, ranging from the extraction of raw materials to the assembly of the pilot plant.

3.2. Life cycle impact assessment (LCIA): Comparative LCA of the proposed treatment scenarios

Once the inventory has been completed and the data have been entered into the software, it is possible to calculate the impact associated with each of the evaluated categories according to the selected LCIA method (Product Environmental Footprint). However, it is important to note that each of the impact categories evaluated has different units, making it inappropriate to compare them directly without first processing the results. This processing has been carried out for the four scenarios, and the obtained results are presented in Fig. 2. Additionally, Table 5 illustrates the characterization results for each scenario.

Table 4
LCI: pilot plant components.

Asset	Component	Description	Value	EcoInvent Process	Data Quality
Feeding tank	–	Linear polyethylene with anti-UV additive	1.5 kg	P1	A
Extractor	Propeller	Acrylonitrile butadiene styrene	0.75 kg	P1	D
	Casing	Polypropylene	0.75 kg	P2	
Feed pump	Impeller	Stainless steel	2.26 kg	P3	B
	Casing	Cast iron	9.04 kg	P4	
Safety filter	Head	Polypropylene	0.59 kg	P2	C
	Vessel	Styrene acrylonitrile	0.59 kg	P2	
	Gasket	Nitrile rubber	0.02 kg	P2	
Heat Exchanger	Shell Tubes	Titanium	1.70 kg	P2	A
Ozonator	Ozone generation core	Stainless steel	1 kg	P3	E
	Venturi tube	Polyvinylidene fluoride	0.50 kg	P2	
Photochemical unit	Electronic control and touch screen	LCD screen	0.30 kg	P5	C
	Ducting	Polypropylene	0.20 kg	P2	
	Casing	Acrylonitrile butadiene styrene	10 kg	P1	
	Others	Stainless steel	2 kg	P3	
	Protective casing	Polylactic filament	0.81 kg	P1	
	Reactor	Borosilicate glass	0.91 kg	P6	
	Lamp protective glass	Borosilicate glass	0.38 kg	P6	
	Trigger strip	Aluminum	0.05 kg	P7	
	LED bulb	–	1ud	P8	
	Ventilation system casing	Polylactic filament	0.33 kg	P1	
	Glass connection	Polyethylene	0.08 kg	P2	
	Fan	Nylon	0.16 kg	P2	
Electrical cabinet	Cabinet	Fiber-reinforced polyester	22.7 kg	P1	E
		Steel	0.78 kg	P3	
		Zinc	0.38 kg	P4	
		Nylon	0.38 kg	P2	
	Control panel	LCD screen	0.30 kg	P5	
Wiring	Aluminum and steel bimetallic cable	50 m	–	E	
Frame	–	Anodized aluminum	78 kg	P7	C
Instrumentation	Level sensors, flowmeters, temperature probes, pressure gauges, pH transmitters and their indicators	Stainless steel	0.50 kg	P3	E
		Aluminium	0.50 kg	P7	
		Polyvinyl chloride	0.50 kg	P1	
		Polyvinyl chloride	4.05 kg	P1	
Piping	–	Stainless steel	1.00 kg	P3	E
Screws	–	Light commercial vehicle transportation	420 km	–	C

* EcoInvent processes: P1 - Blow moulding, P2 - Injection moulding, P3 - Metal working, average for chromium steel product manufacturing, P4 - Metal working, average for metal product manufacturing, P5 - Assembly, LCD screen, P6 - Extrusion, plastic film, P7 - Impact extrusion of aluminium, cold, tempering, P8 - Ultraviolet lamp | ultraviolet lamp production, for water disinfection.

