



# Treatment of laundry wastewater by different processes: Optimization and life cycle assessment

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## ABSTRACT

Wastewater from industrial laundries is often difficult to treat because it usually presents high turbidity and chemical oxygen demand (COD). We studied several processes for the treatment of laundry wastewater which was provided by a hotel in the south of Gran Canaria, Spain. More specifically, we studied coagulation with iron (III) sulphate, Fenton, photo-Fenton and a biological treatment (using a biofilter, and a granular activated carbon, GAC, filtration). The coagulation and Fenton processes produced large amounts of sludge and could not meet the required standards for water reuse in Spain. The use of photo-Fenton and the BF resulted in complete turbidity removal and high COD removal. However, we found that the effluent from the BF did not meet the COD removal criteria for water reuse, and thus a GAC filtration post-treatment was employed to reduce COD to acceptable levels. The photo-Fenton process alone did meet the criteria for water reuse. The estimated cost to treat 1 m<sup>3</sup> of wastewater was 6.72 € for photo-Fenton and 0.71 € for BF + GAC. The cost and life cycle assessment analyses that were also performed revealed that the acquisition of the necessary reagents is the main contribution to the overall economic and environmental costs for both options, and that the BF + GAC option is notably cheaper. Additionally, this option also causes much lower environmental impacts than photo-Fenton.

## 1. Introduction

Industrial laundries cover several sectors including, for example, hospitality, hospitals, and pre-marketed textiles. The European Union, EU, industrial laundry sector washes 2.7 billion kg of textiles per year (wet weight) and uses 42 million m<sup>3</sup> of water, which ends up in the sewage as wastewater [1]. This wastewater can contain over 20000 mg·L<sup>-1</sup> COD [2], and commonly has basic pH and high levels of turbidity, suspended solids, phosphorous compounds and allergenic fragrance compounds, among others.

In the EU, Regulation EC 648/2004, which was later modified by Regulation EU 259/2012, establishes the maximum concentration of phosphorous compounds and allergenic fragrance substances in laundry detergents, and the biodegradability of these products (at least 60% in 28 days).

In addition, several organisms, such as the EU, have launched certificates, such as the EU Ecolabel, to promote the consumption of more environmentally friendly products. In this sense, the criteria for a product to be certified with EU Ecolabel includes the limitation of the use of some toxic substances, fragrances, colouring agents, etc. in the formulations. The products must also meet some criteria based on other factors, such as the information given to the consumer, or the packaging system [3]. The current criteria to receive the EU Ecolabel certificate are included in Decision EU 2018/418. In March 2019, only 256 laundry detergents were certified with EU Ecolabel. This number increased to 932 by September 2022 [4].

Despite this, fragrance allergens and surfactants and microfibers have been found at low concentrations in the effluents of conventional wastewater treatment plants [5–8], potentially leading to environmental toxicity [9,10] and they have been found to be persistent in

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water bodies [11].

Several studies have reported on the treatment of industrial laundry wastewater using a wide range of techniques including biological, chemical, and advanced oxidation processes. Frequently, a combination of techniques is needed to achieve the desired water quality. From a previous review, we concluded that moving-bed bioreactors (MBBR), catalytic ozonation and Fenton are three promising methods for laundry wastewater treatment because mineralization is achieved [12].

Regarding aerobic biological processes, the main handicap for this option is that anionic surfactants at concentrations above 30 mg·L<sup>-1</sup>, such as linear alkylbenzene sulfonates (LAS), can hamper the efficiency of sewage treatment plants by reducing the respiration rate of bacteria and preventing enzymatic reactions [13]. Thus, LAS has proven to have toxic effects on the microbiota, although adding more readily biodegradable co-substrates such as linear alcohol ethoxylates (LAE) and ethanol to the media have been shown to reactivate microbial activity and increase biodegradation by co-metabolism [14]. Very few studies have been published on the use of biofilters and/or constructed wetlands (CWs) for the treatment of laundry wastewaters. Among these, Lutterbeck et al. (2020) found that the raw effluents of hospital laundry wastewater presented high phytotoxicity and genotoxicity. The application of microbial fuel cells and CWs rendered an efficient reduction of the overall toxicity, but with a hydraulic retention time (HRT) of 14 days [15].

However, some components found in detergents, such as fragrances, which are considered emerging contaminants, persist in the environment because of their low biodegradability [16]. Additionally, surfactants and their metabolites exert negative effects on the environment at concentrations below 1 mg·L<sup>-1</sup> [17,18], and it is known that traces of surfactants are found in effluents from biological processes [6]. Several authors have reported that additional more refined biological treatments are necessary when treating laundry wastewater [19,20].

Advanced oxidation techniques, such as ozonation, Fenton or others, have traditionally been an alternative for wastewaters containing recalcitrant contaminants. The hydroxyl radicals formed in these processes have the potential to oxidize organic matter non-specifically.

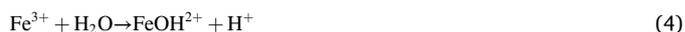
Among these, ozonation has been widely studied for laundry wastewater reclamation and results indicate that assisted ozonation (with iron and hydrogen peroxide, or TiO<sub>2</sub>, for example) enhances the production of hydroxyl radicals and, thus, the degradation rate of the contaminants [21,22]. This technique has been studied for laundry wastewater from hospitals in particular because, in that scenario, other recalcitrant contaminants such as pharmaceuticals may be present. In this sense, wastewater from hospital laundries has been reported to be extremely hazardous in terms of *D. magna* and *A. cepa* toxicity, and ozonation was found to be an option to achieve detoxification [21]. However, ozonation usually requires high energy consumption, which is an environmental burden if non-renewable sources are used. For this reason, treatment systems with high energy requirements should be avoided even if high pollutant removal is achieved [23].

We found very few studies on application of the Fenton process for laundry wastewater treatment [24–26], and only one paper reported photo-Fenton studies [20], in which only a solution of sodium dodecyl sulphate in deionized water and a synthetic wastewater were used. To our knowledge, studies on photo-Fenton applied to the treatment of real laundry wastewater have not been published yet.

The Fenton reaction consists of the production of hydroxyl radicals (·OH) using iron salts and hydrogen peroxide, according to reactions 1–3. The hydroxyl radicals react and oxidize the organic matter present in the media.



Reaction 2 is very slow at ambient temperature [27]. To increase the reaction rate and produce more hydroxyl radicals, UV radiation can be added. The process then becomes catalytic, following reactions 4 and 5:



It should be noted that these reactions are greatly influenced by pH, according to the speciation of iron with pH. In this sense, the optimized pH for Fenton-based reactions has been reported as 2.8 [28]. In addition, the concentrations of iron salts and hydrogen peroxide must be rigorously and experimentally optimized because an excess of reagents can lead to the scavenging of hydroxyl radicals by Fe(II) and/or hydrogen peroxide.

The present work focuses on the optimization of the treatment of laundry wastewater. For this purpose, we compared several options; namely, coagulation with iron salts, Fenton, photo-Fenton and a biological treatment in a biofilter (BF). This work includes: (i) a study of chemical oxygen demand (COD) removal and the reduction of other parameters with the different processes, as well as the monitoring of some by-products, (ii) an economic assessment of two options, and (iii) a life cycle assessment (LCA) analysis. On the basis of the above, the most adequate treatment for this wastewater can be chosen from both a techno-economic and an environmental point of view.

## 2. Materials and methods

### 2.1. Reagents/chemicals

The main reagents employed in this study were: Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·5 H<sub>2</sub>O (97% purity) and H<sub>2</sub>O<sub>2</sub> (30% w/v) from Panreac. When needed, dilute H<sub>2</sub>SO<sub>4</sub> and NaOH were used to adjust pH. Catalase from bovine liver supplied by Fluka was used to remove excess hydrogen peroxide from the water samples.

### 2.2. Analytical determinations

COD was determined following Standard Method 5220 D (USEPA). TNTplus reagent sets, valid for a COD range of 20 – 1500 mg/L, were purchased from Hach® and the digestion of samples was carried out in a SRB200 Hach® reactor. Digested samples were then measured with an Agilent Cary 60® UV-Vis spectrophotometer at 620 nm. It should be noted that hydrogen peroxide acts as a positive interferent in this analytical method [29], and, thus, hydrogen peroxide was removed from the samples using catalase. A fixed concentration of catalase was used for this purpose, and the COD related to the addition of catalase was subtracted from all results.

Anionic detergents (measured as LAS) were determined following the crystal violet method [30]. Non-ionic detergents were determined through the bismuth active substance (BiAS) method [31].

Total phosphorous was measured following the ascorbic acid method (Standard Methods 4500 P) with acid persulfate digestion. The digestion of samples was carried out in an SRB200 Hach® reactor.

Turbidity was measured with a Hach® 2100 P turbidimeter.

Carboxylic acids were measured through high performance liquid chromatography (HPLC). A UV detector was employed with λ set at 210 nm. The column used was Supelcogel C610-H (30 cm × 7.8 mm ID) and the mobile phase was a 0.1% phosphoric acid solution at a 0.5 mL·min<sup>-1</sup> flowrate.

Dissolved iron was determined with the o-phenanthroline method [32]. The orange ferrous complex was measured spectrophotometrically at 510 nm. The detection and quantification limits were 0.12 mg/L and 0.36 mg/L, respectively and the adjusted R<sup>2</sup> was 0.99.

Hydrogen peroxide was determined according to [33]. The yellow titanium(IV)-peroxide complex formed was spectrophotometrically

measured at 400 nm. The detection and quantification limits were 0.48 mg/L and 1.45 mg/L, respectively, and the adjusted  $R^2$  was 0.99.

Sedimentable solids were determined according to Standard Method 2540 F. Suspended solids were determined according to Standard Method 2540D. Total nitrogen (TN) was determined following Standard Method 4500 NC. Ammonia was determined using Standard Method 4500-NH<sub>3</sub>-C. *E. coli* was determined using Standard Method 9222B. BOD<sub>5</sub> was determined with Standard Method 5210B [34].

All samples, except those used to determine sedimentable and suspended solids, were centrifuged before measurement to remove the flocs formed during the reactions in which Fe(III) was used. Additionally, samples were also filtered (0.45 μm) before measuring the concentration of hydrogen peroxide along the different reactions in which this substance was added. Finally, due to the turbidity interference of the raw wastewater, nitrate and nitrite were determined qualitatively using JBL ProAquaTest strips.

### 2.3. Experimental design

The experimental design for the coagulation and photo-Fenton experiments was carried out following the response surface methodology. The Minitab v.16® software was used to obtain the response surfaces.

### 2.4. Experimental conditions

The wastewater was collected from a hotel in the south of Gran Canaria, Spain.

For coagulation and photo-Fenton, tests were carried out in 1 L Pyrex glass batch reactors and a Tecnylab® Jar Test FC6S flocculator was used as stirring equipment.

For coagulation experiments, pH was set at different values for different experiments. Next, varying concentrations of Fe(III) as Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·5 H<sub>2</sub>O were added. The jar-test method was used according to ASTM Standard D2035. Once the coagulant was added to the wastewater, mixing speed was kept at 120 rpm for 1 min, and then kept at 15 rpm for 20 min. Finally, flocs were allowed to settle for 24 h.

For photo-Fenton experiments, the initial pH<sub>0</sub> was set at 2.8 and varying concentrations of Fe(III) as Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·5 H<sub>2</sub>O and H<sub>2</sub>O<sub>2</sub> were added. For comparison purposes, a coagulation experiment was carried out at pH<sub>0</sub> 2.8 with the optimal Fe(III) load that was determined from the photo-Fenton results. Additionally, a Fenton experiment was also done with the optimal Fe(III) and H<sub>2</sub>O<sub>2</sub> loads. Finally, photolysis and UV/H<sub>2</sub>O<sub>2</sub> experiments were also performed adding only the optimal concentration of H<sub>2</sub>O<sub>2</sub> as blank experiments.

The experimental procedure was equivalent to that employed in the coagulation experiments but stirring at 15 rpm was maintained for 240 min. For the photo-Fenton, photolysis and UV/H<sub>2</sub>O<sub>2</sub> experiments, a 60 W Philips Solarium HB175 equipped with four 15 W Philips CLEO fluorescent tubes with emission spectrum from 300 to 400 nm (maximum around 365 nm) and with an average irradiation of about 90 W·m<sup>-2</sup> was used.

