

Contents lists available at ScienceDirect

Sustainable Materials and Technologies



journal homepage: www.elsevier.com/locate/susmat

# Environmental life cycle assessment of UV-C LEDs vs. mercury lamps and oxidant selection for diclofenac degradation

Raffaella Pizzichetti <sup>a,b</sup>, Mario Martín-Gamboa <sup>c</sup>, Cristina Pablos <sup>a,\*</sup>, Ken Reynolds <sup>b</sup>, Simon Stanley <sup>b</sup>, Javier Dufour <sup>c,d</sup>, Javier Marugán <sup>a,e</sup>

<sup>a</sup> Department of Chemical and Environmental Technology, ESCET, Universidad Rey Juan Carlos, C/Tulipán s/n, 28933 Móstoles, Madrid, Spain

<sup>b</sup> ProPhotonix IRL LTD, 3020 Euro Business Park, Little Island, Cork T45 X211, Ireland

<sup>c</sup> Department of Chemical, Energy and Mechanical Technology, ESCET, Universidad Rey Juan Carlos, C/Tulipán s/n, 28933 Móstoles, Madrid, Spain

<sup>d</sup> IMDEA Energy, Systems Analysis Unit, 28935 Móstoles, Spain

<sup>e</sup> Instituto de Tecnologías para la Sostenibilidad, Universidad Rey Juan Carlos, C/ Tulipán s/n, 28933 Mostoles, Madrid, Spain

#### ARTICLE INFO

Keywords: Advanced oxidation processes (AOPs) Environmental footprint (EF) Light-emitting diodes (LEDs) UV-based water treatment Free chlorine (FC) Hydrogen peroxide

## ABSTRACT

This study is the first environmental comparison between a UV-C LED lamp (emitting at 265 nm) and mercury lamps employed in a lab-scale photoreactor for water treatment purification purposes, using the removal of diclofenac as a case study. Ex-ante life cycle assessment (LCA) methodology was used as a robust method to identify hotspots and recommendations at the early stage of the UV-C LEDs technology. The functional unit was defined as "the treatment of 1 L of polluted water with 20 mg  $L^{-1}$  of diclofenac to achieve a 90% removal of the contaminant", while the system boundaries include the production and the operation of the photoreactors, following a cradle-to-gate approach. Several scenarios were explored, and overall, the UV-C LED lamp shows a promising environmental performance, with less or similar potential impacts than the mercury lamps in the 16 categories selected from the Environmental Footprint (EF) method. In particular, it reveals less impact in "human toxicity non-cancer" and "resource use minerals and metals" and presents electricity as the main source of impact. Given the higher efficacy of the UV-driven advanced oxidation processes compared to the UV irradiation alone, and since no studies have previously been conducted on the sustainability of free chlorine (FC) as an oxidant in water treatment, a comparison between UV-C, UV-C/H<sub>2</sub>O<sub>2</sub>, and UV-C/FC while employing the 265 nm UV-C LED lamp was also assessed. UV-C/H<sub>2</sub>O<sub>2</sub> was more sustainable than UV-C/FC for the same treatment time, but both led to an overall impact reduction of 35% and 30%, respectively. To increase sustainability, employing cleaner energy sources such as photovoltaic or wind energy also resulted in an 80% and 93% reduction in the "climate change" category. Overall, this study demonstrates that using UV-C LEDs and the selected oxidants for water purification is beneficial and encourages the scale-up of the system.

## 1. Introduction

Over the last decade, light-emitting diodes (LEDs) have gained significant attention as an alternative mercury-free source [1]. In particular, LEDs in the UV-C (200-280 nm) range, other than being effective for water disinfection [2,3], also proven to be effective in the degradation of many contaminants of emerging concern (CECs) [4–6]. Replacing the conventional low-pressure (LP) mercury lamps with innovative UV- C LEDs could lead to interesting breakthroughs in water treatments. The major advantages of LEDs are that they have tuneable wavelength [7], instant on-off [8], and adjustable intensity [9]. In addition, their small size allows for high design flexibility [10], and the perspective of their exponential growth, similar to the LEDs in the visible and UV-A range, makes them very attractive for the design of any water treatment [11]. Nonetheless, UV-C LEDs are still a relatively new technology with low efficiency compared to conventional mercury lamps [12], hence the

\* Corresponding author.

E-mail address: cristina.pablos@urjc.es (C. Pablos).

https://doi.org/10.1016/j.susmat.2024.e01002

Received 9 February 2024; Received in revised form 6 May 2024; Accepted 31 May 2024 Available online 3 June 2024

2214-9937/© 2024 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

Abbreviations: AOP, Advanced oxidation process; CEC, Contaminant of emerging concern; DCF, Diclofenac; DWTP, Drinking water treatment plant; EEO, Electrical energy per order; EF, Environmental footprint; FC, Free chlorine; FU, Functional unit; ISO, International standardisation organisation; LCA, Life cycle assessment; LCI, Life cycle inventory; LCIA, Life cycle impact assessment; LED, Light-emitting diode; LP, Low-pressure; UV, Ultraviolet; WWTP, Wastewater treatment plant.

interest in evaluating the current potential impacts of the two systems.

The environmental life cycle assessment (LCA), thanks to its international recognition and widespread use, is the preferred method to explore and compare the potential impacts of these systems [13]. In particular, the ex-ante LCA is gaining more and more attention since it investigates the effects of emerging technologies in the early development stages, at the experimental proof of concept, or on validation in a lab or pilot plants [14]. It consists of exploring and evaluating a range of possible scenarios that define the space in which the emerging technology may operate, often in comparison to the conventional technology, with the main objective of recognising hotspots, trade-offs, and opportunities to reduce potential impact.

In the context of UV-based water treatment, previous LCA studies mainly focused on lab or pilot scale applications, primarily employing LP mercury lamps [15-18] and UV-A LEDs [17,19]. Therefore, to the best of the authors' knowledge, this is the first comparative study between a UV-C LED lamp (emitting at 265 nm) and conventional LP mercury lamps employed in a lab-scale reactor for water remediation purposes. Furthermore, the use of advanced oxidation processes (AOPs) coupled with UV treatment was also investigated, as it is a common strategy to decrease the treatment time and increase water quality [20]. AOPs exploit the use of chemical oxidants to generate highly reactive radical species, which can effectively degrade organic pollutants besides fostering the disinfection treatment. However, evaluating the sustainability of the oxidant is essential as its production might account for the majority of the environmental footprint of the treatment, as in the case of sulfate radicals-based AOPs [16,21]. Even if the process is effective, from the environmental perspective, the use of more sustainable options, like  $H_2O_2$ , is recommended [16]. On the other hand, studies on the environmental sustainability of free chlorine (FC)-based AOPs have not been reported yet, despite FC is a widely available disinfection agent [22,23]. Therefore, the second part of this study investigates the relative environmental impact between UV alone, UV/H2O2 and UV/FC treatments when employing a UV-C LED lamp emitting at 265 nm. Finally, as many studies address electricity as the main hotspot for UV-based technologies [24,25], a scenario using cleaner energy sources was evaluated to explore possible improvements.