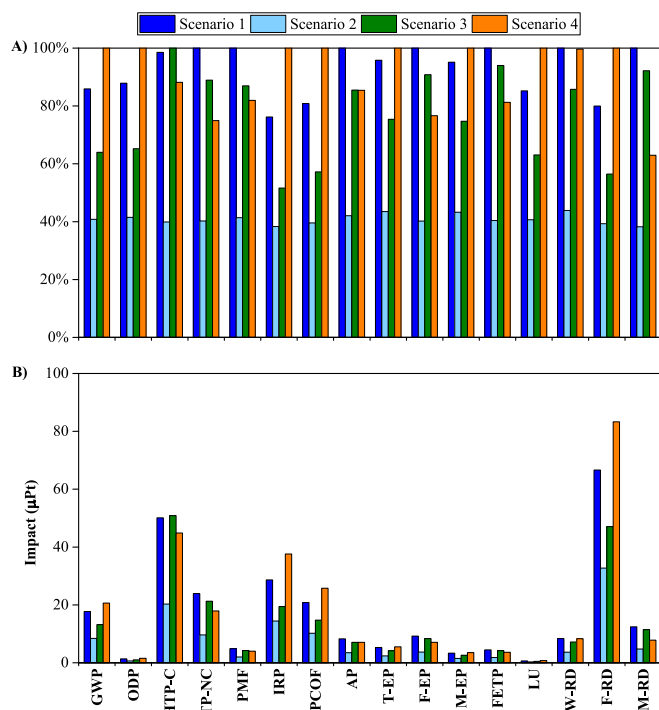


Fig. 2. Quantification of the impacts associated with each scenario: A) In terms of characterization B) Normalized values.

Table 5

LCIA results of the scenario comparison.

Environmental impact	Units	Scenario 1	Scenario 2	Scenario 3	Scenario 4
GWP	kg CO ₂ eq	1.37 10 ⁻¹	6.53 10 ⁻²	1.02 10 ⁻¹	1.60 10 ⁻¹
ODP	kg CFC11 eq	3.18 10 ⁻⁸	1.50 10 ⁻⁸	2.36 10 ⁻⁸	3.62 10 ⁻⁸
HTP-C	CTUh	1.93 10 ⁻⁹	7.82 10 ⁻¹⁰	1.96 10 ⁻⁹	1.73 10 ⁻⁹
HTP-NC	CTUh	1.14 10 ⁻⁸	4.57 10 ⁻⁹	1.01 10 ⁻⁸	8.51 10 ⁻⁹
IRP	kBq U-235 eq	1.21 10 ⁻¹	6.08 10 ⁻²	8.19 10 ⁻²	1.59 10 ⁻¹
AP	mol H ⁺ eq	4.61 10 ⁻⁴	1.94 10 ⁻⁴	3.94 10 ⁻⁴	3.93 10 ⁻⁴
FETP	CTUe	5.26 10 ⁻²	2.13 10 ⁻²	4.95 10 ⁻²	4.28 10 ⁻²
F-EP	kg P eq	2.35 10 ⁻⁵	9.45 10 ⁻⁶	2.14 10 ⁻⁵	1.80 10 ⁻⁵
M-EP	kg N eq	9.42 10 ⁻⁵	4.29 10 ⁻⁵	7.40 10 ⁻⁵	9.91 10 ⁻⁵
T-EP	mol N eq	9.34 10 ⁻⁴	4.24 10 ⁻⁴	7.35 10 ⁻⁴	9.75 10 ⁻⁴
LOP	Pt	8.57 10 ⁻¹	4.09 10 ⁻¹	6.34 10 ⁻¹	1.01 10 ⁻¹
POCP	kg NMVOC eq	8.45 10 ⁻⁴	4.14 10 ⁻⁴	5.99 10 ⁻⁴	1.05 10 ⁻³
F-RD	MJ	4.35	2.14	3.07	5.44
M-RD	kg Sb eq	7.19 10 ⁻⁷	2.75 10 ⁻⁷	6.62 10 ⁻⁷	4.53 10 ⁻⁷
PMF	disease inc.	3.10 10 ⁻⁹	1.28 10 ⁻⁹	2.69 10 ⁻⁹	2.54 10 ⁻⁹
W-RD	m ³ depriv.	9.59 10 ⁻²	4.21 10 ⁻²	8.22 10 ⁻²	9.55 10 ⁻²