For the biological treatment, a lab-scale biofilter (BF) without vegetation was used. The BF comprised a plastic box (height: 13.5 cm, length: 37 cm, width: 26 cm) with a total capacity of 4 L. The substrate was gravel (Ø= 3–5 mm). Aerobic conditions were maintained with a recirculation pump of 2140 mL·min<sup>-1</sup>. The BF was inoculated with the effluent of the primary treatment of a WWTP in Gran Canaria. The pH of the laundry wastewater was adjusted to 7 and the reactor was filled with 3 L of laundry wastewater, operated in batch mode. The maximum HRT studied was 52 h. Water flow was kept subsurface to prevent the proliferation of mosquitoes and algae. A blank experiment was also performed with a similar BF containing sterilized gravel, so that the effect of adsorption and volatilization could be measured.

The effluent from the biological treatment was subjected to post-treatment with activated carbon. The activated carbon used in this study was a granular material with 0.5–3.15 mm granular size,

150 g·L<sup>-1</sup> bulk density, BET 842.82 m<sup>2</sup>·g<sup>-1</sup>, average pore diameter 2.84 nm, total pore volume of 0.60 cm<sup>3</sup>·g<sup>-1</sup> and micropore volume of 0.22 cm<sup>3</sup>·g<sup>-1</sup>.

## 3. Results and discussion

### 3.1. Wastewater characterisation

Wastewater samples were collected from a hotel in the south of Gran Canaria from September 2020 to May 2021 and from May to July 2022. In this hotel, most of the linen and all towels are washed in an external industrial laundry. However, the hotel has an internal laundry with a 12 kg washing machine and a 15 kg drier, which are used to wash and dry curtains, cushions, quilts, and other textiles that are not washed daily or every other day. Additionally, mops and cloths are washed every day in this internal laundry. The wastewater from the mentioned washing machine is currently disposed together with the rest of the wastewater produced in the other services of the hotel. To take the wastewater samples for this study, we collected the wastewater directly from the drain tube of the washing machine.

Taxat Super Silex® was the detergent used in the internal laundry. According to the technical sheet of the product and the characteristics of the water in the hotel, the dosing of this detergent, which is done manually, is 23 g of detergent per kg of textile. This detergent is commonly used in industrial laundering facilities.

According to the safety datasheet, Taxat Super Silex® is an enzymatic detergent based on a combination of LAS and non-ionic surfactants (C13–15 branched and linear alcohol ethoxylates – LAE-). As bleaching agent, it contains sodium percarbonate. It is known that sodium percarbonate forms hydrogen peroxide and sodium carbonate in the washing process [35]. It also contains fluorescent whitening agents (composition not specified in the technical sheet). Zeolites and phosphonates are also present as sequestering agents to reduce water hardness. Finally, d-limonene and linalool are present in the formulation as fragrance compounds.

The wastewater characterisation is shown in Table 1. The legal maximum contaminant level (MCL) for each parameter is also shown, both for the disposal of wastewater in the sewage system and for its reuse.

Several parameters are above the MCL for disposal: total surfactants, COD, TSS and pH. Additionally, turbidity and total phosphorous (TP) are above the limit for reuse. Hydrogen peroxide could not be

**Table 1**

Characterisation of the wastewater samples collected from the washing machine drainage in Servatur Casablanca Suites & Spa and MCLs in Spain.

Parameter	Wastewater	MCL for reuse <sup>a</sup>	MCL for disposal in the sewage <sup>b</sup>
pH	11.07 ± 1.30	6.5–8.5	5.5–9.5
TSS (mg·L <sup>-1</sup> )	2900 ± 326	20	1000
COD (mg·L <sup>-1</sup> )	1920 ± 220	160	1600
BOD <sub>5</sub> (mg·L <sup>-1</sup> )	750 ± 80	30	1000
Turbidity (NTU)	1290 ± 105	10	-
Total Fe (mg·L <sup>-1</sup> )	0.22 ± 0.12	2	10
Total phosphorous (mg·L <sup>-1</sup> )	3.25 ± 0.10	10	20
Total N (mg·L <sup>-1</sup> )	36 ± 2.50	-	-
NH <sub>4</sub> <sup>+</sup> (mg·L <sup>-1</sup> )	-	15	50
NO <sub>3</sub> (mg·L <sup>-1</sup> )	< 10	45	30
Total surfactants (mg·L <sup>-1</sup> ) (LAS)	9.42 ± 0.85	2	6
	37.36 ± 0.17 (BiAS)		
<i>E. coli</i> (CFU in 100 mL)	< 200	200	-

<sup>a</sup> According to Royal Decree 1620/2007 (Spain) and Decree 174/1994 (Canary Islands, Spain). Reuse for irrigation of urban green areas.

<sup>b</sup> According to Decree 2/2019 (Gran Canaria, Canary Islands, Spain).

determined in the raw wastewater because the turbidity of the wastewater interfered with the analytical method. For this same reason, the concentration of total iron presented in Table 1 corresponds to that of the tap water in the hotel and was provided by the water supply company. For the treatment of this wastewater we studied three alternatives: coagulation, photo-Fenton and BF. The results for each option are given below.

### 3.2. Coagulation

Originally, inorganic coagulants such as aluminium or iron salts were employed for water treatment. However, coagulation processes produce flocs (sludge residue) that may be considered toxic residues according to the European List of Waste if hazardous substances included in Regulation EC 1272/2008 are present in the sludge (residue code 190205 \*). Nowadays, organic natural coagulants are preferred to inorganic or synthetic ones because they present lower toxicity [36]. Moreover, low molecular weight organic coagulants have reported better results for laundry wastewater treatment than inorganic ones [37].

Over 80% COD removal has been reported for laundry wastewater using natural coagulants [37,38]. However, the sludge formed in these processes contains detergent residues that may be toxic according to Regulation EC 1272/2008. For this reason, the authors believe that other options that ensure mineralization of the organic matter should be considered for laundry wastewater reclamation.

In this work, we studied the coagulation of laundry wastewater using an iron salt to determine the effect of adding iron to this water because in the Fenton and photo-Fenton reactions (see next section) iron is employed. We used ferric sulphate because of the wide working pH range of this coagulant (pH 4–11) [39].

It is known that the factors affecting coagulation processes are the type of coagulant, the coagulant load, the pH, the mixing speed and the sedimentation time. A two-level factorial design with two variables (pH and ferric sulphate load) was applied to determine the optimal operating conditions for the treatment of laundry wastewater with ferric sulphate. The factorial design is described in Table 2. The experiments were performed randomly to ensure statistical significance and all experiments were carried out twice. The objective function was percent of COD/turbidity removal after the coagulation process.

The low and high levels for pH were chosen according to the pH range of the coagulant, while those for the concentration of Fe(III) were chosen after a bibliographic revision of other works in which Fenton was used to treat laundry wastewater. In this regard, Vilve et al. reported, for a wastewater with 530–560 mg·L<sup>-1</sup> initial COD, between 73% and 80% COD removal when coagulation or Fenton were used, respectively [24]. In both processes, the concentration of iron was 824 mg·L<sup>-1</sup>. Another study reported COD removal of 70% for a wastewater with 365 mg·L<sup>-1</sup> initial COD when adding 25 mg·L<sup>-1</sup> iron in a Fenton reaction [40]. Thus, the amount of iron added in the above references differs considerably, between 1.5 and 0.07 mg Fe(III)·mg COD<sup>-1</sup>. In order to keep the concentration of chemicals added to the process as low as possible, we decided to choose 50 and 500 mg·L<sup>-1</sup> as low and high levels for Fe(III), respectively.

The experimental results returned the response shown in Eq.(6). It must be noted that only the significant variables were included (p-value < 0.05). The only significant factor was pH. The adjusted R<sup>2</sup> was 92.44%.

$$\% \text{Turbidity removal} = -5.12 \cdot \text{pH} + 77.99 \quad (6)$$

**Table 2**

Levels of the experimental factors: 2<sup>2</sup> factorial design.

Factor	Low level (-1)	High level (+1)
pH	4	10
Fe(III) (mg·L <sup>-1</sup> )	50	500

According to Eq.(6), we found that turbidity removal increased as pH decreased. The experimental and predicted values for the experimental points included in the analysis are shown in Fig. 1. It can be seen that all the values fall in the region of interest, which was fixed at ± 10% deviation between experimental and predicted values.

To validate this model, we must confirm that the relationship between the confidence interval, CI, for the centre points of the model and the curvature, C, is above 1 [41]. CI and C are determined following Eq. (7) and Eq.(8), respectively.

$$CI = \frac{t \cdot s}{\sqrt{N}} \quad (7)$$

where *t* is Student's *t*, *s* is the standard deviation of the results obtained for the central points of the design of experiments and *N* is the number of replicates of the central point.

$$C = \bar{Z}_{cp} - \bar{Z} \quad (8)$$

where  $\bar{Z}_{cp}$  is the mean result of the replicates of the central points and  $\bar{Z}$  is the mean result for all experiments included in the design.

In this study, we confirmed that CI (36.49) > C (10.56) and, therefore, we concluded that the obtained response surface is adequate to predict turbidity removal from the treatment of laundry wastewater using coagulation with ferric sulphate at a given pH value.

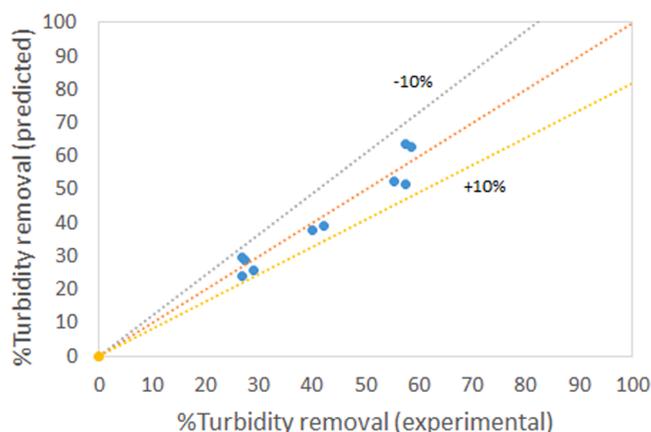
For COD removal, the response is given in Eq.(9). When using COD removal as response variable, the adjusted R<sup>2</sup> was 94.40%. In this occasion, the value of CI (12.52) was also higher than that of C (4.71).

$$\% \text{COD removal} = -1.72 \cdot \text{pH} + 36.25 \quad (9)$$

The highest turbidity removal we achieved in our work with the coagulation experiments was 63.62% (29.69% COD removal). This result was obtained for the experiment at pH 4 with the highest Fe(III) load; that is, 500 mg·L<sup>-1</sup>. Reducing the Fe(III) load ten times returned a 51.39% turbidity removal (29.06% COD removal). Thus, in the studied interval, large variations in the coagulant dosing imply only small variations of the final result (regarding turbidity). When pH was increased to 7 or 10, turbidity removal fell to around 25–30% (18–20% COD removal), which indicates that the coagulation process was favoured at lower pH values. This is in line with the results reported by another author who studied the coagulation of laundry wastewater with an iron-based coagulant [24].

### 3.3. Photo-Fenton

For the photo-Fenton studies, we first did some preliminary experiments at pH 4 with 500 mg·L<sup>-1</sup> Fe(III) and 408–4080 mg·L<sup>-1</sup> H<sub>2</sub>O<sub>2</sub> and



**Fig. 1.** Correlation between experimental and predicted values for turbidity removal using the coagulation process.

found that turbidity and COD removals were very similar to those obtained in the coagulation process alone. For this reason, we decided to set the working pH at 2.8, which is the optimal pH for this process, as mentioned above.

The variables studied in the photo-Fenton experiments were Fe(III) and H<sub>2</sub>O<sub>2</sub> concentrations. The H<sub>2</sub>O<sub>2</sub> concentrations varied between 408 and 4080 mg·L<sup>-1</sup>, and were chosen within the range of 10% and 100% of the theoretical stoichiometric amount relative to COD (2.125 gH<sub>2</sub>O<sub>2</sub>:g<sup>-1</sup> COD) [40]. The mean initial COD of the laundry wastewater studied in this work was 1920 mg·L<sup>-1</sup>. This value was taken for calculations.

Fe(III) concentrations were kept between 33.5 and 500 mg·L<sup>-1</sup>. The high level was kept the same as in the coagulation process. Although the optimum Fe:H<sub>2</sub>O<sub>2</sub> ratio will depend on the characteristics of the wastewater to treat, Vilve et al. reported an optimum Fe:H<sub>2</sub>O<sub>2</sub> molar ratio of 1:2 for the Fenton reaction applied to laundry wastewater [24]. It is known that the photo-Fenton system requires lower iron loads than the Fenton reaction. For this reason, we decided to set the lower Fe(III) concentration to meet a ten times lower ratio than that reported by the mentioned authors, that is, a 1:20 Fe:H<sub>2</sub>O<sub>2</sub> molar ratio. This calculation was applied considering the lowest H<sub>2</sub>O<sub>2</sub> concentration, that is, 408 mg·L<sup>-1</sup>. The values chosen in this work vary between 1:1.3 and 1:200 Fe:H<sub>2</sub>O<sub>2</sub> molar ratios.

