Synthesis and optimization of a selective treatment network of wastewater streams contaminated with paracetamol

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Abstract: The synthesis and optimization of a selective treatment system for contaminated wastewater using Fenton and sono-Fenton oxidation processes is addressed in this work. The problem investigated can be useful for the selective treatment of hospital effluents contaminated with traces of some drug, and the removal of paracetamol is used as a study case. A nonlinear programming model (NLP) that uses a single treatment unit with a defined volume is proposed for the removal of paracetamol by using reported degradation kinetics for each advanced oxidation process evaluated. The cost of the treatment system is considered to be directly proportional to the treated flow. The use of the proposed model is illustrated with the solution of a case study that shows its versatility to achieve optimal treatment systems. Results show that, for the same set of effluents to be treated, the sono-Fenton process exhibits a better paracetamol removal efficiency, i.e., requires less flow through the treatment unit. As higher concentrations of paracetamol are demanded in the discharge, the lesser is the flow of contaminant effluent that is sent to the treatment unit as well. Whilst, as lower concentrations of paracetamol are demanded in the discharge, an opposing effect is observed. Finally, a discussion of the removal ratio of paracetamol in the overall process and in the treatment unit as a function of the discharged limit of paracetamol in the effluent is addressed.

Keywords: optimal design; nonlinear programming model; selective treatment of effluents; wastewater treatment optimization.

Introduction

Nowadays, there are a large number of drugs that are being used in human and veterinary medicine, and their accumulation in the aqueous medium has become an ecological problem because they can cause changes in ecosystems, affecting living beings. Unregulated pharmaceutical waste and other chemicals when are discarded and incorporated into the environment are commonly called emerging pollutants (Bolong *et al.*, 2009; Petrovic *et al.*, 2003; Rout *et al.*, 2021; Sivaranjanee *et al.*,2021; Parida *et al.*, 2021; Prieto-Rodriguez *et al.*, 2012; Karthik *et al.*, 2022).

The regulation, treatment and emission of these substances have received little attention in most countries. Their inclusion as hazardous waste depends on the damage that they can cause to human health, as well as their presence and dispersal capacity in water resources (Petrovic *et al.*, 2003). Thus, there is the possibility that in the near future some drugs will be included in the list of substances of priority interest in the European Directive 2013/39/EC, or in the list of contaminants established by the Environmental Protection Agency (EPA) of the United States. Therefore, new legislations that regulate the discharge of effluents contaminated with specific pharmaceutical products could arise and it is necessary to explore new technologies for the removal of this contaminants.

Among the great diversity of existing drugs, paracetamol is one of the most frequently detected in higher concentrations in different bodies of water with average concentrations of 45-163 ppm for municipal wastewater, 3-16 ppm for treated wastewater, 10-15.5 ppm for surface water and less than 0.298 ppm for drinking water (Lee *et al.*, 2020; Hou *et al.*, 2022). However, conventional wastewater treatment plants have not been designed for removing this drug. It has been reported that paracetamol is transformed during the chlorination stage in conventional treatment plants to the compounds 1-4 benzoquinone and N-acetyl-p-benzoquinone imine (NAPQI), which are more toxic and persistent products than paracetamol itself (Bedner *et al.*, 2006).

Among the various existing alternatives for the removal of pharmaceutical drugs, the called Advanced Oxidation Processes (AOP) are considered a viable alternative to solve the deficiency of conventional treatment plants. AOP involve the generation of hydroxyl radicals (•OH) in sufficient quantities in order to oxidize, and even mineralize, organic contaminants and thus purify water (Glaze *et al.*, 1987). The most common AOP are: Fenton reactions, ozonation, electrolysis, photocatalysis and ultrasound. These processes can be implemented alone or in combination, but it has been reported in the literature that the best results in terms of degradation rates, without considering economic aspects, are obtained by combining two or more of these processes (Morin-Crini *et al.*, 2022; Ahmed *et al.*, 2021).

There are many studies reporting studies on AOP that are effective for the degradation of paracetamol. Some of them, that are considered most relevant in the context and scope of this work are mentioned below.

De Luna *et al.* (2013), studied the degradation of paracetamol at 5 mM (755.85 ppm) in a fluidized bed reactor through the Fenton oxidation process, which involves the use of Fe^{2+} source and hydrogen peroxide (H_2O_2) for the generation of •OH radicals. They reported that, for concentrations of Fe^{2+} between 0.05 and 0.1 mM, this process allows the oxidation of the drug in an approximate time of 40 min, reaching degradation percentages greater than 80%. For the operating conditions that they evaluated, the paracetamol degradation process follows a pseudo-second order kinetics with a rate constant that is a function of the concentrations of paracetamol, Fe^{2+} and H_2O_2. Dalmazio *et al.* (2008) studied the degradation of paracetamol with an initial concentration of 0.1 mM (15.17 ppm) by photocatalysis (TiO_2/UV) in an aqueous medium and reported that in 120 min, around 90% of the drug was degraded and the degree of mineralization was 35%.