Among the CECs, the removal of diclofenac (DCF), a nonsteroidal anti-inflammatory drug commonly used for pain relief, was used as a case study for the comparisons since it was previously examined by the same research group [9]. DCF has captured significant attention for its known toxicity towards wildlife and its potential consequences on aquatic ecosystems. Indeed, it has been detected up to  $\mu$ g L<sup>-1</sup> in water bodies [26–28], and it was introduced in the first *watch list* established by the European Commission [29].

The results of this study are organised into three main sections: the comparison between the two photoreactors (UV-C LED lamp vs. LP mercury lamp) with sensitivity analysis on the key parameters of the system, the comparison between UV alone,  $UV/H_2O_2$  and UV/FC by employing the UV-C LED lamp, and finally the comparison between the electricity mix with the use of cleaner energy sources. The results are especially valuable in identifying the growing UV-C LED technology's weaknesses and strengths, however, this study only gives recommendations at the deployment level since many factors are involved during the scale-up of the laboratory system.

#### 2. Materials and methods

#### 2.1. Description of the system

Two similar photoreactors were compared. The first photoreactor included a UV-C LEDs system emitting at 265 nm, where the UV-C LED lamp was a COBRA Clean FX1 device manufactured by ProPhotonix. On the other hand, the second photoreactor contained two mercury lamps (6 W) purchased by OSRAM emitting at 254 nm. The two photoreactors had the same laboratory equipment and only differed in the power input, the energy emitted by the lamps at the same distance from the quartz tube, and the working wavelength. It is also important to note that for the mercury lamp photoreactor, the height irradiated was 160 mm compared to the 108 mm irradiated from the LED lamp, given the different geometries of the two devices, while the quartz tube had the same diameter of 20 mm. The irradiance in both cases was evaluated experimentally through chemical ferrioxalate actinometry following the protocol described in the literature [30], which resulted in 220 W m<sup>-2</sup> for the UV-C LED lamp and 150.70 W m<sup>-2</sup> for the two mercury lamps employed in the system. In the case of the UV-C LED lamp, no preheating time was needed, whereas the mercury lamp must first heat up for 15 min as per standard procedures. The system then operates by turning on the centrifugal pump and the magnetic stirrer. The two photoreactors are schematised in Fig. S1 of the Supplementary information.

# 2.2. Life cycle assessment

The methodology standardised by the International Standardisation Organisation (ISO) was followed for the LCA study. The method involves four main interrelated stages, defined by ISO 14040 and 14044 [13,31]. The first phase is "Goal and scope", where the functional unit (FU) and the system boundaries are defined, as well as the reasons and limitations for carrying out the study. The second phase is the "Life cycle inventory (LCI)", which consists of the data collection of the inputs and outputs concerning raw material and energy, products and co-products, waste, emissions, and other environmental aspects within the system boundaries (foreground and background processes). The third phase is the "Life cycle impact assessment (LCIA)", which involves the quantification and classification of the substances of the inventory into impact categories and common units to allow the comparison. The fourth and final stage of the LCA is "Interpretation", which involves summarising and discussing the results of the analysis.

#### 2.2.1. Goal and scope

Ex-ante environmental LCA was performed to compare the innovative UV-C LED lamp to the conventional LP mercury lamp working in a small-scale photoreactor for water treatment. While UV-C LED lamps are a new technology with a relatively low efficiency, mercury lamps are a mature technology, but the presence of mercury is highly concerning. The removal of DCF, previously studied by the same research group [9], was selected as the function of both systems to enable the comparison. Furthermore, this study also aims to evaluate the impact of the oxidant ( $H_2O_2$  vs. FC) added to the UV-C LED process. This study was conducted at a lab scale due to the challenges and uncertainties involved in modelling the system at pilot or large-scale applications. The results and recommendations may vary at full scale, nonetheless, the ex-ante LCA provides valuable insights to ensure responsible and sustainable technology growth.

The FU was defined considering the objective of the investigation, "the treatment of 1 L of polluted water with 20 mg L<sup>-1</sup> of diclofenac to achieve a 90% removal of the contaminant". The FU provides the reference unit, and 1 L of polluted water was selected, given that the final aim is water treatment for final end-user consumption. Furthermore, the removal of 90% of a contaminant is a standard approach [17].

The system boundaries for each process were also determined and shown in Fig. 1. The study takes into consideration the cradle-to-gate approach, similar to previous studies [15,18,19]. The system boundaries included the production of the two photoreactors (the lamps and the lab equipment), including resources, energy, and transport of the main materials, the emission to air, water, and soil during their production, and the energy needed during the operation of both systems. In contrast, the end-of-life of the photoreactors was omitted due to the uncertainty regarding the disposal of the lamps [32].



Fig. 1. Scheme of the system boundaries for the two photoreactors.

# 2.2.2. Life cycle inventory

The material and process inputs for 1 unit of UV-C LED lamp, 1 unit of LP mercury lamp of 6 W, and the lab equipment employed in the two photoreactors are shown in Table 1. The UV-C LED lamp materials were directly quantified by opening and dissecting the lamp. In contrast, the material inputs for the mercury lamp were estimated from the ecoinvent database [33]. In this regard, a few modifications were applied to consider the manufacturing processes for aluminium, plastic, and steel and avoid using primary materials such as "Aluminium, primary, ingot". Furthermore, the amounts were remodelled for the mercury lamp system under study by considering a mass proportion between the ecoinvent amounts (380 g) and the lamp employed of 20 g. A ballast was added to each lamp to provide the correct starting and operating voltage and current, as recommended by the lamp manufacturer. Also, in agreement with the ecoinvent database [33], silica sand was assumed to be equivalent to quartz in both lamps' inventories.

To compile the inventory the ecoinvent v3.8 was used as a secondary data source [33], and the cut-off options for the unit process were selected. The specific input data were collected by selecting the global market {GLO} for the material flows, which represents the consumption mix of the product and accounts for the trade between producers and consumers. The global situation is considered to measure the average distances, modes of transport required, and the product losses occurring during transport, loading, and unloading. However, the European market {RER} was selected for the manufacturing process, assuming that the production occurs in Europe once the material is available. Also, the electricity employed was selected for a small-scale application (low voltage) from the European market. The selection was made to create an average supply chain at the European level and extrapolate conclusions for all European countries.

Following, each part's lifetime was defined. Both the LED lamp and the mercury lamp photoreactors were considered to have a life span of 8,000 h. The former experimentally showed a lifetime between 6,000 h to reach 80% and 10,000 h to reach 60% of the initial intensity; therefore, an average value was taken. While, the manufacturer data was taken for the mercury lamp. Regarding the lab equipment, 15 years, corresponding to the lowest lifetime expected for electromechanical equipment [16,18], were considered for the magnetic stirrer, the magnetic bar, the centrifugal pump, and the aluminium support base. 3 years were considered for the quartz tube and the plastic components since

they are more sensitive to breakage, and the contaminant could affect the plastic.