A two-level factorial design was studied in a first approach but we found that the curvature was significant. Therefore, we next applied a two-level central composite design (CCD). The levels of the experimental factors for this design are included in Table 3. Here, the experiments were done randomly and the central point was replicated five times to ensure statistical significance.

In this set of experiments, we found that turbidity was almost completely removed (> 80%) for all experiments except those with the lowest Fe(III) addition. For the experiments with 33.50 mg·L<sup>-1</sup> Fe(III), turbidity removal varied between 3% and 11%.

COD removal was also low for the lowest Fe(III) level (1–22%) and for the lowest H<sub>2</sub>O<sub>2</sub> level (up to 53%). For the rest of the experimental dataset, COD removals between 86% and 90% were obtained. The response surface for COD removal is shown in Eq.(10). The factors are expressed in coded units, x<sub>1</sub> and x<sub>2</sub>. The coded units were calculated as detailed in Eqs. (11) and (12), with the Fe(III) and H<sub>2</sub>O<sub>2</sub> concentrations expressed in mg·L<sup>-1</sup>. As mentioned before, only the significant variables were included (p-value < 0.05). The adjusted R<sup>2</sup> was 99.06%.

$$\% \text{COD removal} = -34.10 \cdot x_1^2 - 19.01 \cdot x_2^2 + 29.81 \cdot x_1 + 16.67 \cdot x_2 + 10.76 \cdot x_1 \cdot x_2 + 87.64 \quad (10)$$

$$x_1 = \left[ \frac{[\text{Fe(III)}] - 33.5}{500 - 33.5} \right] - 1 \quad (11)$$

$$x_2 = \left[ \frac{[\text{H}_2\text{O}_2] - 408}{4080 - 408} \right] - 1 \quad (12)$$

It can be observed from Eq.(10) that both the concentration of Fe(III) and H<sub>2</sub>O<sub>2</sub> influence COD removal. This also occurs if turbidity removal is chosen as the response variable. In that case, the response followed Eq. (13) with an adjusted R<sup>2</sup> of 99.70%.

$$\% \text{Turbidity removal} = -47.36 \cdot x_1^2 - 2.98 \cdot x_2^2 + 41.98 \cdot x_1 + 2.58 \cdot x_2 + 4.36 \cdot x_1 \cdot x_2 + 98.85 \quad (13)$$

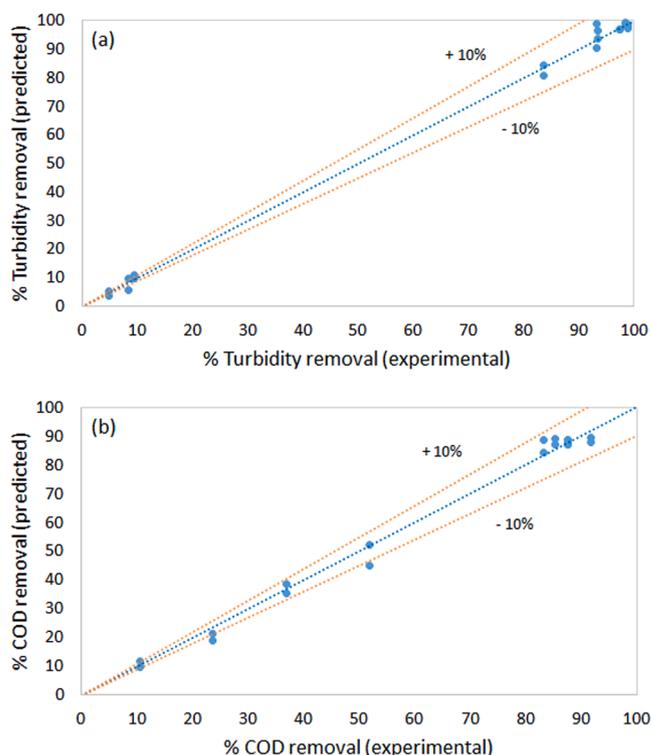
Fig. 2 illustrates the experimental and predicted values for the points included in the analysis.

The overall interactions among the studied variables are shown in Fig. 3. Axis values are expressed as uncoded variables.

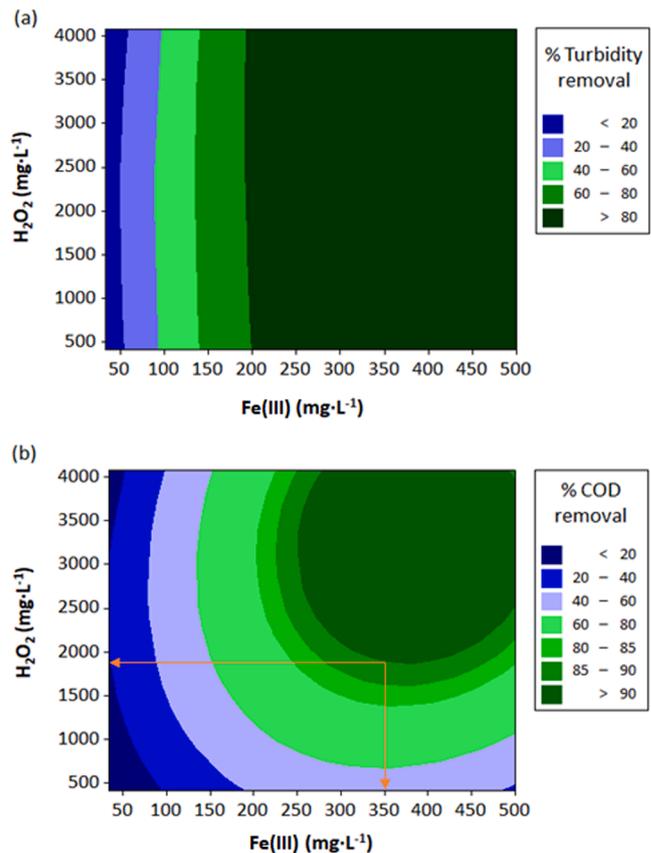
**Table 3**

Levels of the experimental factors: CCD design.

Factor	Low level (-1)	Central level (0)	High level (+1)
Fe(III) (mg·L <sup>-1</sup> )	33.50	266.75	500
H <sub>2</sub> O <sub>2</sub> (mg·L <sup>-1</sup> )	408	2244	4080



**Fig. 2.** Correlation between experimental and predicted values for (a) turbidity and (b) COD removal using the photo-Fenton process.



**Fig. 3.** Response surfaces for the effect of iron and hydrogen peroxide on (a) turbidity and (b) COD removal using the photo-Fenton process.

The COD MCL for the reuse of the treated wastewater is  $160 \text{ mg}\cdot\text{L}^{-1}$  and the target wastewater studied here presents  $1920 \pm 220 \text{ mg}\cdot\text{L}^{-1}$  COD (Table 1). Thus, a COD removal higher than 90% must be achieved if the treated water is to be reused. For this reason, we chose as optimal the lower iron and hydrogen peroxide concentrations that yielded higher than 90% COD reduction according to the results shown in Fig. 3b ( $350 \text{ mg}\cdot\text{L}^{-1}$  Fe(III) and  $1950 \text{ mg}\cdot\text{L}^{-1}$   $\text{H}_2\text{O}_2$ ). The substitution of these values in Eqs.(10–12) results in a predicted COD removal of 90.16%. Recently, Cüce et al. reported an optimized iron concentration of  $400 \text{ mg}\cdot\text{L}^{-1}$  and  $\text{H}_2\text{O}_2$  addition of  $900 \text{ mg}\cdot\text{L}^{-1}$  for laundry wastewater with an initial COD of  $1077 \text{ mg}\cdot\text{L}^{-1}$  [42], although only COD was followed in that study because the purpose of the authors was to analyse different models to predict and optimize Fenton processes rather than to study the treatment of laundry wastewater.

Next, we used the optimized Fe(III) and  $\text{H}_2\text{O}_2$  concentrations and followed COD removal at different intervals in order to optimize the necessary reaction time. To follow the reaction adequately, samples were centrifuged to remove the flocs formed. As mentioned above, for COD determinations, catalase was used to remove the excess hydrogen peroxide from the samples. Additionally, samples were taken along the reaction to determine the remaining concentration of hydrogen peroxide at each time to follow the consumption rate of this reagent.

For comparison purposes, additional experiments were carried out to determine the possible effect of the different variables present in the reaction: UV illumination – 365 nm, as used in the photo-Fenton process, Fe(III) and hydrogen peroxide. Thus, in addition to the photo-Fenton reaction, the following experiments were carried out:

- Fenton, using the Fe(III) and hydrogen peroxide concentrations that were previously optimized for the photo-Fenton process (no UV illumination)
- Coagulation, using the same Fe(III) concentration as in the photo-Fenton reaction (no UV illumination)
- UV (365 nm)/ $\text{H}_2\text{O}_2$ , using the same hydrogen peroxide concentration as in the optimized photo-Fenton process
- Photolysis (only UV, 365 nm)

All experiments were carried out at the same initial pH (2.8). It must be noted that pH only varied slightly during the above-mentioned reactions, being the pH along the reactions between 1.80 and 2.80.

The results for COD removal in the above-mentioned processes are shown in Fig. 4. It should be noted that no COD removal was observed in the UV (365 nm)/ $\text{H}_2\text{O}_2$  and photolysis (365 nm) control experiments. In this sense, Souza et al. reported that the optimized conditions for laundry wastewater treatment using UV/ $\text{H}_2\text{O}_2$  is to work at pH 9 with a COD: $\text{H}_2\text{O}_2$  ratio of 1:2.5, but they found that this treatment was not

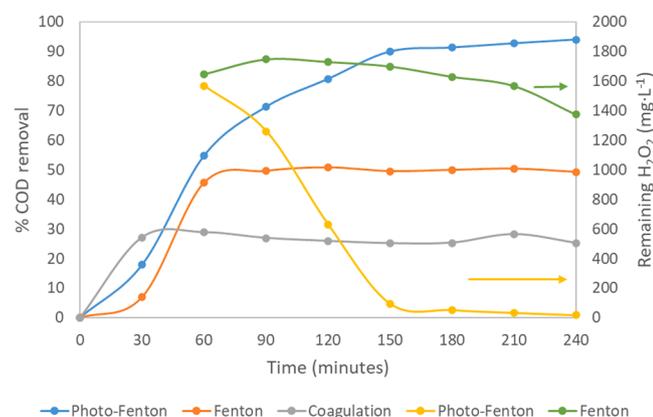


Fig. 4. COD removal (%) using coagulation, Fenton and photo-Fenton to treat laundry wastewater. Consumption of hydrogen peroxide along the photo-Fenton reaction.

efficient in reducing turbidity [43].

The photo-Fenton process returned higher COD removal than the coagulation and Fenton processes. The Fenton reaction returned 10–20% higher COD removals than coagulation for reaction times up to 240 min, which is in line with that reported by other authors [24]. The only process that achieved over 90% COD removal in 240 min was photo-Fenton.

After 240 min of reaction the remaining concentration of hydrogen peroxide in the Fenton reaction was still above  $1000 \text{ mg}\cdot\text{L}^{-1}$ . In this regard, it should be noted that hydrogen peroxide has proven to be toxic to aquatic organisms [44], and we must therefore ensure that this reagent is completely consumed along the reaction.

Another author reported that raising the reaction temperature in the Fenton process from  $50 \text{ }^\circ\text{C}$  to  $90 \text{ }^\circ\text{C}$  can increase the reaction rate by 8 times [40]. However, laundry wastewater from the laundering in hotels is normally at  $40\text{--}60 \text{ }^\circ\text{C}$ , and this would imply adding more energy requirements to the process.

At the end of the studied processes, pH was adjusted to 6.5 to, firstly, comply with water reuse legislation requirements (in Spain, Royal Decree 1620/2007) and, secondly, to precipitate iron. Additionally, the treated water was left to settle for 24 h to remove the flocs that were produced during the process. The concentration of different parameters in the end-treatment effluent, that is, the treated wastewater at pH 6.5 and free of flocs, are shown in Table 4.