Fig. 2A enables a comparison of the different scenarios by expressing the impacts for each category relative to the worst result. To achieve this, the highest value obtained in each category among the four

evaluated scenarios has been designated as 100%, with the impacts on the other scenarios expressed as a percentage of the maximum. Scenario 4 emerges as the worst environmental performer, exhibiting the highest impact values in 8 out of the 16 categories. This outcome can be attributed to its utilization of a lower concentration of reagents compared to other scenarios, leading to a higher consumption of electrical energy due to the longer treatment duration. Consequently, certain categories where Scenario 4 exhibits the greatest detriment include global warming and the use of fossil resources. In contrast, scenario 1 demonstrates the highest values in 7 categories. However, scenario 3 surpasses the others in terms of its impact on the category concerning carcinogenic potential in humans (HTP-C), which can be attributed to its use of the greatest number of reagents (1.5 mM PMS and H₂O₂). As anticipated, scenario 2 demonstrated the lowest associated impacts in all categories, as it utilizes the lowest dose of reagents while still achieving the same disinfection outcome as the other proposed treatments, thereby requiring the least amount of energy.

The same results are depicted in Fig. 2B, albeit this time they have been normalized using the normalization values proposed in the methodology. While normalization is an optional step in conducting an LCA, it resolves the issue of different units and scales by generating dimensionless values that facilitate category comparisons [34]. For the normalization process, the normalization factors specified in the EICV EF methodology have been applied. In accordance with the standard outlined in Annex B of the Product Environmental Footprint Category Rules Guidance [35], the EICV results are divided by the per capita impact for the category at a global [36]. Based on the outcomes, it can be affirmed that the categories with the most significant impacts in all scenarios are the depletion of fossil resources, which is associated with energy generation, and human toxicity (carcinogenic effects).

While the high score obtained in relation to human health may appear noteworthy, it has been observed that other authors have reported similar findings when assessing the environmental impacts of different AOPs. For instance, Chatzisyseon et al. [21] identified a significant impact in this category when evaluating various UV/TiO₂-based treatments. In their work, this contribution was primarily attributed to atmospheric emissions resulting from energy generation, given the high reliance on fossil resources, particularly coal, in the country under investigation (Greece). Similarly, Maniakova et al. [37] examined the environmental impacts of removing emerging contaminants using a sequential treatment system based on sunlight/H₂O₂ and solar photo-Fenton with EDDS in an RPR-type reactor, comparing it to conventional ozone treatment. They concluded that energy consumption plays a crucial role in the sustainability of the treatment system. Consequently, the sequential system based on AOPs was deemed more sustainable, as it utilizes solar radiation to facilitate radical generation reactions, while the ozone process requires electrical energy for ozone generation. This aligns with the conclusion drawn in the present investigation when comparing SR-AOPs at the pilot plant scale. Furthermore, Sbardella et al. [27] also identified human toxicity as one of the top 5 relevant categories in their study, where they evaluated two SR-AOPs that exhibited high human toxicity. However, in their case, they determined that the increase in this impact was not solely attributed to electricity generation but also to the detrimental effect of the persulfate synthesis process.

The calculation of a single impact score confirms what has been previously mentioned, indicating that scenario 2 exhibits the lowest associated impacts with a weighted score of 7.72 μPt. On the opposite end, scenario 4 obtains a score of 18.5 μPt (Fig. 3). This aggregated value serves as a singular indicator. It is important to note that this value may not be entirely reliable as it can vary depending on the specific LCIA method employed. Each method utilizes its own normalization and weighting factors, assigning varying importance to different categories. Nevertheless, this single indicator allows for a comparison among the different scenarios, enabling them to be ranked according to their overall environmental impact as follows: **Scenario2 < Scenario3 <**