Another AOP used for the degradation of paracetamol is the Fenton oxidation coupled with ultrasound, commonly called the sono-Fenton process. In the sono-Fenton process, the degradation takes place due to two conditions: the non-selective attack of •OH radicals on the contaminant and the thermal decomposition of the molecule (Ying-Shih, 2012). Cruz-González *et al.* (2015) studied the removal of paracetamol at concentration of 100 ppm by means of a sono-Fenton process and evaluated the effect of various parameters – ultrasonic power and frequency, pH and initial concentrations of Fe^{2+}/ H_2O_2. These authors found that all these parameters significantly affect the removal efficiency and reported that the removal rate of the paracetamol follows a pseudo-first order kinetics. Andreozzi *et al.* (2003) reported that with ozonation and H_2O_2 photolysis processes is possible to oxidize completely a solution of paracetamol with concentration of 6.0 ppm within the first 60 min and to obtain mineralization degrees between 30% and 40%.

One of the main disadvantages of AOP is their cost, which is significantly higher when is compared with conventional wastewater treatment. Despite this, there is little information reported in the literature about the economic factors involved in such systems. Therefore, studies focused on solving this problem, which is the goal of this work, are required. In order to develop treatment strategies that allow include the process cost, a methodology based on the use of a basic superstructure for the treatment of a set of effluent streams using a single treatment unit with a defined volume is investigated in this work.

Usually, the treatment received by both industrial and hospital effluents are carried out in centralized systems. The combination of effluents of different concentrations and flows, increases the pollutant concentrations in the more diluted streams increasing the treatment costs. In contrast, distributed or decentralized treatment systems allow to treat contaminated effluents separately and more selectively, reducing thus the effluent flows to be treated, and consequently the costs of the treatment system (Wang *et al.*, 1994; Gálan *et al.*, 1999; Zamora *et al.*, 2004; López-Yañez *et al.*, 2018).

The design of treatment systems involving a single contaminant and constant removal rates can be efficiently addressed with point-of-fold methodologies (Wang *et al.*, 1994, Bagajewicz M., 2000) or with mathematical programming methods developed from superstructures of the treatment system that expand a wide set of feasible solutions (Zamora *et al.*, 2004; Gálan *et al.*, 1999; Grossmann *et al.*, 1999; Hernández *et al.*, 2004, Lopez-Yañez *et al.*, 2018). In mathematical programming method, each network superstructure is associated with a NLP model or integer-mixed nonlinear programming (NLP-EM), whose solution allows obtaining optimal designs for treatment networks. This method has been used for selective treatment of waste streams contaminated with trace amounts of amoxicillin (López-Yañez *et al.*, 2016) and hexavalent chromium (López-Yañez *et al.* 2018). This paper deals with the problem of designing selective treatment systems for effluent streams contaminated with traces of paracetamol. The mathematical programming model is solved for two different removal kinetics: Fenton and the sono-Fenton process.

Materials and Methods

Outline of the Proposed Model

For the design of a treatment system capable of degrading pharmaceutical compounds through advanced oxidation technology, the development of a mathematical programming model based on a basic superstructure (Zamora *et al.*, 2004; Lopez-Yañez *et al.*, 2016; Lopez-Yañez *et al.*, 2018) is proposed. In the problem studied, the mass flows of the effluent currents (S_i) and the concentrations of paracetamol in them ($C_{i,A}$) are assumed to be known. The solution of the nonlinear model with each variable removal kinetics determines the optimal values of the flow variables ($t, f_i, f_{i,e}$) and concentrations ($cin_A, cout_A$ and $c_{e,A}$), under the restriction that the final discharge concentration of paracetamol ($c_{e,A}$) must satisfice the limit for the effluent discharge ($c_{e,A}^U$). The superstructure shown in Figure 1 makes it possible since include a significant number of treatment designs. The superstructure considers initial splitters for the effluent streams entering to the treatment system, a mixer for the effluent streams entering to the treatment, a treatment unit (UT) that operates with a variable removal rate that incorporates the degradation kinetics of the contaminant, and a mixer preceding the discharge point.