While Table 1 represents the construction phase, the operational phase is considered through the electricity demand of the UV source, the centrifugal pump, the magnetic stirrer, and the use of the two photo-reactors. The operational phase depends on the treatment time and, therefore, the system's efficiency. Data obtained from the work published by the same research group showed that the 265 nm LED lamp under only photolysis reached 90% removal of DCF after 29.8 min of the operation time [9], which is the time considered for the energy costs associated with the system.

To measure the 90% removal of DCF through the mercury lamp system, the UV fluence-based kinetic constant at 254 nm was evaluated from the 265 nm results ( $k'_{265} = 1.73 \cdot 10^4 \text{ J}^{-1} \text{ m}^2$ ) according to Eq. (1) [34].

$$A \xrightarrow{\mu\nu} B \qquad r_{A,\lambda} = \Phi_{A,\lambda} \cdot e^a_{A,\lambda} = \Phi_{A,\lambda} \cdot \varepsilon_{A,\lambda} \cdot [A] \cdot G_\lambda = k_{A,\lambda} \cdot [A] \cdot G_\lambda \qquad (1)$$

Considering the molar absorption coefficient ( $\varepsilon$ ) at 254 nm and assuming the same quantum yield ( $\Phi$ ) at the two wavelengths, the UV fluence-based kinetic constant at 254,  $k'_{254}$ , was found to be  $1.10 \cdot 10^4$  J<sup>-1</sup> m<sup>2</sup>. Afterwards, following Eq. (2), the operation time of 46.0 min was calculated, taking into account the mercury lamp irradiance of 150.70 W m<sup>-2</sup>, the pseudo-first order degradation, and the active volume over the total volume.

$$ln\left(\frac{C}{C_0}\right) = -k \cdot t \cdot \frac{V_R}{V_T} \tag{2}$$

Where  $V_T$  was 1 L in the two cases, while  $V_R$  of the mercury lamp photoreactor was 0.050 L against the 0.034 L of the LED photoreactor, given the longer size of the mercury lamp compared to the LED lamp.

Regarding the study of the oxidant, the operation times considered were also taken from the experimental results [9], where 90% of the DCF removal was achieved at 19.8 and 20.4 min for UV-C/H<sub>2</sub>O<sub>2</sub> and UV-C/FC, respectively. The higher efficiency is explained by the concurrent attack of the free radical in addition to the direct photolysis of DCF, HO<sup>•</sup> during the UV-C/H<sub>2</sub>O<sub>2</sub> and mainly HO<sup>•</sup>, Cl<sup>•</sup> during the UV-C/FC treatment. In the authors' previous work [9], the detailed mechanisms with the main reactions involved are rigorously described.

Within SimaPro modelling, the corresponding ecoinvent datasets of

#### Table 1

Raw materials and processes input for 1 unit of UV-C LED lamp, 1 unit of LP mercury lamp (ML) 6 W (20 g), and 1 unit of lab equipment, which is equal for the two photoreactors.

Component	Materials and processes	ecoinvent dataset selected	Amount
	16 LEDs	Light emitting diode {GLO}  market for  Cut-off, U	0.31 g
	Heatsink	Aluminium alloy, AlMg3 {GLO}  market for   Cut-off, U	127.59 g
	Lamp case	Aluminium alloy, AlMg3 {GLO}  market for  Cut-off, U	130.70 g
	Screws/spring	Aluminium alloy, AlMg3 {GLO}  market for  Cut-off, U	5.78 g
	Aluminium	Metal working, average for aluminium product	264.07
1 UV-C LED lamp	processing	manufacturing {RER}	g
-	Quartz glasses	Silica sand {GLO}  market for	5.42 g
	Fans	Fan, for power supply unit, desktop computer {GLO}	23.47 g
		market for   Cut-off, U	
	PLC-	supply unit, desktop computer,	00.00
	logic controller	Pb free {GLO}  market for   Cut- off. U	20.00 g
		Aluminium, wrought alloy	1 50
	Aluminium parts	{GLO}  market for   Cut-off, U Metal working, average for	1.72 g
		aluminium product	1 72 a
		manufacturing {RER}	1.725
	Mercury	Mercury {GLO}  market for	0 02 g
	Mercury	Cut-off, U Polyethylene high density	0.02 8
115	Plastics parts	granulate {GLO}  market for   Cut-off, U	0.10 g
1 LP mercury lamp 6 W (20 g)	Plastics parts	Polypropylene, granulate {GLO}  market for   Cut-off, U	2.40 g
	Plastic processing	Injection moulding {RER}  processing  Cut-off, U	2.50 g
	Glass	Sanitary ceramics {GLO}  market for   Cut-off, U	6.62 g
	Lamp quartz tube	Silica sand {GLO}  market for   Cut-off, U	8.86 g
	Steel parts	Steel, unalloyed {GLO}  market for   Cut-off, U	0.23 g
		Metal working, average for steel product manufacturing {RER}	0.23 g
		processing   Cut-off, U Electronics, for control units	
	Ballasts	{GLO}  market for  Cut-off, U	55.00 g
	UV Quartz tubes	Silica sand {GLO}  market for   Cut-off, U	130.00 g
		Polyvinylchloride, emulsion polymerised {GLO}  market for	1.00 kg
		Cut-off, U	0
Lab Equip.	PVC tube systems	Extrusion, plastic pipes {RER}  extrusion, plastic pipes   Cut-off,	1.00 kg
		Polyethylene, low density,	200.00
	Plastic container	granulate {GLO}  market for   Cut-off, U	g
		Blow moulding {RER}  blow moulding   Cut-off, U	200.00 g
	Magnetic stirrer	Permanent magnet, for electric motor {GLO}  production   Cut-	0.41 kg
		on, U Steel, low-alloyed {GLO}	745 o
		market for   Cut-off, U Metal working average for steel	, . <del>т</del> ,
	Magnetic bar	product manufacturing {RER}	7.45 g
	Contrifue -1	processing   Cut-off, U Pump, 40 W {GLO}  market for	1
	Centrifugal pump	Cut-off, U	тр

Table 1 (continued)

	,		
Component	Materials and processes	ecoinvent dataset selected	Amount
	Aluminium support base	Aluminium, wrought alloy {GLO}  market for   Cut-off, U Metal working, average for aluminium product manufacturing {RER}  processing   Cut-off, U	2.00 kg 2.00 kg

the oxidants were added to the UV-C LEDs treatment inventory in the amount needed for a final concentration in solution of 20 mg  $L^{-1}$ .

The electrical energy consumption of the two UV photoreactors and for the two UV-C LEDs based AOPs was consequently calculated from the definition of the electrical energy per order (EEO), which is the electric energy in kWh required to reduce the concentration of a contaminant by one order of magnitude (90% removal) in 1 m<sup>3</sup> of water, and it is measured in batch operations according to Eq. (3) [35,36].

$$EEO = \frac{P \cdot t}{V \cdot \log\left(\frac{C_0}{C}\right)} \tag{3}$$

Where *P* is the rated power or energy input of the lamp system, and *V* is the volume of water treated in the time *t*. In this case,  $\log(C_0/C) = 1$  (one order of the pollutant is degraded) and the total volume of 1 L was considered according to the FU.