It should be noted that, after 24 h, the Fenton reaction continued and COD was reduced by 80.5%. Even so, the remaining COD was high ( $375 \text{ mg}\cdot\text{L}^{-1}$ ). The concentration of hydrogen peroxide in the end-treatment effluent was  $79.7 \text{ mg}\cdot\text{L}^{-1}$ . Higher COD reduction may be obtained using the Fenton process if the reagents are optimized but, generally, the required concentration of iron and hydrogen peroxide increase notably if the Fenton process is used rather than the photo-Fenton [45].

The concentration of iron at the end of the coagulation, Fenton and photo-Fenton processes was lower than  $0.36 \text{ mg}\cdot\text{L}^{-1}$  for samples at pH 6.5. It should be noted that, in Spain, no more than  $10 \text{ mg}\cdot\text{L}^{-1}$  iron can be discharged with treated wastewater effluents, and no more than  $2 \text{ mg}\cdot\text{L}^{-1}$  is allowed for water reuse.

Although TP was below the limit for water reuse in the raw wastewater (see Table 1), we observed that this parameter was removed from the wastewater with coagulation, Fenton and also photo-Fenton. It is well known that the phosphonates contained in detergent formulations adsorb on the metal hydroxide flocs that are formed in the Fe(III) coagulation process and, thus, phosphonates are removed from wastewaters by sedimentation of the formed flocs [46]. Due to the speciation of iron in water and the zeta potential of the flocs formed at different pH values, this process occurs preferentially at pH values between 4.5 and 7.0 [46,47].

Lastly, the TN present in the raw wastewater ( $36 \text{ mg}\cdot\text{L}^{-1}$ ) was mainly due to organic nitrogen. Nitrate was present at less than  $10 \text{ mg}\cdot\text{L}^{-1}$  and nitrite at less than  $1 \text{ mg}\cdot\text{L}^{-1}$ . Removal of TN was over 72% during the photo-Fenton process. The TN in the effluent from the photo-Fenton process was less than  $10 \text{ mg}\cdot\text{L}^{-1}$ , and the ammonia and nitrate concentrations complied with legislation for wastewater reuse. Regarding these results, we conclude that nitrogen removal was attained due to

Table 4

Concentration of different parameters after the treatment of laundry wastewater using coagulation, Fenton or photo-Fenton.

Parameter	Concentration in end-treatment effluent		
	Coagulation	Fenton	Photo-Fenton
COD ( $\text{mg}\cdot\text{L}^{-1}$ )	1371	375	142
Turbidity (NTU)	473	32	< 10
Total Fe ( $\text{mg}\cdot\text{L}^{-1}$ )	< 0.36	< 0.36	< 0.36
Total phosphorous ( $\text{mg}\cdot\text{L}^{-1}$ )	< 0.1	< 0.1	< 0.1
Hydrogen peroxide ( $\text{mg}\cdot\text{L}^{-1}$ )	-	79.7	< 1.45

precipitation of nitrogen-containing compounds in the raw wastewater. In this sense, it is known that diethanolamines are frequently employed in detergents [48]. These show average biodegradability [49], but the Fenton process has been shown to increase the biodegradability of these compounds [50].

Regarding COD and turbidity, photo-Fenton was the only process that met the requirements of the legislation for water reuse (COD < 160 mg·L<sup>-1</sup> and turbidity < 10 NTU, as shown in Table 1).

The concentration of hydrogen peroxide in the treated water (at pH 6.5) was 1.2 mg·L<sup>-1</sup>; therefore, 99.9% of the initial concentration was consumed during the photo-Fenton reaction. Additionally, TP was completely removed during this process.

In summary, from the study of different physico-chemical processes to treat laundry wastewater we found that a photo-Fenton process (natural sunlight could be employed) can achieve high COD, TP and turbidity removal with an almost complete consumption of hydrogen peroxide at the end of the process.

Other parameters not included in Table 4 that need to be analysed to determine whether the treated water can be reused in irrigation are suspended solids and the concentration of surfactants. We determined the concentration of these in the end-treatment effluent for the photo-Fenton process.

Regarding surfactants, LAS removal was 82.4% (less than 2 mg·L<sup>-1</sup> remained in the treated water). BiAS removal was over 98.7% (less than 0.5 mg·L<sup>-1</sup> remained in the treated water).

Suspended solids in the supernatant were lower than 20 mg·L<sup>-1</sup> and, thus, the treated water complied in all parameters for water reuse according to Spanish legislation.

On the other hand, the sedimentable solids produced during the photo-Fenton process were 16 mL·L<sup>-1</sup>. It should be noted that this sludge contains almost all the iron used and must be retired and disposed or further treated by a waste management company.

### 3.4. Biological treatment

Fig. 5 shows the removal of COD in the BF with time. It should be noted that at least 24 h are needed to obtain removals above 60%. COD removal was 75% after 40 h. Increasing the reaction time does not produce additional COD removal, as can be seen in Fig. 5. Turbidity was completely removed (over 99% after 24 h) during the biological treatment. The TP concentration was 2 mg·L<sup>-1</sup> after 40 h (37% removal). LAS removal was 98% after 40 h (0.2 mg·L<sup>-1</sup> remained in the treated effluent) and BiAS removal was over 98.7% (less than 0.5 mg·L<sup>-1</sup> remained in the treated water).

It is known that during aerobic wastewater treatment, microbial communities are greatly influenced by nutritional conditions. At the beginning and end of the treatment, TN was 97 and 22 mg·L<sup>-1</sup>, respectively. Consequently, 77.3% of TN was removed during the process. The COD:N ratio was 20:1 throughout the process, which is, in general, an acceptable ratio [51]. The effect of COD:N ratios on the

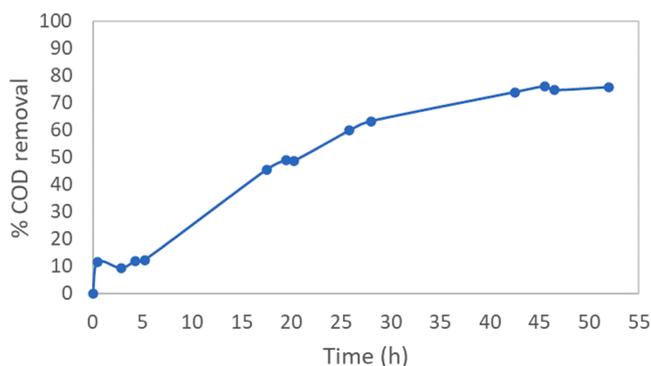


Fig. 5. COD removal for laundry wastewater treated with a batch BF.

performance of wetlands also depends on the type of organic matter found in the wastewater. Generally, high COD:N (10:1–20:1) ratios have proven to increase the diversity of the microbial community and be beneficial in denitrification processes [51,52].

Removal of TP was 42.3%, with a concentration in the effluent of 2 mg·L<sup>-1</sup>. The N:P ratio was 30:1 at the beginning of the water treatment and 11:1 at the end of the process.

Although the required treatment time for the BF was very long compared to those needed for the physicochemical processes, it should be noted that the required time in this study (40 h) was much shorter than the 14 days needed by Lutterbeck et al. for hospital laundry wastewater treatment [15]. This could be attributed to the aeration effect of the recirculation. Unlike the other methods tested, the BF reactor did not use any type of chemical reagent, and its only expense was that of the recirculation pump.

However, it was observed that COD reduction was 15% lower for the BF system compared to the photo-Fenton process. COD remained over 450 mg·L<sup>-1</sup> in the effluent of the BF after 40 h. The BF alone was not enough to reduce COD to acceptable values for water reuse, although the treated water complies with the legislation for disposal (see Table 1). Similarly, the concentration of *E. coli* in the BF effluent was higher than 200 FCU/100 mL, which is the established limit for water reuse (see Table 1). Therefore, the BF process needs to be assisted with a post-treatment for water to be reused. Even though, it must be noted that other authors have reported that biological treatments applied to laundry wastewater, such as a combination of microbial fuel cells and constructed wetlands, enable significant reductions of genotoxicity [53].

In order to remove the bacteria and reduce COD to meet the required standards for water reuse, the effluent from the biofilter was filtered (0.45 μm) and further treated with granulated activated carbon (GAC) under continuous stirring. The concentration of activated carbon employed in the experiences varied between 1 and 20 g·L<sup>-1</sup>.

The COD removal for different activated carbon loads is shown in Fig. 6. Results show that adsorption of COD and the consequent COD removal increases with activated carbon loads up to 15 g·L<sup>-1</sup>.

Activated carbon has been reported as posttreatment in other studies to treat laundry wastewater. Most reports use activated carbon after a coagulation or electrocoagulation processes to increase COD removal [54,55], although some studies also report the use of this adsorbent after a biological and membrane process [56]. The use of activated carbon on its own to treat laundry wastewater has also been studied, but this resulted in low COD removal (41.9% for a raw wastewater with 240.8 mg·L<sup>-1</sup> COD) [57].

The concentration of different parameters in the treated wastewater (end-treatment effluent) for the BF alone and the BF + GAC treatment system are shown in Table 5.

It should be noted that, for the BF + GAC system, the other parameters included in Table 1 but not in Table 5 were all acceptable for

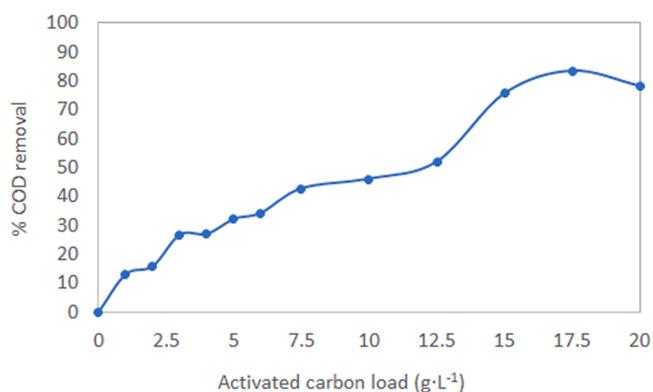


Fig. 6. COD removal for the BF effluent after adsorption with granular activated carbon for 24 h.

**Table 5**

Concentration of different parameters after the treatment of laundry wastewater using BF (40 h) and BF followed by adsorption on 15 g·L<sup>-1</sup> activated carbon (GAC).

Parameter	Concentration in end-treatment effluent	
	BF	BF + GAC
COD (mg·L <sup>-1</sup> )	450	103
Turbidity (NTU)	< 10	< 10
Total phosphorous (mg·L <sup>-1</sup> )	2	-

wastewater reuse in irrigation.

### 3.5. Degradation by-products

Although both the photo-Fenton and BF systems yield high COD removals, the corresponding degradation pathways and remaining by-products differ.

In this sense, it is known that some fragrance compounds present resistance to biodegradation, although limonene and linalool, which are those present in the detergent considered in this study, can be removed during biological treatments, as reported by other authors [16].

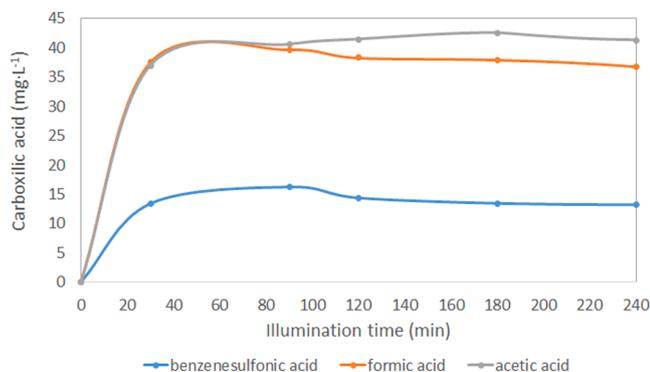
However, the detergent used in the laundry considered in this study contains fluorescent whitening agents (FWA), and these have been reported to be recalcitrant to biodegradation, although they undergo photochemical degradation [58]. Thus, these products may be present in the effluent from the biological process but probably not in the photo-Fenton effluent.

The metabolic pathway for LAS degradation produces n-alkanes, benzoate, catechol and carboxylic acids, while the metabolic pathway for LAE degradation produces n-alkanes, polyethylene glycol, some carboxylic acids and ethanol [14].

It is known that the main degradation products formed when photo-Fenton is used are carboxylic acids. In this sense, the degradation of LAS by means of hydrogen peroxide has proven to give hydroxylic derivatives, hydroxy acids of benzenesulfonic acid and aliphatic aldehydes [59].