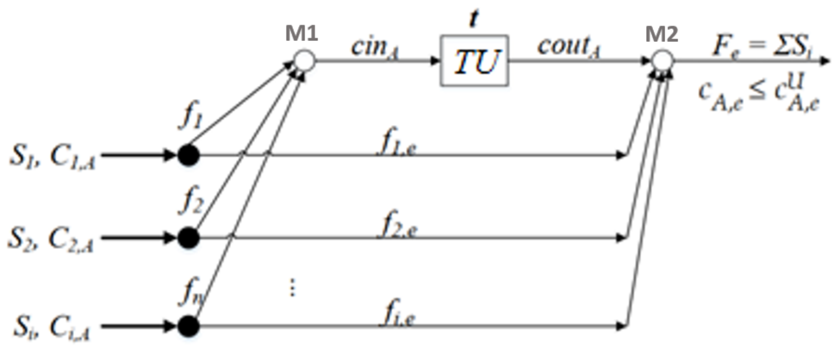

Fig. 1. Basic superstructure for the effluent treatment (Zamora *et al.*, 2004).

Kinetic models

The reaction rate law that describes the drug oxidation process with sufficient precision is established. As mentioned above, this paper deals with the paracetamol degradation by the Fenton and sono-Fenton oxidation processes. Pseudo-second order kinetics describing the Fenton oxidation process, which has been previously developed by De Luna *et al.* (2013), is presented in Equations (1) and (2). Where C_A represents the concentration of paracetamol, C_B is the concentration of Fe^{2+} and C_C is the concentration of hydrogen peroxide (H_2O_2). On the other hand, the pseudo-first order kinetics for the sono-Fenton oxidation process, reported by Cruz-González *et al.* (2015), is presented in Equation (3), and the expression of the associated rate constant in (4); this kinetic constant was also obtained in this work from the data reported by them.

Fenton oxidation (De Luna *et al.*, 2013):

$$-\frac{dC_A}{dt} = k_1 C_A^2 \quad \textbf{(1)}$$

$$k_1 = e^{\left(37.998[C_B]-226.509\frac{[C_B]}{[C_C]}-4.28\right)} \quad \textbf{(2)}$$

Valid for Fenton reagent concentration range: $0.05 \leq C_B \leq 0.01$ y $5 \leq C_C \leq 20$

Sono-Fenton oxidation (Cruz-González *et al.*, 2015):

$$-\frac{dC_A}{dt} = k_2 C_A^2 \quad \textbf{(3)}$$

$$k_2 = 0.05698\, e^{(0.00132\, C_B)} \quad \textbf{(4)}$$

Valid for Fenton reagent concentration conditions: $0.7 \leq C_B \leq 3.5$ y $C_C = 6.9$

All concentrations are given in mmol L^{-1}.

Development of the mathematical model

A NLP model is developed for the selective treatment of effluents contaminated with paracetamol, where the objective function minimizes the cost of the treatment system, a cost that in this work is considered directly proportional to the flow of effluent sent to treatment (t). The decision variables (positive variables) obtained in the solution of the optimization model, determine the optimal design of the treatment system.

The model constraints include balances of water and balances of paracetamol at each element of the superstructure. Treatment technology for paracetamol degradation through oxidation kinetics that describes each process is also included. The formulated model is implemented and solved in the environment of GAMS-CONOPT ver. 21.4.
The nomenclature used in this paper is described below:

Abbreviations:
LP = Linear programming
NLP = Nonlinear programming
TAC = Total annual cost
in = Inlet
S = Sink
P = Parameters
FO = Objective function
D = Constraints

Symbols:
M_1 = Mixer 1
M_2 = Mixer 2
TU = Treatment units

Indexes:
i = Index for wastewater streams
A = Paracetamol
B = $FeSO_4 \cdot 7H_2O$
C = H_2O_2
e = Index of discharge point

Sets:
I = {*i* : *i* is a wastewater stream in the basic superstructure}

Parameters:
$S_i =$ Wastewater flow rate ($L \cdot min^{-1}$)
$C_{i,A} =$ Concentration of paracetamol in each wastewater stream ($mmol \cdot L^{-1}$)
$c_{e,A}^U =$ Discharge limit for the paracetamol, ($mmol \cdot L^{-1}$)
$\Delta m_A^U =$ Upper limit for mass load (mol min^{-1}), $\Delta m_j^U = \sum_{i \in I} S_i C_{i,j}$

$V =$ Reactor volume (L)
$Fe =$ Total flowrate in discharge point (L min^{-1}), $F_e = \sum_{i \in I} S_i$