Table 2 summarises the electricity and equipment required in the operation phase for the two photoreactors and for the two UV-C LEDs based AOPs. The centrifugal pump and the magnetic stirrer were the same for the two photoreactors, and the power inputs were taken from the manufacturer data, respectively 51 and 90 W. However, only half power was considered for the latter since the stirring was used approximately at half of the maximum rpm allowed. For the mercury lamp system, the time the lamps need to warm up (15 min as per standard procedures) was also taken into account.

#### 2.2.3. Life cycle impact assessment

The Environmental Footprint (EF) 3.0 method was employed as it is the reference method proposed by the European Commission [37], with the scope of bringing together different methods under one. For instance, regarding toxicity, the EF method includes the USEtox model, which calculates the impacts of chemicals on ecosystems and human health [38]. The EF method assesses the environmental impacts through 16 midpoint impact categories; indeed, the midpoint level is generally considered to be more accurate than the endpoint level [39]. All the 16 impact categories covered in the EF method were evaluated: climate change, ozone depletion, ionising radiation, photochemical ozone formation, particulate matter, human toxicity cancer and non-cancer effects, acidification, eutrophication freshwater, marine, and terrestrial, ecotoxicity freshwater, land use, water use, resource use fossils, and resource use minerals and metals [40]. SimaPro 9.4 was employed to compile the inventory data referred to the FU and solved through the EF method to quantify the LCA results [41].

#### 2.2.4. Description of sensitivity analysis

A sensitivity analysis was conducted to evaluate the robustness of the results by examining the impact of variations in key parameters or assumptions on the outcomes, thereby enhancing the reliability of these findings.

Within the comparison between the two photoreactors in Section 3.1, three scenarios were evaluated: one base case and two others. The base case is influenced by the different characteristics of the two lamps. The LED lamp has higher irradiance, while the mercury lamps have a larger active volume, reducing the circulation time. Nevertheless, the

#### Table 2

LCI of the operational phase of the two photoreactors and the two UV-C LED oxidation processes.

Treatment	Input	ecoinvent dataset selected	Amount
UV-C LED lamp	Electricity for the lamp Electricity for the centrifugal pump Electricity for the magnetic stirrer	Electricity, low voltage {RER}  market group for  Cut-off, U*	17.95 Wh 25.33 Wh 22.35 Wh
	UV-C LED lamp Quartz tube, plastic tubes, and plastic container Magnetic stirrer, magnetic bar, centrifugal pump, and aluminium support base.	Own model (Table 1)	$6.2 \cdot 10^{-5}$ p $1.9 \cdot 10^{-5}$ p $3.8 \cdot 10^{-6}$ p
Mercury lamp	Electricity for the lamp Electricity for the centrifugal pump Electricity for the magnetic stirrer	Electricity, low voltage {RER}  market group for  Cut-off, U*	12.19 Wh 39.06 Wh 34.47 Wh
	Mercury lamps Quartz tube, plastic tubes, and plastic container Magnetic stirrer, magnetic bar, centrifugal pump, and aluminium support base.	Own model (Table 1)	$2.5 \cdot 10^{-4}$ p $2.9 \cdot 10^{-5}$ p $5.8 \cdot 10^{-6}$ p
UV-C LED lamp / H <sub>2</sub> O <sub>2</sub>	Oxidant addition Electricity for the lamp Electricity for the centrifugal pump Electricity for the magnetic stirrer	Hydrogen peroxide, without water, in 50% solution state {RER}  market for   Cut-off, U Electricity, low voltage {RER}  market group for  Cut-off, U	40 mg 11.73 Wh 16.56 Wh 14.61 Wh
	UV-C LED lamp Quartz tube, plastic tubes and plastic container Magnetic stirrer, magnetic bar, centrifugal pump, and aluminium support base.	Own model (Table 1)	$4.1 \cdot 10^{-5}$ p 1.2 \cdot 10^{-5} p 2.5 \cdot 10^{-6} p
UV-C LED lamp / FC_	Oxidant addition Electricity for the lamp Electricity for the centrifugal pump Electricity for the magnetic stirrer	Sodium hypochlorite, without water, in 15% solution state {RER}  market for   Cut-off, U Electricity, low voltage {RER}  market group for  Cut-off, U	133.33 mg 12.16 Wh 17.15 Wh 15.13 Wh 4.2.10 <sup>-5</sup>
	UV-C LED lamp Quartz tube, plastic tubes and plastic container Magnetic stirrer, magnetic bar, centrifugal pump, and	Own model (Table 1)	p 1.3·10 <sup>-5</sup> p 2.6·10 <sup>-6</sup> p

Table 2 (continued)

Treatment	Input	ecoinvent dataset selected	Amount
	aluminium support base.		

LED's wavelength at 265 nm is more effective than the LP mercury lamp's 254 nm, resulting in overall more efficient treatment and shorter treatment times. While the LED lamp itself is more energy-intensive than the mercury lamp, the total energy consumption is determined by the entire system, including the pump and the magnetic stirrer; therefore, the total treatment time highly affects the photoreactor impacts. In the first alternative scenario, the LED lamp's irradiance is adjusted to the mercury lamps' irradiance, to have similar treatment times. This allows a more direct comparison of the UV source's impact, independent of other auxiliary components' energy consumption. The second alternative scenario explores the potential future pathway of UV-C LEDs experiencing an increase in efficiency. By analysing this scenario, the potential impacts of technological advancements on the overall performance of the photoreactors are highlighted.

Moreover, additional alternative scenarios, which imply reducing treatment time through the use of oxidants and adopting cleaner energy sources for electricity consumption, were evaluated in Sections 3.2 and 3.3 to explore the operating range of the UV-C LED lamp.

# 3. Results and discussion

The following results are divided into three sections relative to the comparison between the two photoreactors (UV-C LED lamp vs. LP mercury lamps), the comparison between the oxidants ( $H_2O_2$  and FC) by employing the UV-C LED lamp, and finally, the use of cleaner sources instead of the European energy mix.

## 3.1. UV-C LEDs photoreactor vs. LP mercury lamps photoreactor

Three scenarios were evaluated as a result of the sensitivity analysis carried out by varying system key parameters to help identify the main environmental benefits and hotspots of the UV-C LEDs compared to the mercury lamps reactor. Appendix B of the Supplementary Information reports the absolute value for each category in all cases. Fig. 2 shows the relative impacts of the two systems in the base case for the 16 midpoint categories.

From Fig. 2, it is noticeable that in these conditions the LED system outperformed in all potential environmental impact categories, having around 25% less impact in almost all categories. The exceptions were in "human toxicity non-cancer" and "resource use minerals and metals", where the impacts were even lower, 38% and 47%, respectively. Therefore, even if the LED lamp consumed 17.95 Wh compared to the mercury lamp's 12.19 Wh as detailed in Table 2; on the whole, the total energy consumption, including the pump and the magnetic stirrer, was lower (65.63 Wh vs 85.72 Wh) due to shorter treatment times.