In this work, we monitored the formation of some carboxylic acids which were detected through HPLC. The detected intermediates were benzenesulfonic, acetic and formic acids for the photo-Fenton reaction, and formic acid for the BF process. The results for the photo-Fenton process are shown in Fig. 7.

Benzenesulfonic acid is the simplest aromatic sulfonic acid that is produced as a by-product of LAS. Maleic acid should next form from the opening of the aromatic ring. In the photo-Fenton process, further hydroxyl attack produces lower chain carboxylic acids, such as acetic or formic acids. The reaction rate between acetic acid and hydroxyl radicals has been reported to be low [60]. For this reason, we see that acetic acid is produced during the first 20 min of illumination and then



**Fig. 7.** Evolution of benzenesulfonic, formic and acetic acids during the illumination stage of the treatment of laundry wastewater with photo-Fenton.

accumulates.

Formic acid is produced and slowly consumed in the photo-Fenton process, as can be seen in Fig. 7. In this sense, formic acid has been reported to react with hydroxyl radicals to form formate radicals, that can react with oxygen to produce CO<sub>2</sub> or with hydrogen peroxide to form formic acid again [60]. Formic acid is totally consumed when the effluent from the photo-Fenton reaction is left to settle for 24 h. This indicates that a Fenton reaction continues once the UV illumination is stopped.

The end-treatment effluent contained 38.05 mg·L<sup>-1</sup> acetic acid and 14.26 mg·L<sup>-1</sup> benzenesulfonic acid, which correspond to 40.59 mg·L<sup>-1</sup> and 21.66 mg·L<sup>-1</sup> COD, respectively. This represents 43.83% of the COD that remains after the photo-Fenton process. Specifically, 28.58% of the COD in the effluent corresponds to acetic acid and 15.25% to benzenesulfonic acid.

Regarding the biological system, of the analysed carboxylic acids, formic acid was the only one found in the BF effluent. Formic acid was present in a concentration of 4.68 mg·L<sup>-1</sup>, which corresponds to just 0.36% of total COD.

The authors believe that further future work, which is beyond the scope of this study, is needed to study in detail the by-products that remain in the treated water and their toxicity.

### 3.6. Biological & activated carbon treatment vs. photo-Fenton

A comparison of the COD and turbidity results obtained from the photo-Fenton and the BF + GAC processes is shown in Table 6.

As described in the above sections, the rest of the parameters required for water reuse were acceptable.

Based on the results shown above and given that the aim of this work is the treatment of laundry wastewater for reuse in irrigation, we present an economic evaluation and LCA analysis for the photo-Fenton and BF + GAC options.

### 3.7. Economic evaluation

The flow diagrams of the optimized processes taken into consideration for the economic evaluation are shown in Fig. 8.

For the photo-Fenton process (see Fig. 8a), the laundry wastewater is introduced into a UV reactor equipped with a mechanical stirrer (R1). With help of a pH controller and dosing pump (P1), the pH of the wastewater needs to be regulated to 2.8. According to the amount of H<sub>2</sub>SO<sub>4</sub> (96% purity) needed in the laboratory experiments, we estimate that approximately 2 L of this solution is required. Next, to treat 1 m<sup>3</sup> of wastewater, 6.5 L hydrogen peroxide 30% w/v and 1.53 kg iron (III) sulphate pentahydrate must be added. At this moment, the UV lamps are connected. According to the experimental conditions and results, the necessary energy for the process is 25.92 kJ·L<sup>-1</sup> (25920 kJ·m<sup>-3</sup>), as calculated from Eq. (14). This is equivalent to 7.2 kWh·m<sup>-3</sup>.

$$Q_{UV} = \frac{A_r \cdot UV_{GL} \cdot t}{V} \quad (14)$$

where:

**Table 6**

Concentration of different parameters after the treatment of laundry wastewater using photo-Fenton or the biological batch reactor.

Parameter	Concentration in end-treatment effluent		Acceptable for reuse? <sup>a</sup>	
	Photo-Fenton <sup>b</sup>	BF + GAC	Photo-Fenton	BF + GAC
COD (mg·L <sup>-1</sup> )	142	103	Yes	Yes
Turbidity (NTU)	< 10	< 10	Yes	Yes

<sup>a</sup> According to Royal Decree 1620/2007 (Spain). Reuse for irrigation of urban green areas.

<sup>b</sup> Sample at pH 6,5 to meet the pH requirements for water reuse (see Table 1).

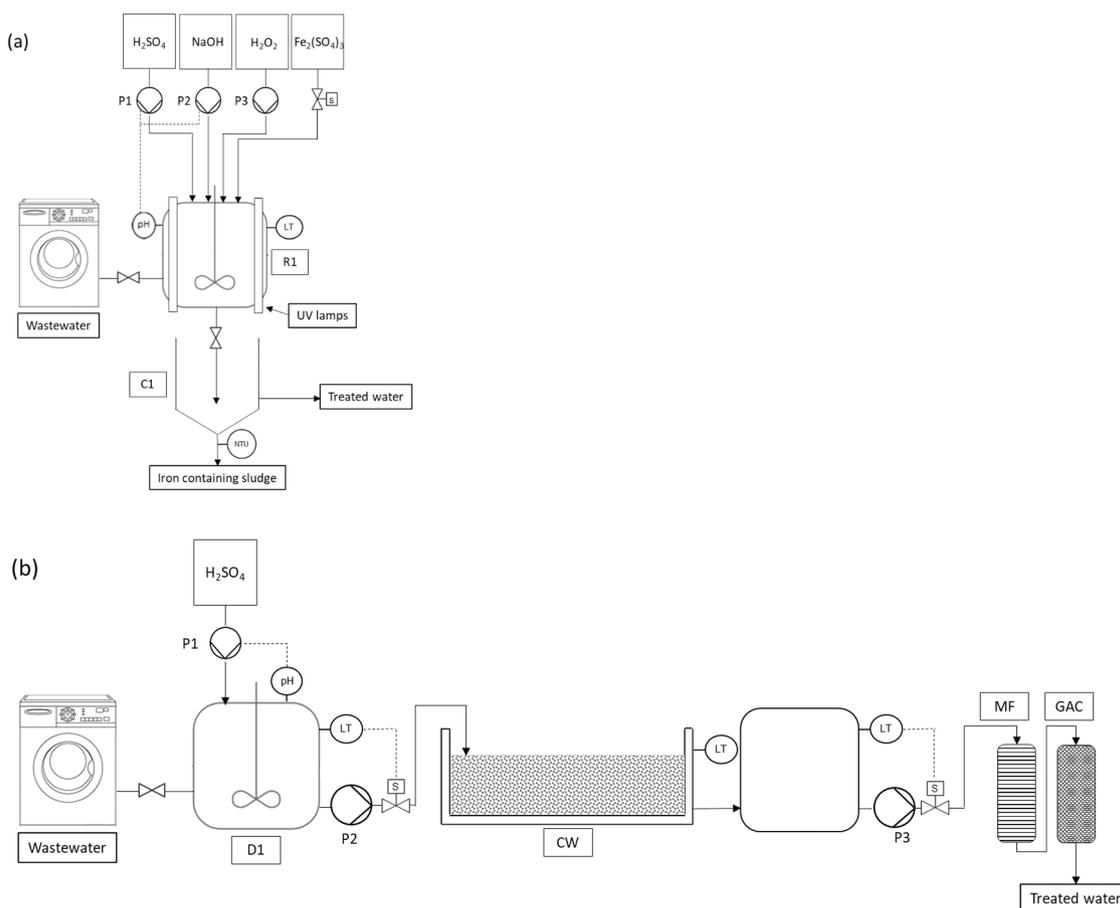


Fig. 8. Flow diagrams for laundry wastewater treatment using (a) photo-Fenton and (b) BF + GAC.

- $Q_{UV}$  ( $J \cdot L^{-1}$ ) is the energy required in the photo-Fenton process to guarantee a COD removal equivalent to that obtained in the laboratory for the optimized process
- $A_r$  is the UV catchment area ( $m^2$ ), which was  $0.02 m^2$  for the laboratory 1 L batch reactors
- $UV_{GL}$  ( $W \cdot m^{-2}$ ) is the irradiance, which was  $90 W \cdot m^{-2}$  for the lamps used in the laboratory experiences
- $t$  is the reaction time (s), which was 240 min (14400 s) for the optimized photo-Fenton reaction studied in the laboratory

After completing the reaction, the pH must be regulated to 6.5 (using pump P2) to comply with the legislation for wastewater reuse. We estimate that about 0.5 L of a NaOH 10 M solution is needed in this operation. Lastly, the treated water is introduced by gravity into a sedimentation tank (C1). Here, the flocs form during the reaction settle and are separated from the treated water. As mentioned above, the amount of sludge produced in the process is 16 L per  $m^3$  of treated wastewater.

For the BF + GAC option (see Fig. 8b), the wastewater is first introduced into a tank where the pH is regulated to 7 (P1). We estimate that about 0.5 L of  $H_2SO_4$  (96% purity) per  $m^3$  of wastewater is enough for this purpose. Next, the wastewater is recirculated in the BF for an HRT of 40 h, which was the optimized HRT found in the laboratory experiments. After this, the wastewater flows into a water tank and is then pumped through a microfiltration cartridge ( $0.45 \mu m$ ) and a GAC filter (15 kg per  $m^3$  of treated wastewater).

In this study, we calculated the operating costs for the photo-Fenton and BF + GAC systems when employed to treat  $1 m^3$  of wastewater. The operating costs are divided into cost of reagents, waste disposal, energy consumption and maintenance.

Reagent costs were calculated from a mean price from several local providers and considering the optimized reagent amounts mentioned above.

Electricity costs were estimated for the UV lamps (photo-Fenton process) and the filtration pump (P3) (BF + GAC process). For the P3 pump, we considered that the necessary energy was  $0.28 kWh \cdot m^{-3}$  [61]. The electricity consumption of the remaining electrical equipment was considered negligible, and, for this reason, was not included in the analysis. The mean cost of a kWh was taken as  $0.18 \text{ €} \cdot kWh^{-1}$ , as reported by Eurostat for the first 6 months of 2022 [62].

Lastly, maintenance costs were estimated to be  $0.05 \text{ €} \cdot m^{-3}$ , as calculated in [63] for several wastewater treatment plants. The summary of the total costs is given in Table 7.

The total operating cost was  $6.71 \text{ €} \cdot m^{-3}$  for the photo-Fenton system and  $0.71 \text{ €} \cdot m^{-3}$  for the BF + GAC option. Considering the COD removed in each process, the cost was  $3.77 \text{ €} \cdot CODr^{-1}$  for the photo-Fenton option and  $0.39 \text{ €} \cdot CODr^{-1}$  for the BF + GAC. CODr refers to the  $kg \cdot m^{-3}$  COD removed in each process.

For the BF + GAC alternative, reagents and GAC replacement amounted to 76% of the total cost. For the photo-Fenton alternative, over 75% of the total cost was due to the acquisition of the necessary reagents and almost 20% to the use of UV lamps. Among the cost of the reagents, 39.5% (29.8% of the total cost) was due to the acquisition of acid and base, and 60.5% (45.6% of the total cost) was due to the iron salt and hydrogen peroxide.

The cost of applying photo-Fenton systems depends mainly on Fe(III) and hydrogen peroxide costs. Additionally, the UV source also influences the total operating cost. In this sense, the total operating cost for photo-Fenton, for different wastewaters, can vary between  $2.06 \text{ €} \cdot m^{-3}$  for solar systems and  $10.36 \text{ €} \cdot m^{-3}$  for systems where UV lamps are

**Table 7**  
Estimated operating costs for the treatment of 1 m<sup>3</sup> of wastewater.

	€/m <sup>3</sup> of wastewater	
	Photo-Fenton	BF + GAC
<b>Reagents</b>		
Iron (III) sulphate	0.72	-
Hydrogen peroxide	2.34	-
Sulphuric acid and sodium hydroxide for pH regulation	2.00	0.34
Total reagents	5.06	0.34
<b>Filtering system</b>		
GAC replacement <sup>a</sup>	-	0.20
<b>Waste disposal</b>		
Sludge/GAC	0.26	0.12
<b>Energy</b>		
Electricity (pumping system)	-	0.05
Electricity (UV lamps)	1.30	-
<b>Maintenance</b>	0.05	0.05
<b>TOTAL OPERATIONAL COST</b>	<b>6.71</b>	<b>0.71</b>

<sup>a</sup> This data is based on other references in which GAC was used for the removal of TOC and micropollutants in treated grey water [64,65].

employed [66,67].