Positive continuous variables:
$f_i =$ Segregated flowrate for process stream to treatment unit (L·min^{-1})
$f_{i,e} =$ Segregated flowrate discharge into the environment (L·min^{-1})
$t =$ Treatment flow rate through the treatment unit (L·min^{-1})
$cin_A =$ Input concentration of the pollutant in each treatment unit (mmol·L^{-1})
$cout_A =$ Pollutant concentration obtained in discharge point (mmol·L^{-1})
$c_{e,A} =$ Pollutant concentration obtained in discharge point (mmol·L^{-1})
$R_A =$ Removal ratio of paracetamol in the treatment unit (%)
$r_A =$ Reaction rate kinetic (mmol L^{-1} min^{-1})
$k_1 =$ Rate constant for Fenton oxidation process (L $mmol^{-1}$ min^{-1})
$k_2 =$ Rate constant for sono-Fenton oxidation process (min^{-1})

Nonlinear programming (NLP) model

The continuous mass balance model with a variable reaction rate proposed is shown in (5). It incorporates the terms of input and output flux, steady state and the reaction term that is function of the paracetamol (A) concentration.

$$0 = \frac{t}{V} cin_A - \frac{t}{V} cout_A + r_A \tag{5}$$

The objective function goes in the direction of minimizing the treatment flow rate through the treatment unit, which is directly proportional to operating costs (CO), as shown in (Equations 6 and 7):

$$Min\ f(x) = Operating\ Costs \tag{6}$$

$$Min\ f(x) = CO \cdot t \tag{7}$$

Being: $X = t$

With the previous balances, the problem to solve is minimizing the treatment flow rate through the treatment unit, given by the Objective Function (Equation 8):

$$Minimize\ f(x) = t \tag{8}$$

Subjected to

Splitter units:

$$S_2 = f_i + f_{i,e} \qquad i \in I \tag{9}$$

Balance of water in the mixer that precedes at UT:

$$M_1: \sum_{i \in I} f_i = t \tag{10}$$

Balance of water in the mixer after of UT:

$$M_2: \sum_{i \in I} f_{i,e} + t = F_e \tag{11}$$

Mass load of pollutant to be removed at UT:

$$\Delta m_A = 0.001 \cdot R_A \cdot t \cdot cin_A \tag{12}$$

Pollutant concentration A at the inlet for each UT:

$$M_1: \sum_{i \in I} f_i C_{i,A} = t \cdot cin_A \tag{13}$$

Pollutant concentration A at the outlet for each UT:

$$M_2: \sum_{i \in I} f_{i,e} C_{i,A} + t \cdot cout_A = F_e c_{e,A} \tag{14}$$

Variable pollutant removal ratio in UT:

$$R_A = 100 \times \left(\frac{cin_A - cout_A}{cin_A} \right) \tag{15}$$

Pseudo-second order kinetics model for Fenton oxidation process in UT:

$$-r_A = k_1 cout_A^2 \tag{16}$$

Pseudo-first order kinetics for the sono-Fenton oxidation process in UT:

$$-r_A = k_2 cout_A \tag{17}$$

Rate constant for Fenton oxidation process in UT:

$$k_1 = e^{\left(37.998[C_B] - 226.509\frac{[C_B]}{[C_C]} - 4.28\right)} \tag{18}$$

Rate constant for sono-Fenton oxidation process in UT:

$$k_2 = 0.05698\, e^{(0.00132\, C_B)} \tag{19}$$

Steady state mass balance in each UT_k:

$$0 = \frac{t}{V} cin_A - \frac{t}{V} cout_A + r_A \tag{20}$$

Boundaries for variables of the NLP model:

$$0 \leq f_i, f_{i,e} \leq S_i \qquad i \in I \tag{21}$$

$$0 \leq t \leq F_e \tag{22}$$

$$c_{e,A} \leq c_{e,A}^U \tag{23}$$

$$0 \leq R_A \leq 1 \tag{24}$$

$$\Delta m_A^U \leq \sum_{i \in I} S_i C_{i,A} \tag{25}$$

$$C_B^L \leq C_B \leq C_B^U \tag{26}$$

$$C_C^L \leq C_C \leq C_C^U \tag{27}$$

Results and Discussion

Case of Study

The results obtained from the design of a treatment system for effluents contaminated with paracetamol are discussed below. The input data for flowrates and pollutant concentration present in seven streams are summarized in Table 1. Emerging pollutants are not controlled by any standard, for that reason a concentration was arbitrarily established in the discharge point: $c_{e,A}^{U} = 3.2$ mmol L^{-1}, and the reaction volume is 800 L. The results obtained for the Fenton and sono-Fenton process was compared with the same input data. On the other hand, a parametric analysis is developed only for the Fenton treatment system. The results that could be expected for the sono-Fenton process would be qualitatively similar, therefore they are not included in this work.