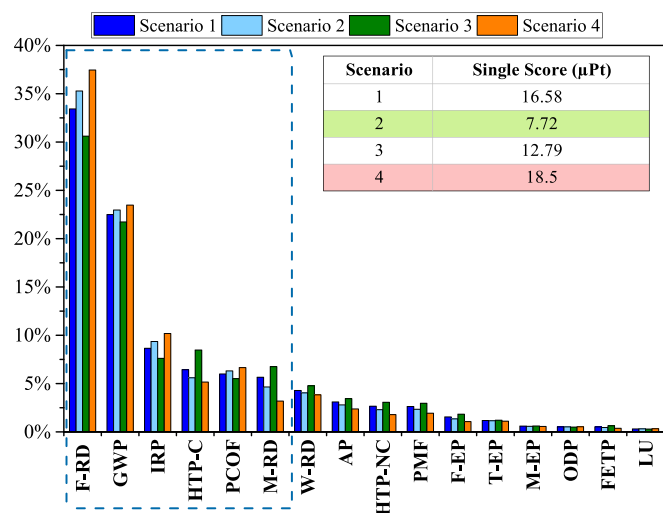


Fig. 3. Overall impact of each scenario (single score) and contribution of each of the categories.

Scenario1 < Scenario4.

As demonstrated in this paper, Arzate et al. [24] determined that a tertiary treatment based on ozonation results in fewer impacts compared to other treatments, such as photo-Fenton, primarily due to its lack of reagents requirements. In the evaluated treatment, ozone has been combined with PMS, but both the reagent dosage and required time are lower compared to alternative treatments. On the contrary, Tarpani and Azapagic [25] concluded in their research that ozone represented the worst environmental treatment among those they compared. However, it should be noted that, among the scenarios evaluated by these authors, ozonation is the only treatment that involves energy consumption.

According to the PEF methodology, the impacts whose sum contributes 80% to the overall impact are considered relevant, while the rest of the categories are considered not relevant. Fig. 3 highlights the most relevant categories in this study, which are: depletion of fossil resources (F-RD), climate change (GWP), ionizing radiation potential (IRP), human toxicity (carcinogenic effects) (HTP-C), photochemical ozone formation (PCOF) and depletion of mineral resources (M-RD). Although there are six relevant categories, it is worth noting that the first two account for more than 50% of the overall impact in all scenarios. This is due to the high score calculated for fossil resource depletion, as well as the high importance given to climate change in this method. Climate change contributes to the single score with a weighting factor of 21.06%.

Both categories are highly related to electricity consumption and, once again, it is confirmed that electricity is by far the largest contributor to the overall impact associated with the different treatments as can be seen in Table 6. In the longest treatment, represented in scenario 4, this contribution is over 90%. Although the proportion decreases for the shorter treatments and/or as the dose of reagents required increases, it always remains above 65%. On the contrary, despite the H_2O_2 concentration used in scenario 3 being the same as that of PMS, its contribution to the overall impact is negligible, as also stated by Pesqueira et al. [26].

Electricity is the most influential element in all the studies consulted in the literature [13,38]. However, this contribution is not always due to the use phase, as some photocatalytic treatments depend on the prior

Table 6

Contribution of the elements involved in the treatment to the overall impact of each scenario.

	Scenario 1	Scenario 2	Scenario 3	Scenario 4
Electricity (%)	75.8	82.7	65.6	90.77
PMS (%)	24.2	17.3	31.3	7.23
H_2O_2 (%)	0	0	3.1	2.07

synthesis of nanoparticles. This process is associated with high electricity consumption and accounts for a significant part of the overall impact [39]. Regarding the energy consumption derived from the use of lamps, solar treatments are a terrific way to tackle this problem and reduce the impacts [13,40]. Feijoo et al. [41] compare the results obtained in their electrochemical oxidation study with the results reported by other authors for the treatment of wastewater using different advanced treatments. The conclusion reached is analogous to the one reported in this research, which states that energy consumption is the factor that most affects the final environmental impacts. In the case of photo-assisted treatments, the use of solar radiation greatly reduces the value of the GWP impact category.