In Spain, the cost of treating 1 m<sup>3</sup> of water in municipal conventional wastewater treatment plants (for wastewater disposal) was 0.80 €·m<sup>-3</sup> and the cost of potable water production was 1.14 €·m<sup>-3</sup> in 2020 [68]. The cost of the photo-Fenton treatment studied in this work as a technique for laundry wastewater reuse is almost 6 times higher than the cost of potable water. The purchase of reagents and electricity consumption are responsible for the higher cost of the photo-Fenton process. However, the estimated cost of the BF + GAC process aimed at laundry wastewater reuse was 11% lower than the cost of conventional wastewater treatment in Spain in 2020. Thus, we can affirm that the BF + GAC system is currently a cost-effective alternative for laundry wastewater treatment and reuse.

A few studies have provided an economic evaluation of the treatment of laundry wastewater. Among the techniques with a reported cost assessment, we find coagulation [69], electrocoagulation [70], adsorption on activated carbon [2], ultrafiltration/reverse osmosis [71], membrane bioreactors [72] and combined systems, such as coagulation, ozonation, adsorption on activated carbon and ultrafiltration [69], or biological treatment, UV/O<sub>3</sub> and filtration [73]. The reported cost per cubic meter of treated water varies from 0.09 to 1.50 €·m<sup>-3</sup> and from 0.89 to 4.87 €·CODr<sup>-1</sup>. The highest cost was reported for the ultrafiltration/reverse osmosis system. The BF + GAC process studied in this work (0.39 €·CODr<sup>-1</sup>) is found to be the economically most feasible option when considering the cost in €·CODr<sup>-1</sup> units. However, it should be noted that the referenced studies were published in different years, indeed in some cases in different decades, and considerable care therefore needs to be taken when comparing costs due to possible differences in electricity and material prices.

### 3.8. LCA analysis

For the LCA analysis, we followed standards ISO 14040 and ISO 14044. The chosen functional unit was the treatment of 1 m<sup>3</sup> of wastewater. System boundaries were restricted to the operational phase, namely the consumption of reagents, GAC and electricity. The possible regeneration of GAC was not included in the analysis. The wastewater inventory included COD, total surfactants and total phosphorous, according to the experimental data given in this work. The waste disposal of each process was also considered. These were iron hydroxide sludge for the photo-Fenton and used activated carbon for the BF + GAC.

OpenLCA (version 1.10.3) was used as LCA software. For the life cycle inventory (LCI) we used product environmental footprint (PEF) secondary datasets [74]. We used the environmental footprint 2.0

midpoint Life Cycle Impact Assessment (LCIA) methodology [75].

The detailed inputs and outputs, according to the experimental results obtained in this work using photo-Fenton and BF + GAC are shown in Tables 8 and 9, respectively.

Table 10 shows the indicators that were used to evaluate the impacts in this work and the LCA results for the treatment of 1 m<sup>3</sup> of laundry wastewater. The main impacts concern climate change, ecotoxicity (freshwater) and fossil fuel resource use.

The corresponding results for the two processes are shown in Fig. 9. It should be noted that photo-Fenton has a higher impact than BF + GAC on all indicators except for eutrophication and human toxicity (non-cancer).

The relative contributions of each input (reagents and electricity) are shown in Fig. 10 for the two processes. In general, the production of the necessary reagents for the process is the factor with the highest impact on most indicators. Electricity has a high impact on acidification, climate change, photochemical ozone formation and resource use (fossils). In this analysis, electricity was considered to be produced from fossil fuels (fuel oil). The impact on the aforementioned indicators could be reduced if renewable energy sources were used.

This is in line with the very few reports on laundry wastewater treatment that include an LCA analysis. Other authors have concluded that processes with higher electricity consumption (when using non-renewable sources) have higher environmental impacts [23,78].

## 4. Conclusions

In this work, we studied and compared different options for the treatment of laundry wastewater. This type of wastewater normally presents high turbidity, pH and organic matter. Moreover, detergent formulations often include several substances, including anionic and non-ionic surfactants, FWAs, bleaching agents, etc., that may make treatment of the wastewater more difficult.

Although EU regulations have limited the amount of phosphorous in detergents and promoted the biodegradability of these formulations, we found that a biological treatment alone, using a BF, was not enough to remove the necessary COD to reuse the treated water. The BF was able to reduce turbidity and the concentration of anionic and non-ionic surfactants by almost 100%, total phosphorous by 42.3% and COD by almost 80%. However, the final COD found in the effluent was

**Table 8**  
Inputs and outputs for LCA analysis for the treatment of 1 m<sup>3</sup> of laundry wastewater using photo-Fenton.

Parameter	Input	Output	Ecoinvent unit process
COD (kg)	1.92	0.142	
Total phosphorous (kg)	0.003	0.0001	
Total detergents (kg)	0.047	0.002	
Iron sulphate (kg)	0.95	-	Adapted from: iron (II) sulphate production, production mix, at plant, technology mix, 100% active substance, as Fe <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> can be produced from the oxidation of FeSO <sub>4</sub> with sulphuric acid [76]
Hydrogen peroxide 50% (kg)	1.17	-	Hydrogen peroxide, 50% production, production mix, at plant, technology mix, 100% active substance
Sulphuric acid (kg)	3.51	-	Sulphuric acid production, production mix, at plant, technology mix, 100% active substance
Sodium hydroxide (kg)	0.20	-	Sodium hydroxide production, production mix, at plant, technology mix, 100% active substance
Sludge (kg)	-	16	
Electricity (kWh)	7.2	-	Electricity from heavy fuel oil (HFO), production mix, at power plant, AC
Wastewater (m <sup>3</sup> )	1	0.84	

**Table 9**

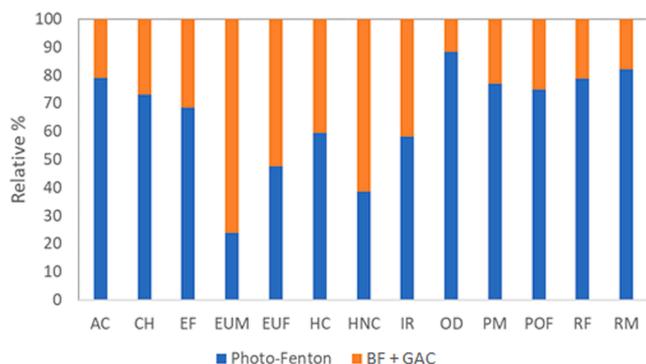
Inputs and outputs for LCA analysis for the treatment of 1 m<sup>3</sup> of laundry wastewater using BF + GAC.

Parameter	Input	Output	Ecoinvent unit process
COD (kg)	1.92	0.103	
Total phosphorous (kg)	0.003	0.002	
Total detergents (kg)	0.047	0.002	
GAC (kg)	15	-	Supposing replacement. Production process configured taking GAC from coconut shell as reference[77].
Waste (replaced GAC) (kg)	-	15	
Sulphuric acid (kg)	0.88	-	Sulphuric acid production, production mix, at plant, technology mix, 100% active substance
Electricity (kWh)	0.28	-	Electricity from heavy fuel oil (HFO), production mix, at power plant, AC
Wastewater (m <sup>3</sup> )	1	≈ 1	

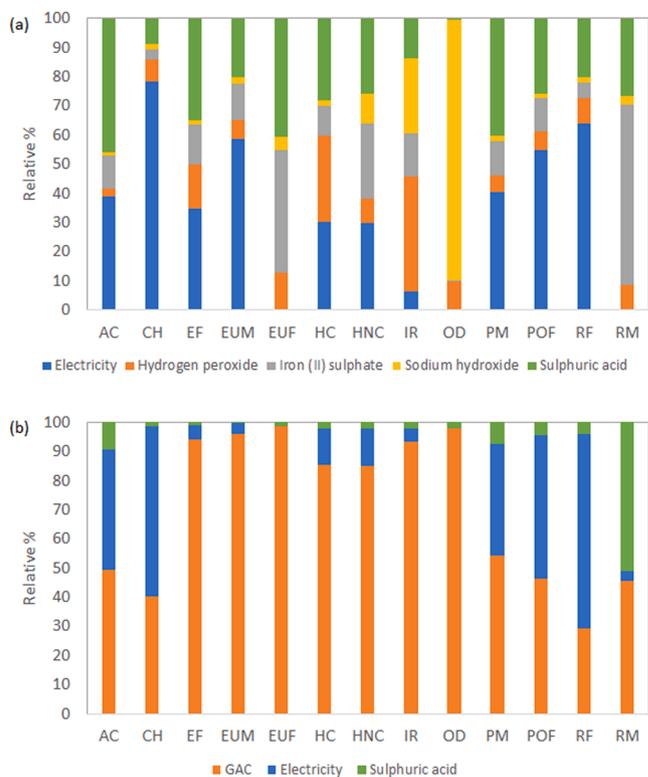
**Table 10**

Contributions of the photo-Fenton process and the BF + GAC process to the selected indicators using the environmental footprint 2.0 midpoint LCIA method.

Impact category	Reference unit	Photo-Fenton	GAC + BF
Acidification (AC)	mol H <sup>+</sup> eq.	0.45	0.12
Climate change (CH)	kg CO <sub>2</sub> eq.	53.2	19.5
Ecotoxicity, freshwater (EF)	CTUe	23.1	10.6
Eutrophication, marine (EUM)	kg N eq.	0.041	0.13
Eutrophication, freshwater (EUF)	kg P eq.	2.85·10 <sup>-3</sup>	3.15·10 <sup>-3</sup>
Human toxicity, cancer (HC)	CTUh	8.11·10 <sup>-7</sup>	5.56·10 <sup>-7</sup>
Human toxicity, non-cancer (HNC)	CTUh	3.40·10 <sup>-6</sup>	5.41·10 <sup>-6</sup>
Ionising radiation, human health (IR)	kBq U <sup>235</sup>	1.21	0.87
Ozone depletion (OD)	kg CFC11 eq.	9.22·10 <sup>-7</sup>	1.21·10 <sup>-7</sup>
Particulate matter (PM)	Item(s)	3.43·10 <sup>-6</sup>	1.02·10 <sup>-6</sup>
Photochemical ozone formation (POF)	kg non-methane volatile organic compounds (NMVOC) eq.	0.15	0.05
Resource use, fossils (RF)	MJ	798.73	217.01
Resource use: minerals and metals (RM)	kg Sb eq.	2.42·10 <sup>-4</sup>	5.24·10 <sup>-5</sup>



**Fig. 9.** Relative contribution of the photo-Fenton or GAC + BF process to the indicators considered in the LCA assessment.



**Fig. 10.** Relative contributions of the inputs (reagents and electricity) of the (a) photo-Fenton and (b) BF + GAC processes to the selected indicators using the environmental footprint 2.0 midpoint LCIA method.

450 mg·L<sup>-1</sup> and regulations in Spain limit COD to 160 mg·L<sup>-1</sup> for wastewater reuse. Therefore, this effluent needs to undergo a post-treatment to further eliminate the remaining organic matter. The chosen post-treatment consisted of microfiltration and GAC filtration. The BF + GAC system achieved higher COD reduction, with final COD concentrations in the effluent of 103 mg·L<sup>-1</sup>.

Coagulation with iron (III) sulphate, Fenton and photo-Fenton processes were also studied to treat the raw wastewater. We found that coagulation and Fenton were not suitable to meet the water reuse standards. However, when photo-Fenton was applied, the resulting wastewater met the criteria for water reuse in all parameters. We found that 43.83% of the COD in the filtered effluent was due to the presence of short-chain carboxylic acids.

Organic matter remains after both the BF + GAC and the photo-Fenton plus filtration processes. The by-products in the final effluents should be studied in depth in future works to enable a detailed impact assessment of both options.

Following the experimental results, we also carried out an economic and LCA analysis of the photo-Fenton option and found that the total operating costs were 6.71 €·m<sup>-3</sup> for the photo-Fenton option and 0.71 €·m<sup>-3</sup> for the BF + GAC. About 75% of the cost can be attributed to the acquisition of reagents for both processes. The LCA analysis revealed that the contribution to environmental impacts was, in general, much lower for the BF + GAC system and that the main impacts were also caused by the reagents used in the processes.

#### Ethics approval and consent to participate

Not applicable.

#### Consent for publication

Not applicable.

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## Author contributions

All authors contributed to the study conception and design. Material preparation was performed by all authors. Data collection and analysis were performed by E. Leon, E. Pulido Melián, J. Vaswani Reboso and Dunia E. Santiago. The first draft of the manuscript was written by Dunia E. Santiago and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

## Declaration of Competing Interest

The authors have no relevant financial or non-financial interests to disclose.