Table 1. Wastewater streams data for the study cases.

i	S_i (L min^{-1})	$C_{i,A}$ (mmol L^{-1})	$m_{i,j} = S_{i,A}C_{i,A}$ (mmol min^{-1})
1	6	5.2	31.2
2	4	7	28.0
3	4	8.4	33.6
4	2.5	6.1	15.25
5	2	12	24.0
6	5.5	3.8	20.9
7	6	4.5	27.0
	$\Delta m_A^U = \sum m_A$		179.95

Comparison between Fenton and sono-Fenton treatment systems

The optimal treatment system illustrated in Figure 2 is obtained solving the NLP model considering the Fenton oxidation process kinetic (Eqs. (16), (18), (26) and (27)) and restrictions for the experimental conditions addressed by de Luna *et al.* [4]. In this case, streams 2, 3, 4 and 5 were treated in UT with $t = 11.04$ L min^{-1}, and flowrate 1, 6 and 7 were bypassed directly to the discharge point where it is mixed with flowrate t. The lower treatment flowrate in UT comes from the fact that the higher flow streams have lower paracetamol concentrations. Thus, the solution of the optimization model yields $cout_A = 0.73$ mmol L^{-1} for the paracetamol concentration obtained in discharge point, and it is below the required environmental limit concentration of 3.2 mmol L^{-1}. The obtained removal ratio of paracetamol in the treatment unit with this process is $R_A = 91.3\%$.

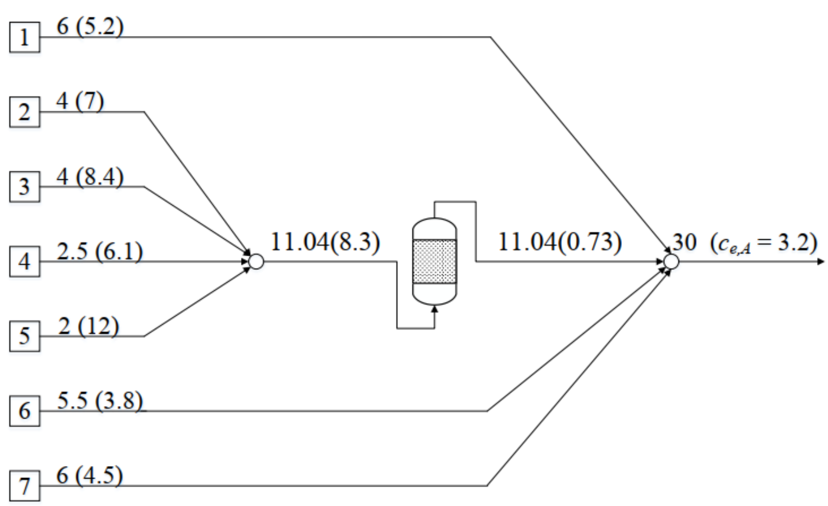

Figure 2. Optimal design of the treatment system for effluents contaminated with paracetamol operating with Fenton oxidation, $R_A = 91.3\%$.

When sono-Fenton oxidation (Eqs. (17), (19), (26), (27)) and restrictions for the experimental conditions addressed by Cruz *et al.* [7], the treatment system illustrated in Figure 3 is obtained. Different to the previously discussed case, this treatment scheme presents three possibilities, these are: total treatment, partial or be discharged bypassing the treatment unit. The streams 2, 3, 5 and a fraction of stream 4 were treated in UT and flowrate 1, 6, 7 and the untreated fraction of flowrate 4 were bypassed directly to the discharge point where it is mixed with flowrate $t = 10.62$ L min^{-1}. The removal ratio of paracetamol in the treatment unit with sono-Fenton process was $R_A = 93.9\%$, which is 2.6% higher than the Fenton process. Furthermore, it demands a lower treatment flowrate compared to the Fenton process presented (see Figure 2), and in principle, if the cost is directly related to the flow, it would be more economical. However, it is recommended to include the costs of reagents and ultrasound in the objective function, but, this is beyond the scope of this work. The better performance of the sono-Fenton process than the Fenton ones could be attributed to the fact that this technology involves two oxidation technologies, i.e., ultrasound and oxidation Fenton.