Sbardella et al. [27] obtained a significantly different result in their study of the persulfate/UV-A system. They determined that PMS production accounts for 85% of the overall impact. However, this difference can be explained by the high concentration of reagent used (0.4 mM) compared to the low operation times and thus low electrical power consumption (<4 min).

Due to the immense importance of this element in determining the impact of the treatments, a sensitivity analysis has been performed to evaluate the influence of the energy mix used in power generation. The results are presented in section 3.4.

3.3. LCIA: Pilot plant life cycle

There is no uniformity in the literature regarding the inclusion of infrastructure in the analysis of treatments impacts [13]. However, several authors, especially in recent publications, do take it into account [24,38,40]. The impact of the treatment is not solely determined by the use phase but also by the impact associated with the facility where it takes place. Therefore, an LCA of the pilot plant, in which the tests were conducted (using one pilot plant as a FU in this case) was also performed. This additional analysis allowed for the calculation of a new

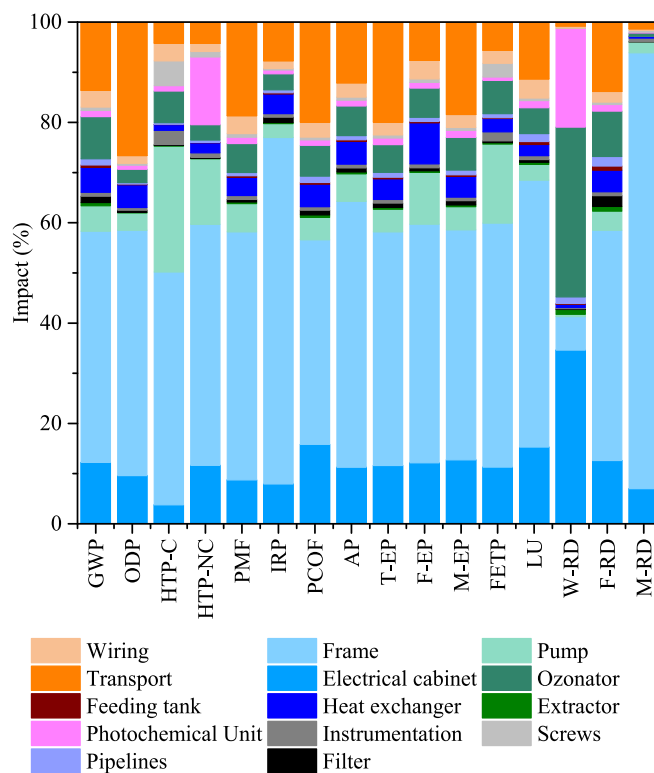


Fig. 4. LCA of the pilot plant: contribution of the main components to the different impact categories (the total length of the bar represents 100% of the impact within each category).

impact of the treatments, considering this new contribution.

The results of the LCA performed on the pilot plant are presented in Fig. 4. The contribution of the different elements that constitute the plant has been determined for each category, and the importance of the aluminium frame stands out in most of the categories. This finding is not surprising, since, as the inventory shows that the mass of the aluminium frame accounts for 45% of the total mass of the plant.

As in the previous LCA, after normalization, it is possible to determine the most relevant impact categories in terms of the global impact of the plant. In this case, the most significant category is the use of mineral resources, which represents 27.5% of the total impact. Although this category was also important in the LCA for treatments, it accounted for approximately 5% in that case. The higher value in this occasion is primarily due to the extraction of materials required for the manufacturing of the plant, primarily aluminium.

To summarize the impact of the plant amortized in each treatment, it is necessary to establish a relationship between the FU used in the two LCAs. For this purpose, it has been determined that the plant will be considered amortized over a period of 10 years. During this time, it will operate for 5 h a day, 5 days a week, for a total of 53 weeks per year. The number of treatments that can be performed in each scenario has been determined based on the time required, as specified in the FU of the comparative LCA, for the elimination of 10^5 CFU·mL⁻¹ of *Enterococcus faecalis* from 35 L of wastewater (Table 7). Furthermore, since the presence of the ozonator has been shown to have a significant impact on some of the impact categories, it has been assumed that the plant is built without ozonator for scenarios 1, 3 and 4, where it is not used. The inclusion of the ozonator is considered only in scenario 2.