Overall, these results are very promising for scaling this type of system, but the uncertainty associated with these conclusions should also be recognised because the stirrer and the pump can have different impacts in a large-scale photoreactor.

# 3.1.1. Sensitivity analysis

3.1.1.1. Same treatment time. The LED lamp irradiance was decreased to 150.70 W m<sup>-2</sup> (68% lamp intensity), corresponding to an operation time of 43.38 min and a power input of 24.79 W, taking advantage of the linear relationship between LED lamp irradiance, DCF degradation, and lamp power [9]. Under these conditions, the overall time in the two photoreactors was comparable, and the energy inputs per FU for the LEDs photoreactor were 17.92 Wh for the lamp, 36.88 Wh for the centrifugal pump, and 32.54 Wh for the magnetic stirrer.



Fig. 2. Relative impacts in percentage of the UV-C LEDs (orange) and the LP mercury lamp (red) photoreactors referred to the functional unit on the 16 categories of the Environmental Footprint method.



**Fig. 3.** Relative impacts in percentage of the UV-C LEDs (crossed orange) and the LP mercury lamp (red) photoreactors referred to the FU on the 16 categories of the EF method when the lamps are emitting at the same irradiation of 150.70 W  $m^{-2}$  (a), and the percentage contributions referred to each component: UV-C source (blue), lab equipment (violet), and electricity (teal) for "climate change" and "human toxicity non-cancer" categories (b).

In this case, with energy inputs for the centrifugal pump and magnetic stirrer being equal, attention was drawn to the greater energy demand of the LED lamp compared to the mercury lamps. As shown in Fig. 3a, this time the impacts were very comparable, and it is less straightforward to state the overall best performance. Even so, the categories mentioned earlier, "human toxicity non-cancer" and "resource use minerals and metals", were still more affected by the mercury lamp photoreactor, where the LED system had 16% and 28% less impact, respectively. Additionally, "human toxicity cancer" and "ecotoxicity freshwater" were also more affected by the mercury lamp photoreactor, with a  $\sim$ 4% difference in both cases. All these categories are negatively affected by the higher environmental burden of the mercury lamps compared to the LEDs, mainly due to mercury production, which is found to release as emission to air 16% of mercury for each kg produced, according to the ecoinvent dataset [33]. For the rest of the categories, the differences were between 1 and 2% in favour of the mercury lamp, a percentage that can be considered inside the dataset's uncertainty and due to the higher energy inputs required for the UV-C LED lamp.

Given the similarities of the results in this case and the interest in understanding the hotspots of the systems, the individual percentage contributions for the three main parts: the UV source, the lab equipment, and the overall electricity employed during the treatment, were evaluated for the two photoreactors separately. Fig. 3b shows the relative contribution of each part for "climate change" and "human toxicity noncancer". These categories were chosen as representative of the entire dataset, other than being particularly important for decision-making. The contribution within "climate change" highly resembles the contribution within most of the categories, while between "human toxicity non-cancer" and "resource use minerals and metals", where the mercury lamp was remarkably more impactful than the UV-C LEDs, only the former was shown for discussion since they are showing similar outcomes.

From Fig. 3b, it is clear that electricity is the main contributor in all categories. In the case of "climate change", where the total values were 0.0352 and 0.0346 kg CO<sub>2</sub> equivalent for the UV-C LED and the mercury lamp, respectively, >97% was attributed to the energy consumption in both cases. On the other hand, for "human toxicity non-cancer", with values respectively of  $4.84 \cdot 10^{-10}$  and  $5.77 \cdot 10^{-10}$  CTUh for the UV-C

LEDs and the mercury lamp, the impact of the latter in percentage was more prominent given the toxicity of mercury, reaching almost one-fifth of the entire category, whereas, in the case of the UV-C LEDs, the impact was ten times less.

*3.1.1.2.* Varying LED lamp efficiency. Another scenario was evaluated considering the predicted trend of the UV-C LED efficiency in the coming years. The power input for the UV-C LED lamp was assumed to be 20% lower, a value expected to be reached by 2030 [11]. Therefore, Fig. 4 compares the mercury lamp photoreactor, whose maturity can be considered stable [11], with the base case at 100% intensity of the UV-C LEDs and when working at 68% of the LED lamp, both with 20% lower power inputs, therefore considering 28.92 and 19.83 W instead of 36.14 and 24.79 W, respectively.

With only 20% higher efficiency, Fig. 4 shows that UV-C LEDs performed better than mercury lamps with lower relative impacts in all categories, also when working at a lower intensity. The UV-C LED lamp at 68% intensity had around 3% less impact in all categories, except for the ones it was already outstanding in the second scenario, where the impact was 20% and 30% lower. At the same time, the UV-C LED lamp at 100% intensity had >27% less impact in all categories. Assuming that this higher efficiency will be reached in the next years, it can be concluded that the results are very promising for the implementation of the UV-C LED lamp in water treatment systems. These findings align with prior research demonstrating that UV-C LED technology shows promise as a sustainable method for breaking down various organic compounds [42], and it outperformed LP mercury lamp systems at the bench-scale [43]. However, for the system analysed in this study, it is specifically recommended to prioritise working at a higher intensity and reduce the overall treatment time, whether possible.

# 3.2. Comparison between UV-C, UV-C/H<sub>2</sub>O<sub>2</sub>, and UV-C/FC

The addition of an oxidant also provides a strategy to reduce the required operation time, but the contribution of the oxidant to the impact categories should also be considered. Pesqueira et al. [16] evaluated the sustainability of three oxidants,  $H_2O_2$ , peroximonosulfate, and persulfate, and found the unsuitability of the last two, even if shorter



Fig. 4. Relative impacts in percentage of the UV-C LEDs photoreactor at 100% intensity (orange) and 68% intensity (crossed orange) assuming a 20% improvement in the energy efficiency of the lamp, and the LP mercury lamp photoreactor (red). The results refer to the FU on the 16 categories of the EF method employed.

times and higher efficiencies were achieved. Therefore, in this section, the life cycle impacts of combining UV-C with  $H_2O_2$  and FC were investigated with the same FU, where the UV source was the UV-C LED lamp at 265 nm. The results are shown in Fig. 5, and the absolute values for each category have been reported in Appendix C of the Supplementary Information.

As displayed in Fig. 5, the addition of  $H_2O_2$  or FC led to an average relative reduction of around 35% and 30%, respectively, in all the impact categories. Therefore, in the photoreactor under study, adding the oxidants generated less impact on the environment than the extra electricity required to work for longer times when no oxidant is added. When comparing the two UV-based AOPs with each other,  $H_2O_2$ showed, on average, 5% lower impacts, except for "ozone depletion" and "ecotoxicity freshwater", where the impact differences were 19.5% and 12.5%, respectively, being these last two categories the most affected by FC. Fig. 6 shows the relative impacts of the oxidants, UV-C LED lamp, lab equipment, and electricity in the three cases for four selected categories: "climate change" and "human toxicity non-cancer", as previously selected and quite representative of the distribution in all the other categories, and "ozone depletion" and "ecotoxicity freshwater", since the most affected by the oxidant selection.