## Data Availability

Data will be made available on request.

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## References

- Federatie van de Belgische Textielverzorging, Sustainable Measures for Industrial Laundry Expansion Strategies: SMART LAUNDRY-2015, CORDIS EU Research Results. (2015) 1. (<https://cordis.europa.eu/project/id/217809/es>) (accessed June 7, 2021).
- M.C. Collivignarelli, M. Carnevale Miino, M. Baldi, S. Manzi, A. Abbà, G. Bertanza, Removal of non-ionic and anionic surfactants from real laundry wastewater by means of a full-scale treatment system, *Process Saf. Environ. Prot.* 132 (2019) 105–115, <https://doi.org/10.1016/j.psep.2019.10.022>.
- E. Commission, J.R. Centre, R. Kaps, A. Boyano, E. Sims, J. Arendorf, R. Menkveld, A. Gaasbeek, L. Golsteijn, K. Bojczuk, G. Medina, Revision of the European ecolabel criteria for laundry detergents and industrial and institutional laundry detergents: preliminary report, European Commission, Jt. Res. Cent. (2015) <https://doi.org/10.2791/0171>.
- European Commission, Evolution of products per product group (2014 - 2022), (2022). ([https://environment.ec.europa.eu/topics/circular-economy/eu-ecolabel-home/business/ecolabel-facts-and-figures\\_en](https://environment.ec.europa.eu/topics/circular-economy/eu-ecolabel-home/business/ecolabel-facts-and-figures_en)) (accessed December 24, 2022).
- A. Godayol, E. Besalú, E. Anticó, J.M. Sanchez, Monitoring of sixteen fragrance allergens and two polycyclic musks in wastewater treatment plants by solid phase microextraction coupled to gas chromatography, *Chemosphere* 119 (2015) 363–370, <https://doi.org/10.1016/j.chemosphere.2014.06.072>.
- A.C. Ahmia, M. Idouhar, O. Arous, K. Sini, A. Ferradj, A. Tazerouti, Monitoring of anionic surfactants in a wastewater treatment plant of algiers western region by a simplified spectrophotometric method, *J. Surfactants Deterg.* 19 (2016) 1305–1314, <https://doi.org/10.1007/s11743-016-1884-x>.
- C.F. Granatto, G.M. Grosselli, I.K. Sakamoto, P.S. Fadini, M.B.A. Varesche, Influence of cosubstrate and hydraulic retention time on the removal of drugs and hygiene products in sanitary sewage in an anaerobic Expanded Granular Sludge Bed reactor, *J. Environ. Manag.* 299 (2021), <https://doi.org/10.1016/j.jenvman.2021.113532>.
- L.T. Le, K.Q.N. Nguyen, P.T. Nguyen, H.C. Duong, X.T. Bui, N.B. Hoang, L. D. Nghiem, Microfibers in laundry wastewater: problem and solution, *Sci. Total Environ.* 852 (2022), <https://doi.org/10.1016/j.scitotenv.2022.158412>.
- J. Blasco, M. Hampel, I. Moreno-Garrido, Toxicity of Surfactants, in: *Analysis and Fate of Surfactants and the Aquatic Environment*, Elsevier, 2003, pp. 827–925, [https://doi.org/10.1016/S0166-526X\(03\)40010-X](https://doi.org/10.1016/S0166-526X(03)40010-X).
- S.O. Badmus, H.K. Amusa, T.A. Oyeohan, T.A. Saleh, Environmental risks and toxicity of surfactants: overview of analysis, assessment, and remediation techniques, *Environ. Sci. Pollut. Res.* 28 (2021) 62085–62104, <https://doi.org/10.1007/s11356-021-16483-w/Published>.
- M. Stuart, D. Lapworth, E. Crane, A. Hart, Review of risk from potential emerging contaminants in UK groundwater, *Sci. Total Environ.* 416 (2012) 1–21, <https://doi.org/10.1016/j.scitotenv.2011.11.072>.
- D.E. Santiago, M.J. Hernández-Rodríguez, E. Pulido Melián, Laundry wastewater treatment: review and life cycle assessment, *J. Environ. Eng.* 147 (2021), 3121001, [https://doi.org/10.1061/\(ASCE\)EE.1943-7870.0001902](https://doi.org/10.1061/(ASCE)EE.1943-7870.0001902).
- S.A. Mousavi, F. Khodadoost, Effects of detergents on natural ecosystems and wastewater treatment processes: a review, *Environ. Sci. Pollut. Res.* 26 (2019) 26439–26448, <https://doi.org/10.1007/s11356-019-05802-x>.
- F. Motteran, B.M. Nadai, J.K. Braga, E.L. Silva, M.B.A. Varesche, Metabolic routes involved in the removal of linear alkylbenzene sulfonate (LAS) employing linear alcohol ethoxylated and ethanol as co-substrates in enlarged scale fluidized bed reactor, *Sci. Total Environ.* 640–641 (2018) 1411–1423, <https://doi.org/10.1016/j.scitotenv.2018.05.375>.
- C.A. Lutterbeck, Ê.L. Machado, A. Sanchez-Barrios, E.O. Silveira, D. Layton, A. Rieger, E.A. Lobo, Toxicity evaluation of hospital laundry wastewaters treated by microbial fuel cells and constructed wetlands, *Sci. Total Environ.* 729 (2020), 138816, <https://doi.org/10.1016/j.scitotenv.2020.138816>.
- A. Godayol, E. Besalú, E. Anticó, J.M. Sanchez, Monitoring of sixteen fragrance allergens and two polycyclic musks in wastewater treatment plants by solid phase microextraction coupled to gas chromatography, *Chemosphere* 119 (2015) 363–370, <https://doi.org/10.1016/j.chemosphere.2014.06.072>.
- J. Blasco, M. Hampel, I. Moreno-Garrido, Toxicity of Surfactants, in: *Analysis and Fate of Surfactants and the Aquatic Environment*, Elsevier, 2003, pp. 827–925, [https://doi.org/10.1016/S0166-526X\(03\)40010-X](https://doi.org/10.1016/S0166-526X(03)40010-X).
- K. Jardak, P. Drogui, R. Daghrir, Surfactants in aquatic and terrestrial environment: occurrence, behavior, and treatment processes, *Environ. Sci. Pollut. Res.* 23 (2016) 3195–3216, <https://doi.org/10.1007/s11356-015-5803-x>.
- S. Bering, J. Mazur, K. Tarnowski, M. Janus, S. Mozia, A.W. Morawski, The application of moving bed bio-reactor (MBBR) in commercial laundry wastewater treatment, *Sci. Total Environ.* 627 (2018) 1638–1643, <https://doi.org/10.1016/j.scitotenv.2018.02.029>.
- A. Belén, E. García, K. Szyma, S. Mozia, Treatment of laundry wastewater by solar photo-Fenton process at pilot plant scale, *Environ. Sci. Pollut. Res.* 28 (2020) 8576–8584, <https://doi.org/10.1007/s11356-020-11151-x>.
- D.I. Kern, R. de, O. Schwaickhardt, G. Mohr, E.A. Lobo, L.T. Kist, Ê.L. Machado, Toxicity and genotoxicity of hospital laundry wastewaters treated with photocatalytic ozonation, *Sci. Total Environ.* 443 (2013) 566–572, <https://doi.org/10.1016/j.scitotenv.2012.11.023>.
- L.T. Kist, C. Albrech, Ê.L. Machado, Hospital laundry wastewater disinfection with catalytic photoozonation, *Clean. (Weinh.)* 36 (2008) 775–780, <https://doi.org/10.1002/clen.200700175>.
- C.A. Lutterbeck, G.S. Colares, N. Dell’Osbel, F.P. da Silva, L.T. Kist, Ê.L. Machado, Hospital laundry wastewaters: a review on treatment alternatives, life cycle assessment and prognosis scenarios, *J. Clean. Prod.* 273 (2020), 122851, <https://doi.org/10.1016/j.jclepro.2020.122851>.
- M. Vilve, A. Hirvonen, M. Sillanpää, Effects of reaction conditions on nuclear laundry water treatment in Fenton process, *J. Hazard Mater.* 164 (2009) 1468–1473, <https://doi.org/10.1016/j.jhazmat.2008.09.058>.
- M. Munoz, P. García-Muñoz, G. Pliego, Z.M. de Pedro, J.A. Zazo, J.A. Casas, J. J. Rodriguez, Application of intensified Fenton oxidation to the treatment of hospital wastewater: kinetics, ecotoxicity and disinfection, *J. Environ. Chem. Eng.* 4 (2016) 4107–4112, <https://doi.org/10.1016/j.jece.2016.09.019>.
- A. Belén, E. García, K. Szyma, S. Mozia, Treatment of laundry wastewater by solar photo-Fenton process at pilot plant scale, *Environ. Sci. Pollut. Res.* 28 (2020) 8576–8584, <https://doi.org/10.1007/s11356-020-11151-x>.
- J. De Laat, G.T. Le, B. Legube, A comparative study of the effects of chloride, sulfate and nitrate ions on the rates of decomposition of H<sub>2</sub>O<sub>2</sub> and organic compounds by Fe (II)/H<sub>2</sub>O<sub>2</sub> and Fe (III)/H<sub>2</sub>O<sub>2</sub>, 55 (2004) 715–723. (<https://doi.org/10.1016/j.chemosphere.2003.11.021>).
- J.J. Pignatello, E. Oliveros, A. MacKay, Advanced oxidation processes for organic contaminant destruction based on the fenton reaction and related chemistry, *Crit. Rev. Environ. Sci. Technol.* 36 (2006) 1–84, <https://doi.org/10.1080/10643380500326564>.
- S.H. Lin, C.M. Lin, H.G. Leu, Operating characteristics and kinetic studies of surfactant wastewater treatment by fenton oxidation, *Water Res.* 33 (1999) 1735–1741, [https://doi.org/10.1016/S0043-1354\(98\)00403-5](https://doi.org/10.1016/S0043-1354(98)00403-5).
- C.E. Hedrick, B.A. Berger, Extraction of anions using triphenylmethane dyes, *Anal. Chem.* 38 (1966) 791–793, <https://doi.org/10.1021/ac60238a036>.
- ISO, International Standard ISO 7875–2. Water quality - Determination of surfactants - Part 2: Determination of non-ionic surfactants using Dragendorff reagent, (1984) 8.
- L.G. Saywell, B.B. Cunningham, Determination of iron: colorimetric o-phenanthroline method, *Ind. Eng. Chem. Anal. Ed.* 9 (1937) 67–69, <https://doi.org/10.1021/ac50106a005>.
- R.M. Sellers, Spectrophotometric determination of hydrogen peroxide using potassium titanium(IV) oxalate, *Analyst* 105 (1980) 950–954, <https://doi.org/10.1039/an9800500950>.
- APHA-AWWA-WEF, Standard Methods for the Examination of Water and Wastewater, (2005).
- J. Steber, 3 - The Ecotoxicity of Cleaning Product Ingredients, in: I. Johansson, P.B. T.-H. for C. of S. Somasundaran (Eds.), *Handbook for Cleaning/Decontamination of Surfaces*, Elsevier Science B.V., Amsterdam, 2007: pp. 721–746. <https://doi.org/https://doi.org/10.1016/B978-044451664-0/50022-X>.
- T. V.K. Vijayaraghavan, G. Sivakumar, Application of plant based coagulants for waste water treatment, *Int. J. Adv. Eng. Res. Stud.* 1 (2011) 88–92.
- J. Dotto, M.R. Fagundes-Klen, M.T. Veit, S.M. Palácio, R. Bergamasco, Performance of different coagulants in the coagulation/flocculation process of textile wastewater, *J. Clean. Prod.* 208 (2019) 656–665, <https://doi.org/10.1016/j.jclepro.2018.10.112>.
- S. Deressa, H. Endale, D. Dadi, S.A. Kite, Removal of anionic surfactant from residential laundry wastewater using jackfruit (*Artocarpus heterophyllus*) seeds,