When analyzing the stage of use independently, the scenario with the greatest impact varied for each category (Fig. 2), with scenario 4 (due to electricity consumption) and 1 (due to the high dose of PMS required) being the most frequently observed. However, when considering the impact associated with the plant, scenario 4 consistently emerges as the worst for all categories (Fig. 5). This is because scenario 4 has the longest duration with the estimated 10-year amortization period, allowing for fewer treatments to be carried out. As a result, a greater proportion of the overall impact is amortized in each of them.

Although these results cannot be extrapolated to a large facility where the studied processes would potentially be implemented due to the variations in configuration and materials used, there are still some conclusions that can be drawn. A shorter treatment not only leads to fewer impacts associated with energy generation but also allows a greater number of treatments within the amortization time (10 years in this case). This, in turn, results in lower overall impact of the facility associated with the FU used.

To gain a more comprehensive understanding of the environmental impact of tertiary treatment, it is insufficient to solely examine the impact of the process or installation, which inherently involves the use of resources and energy. It is also important to consider the positive impact of removing pollutants from the water and enabling its reuse. Tarpani and Azapagic [25] took into account the benefit of pollutant removal in their study, although they considered the effluent's purpose to be discharge into the environment. In their study, even though the ozone treatment from air exhibited the lowest impact among the treatments studied, the positive impact associated with the removal of CECs was overshadowed by the overall impacts of the treatment, resulting in a

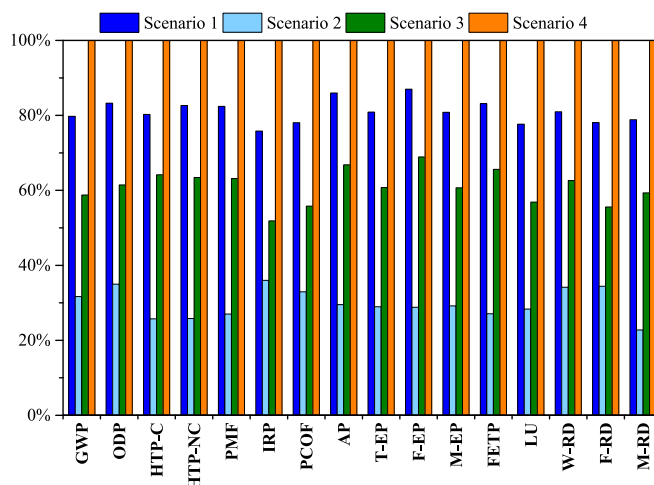


Fig. 5. Quantification of the impacts associated with each scenario (in terms of characterization) considering the impact associated with pilot plant manufacturing.

negative overall balance. A similar conclusion was reached by Risch et al. [42], who stated that the direct benefits derived from improved water quality were outweighed by the impacts stemming from energy and resource consumption. However, it should be noted that both papers primarily focused on the benefits of removing specific target pollutants. By implementing tertiary treatment, numerous other pollutants that are not being considered would also be removed, thereby increasing the positive impact of the treatment.

On the other hand, none of these studies take into consideration the potential for treated effluent to be reused, which can lead to a reduction in water resources consumption if the analysis expands its focus. In fact, in an LCA conducted by Canaj et al. [43] on wastewater treatment for crop irrigation, it was discovered that the impact associated with the application of conventional tertiary treatment followed by was lower compared to the discharge of secondary effluent. Unfortunately, a comprehensive study specifically addressing the application of AOPs has not been conducted thus far. However, it is expected that when such a study is conducted, the results obtained would be more favourable.