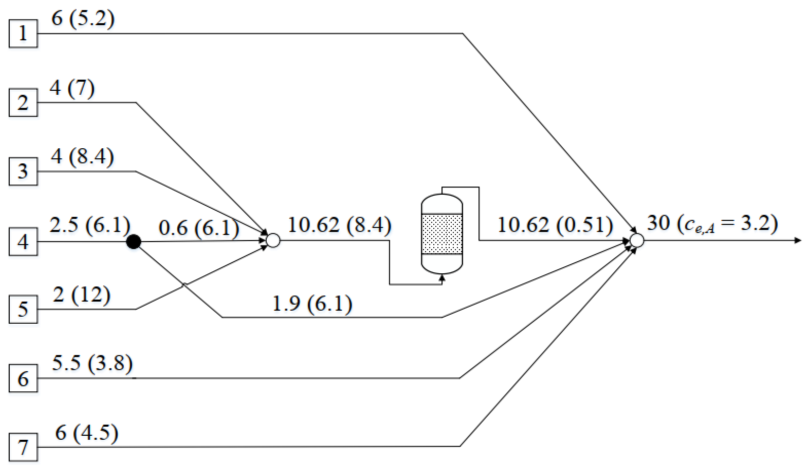

Figure 3. Optimal design of the treatment system for effluents contaminated with paracetamol operating with sono-Fenton oxidation, $R_A = 93.9\%$.

On the other hand, under the premise that the cost depends only on the effluent flow that is sent to treatment, it is of interest to determine how the cost and the topology of the treatment system vary when a stricter concentration limit is imposed on the discharge. To this end, a parametric sensitivity analysis is addressed in the next section, for which the Fenton process was chosen.

Parametric analysis on the paracetamol concentration in the topology of the Fenton system

If it is assumed that in the future an environmental norm that regulates the levels of paracetamol present in effluents is decreed, result of interest to carry out a parametric sensitivity study on the cost and topology of the treatment system for effluent currents contaminated with this drug for different scenarios of the limit concentration imposed in the discharge. The scenarios for the limit concentration of paracetamol in the discharge are established in the range 3.2 mmol L^{-1} to 1 mmol L^{-1} with a step size of 0.1 mmol L^{-1}. For this sensitivity study, only the Fenton oxidation process is considered, the results that would be expected to be obtained for the sono-Fenton process would have a similar qualitative behavior. Under the established requirements, 23 different designs were proposed for the treatment system, some of them with the same topology, although with different costs and operating conditions. Figure 4 shows the relationship between the limit established for the concentration of paracetamol in the discharge of the treatment system and the minimum flowrate in the treatment unit. As can be seen, as the limit concentration in the output is reduced (abscissa axis), the minimum flowrate to be treated increases linearly (ordinate axis), and therefore, a negative slope is obtained. Sections I, II, III and IV indicate regions where the configuration of the optimal treatment structure does not change.

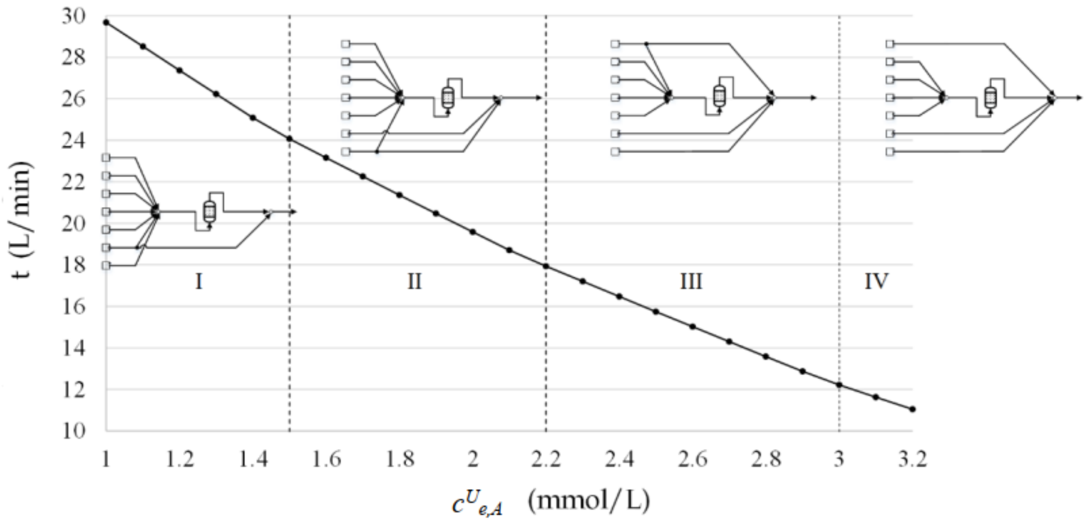

Figure 4. Sensitivity analysis for the effect of $c_{e,A}^{U}$ on the cost and optimal topology of the Fenton system.