- Adv. Environ. Technol. 5 (2019) 47–53, <https://doi.org/10.22104/aet.2020.3841.1189>.
- [39] A.J.E. Kerslake, H.C. Schmitt, N.A. Thomas, B.J.E. Kerslake, H.C. Schmitt, N.A. Thomas, Iron Salts as Coagulants, 38 (1946) 1161–1169.
- [40] M. Muñoz, P. García-Muñoz, G. Pliego, Z.M. De Pedro, J.A. Zazo, J.A. Casas, J. Rodríguez, Application of intensified Fenton oxidation to the treatment of hospital wastewater: kinetics, ecotoxicity and disinfection, *J. Environ. Chem. Eng.* 4 (2016) 4107–4112, <https://doi.org/10.1016/j.jece.2016.09.019>.
- [41] S. Addelman, Statistics for experimenters, *Technometrics* 21 (1979) 387–388, <https://doi.org/10.1080/00401706.1979.10489788>.
- [42] H. Cüce, F.A. Temel, O.C. Yolcu, Modelling and optimization of Fenton processes through neural network and genetic algorithm, *Korean J. Chem. Eng.* 38 (2021) 2265–2278, <https://doi.org/10.1007/s11814-021-0867-4>.
- [43] R.C. Souza, T.L. da Silva, A.Z. dos Santos, C.R.G. Tavares, Wastewater treatment of hospital laundry by advanced oxidation process: UV/H<sub>2</sub>O<sub>2</sub>, *Eng. Sanit. Ambient.* 24 (2019) 601–611, <https://doi.org/10.1590/s1413-41522019092923>.
- [44] M. Abdollahi, A. Hosseini, Hydrogen Peroxide, in: P.B.T.-E. of T. (Third E. Wexler (Ed.), *Encyclopedia of Toxicology* (Third Edition), Academic Press, Oxford, 2014: pp. 967–970. <https://doi.org/https://doi.org/10.1016/B978-0-12-386454-3.00736-3>.
- [45] D.E. Santiago, O.M. Gonzalez-Diaz, F.J.A. Mesa, E.P. Melián, J.P. Peña, J.M. Doña-Rodríguez, Factorial experimental design of imazalil-containing wastewater to be treated by Fenton-based processes, *J. Photochem. Photobio. A Chem.* 353 (2018), <https://doi.org/10.1016/j.jphotochem.2017.11.038>.
- [46] E. Rott, R. Minke, H. Steinmetz, Removal of phosphorus from phosphonate-loaded industrial wastewaters via precipitation/flocculation, *J. Water Process Eng.* 17 (2017) 188–196, <https://doi.org/10.1016/j.jwpe.2017.04.008>.
- [47] S. Sun, S. Wang, Y. Ye, B. Pan, Highly efficient removal of phosphonates from water by a combined Fe(III)/UV/co-precipitation process, *Water Res.* 153 (2019) 21–28, <https://doi.org/10.1016/j.watres.2019.01.007>.
- [48] B. Wang, D.E. Amacher, M.H. Whittaker, Derivation of a No-Significant-Risk-Level (NSRL) for diethanolamine (DEA), *Regul. Toxicol. Pharmacol.* 68 (2014) 76–84, <https://doi.org/10.1016/j.yrtph.2013.11.009>.
- [49] A.I. Martiryan, G.A. Shahinyan, V. v Vardapetyan, Antiradical activity, base-catalyzed hydrolysis and partition coefficients of some surfactants, *Colloids Interface Sci. Commun.* 50 (2022), <https://doi.org/10.1016/j.colcom.2022.100653>.
- [50] A. Durán-Moreno, S.A. García-González, M.R. Gutiérrez-Lara, F. Rigas, R. M. Ramírez-Zamora, Assessment of Fenton's reagent and ozonation as pre-treatments for increasing the biodegradability of aqueous diethanolamine solutions from an oil refinery gas sweetening process, *J. Hazard Mater.* 186 (2011) 1652–1659, <https://doi.org/10.1016/j.jhazmat.2010.12.043>.
- [51] W. Lyu, L. Huang, G. Xiao, Y. Chen, Effects of carbon sources and COD/N ratio on N<sub>2</sub>O emissions in subsurface flow constructed wetlands, *Bioresour. Technol.* 245 (2017) 171–181, <https://doi.org/10.1016/j.biortech.2017.08.056>.
- [52] X. Lai, Y. Zhao, F. Pan, B. Yang, H. Wang, S. Wang, F. He, Enhanced optimal removal of nitrogen and organics from intermittently aerated vertical flow constructed wetlands: relative COD/N ratios and microbial responses, *Chemosphere* 244 (2020), <https://doi.org/10.1016/j.chemosphere.2019.125556>.
- [53] C.A. Lutterbeck, G.S. Colares, G.A. Oliveira, G. Mohr, F. Beckenkamp, A. Rieger, E. A. Lobo, L.H.R. Rodrigues, É.L. MacHado, Microbial fuel cells and constructed wetlands as a sustainable alternative for the treatment of hospital laundry wastewaters: assessment of load parameters and genotoxicity, *J. Environ. Chem. Eng.* 10 (2022), <https://doi.org/10.1016/j.jece.2022.108105>.
- [54] S. Sostar-Turk, I. Petrić, M. Simonić, Laundry wastewater treatment using coagulation and membrane filtration, *Resour. Conserv Recycl* 44 (2005) 185–196, <https://doi.org/10.1016/j.resconrec.2004.11.002>.
- [55] S. Veli, A. Arslan, C. Gülümser, E. Topkaya, H. Kurtklak, S. Zeybek, A. Dimoglo, M. Işgören, Advanced treatment of pre-treated commercial laundry wastewater by adsorption process: experimental design and cost evaluation, *J. Ecol. Eng.* 20 (2019) 165–171, <https://doi.org/10.12911/22998993/113151>.
- [56] M.C. Collivignarelli, M. Carnevale Miino, M. Baldi, S. Manzi, A. Abbà, G. Bertanza, Removal of non-ionic and anionic surfactants from real laundry wastewater by means of a full-scale treatment system, *Process Saf. Environ. Prot.* 132 (2019) 105–115, <https://doi.org/10.1016/j.psep.2019.10.022>.
- [57] I. Ciabatti, F. Cesaro, L. Faralli, E. Fatarella, F. Tognotti, Demonstration of a treatment system for purification and reuse of laundry wastewater, *Desalination* 245 (2009) 451–459, <https://doi.org/10.1016/j.desal.2009.02.008>.
- [58] T. Poiger, J.A. Field, T.M. Field, H. Siegrist, W. Giger, Behavior of fluorescent whitening agents during sewage treatment, *Water Res* 32 (1998) 1939–1947, [https://doi.org/10.1016/S0043-1354\(97\)00408-9](https://doi.org/10.1016/S0043-1354(97)00408-9).
- [59] A. Cuzzola, A. Raffaelli, P. Salvadori, Linear alkylbenzenesulphonic acids (LAS) oxidation by H<sub>2</sub>O<sub>2</sub> and O<sub>2</sub>: An investigation by gas- and liquid-chromatography coupled with mass spectrometry, *Appl. Catal. B* 59 (2005) 113–120, <https://doi.org/10.1016/j.apcatb.2005.01.008>.
- [60] G.V. Buxton, C.L. Greenstock, W.P. Helman, A.B. Ross, Critical Review of rate constants for reactions of hydrated electrons, hydrogen atoms and hydroxyl radicals ( $\cdot\text{OH}/\text{O}^-$  in Aqueous Solution, *J. Phys. Chem. Ref. Data* 17 (1988) 513–886, <https://doi.org/10.1063/1.555805>.
- [61] D. Larsen, TigerPrints An Evaluation of Energy Consumption Comparing Conventional Water Treatment Plants to Microfiltration and Ultrafiltration Water Treatment Plants, (2022).
- [62] Eurostat, Electricity prices for non-household consumers, Eurostat Data Browser. (2022) 1. ([https://ec.europa.eu/eurostat/databrowser/view/nrg\\_pc\\_205/default/table?lang=en](https://ec.europa.eu/eurostat/databrowser/view/nrg_pc_205/default/table?lang=en)) (accessed October 29, 2022).
- [63] M. Molinos-Senante, F. Hernández-Sancho, R. Sala-Garrido, Economic feasibility study for wastewater treatment: a cost-benefit analysis, *Sci. Total Environ.* 408 (2010) 4396–4402, <https://doi.org/10.1016/j.scitotenv.2010.07.014>.
- [64] L. Hernández-Leal, H. Temmink, G. Zeeman, C.J.N. Buisman, Removal of micropollutants from aerobically treated grey water via ozone and activated carbon, *Water Res.* 45 (2011) 2887–2896, <https://doi.org/10.1016/j.watres.2011.03.009>.
- [65] A. Joss, H. Siegrist, T.A. Ternes, Are we about to upgrade wastewater treatment for removing organic micropollutants, *Water Sci. Technol.* 57 (2008) 251–255, <https://doi.org/10.2166/wst.2008.825>.
- [66] Y. Segura, A. Cruz del Álamo, M. Muñoz, S. Álvarez-Torrellas, J. García, J.A. Casas, Z.M. De Pedro, F. Martínez, A comparative study among catalytic wet air oxidation, Fenton, and Photo-Fenton technologies for the on-site treatment of hospital wastewater, *J. Environ. Manag.* 290 (2021), <https://doi.org/10.1016/j.jenvman.2021.112624>.
- [67] D.E. Santiago, O. González-Díaz, J. Araña, E. Pulido Melián, J. Pérez-Peña, J. M. Doña-Rodríguez, Factorial experimental design of imazalil-containing wastewater to be treated by Fenton-based processes, *J. Photochem. Photobio. A Chem.* 353 (2018) 240–250, <https://doi.org/10.1016/j.jphotochem.2017.11.038>.
- [68] INE, Estadística sobre el Suministro y Saneamiento del Agua, 6 (2018) 1–8. ([https://www.ine.es/dyngs/INEbase/es/operacion.htm?c=Estadistica\\_C&cid=1254736176834&menu=ultiDatos&idp=1254735976602](https://www.ine.es/dyngs/INEbase/es/operacion.htm?c=Estadistica_C&cid=1254736176834&menu=ultiDatos&idp=1254735976602)).
- [69] I. Ciabatti, F. Cesaro, L. Faralli, E. Fatarella, F. Tognotti, Demonstration of a treatment system for purification and reuse of laundry wastewater, *Desalination* 245 (2009) 451–459, <https://doi.org/10.1016/j.desal.2009.02.008>.
- [70] A. Dimoglo, P. Sevim-Elibol, Dinç, K. Gökmen, H. Erdoğan, Electrocoagulation/electroflotation as a combined process for the laundry wastewater purification and reuse, *J. Water Process Eng.* 31 (2019) 1–8, <https://doi.org/10.1016/j.jwpe.2019.100877>.
- [71] S. Sostar-Turk, I. Petrić, M. Simonić, Laundry wastewater treatment using coagulation and membrane filtration, *Resour. Conserv Recycl* 44 (2005) 185–196, <https://doi.org/10.1016/j.resconrec.2004.11.002>.
- [72] C. Nicolaidis, I. Vyrides, Closing the water cycle for industrial laundries: an operational performance and techno-economic evaluation of a full-scale membrane bioreactor system, *Resour. Conserv Recycl* 92 (2014) 128–135, <https://doi.org/10.1016/j.resconrec.2014.09.001>.
- [73] K. Zoroufchi Benis, A. Behnami, E. Aghayani, S. Farabi, M. Pourakbar, Water recovery and on-site reuse of laundry wastewater by a facile and cost-effective system: combined biological and advanced oxidation process, *Sci. Total Environ.* 789 (2021), 148068, <https://doi.org/10.1016/j.scitotenv.2021.148068>.
- [74] F. Recanatì, A. Giroth, Secondary data for openLCA, Berlin, 2019.
- [75] S. Fazio, F. Biganzoli, V. De Laurentiis, L. Zampori, S. Sala, E. Diaconu, Supporting information to the characterisation factors of recommended EF Life Cycle Impact Assessment methods, version 2, from ILCD to EF 3.0, EUR 29600 EN, European Commission, Ispra, 2018, ISBN 978–92–79–98584–3, doi:10.2760/002447, PUBSY No. JRC114822., 2018. (<https://doi.org/10.2760/002447>).
- [76] H. Mattila, Process of preparing ferric sulphate, 1998.
- [77] A. Vilén, P. Laurell, R. Vahala, Comparative life cycle assessment of activated carbon production from various raw materials, *J. Environ. Manag.* 324 (2022), <https://doi.org/10.1016/j.jenvman.2022.116356>.
- [78] R. de Oliveira Schwaickhardt, É.L. Machado, C.A. Lutterbeck, Combined use of VUV and UVC photoreactors for the treatment of hospital laundry wastewaters: reduction of load parameters, detoxification and life cycle assessment of different configurations, *Sci. Total Environ.* 590–591 (2017) 233–241, <https://doi.org/10.1016/j.scitotenv.2017.02.218>.