While  $H_2O_2$  contribution to the environmental impact is almost null at the concentrations employed (below 1% in all categories), according to Fig. 6, the use of FC (in dark red under UV-C/FC treatment) impacted in percentages of 1.6%, 17%, 4.5%, and 9.6% in "climate change", "ozone depletion", "human toxicity non-cancer", and "ecotoxicity freshwater", respectively. The higher impacts were due to sodium hypochlorite production, which involved the release to the water of 0.5 kg of chloride and 0.3 kg of sodium for each kg produced, according to the information provided in the ecoinvent dataset [33]. Therefore, among  $H_2O_2$  and FC, the former is preferred when similar treatment times are required to achieve DCF degradation. However, the use of FC is still recommended in comparison to only photolysis and if it achieves shorter treatment times compared to  $H_2O_2$ .

### 3.3. Renewable electricity

Since, in all cases, electricity is the main hotspot for this technology in the experimental system under investigation, strategies to reduce its environmental burden are necessary; however, further research is needed to verify whether this hotspot persists on a larger scale. The results of employing renewable energy sources to replace the selected average electricity mix for the European market have been examined. As a feasible option for countries like Spain that benefit from high solar exposition all year, the electricity from the photovoltaic energy source was selected from the ecoinvent database: "electricity, low voltage {ES} electricity production, photovoltaic, 3 kWp slanted-roof installation, single-Si, panel, mounted | Cut-off, U". Another feasible option for Spain [44], but also for other countries that do not benefit from so many hours of sun, such as Ireland [45], wind power was also considered, selected in the ecoinvent database: "electricity, high voltage {ES}| electricity production, wind, <1 MW turbine, onshore | Cut-off, U" for the Spanish market as well. For the comparison, the treatment with the 265 nm UV-C LED lamp without any oxidant and at 100% intensity was chosen, and the results referred to the FU are shown in Fig. 7. The absolute values for each category can be found in Appendix D of the Supplementary Information.

From Fig. 7, the relative impacts of using electricity from photovoltaic and wind instead of the electricity mix in the photoreactor under study showed a middle to high impact reduction, except for "resource use minerals and metals" in the case of photovoltaic. The latter saw an increase of 40% mainly due to the copper mine operation, followed by the silver-gold mine operation involved in the production of solar panels, based on the information from the ecoinvent database [33]. In this regard, wind produced less impact in the resources categories compared to solar energy and achieved a reduction in all 16 categories ranging from 30% up to 99% compared to the European electricity mix. Compared to photovoltaic, wind energy had higher impacts only in "human toxicity cancer" and "land use" of 12% and 30% due to the ferrochromium production for the wind turbine and the wind power plant construction, respectively. In all other categories, the impacts were from 60% to 80% lower than photovoltaics, according to the relative impacts of each clean energy source [46]. Photovoltaic and wind energy were also investigated for UV/H2O2 and UV/FC. By comparing only the "climate change" category, the reductions observed were 80% and 93% when photovoltaic and wind energy were employed instead of the electricity mix. Therefore, using cleaner sources during UV-C LEDs and UV-C LEDs-based AOPs greatly improved the environmental sustainability of the photoreactor analysed.



Fig. 5. Relative impacts of the three UV-C LED water treatments; UV-C (orange), UV-C/H<sub>2</sub>O<sub>2</sub> (blue with horizontal white lines) and UV-C/FC (green with black diagonal lines) for the removal of 90% of DCF on the 16 categories of the Environmental Footprint method.



Fig. 6. Relative contribution of the oxidant (dark red), UV-C LED lamp (blue), lab equipment (violet), and electricity (teal) of the three UV-C water treatments per functional unit for the four selected categories of the EF method: "climate change", "ozone depletion", "human toxicity non-cancer", and "ecotoxicity freshwater".



Fig. 7. Relative impacts in percentage of the UV-C LED water treatment referred to the functional unit when employing the electricity mix of the European market (orange) and the electricity from photovoltaic (grey) or wind (light blue) of the Spanish market. The results are shown for the 16 categories of the Environmental Footprint method employed.

# 4. Conclusions

A comprehensive ex-ante environmental life cycle assessment was conducted for the first time to compare a UV-C LED lamp photoreactor with a LP mercury lamp photoreactor while degrading 90% of DCF in water. The analysis included various scenarios to provide a holistic understanding of the environmental implications associated with the UV-C technology. The findings demonstrate that the LEDs performed better, with lower impacts observed when working at higher lamp irradiance or expecting a 20% increase in power efficiency in the coming years. However, the impacts were similar when working at the same lamp irradiance and treatment time due to the present higher photoelectric conversion efficiency of the mercury lamps.

Furthermore, the comparison between UV-C, UV-C/H<sub>2</sub>O<sub>2</sub>, and UV-C/ FC while employing the UV-C LED lamp emitting at 265 nm was also assessed, as no previous studies explored the sustainability of free chlorine. The addition of  $H_2O_2$  and FC reduced the total treatment time, leading to a decrease of around 35% and 30% compared to photolysis alone in all the impact categories. Between the two oxidants, for equal treatment time, the use of  $H_2O_2$  is preferred as less environmentally impactful.

Throughout the analysis, electricity consumption for the lamp and other laboratory equipment was identified as a major hotspot, emphasising the need for further research to validate these findings at a larger scale. Comparing energy sources, it was found that transitioning to cleaner energy, such as photovoltaic and wind energy, significantly improved the overall environmental sustainability of the system.

In conclusion, the present research advances knowledge in the field of water treatment technology. By conducting a comprehensive ex-ante environmental LCA, valuable insights regarding the environmental performance of UV-C LED technology compared to traditional LP mercury lamps are provided, demonstrating the promising applicability of this technology in water treatment applications if continued research efforts are integrated into the scale-up of this technology. Furthermore, incorporating environmental analysis into the initial design phase of emerging systems is revealed as critical for identifying potential environmental impacts, optimising system performance and guiding decision-making for the development of more sustainable water treatment systems.

## CRediT authorship contribution statement

**Raffaella Pizzichetti:** Writing – review & editing, Writing – original draft, Visualization, Software, Investigation, Data curation, Conceptualization. **Mario Martín-Gamboa:** Writing – review & editing, Validation, Software, Methodology, Conceptualization. **Cristina Pablos:** Writing – review & editing, Supervision, Methodology, Funding acquisition. **Ken Reynolds:** Writing – review & editing, Supervision, Funding acquisition. **Simon Stanley:** Writing – review & editing, Supervision, Funding acquisition. **Javier Dufour:** Writing – review & editing, Software, Methodology. **Javier Marugán:** Writing – review & editing, Supervision, Methodology. Javier Marugán: Writing – review & editing, Supervision, Methodology, Funding acquisition.