Figures 5-7 show the systems obtained for the topologies indicated in sections I, II and III of Figure 4, while Figure 2 corresponds to region IV. In general, it can be observed that, at more relaxed limit concentrations of paracetamol in the effluent (Figure 2 or Region IV of Figure 4), only four streams with the highest concentrations of contaminants were treated in the treatment unit. As the limit concentration of paracetamol is reduced, the streams from higher to lower concentration of paracetamol are progressively treated in UT. Finally, for the smallest value of the limit concentration evaluated in this work, most of the effluent streams were treated in UT (Figure 7, Region I of Figure 4), i.e., it is an arrangement that tends to give total treatment of the effluent streams. In principle, the true potential of the methodology proposed in this work, for the case study addressed in Table 1, requires that the treatment system operate with a partial treatment of the effluent flowrates, i.e. at limit concentrations of paracetamol higher than 0.97 mmol L^{-1}.

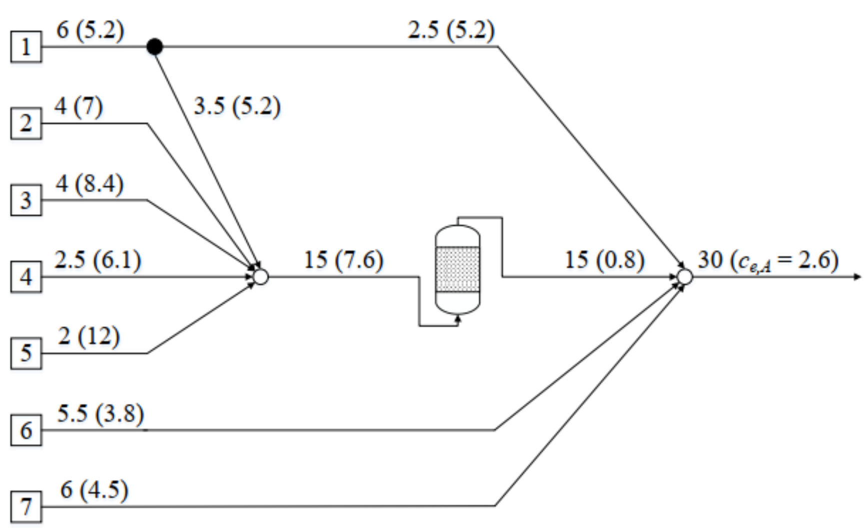

Figure 5. Optimal design of the treatment system that operates with Fenton oxidation, $c_{e,A}^{U} = 2.6$ mmol L^{-1}, $R_A = 89.5\%$.

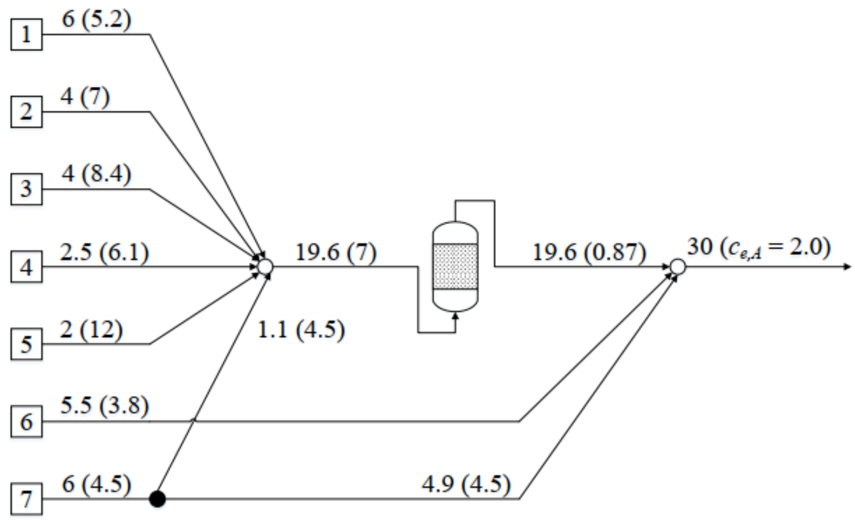

Figure 6. Optimal design of the treatment system operating with Fenton oxidation, $c_{e,A}^{U} = 2.0$ mmol L^{-1}, $R_A = 87.6\%$.