3.4. Sensitivity analysis: Influence of the energy mix

The results obtained in an LCA are highly sensitive to the methodology used and other factors, such as the energy mix in scenarios involving high electricity consumption [26,38]. Considering the significant contribution of energy consumption to the overall impact observed in section 3.1., a sensitivity analysis has been conducted to compare the results obtained using the energy mix of the year 2020 (considered the base case) with the results based on the national energy mix of Spain for the year 2014, as documented in the EcoInvent databases. Additionally, a third scenario called Renewable Mix (or Green Mix) has been included in the comparison. The Renewable Mix represents the target proposed in the National Integrated Energy Climate Plan (PNIEC) for 2030, where 74% of the electricity generation comes from renewable energy source. Within this percentage, 50% corresponds to wind energy generation, while 24% is contributed by photovoltaic and hydroelectric energy source.

Table 8 displays the variations observed in comparison to the base scenario when applying the new energy mix. The utilization of renewable energies results in a general reduction across all impact categories, with the most substantial decrease observed in photochemical ozone formation, reaching approximately 80% for all scenarios. Conversely, a significantly notable increase has been observed when employing the 2014 energy mix, leading to up to 638% and 645% rise in acidification potential and freshwater eutrophication potential, respectively, in

Table 7
Number of treatments performed in 10 years of pilot plant operation.

Scenario	Treatments in 10 years
1	8,833
2	39,750
3	13,250
4	6,625

Table 8

Influence of the energy mix: variation of the different impact categories with respect to the score obtained in the base case (electricity energy mix 2020).

	Scenario 1		Scenario 2		Scenario 3		Scenario 4	
	Green Mix	2014 Mix	Green Mix	2014 Mix	Green Mix	2014 Mix	Green Mix	2014 Mix
GWP	-48%	81%	-51%	87%	-43%	73%	-55%	93%
ODP	-42%	-6%	-45%	-7%	-38%	-6%	-49%	-8%
IRP	-35%	18%	-35%	18%	-34%	17%	-35%	18%
PCOF	-80%	18%	-83%	19%	-75%	17%	-86%	20%
PMF	-11%	121%	-14%	148%	-9%	93%	-18%	197%
HTP-NC	-1%	204%	-2%	257%	-1%	153%	-2%	363%
HTP-C	12%	52%	16%	66%	8%	34%	19%	78%
AP	-29%	409%	-35%	493%	-23%	319%	-45%	638%
F-EP	-5%	371%	-7%	468%	-4%	272%	-9%	645%
M-EP	-39%	278%	-44%	309%	-33%	236%	-50%	352%
T-EP	-42%	300%	-46%	335%	-35%	254%	-53%	383%
FETP	1%	96%	2%	120%	1%	68%	2%	157%
LU	-11%	107%	-12%	114%	-10%	96%	-13%	121%
W-RD	-54%	68%	-62%	78%	-42%	53%	-72%	91%
F-RD	-42%	20%	-44%	21%	-40%	19%	-45%	22%
M-RD	15%	-16%	19%	-22%	10%	-12%	31%	-34%

scenario 4. This considerable disparity is predominantly attributed to the decarbonization efforts that have been undertaken in our country in recent years.

Regardless of the energy mix employed, scenario 2 consistently emerges as the most favorable option when assessing the overall impact (Fig. 6). However, while scenario 4 clearly achieves the highest score when using the 2014 and 2030 energy mixes, scenario 1 exhibits a slightly worse performance (0.1 μ Pt) when considering an energy mix comprising renewables. This observation underscores how, as reliance on fossil resources diminishes, the significance of treatment duration (or energy consumption) decreases, and the dosage of PMS utilized (which is significantly higher in scenario 1) becomes more influential.