#### 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.

## Acknowledgements

The authors acknowledge the financial support of the European Union's Horizon 2020 research and innovation programme in the frame of REWATERGY, Sustainable Reactor Engineering for Applications on the Water-Energy Nexus, MSCA-ITN-EID Project N. 812574. Additionally, Dr. Martín-Gamboa would like to thank the Regional Government of Madrid for financial support (2019-T2/AMB-15713).

# Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.susmat.2024.e01002.

### References

- United Nations Environment Programme, Minamata Convention on Mercury, 2019.
- [2] K. Song, M. Mohseni, F. Taghipour, Application of ultraviolet light-emitting diodes (UV-LEDs) for water disinfection: a review, Water Res. 94 (2016) 341–349, https:// doi.org/10.1016/j.watres.2016.03.003.

- [3] P. Jarvis, O. Autin, E.H. Goslan, F. Hassard, Application of ultraviolet lightemitting diodes, Water 11 (2019) 1–15.
- [4] Z. Wang, V. Srivastava, I. Ambat, Z. Safaei, M. Sillanpää, Degradation of ibuprofen by UV-LED/catalytic advanced oxidation process, J. Water Process Eng. 31 (2019), https://doi.org/10.1016/j.jwpe.2019.100808.
- [5] Y. Cha, T.K. Kim, J. Lee, T. Kim, A.J. Hong, K.D. Zoh, Degradation of iopromide during the UV-LED/chlorine reaction: effect of wavelength, radical contribution, transformation products, and toxicity, J. Hazard. Mater. 437 (2022) 129371, https://doi.org/10.1016/j.jhazmat.2022.129371.
- [6] S.A. Popova, G.G. Matafonova, V.B. Batoev, Dual-wavelength UV degradation of bisphenol a and bezafibrate in aqueous solution using excilamps (222, 282 nm) and LED (365 nm): yes or no synergy? J. Environ. Sci. Heal. - Part A. 58 (2023) 39–52, https://doi.org/10.1080/10934529.2023.2172270.
- [7] Z.C. Gao, Y.L. Lin, B. Xu, Y. Xia, C.Y. Hu, T.C. Cao, X.Y. Zou, N.Y. Gao, Evaluating iopamidol degradation performance and potential dual-wavelength synergy by UV-LED irradiation and UV-LED/chlorine treatment, Chem. Eng. J. 360 (2019) 806–816, https://doi.org/10.1016/j.cej.2018.12.022.
- [8] M.R. Eskandarian, M. Ganjkhanloo, M.H. Rasoulifard, S.A. Hosseini, Energyefficient removal of acid red 14 by UV-LED/persulfate advanced oxidation process: pulsed irradiation, duty cycle, reaction kinetics, and energy consumption, J. Taiwan Inst. Chem. Eng. 127 (2021) 129–139, https://doi.org/10.1016/j. jtice.2021.07.035.
- [9] R. Pizzichetti, K. Reynolds, C. Pablos, C. Casado, E. Moore, S. Stanley, J. Marugán, Removal of diclofenac by UV-B and UV-C light-emitting diodes (LEDs) driven advanced oxidation processes (AOPs): wavelength dependence, kinetic modelling and energy consumption, Chem. Eng. J. 471 (2023) 144520, https://doi.org/ 10.1016/j.cej.2023.144520.
- [10] S. Kang, J. Bae, S. Park, K. Kim, J. Lee, C. Yoon, C. Ryu, Design optimization of a cylindrical UV-C LED reactor for effective water disinfection with numerical simulations and test reactor fabrication, J. Environ. Chem. Eng. 12 (2024) 1–10, https://doi.org/10.1016/j.jece.2024.112366.
- [11] M. Martín-Sómer, C. Pablos, C. Adán, R. Van, J. Marugán, A review on led technology in water photodisinfection, Sci. Total Environ. 885 (2023) 163963, https://doi.org/10.1016/j.scitotenv.2023.163963.
- [12] J. Li, N. Gao, D. Cai, W. Lin, K. Huang, S. Li, J. Kang, Multiple fields manipulation on nitride material structures in ultraviolet light-emitting diodes, Light Sci. Appl. 10 (2021) 129, https://doi.org/10.1038/s41377-021-00563-0.
- [13] International Organization for Standardization, ISO 14040, 2006 Environmental Management - Life cycle assessment - Principles and Framework, 2006.
- [14] S. Cucurachi, C. Van Der Giesen, J. Guinée, Ex-ante LCA of emerging technologies, Procedia CIRP. 69 (2018) 463–468, https://doi.org/10.1016/j.procir.2017.11.005.
- [15] S. Foteinis, A.G.L. Borthwick, Z. Frontistis, D. Mantzavinos, E. Chatzisymeon, Environmental sustainability of light-driven processes for wastewater treatment applications, J. Clean. Prod. 182 (2018) 8–15, https://doi.org/10.1016/j. jclepro.2018.02.038.
- [16] J.F.J.R. Pesqueira, J. Marugán, M.F.R. Pereira, A.M.T. Silva, Selecting the most environmentally friendly oxidant for UVC degradation of micropollutants in urban wastewater by assessing life cycle impacts: hydrogen peroxide, peroxymonosulfate or persulfate? Sci. Total Environ. 808 (2022) 152050 https://doi.org/10.1016/j. scitotenv.2021.152050.
- [17] S.M. McKee, E. Chatzisymeon, Assessing the environmental sustainability of light emitting diodes and conventional blacklight lamps for the treatment of bisphenol-a in water, Environ. Impact Assess. Rev. 97 (2022) 106886, https://doi.org/ 10.1016/j.eiar.2022.106886.
- [18] B. Notarnicola, G. Tassielli, P.A. Renzulli, R. Di Capua, F. Astuto, G. Mascolo, S. Murgolo, C. De Ceglie, M.L. Curri, R. Comparelli, M. Dell'Edera, Life cycle assessment of UV-C based treatment systems for the removal of compounds of emerging concern from urban wastewater, Sci. Total Environ. 857 (2023) 159309, https://doi.org/10.1016/j.scitotenv.2022.159309.
- [19] S. Guerra-Rodríguez, S. Cuesta, J. Pérez, E. Rodríguez, J. Rodríguez-Chueca, Life cycle assessment of sulfate radical based-AOPs for wastewater disinfection, Chem. Eng. J. 474 (2023) 1–11, https://doi.org/10.1016/j.cej.2023.145427.
- [20] I. Alessandretti, C.V.T. Rigueto, M.T. Nazari, M. Rosseto, A. Dettmer, Removal of diclofenac from wastewater: a comprehensive review of detection, characteristics and tertiary treatment techniques, J. Environ. Chem. Eng. 9 (2021) 106743, https://doi.org/10.1016/j.jece.2021.106743.
- [21] L. Sbardella, I.V. Gala, J. Comas, S.M. Carbonell, I. Rodríguez-Roda, W. Gernjak, Integrated assessment of sulfate-based AOPs for pharmaceutical active compound removal from wastewater, J. Clean. Prod. 260 (2020) 121014, https://doi.org/ 10.1016/j.jclepro.2020.121014.
- [22] W.L. Wang, Q.Y. Wu, Z.M. Li, Y. Lu, Y. Du, T. Wang, N. Huang, H.Y. Hu, Lightemitting diodes as an emerging UV source for UV/chlorine oxidation: carbamazepine degradation and toxicity changes, Chem. Eng. J. 310 (2017) 148–156, https://doi.org/10.1016/j.cej.2016.10.097.
- [23] G. Cerreta, M.A. Roccamante, P. Plaza-Bolaños, I. Oller, A. Aguera, S. Malato, L. Rizzo, Advanced treatment of urban wastewater by UV-C/free chlorine process: Micro-pollutants removal and effect of UV-C radiation on trihalomethanes formation, Water Res. 169 (2020) 115220, https://doi.org/10.1016/j. watres.2019.115220.
- [24] D.B. Miklos, C. Remy, M. Jekel, K.G. Linden, J.E. Drewes, U. Hübner, Evaluation of advanced oxidation processes for water and wastewater treatment – a critical review, Water Res. 139 (2018) 118–131, https://doi.org/10.1016/j. watres.2018.03.042.
- [25] J.F.J.R. Pesqueira, M.F.R. Pereira, A.M.T. Silva, Environmental impact assessment of advanced urban wastewater treatment technologies for the removal of priority