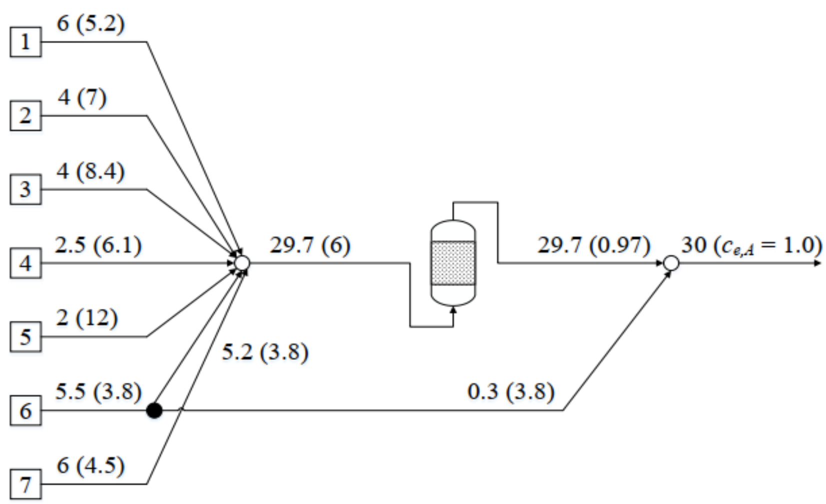

Figure 7. Optimal design of the treatment system that operates with Fenton oxidation, $c_{e,A}^{U} = 1.0$ mmol L^{-1} $R_A = 83.9\%$.

Figure 8 shows the values of the paracetamol removal percentage in the treatment unit and in the process. It is observed that, as the value of the limit concentration of paracetamol in the discharge of the system is reduced, the streams from higher to lower concentrations of the contaminant gradually begin to enter the treatment unit. This causes a decreasing in the concentration of the contaminant entering the Fenton reactor, and thus, also a decreasing the removal rate of paracetamol in the reactor. However, since the mass of contaminant entering the system is the same in all cases examined and the amount of paracetamol that is removed increases, the overall efficiency of the process increases. It can also be seen in Figure 8 that both efficiencies (in the treatment unit and in the overall process) coincide when the total effluent treatment is carried out. For this particular case, the concentration at the outlet of the reactor is the same concentration of paracetamol in the discharged current to the environment, and corresponds to 0.97 mmol L^{-1}.

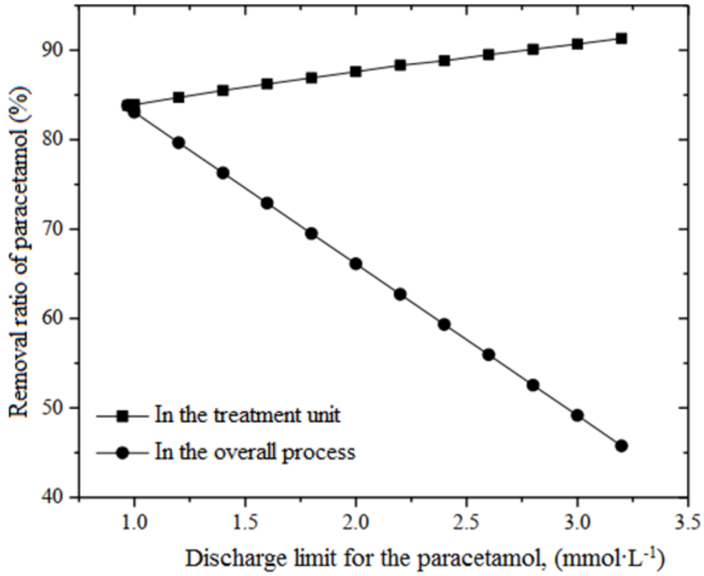

Figure 8. Removal ratio of paracetamol (%) as a function of $c_{e,A}^{U}$.

Conclusions

An optimization model for the synthesis of treatment system of wastewater streams contaminated with paracetamol was developed. The use of a basic superstructure for the treatment is proposed from a set of effluent streams that uses a single treatment unit with a defined volume. The model considers two different technologies: Fenton and sono-Fenton, which have demonstrated to be efficient for removal paracetamol. The proposed model allowed to minimize systematically the treatment flowrate for the specific problems addressed in this work. The objective function includes only the wastewater treatment stream which is directly proportional to the operation cost. Emerging pollutants are not controlled by any norm, and the concentration at the discharge point is arbitrarily stablished, thus a parametric analysis was performed to study the versatility of the nonlinear programming model. It was found that, for the same set of effluents to be treated, the sono-Fenton process exhibits a better remotion efficient than the Fenton one. Results from this study suggest that the programming model is very efficient to evaluate different technologies for removal emerging pollutants.

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Author contributions: A.L.-Y. and M.G.H.-M.: model development and simulations; H.E.G.-B: analysis and interpretation of data and editing; V.M.F.-R. and J.R.-M.: supervision and writing.

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