Foteinis *et al.* [38] conducted a sensitivity analysis to assess the influence of the electricity mix, determining that the use of renewable energies can reduce the environmental impact of AOPs based on UV radiation by up to 87%. In this study, it was observed that for scenario 3 (the most favorable among those utilizing UV-A radiation), electricity consumption is reduced by nearly 60% compared to the result obtained using the 2014 energy mix. However, the decrease observed in this study is less significant due to the fact that the energy mix used by Foteinis *et al.* [38] in Greece (2016) is even more reliant on fossil fuels than the Spanish energy mix. In Spain, renewable sources accounted for over 40% of the energy mix in 2014, whereas in Greece, only 18% of the energy came from renewable sources [44]. A similar study was conducted by Pesqueira *et al.* [26] which evaluated various European

energy mixes. Their findings indicated that higher usage of renewable energies, such as photovoltaic energy, allows for a significant decrease in impacts. Conversely, the use of energy mixes in countries highly dependent on fossil resources, such as Serbia, North Macedonia, or Montenegro, results in a higher impact from the same treatment.

4. Conclusions

In this work, the LCA tool has been utilized to quantify the environmental impacts associated with four different scaled-up and optimized SR-AOPs employed for wastewater disinfection at a pilot scale. The results obtained through LCA will play a crucial role in the decision-making process for the final implementation of these treatments at a real scale. This decision-making process takes into account not only treatment efficiency but also the corresponding environmental impacts. This study is novel as the applied treatments had not been previously documented in the literature, and furthermore, it integrates LCA as a decision-making tool in the technology scaling process. While it is acknowledged that the life cycle inventory may have certain limitations in collecting specific information from various components of the pilot plant, these limitations are not considered to be significantly influential in the final results. Moreover, it should be noted that environmental impacts can vary considerably for the same treatments based on the geographical and temporal context of their implementation.

Therefore, through a comparative LCA, it has been determined that the treatment combining PMS and O₃ is the most environmentally sustainable option, thanks to its low consumption of reagents and electricity. Similarly, it has been found that the scenario employing the PMS (0.5 mM)/H₂O₂ (1.5 mM)/UV-A system exhibits the highest associated impacts in 9 out of the 16 evaluated categories. Thus, in this instance, it can be stated that the most effective disinfection treatment is also the most sustainable. However, it is important to note that this conclusion cannot be extrapolated to other case studies, and it is recommended that the impacts of each specific case be evaluated.

Furthermore, the study found that electricity consumption plays a crucial role in assessing the environmental sustainability of a treatment, contributing between 65% and 90% to the total impact. This finding is also reflected in the impact categories, where fossil resource depletion and climate change consistently emerged as the most significant factors across all studied scenarios.

Additionally, a sensitivity analysis highlighted the significant impact of the energy mix on the overall results. By transitioning from the 2030 energy mix to one predominantly based on renewable energies, the impact of the treatments was reduced by approximately 30% to 42%. Moreover, this analysis demonstrated that by generating energy from renewable sources, the relevance of energy consumption in determining

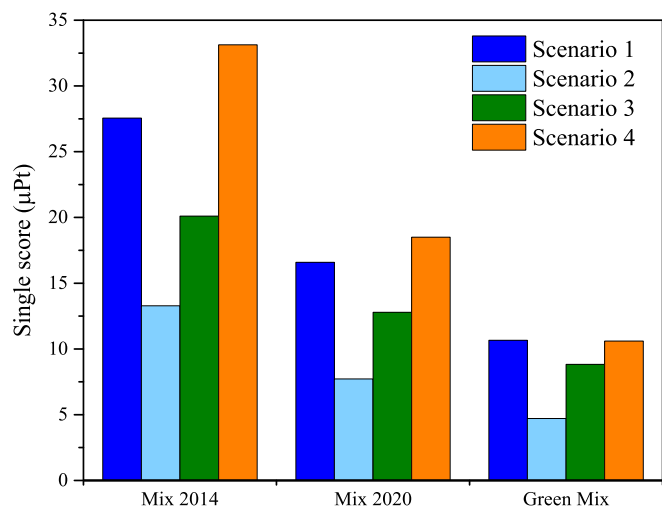


Fig. 6. Influence of the energy mix on the total impact of each scenario.

the overall impact decreased, while the importance of the reagent dosage used became more prominent.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cej.2023.145427>.

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