#### R. Pizzichetti et al.

substances and contaminants of emerging concern: a review, J. Clean. Prod. 261 (2020) 121078, https://doi.org/10.1016/j.jclepro.2020.121078.

- [26] N. Vieno, M. Sillanpää, Fate of diclofenac in municipal wastewater treatment plant - a review, Environ. Int. 69 (2014) 28–39, https://doi.org/10.1016/j. envint.2014.03.021.
- [27] J.C.G. Sousa, A.R. Ribeiro, M.O. Barbosa, C. Ribeiro, M.E. Tiritan, M.F.R. Pereira, A.M.T. Silva, Monitoring of the 17 EU watch list contaminants of emerging concern in the Ave and the Sousa Rivers, Sci. Total Environ. 649 (2019) 1083–1095, https://doi.org/10.1016/j.scitotenv.2018.08.309.
- [28] L. Lonappan, S.K. Brar, R.K. Das, M. Verma, R.Y. Surampalli, Diclofenac and its transformation products: environmental occurrence and toxicity - a review, Environ. Int. 96 (2016) 127–138, https://doi.org/10.1016/j.envint.2016.09.014.
- [29] European Commission, Decision (EU) 2015/495 of 20 March 2015 Establishing a Watch List of Substances for Union-Wide Monitoring in the Field of Water Policy Pursuant to Directive 2008/105/EC of the European Parliament and of the Council, 2015.
- [30] J.R. Bolton, M.I. Stefan, P.S. Shaw, K.R. Lykke, Determination of the quantum yields of the potassium ferrioxalate and potassium iodide-iodate actinometers and a method for the calibration of radiometer detectors, J. Photochem. Photobiol. A Chem. 222 (2011) 166–169, https://doi.org/10.1016/j.jphotochem.2011.05.017.
- [31] International Organization for Standardization, ISO 14044, 2006 Environmental Management - Life cycle assessment - Requirements and Guidelines, 2006.
- [32] L. Tähkämö, M. Puolakka, L. Halonen, G. Zissis, Comparison of life cycle assessments of LED light sources, J. Light. Vis. Environ. 36 (2012) 44–53, https:// doi.org/10.2150/jlve.36.44.
- [33] G. Wernet, C. Bauer, B. Steubing, J. Reinhard, E. Moreno-Ruiz, B. Weidema, The ecoinvent database version 3 (part I): overview and methodology, Int. J. Life Cycle Assess. 21 (2016) 1218–1230, https://doi.org/10.1007/s11367-016-1087-8.
- [34] C. Casado, J. Moreno-SanSegundo, I. De la Obra, B. Esteban García, J.A. Sánchez Pérez, J. Marugán, Mechanistic modelling of wastewater disinfection by the photo-Fenton process at circumneutral pH, Chem. Eng. J. 403 (2021) 126335, https:// doi.org/10.1016/j.cej.2020.126335.
- [35] J.R. Bolton, M.I. Štefan, Fundamental photochemical approach to the concepts of fluence (UV dose) and electrical energy efficiency in photochemical degradation reactions, Res. Chem. Intermed. 28 (2002) 857–870, https://doi.org/10.1163/ 15685670260469474.

- [36] J.R. Bolton, K.G. Bircher, W. Tumas, C.A. Tolman, Figures-of-merit for the technical development and application of advanced oxidation technologies for both electric- and solar-driven systems, Pure Appl. Chem. 73 (2001) 627–637, https://doi.org/10.1351/pac200173040627.
- [37] European Commission, Recommendation (EU) 2021/2279 of 15 December 2021 on the Use of the Environmental Footprint Methods to Measure and Communicate the Life Cycle Environmental Performance of Products and Organisations, 2021.
- [38] S. Sala, F. Biganzoli, E. Sanye, M. Erwan, Toxicity impacts in the environmental footprint method : calculation principles, Int. J. Life Cycle Assess. 2022 (2013) 587–602, https://doi.org/10.1007/s11367-022-02033-0.
- [39] European Commission Joint Research Center Institute for Environment and Sustainability, International Reference Life Cycle Data System (ILCD) Handbook -General guide for Life Cycle Assessment - Detailed Guidance, 2010, https://doi. org/10.2788/38479.
- [40] S. Fazio, S. Castellani, V. Sala, S. Schau, E. Secchi, M. Zampori, Supporting information to the characterisation factors of recommended EF life cycle impact assessment method, Eur. Comm. Jt. Res. Cent. (2018), https://doi.org/10.2760/ 671368.
- [41] M. Goedkoop, M. Oele, J. Leijting, T. Ponsioen, E. Meijer, Introduction to LCA with SimaPro, Pré, 2016.
- [42] D. Pelayo, M.J. Rivero, G. Santos, P. Gómez, I. Ortiz, Techno-economic evaluation of UV light technologies in water remediation, Sci. Total Environ. 868 (2023) 161376, https://doi.org/10.1016/j.scitotenv.2022.161376.
- [43] S.A. MacIsaac, K.D. Rauch, T. Prest, R.M. Simons, G.A. Gagnon, A.K. Stoddart, Improved disinfection performance for 280 nm LEDs over 254 nm low-pressure UV lamps in community wastewater, Sci. Rep. 13 (2023) 7576, https://doi.org/ 10.1038/s41598-023-34633-7.
- [44] F.G. Montoya, M.J. Aguilera, F. Manzano-Agugliaro, Renewable energy production in Spain: a review, Renew. Sustain. Energy Rev. 33 (2014) 509–531, https://doi. org/10.1016/j.rser.2014.01.091.
- [45] Sustainable Energy Authority of Ireland (SEAI), Renewable Energy in Ireland, 2020.
- [46] A. Gargiulo, M.L. Carvalho, P. Girardi, Life cycle assessment of Italian electricity scenarios to 2030, Energies 13 (2020) 3852, https://doi.org/10.3390/ en13153852.