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Title: Sustainability of Treatment Technologies for Industrial Biowastes Effluents

Article Type: VSI: Conversion of Biowaste

Keywords: Sustainability, Fuzzy-Delphi methodology, Biowastes, Physico-chemical methods, Membrane technologies, Biological methods

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Order of Authors: Tejraj M Aminabhavi, PhD; Mohammadreza Kamali, Ph. D.; Maria Elisabete Costa, Ph. D.; Isabel Capela, Ph. D.

Abstract: Despite the huge efforts to develop efficient technologies for the treatment of recalcitrant biowastes and other emerging pollutants, selecting the most sustainable method among the possible alternatives is still a formidable task. This is mainly because of the integration of technical, economic, environmental, and social criteria in decision-making process. Traditionally, various multi-criteria decision-making approaches have been adopted to integrate innumerable criteria for environmental applications. In this study, we have examined the fuzzy-Delphi approach to evaluate seventeen parameters for integrating technical, economic, environmental and social criteria in order to rank the nine treatment technologies divided in two categories (physico-chemical and biological processes). The results of this study indicated that although efficiency of treatment methods is the most important criterion, but contribution of other sustainability criteria should also be considered because they are of high importance for the selection of sustainable wastewater treatment methods. As per our proposed framework on membrane technologies (among the many other physico-chemical methods) and anaerobic sludge blanket technology (among the biological treatment methods) are the most promising approaches for the treatment of highly polluted emerging industrial pollutants. The findings of this study are fully supported by the consensus achieved by a group of fifty experts from nineteen different countries. Opportunities for the improvement of the methods as per data generated are discussed.

Response to Reviewers: Ms. Ref. No.: CEJ-D-19-02276

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Chemical Engineering Journal

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Tejraj M. Aminabhavi

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Google Scholar: <https://scholar.google.co.in/citations?user=66tWHW0AAAAJ&hl=en>

Research Director: http://www.soniapharmacy.org/m2_2.php

Chair Person Advisory Committee: <http://sdpc.co.in/>

March, 26th, 2019

Professor You Fai Tsang, Special Issue Editor

Journal of Chemical Engineering (**under Thematic Special Issue “Thermochemical Conversion of Biowaste on Energy and Resource Recovery and Pollution Abatement”**)

Dear Prof. Tsang

I am herewith submitting the completely revised copy of the manuscript entitled

Sustainability of Treatment Technologies for Industrial Biowastes Effluents

Mohammadreza Kamali^{1,2}, Maria Elisabete Costa², Tejraj M. Aminabhavi^{*3}, Isabel Capela^{*1}

¹ Department of Environment and Planning, Center for Environmental and Marine Studies, CESAM, University of Aveiro, 3810-193 Aveiro, Portugal

² Department of Materials and Ceramics Engineering, Aveiro Institute of Materials, CICECO, University of Aveiro, 3810-193 Aveiro, Portugal

³ Pharmaceutical Engineering, Soniya College of Pharmacy, Dharwad, India 580-002

For further consideration to the publication in CEJ

Conflict of interest:

All the authors have equally contributed towards the preparation of this review and were participated equally during revision of the work. There is no conflict of interest. All of us agree and are responsible to the statements presented in this revised work.

Please address all correspondence concerning this manuscript to Professor Tejraj Aminabhavi at aminabhavit@gmail.com

Thank you very much for your consideration of this review in CEJ special issue.

Sincerely,

Dr. Tejraj Aminabhavi

Professor and Research Director

Soniya College of Pharmacy, Dharwad 580 002, India

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Highlights:

- The most important criteria for the treatments of industrial wastewater are studied.
- Applications of various physico-chemical and biological methods are assessed for the treatment of industrial effluents.
- Opportunities for industrial wastewater treatment methods are discussed.

Sustainability of Treatment Technologies for Industrial Biowastes Effluents

Mohammadreza Kamali^{1,2}, Maria Elisabete Costa², Tejraj M. Aminabhavi^{*3}, Isabel Capela^{*1}

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1. Introduction

Quality of the final discharged emerging effluents from innumerable industrial activities has been the subject of much debate over the past decades for improving the performance of the methods used either by physico-chemical, biological or a combination of these processes. On the other hand, industries around the world, particularly textile or pulp and paper industries are struggling for their economic profitability [1]. In this situation, stringent environmental regulations have forced the industries, especially those releasing the recalcitrant biowastes compounds, to adopt more efficient treatment methods. Therefore, it is vital to consider both the importance of technical and economic factors while selecting the most appropriate treatment technologies [2]. In addition, long-term sustainability considerations enforce the industries to include environmental and health issues [3] along with the social criteria in decision-making process for selecting the best treatment strategies [4]. Integration of all these aspects while selecting the most appropriate techniques for the treatment of recalcitrant biowastes in industrial effluents is a complex task requiring the need for a multi-criteria analysis framework to identify the most suitable technology.

Due to the inherent advantages of multi-criteria decision making (MCDM) approaches [5, 6], such as their strong structure and logic [7], they have been effectively applied to support the decision makers to choose the most appropriate alternative to solve innumerable environmental problems [8]. Delphi, as a MCDM process, is basically conducted through a group of decision-making among the experts with different experiences and knowledge on the same application domain. Two main applications of this process are screening the criteria and forecasting (or evaluating) the performance of a method or technology [9]. Fuzzy-Delphi method (FDM) has thus been developed by the application of traditional Delphi methodology in a fuzzy environment. This technique has been previously employed in sustainable decisions in various fields [10–12]. In this study, we have employed FDM to assess the current opinion of experts in the field of various conventional and emerging technologies for the treatment of recalcitrant biowastes arising from pulp, paper and textile industries.

Selecting the most appropriate technologies to deal with the recalcitrant biowastes and other emerging pollutants in the context of industrial effluents to satisfy the stringent environmental standards while considering various technical, environmental, economic and social aspects is a complex task [13]. Multi-criteria decision analysis, as a set of methods allows to identify the most important criteria to rank the available alternatives [14,15]. Even though these methods have been extensively used to rank the relevant criteria to select the most suitable alternatives in various scientific fields, yet only handful of reports are available on the application of MCDMs for selecting suitable wastewater treatment technologies. Arroyo and Molinos-Senante [2] used choosing-by-advantages (CBA) approach to select the most suitable municipal wastewater treatment technology among the widely used methods (constructed wetlands, pond systems, extended aeration, membrane bioreactor [16], rotating biological contactor, trickling filter and sequencing batch reactor) by considering several criteria including treatment efficiency, energy consumption, land area required, sewage sludge production, potential for water reuse, potential to recover by-products, reliability, odor impact, noise impact, visual impact, public acceptance, and complexity of operation. According to the opinions of nineteen participants in this research, odor impact was identified as the most important sustainability criterion and the extended aeration together with the sequencing batch reactor were ranked as the most promising treatment technologies.

A “technique for order of preference by similarity to ideal solution” (TOPSIS) approach was also used in some studies. For instance, Dursun [17] using this technique indicated that sustainability is the most important parameter among the studied factors (i.e., cost, global warming, eutrophication, land requirement, manpower requirement, reliability, sustainability and flexibility). However, the method of sequencing batch reactors (SBRs) is the best approach among of all the investigated technologies (i.e., activated sludge, up-flow anaerobic sludge blanket followed by a facultative aerated lagoon, sequential batch reactor, constructed wetlands). Analytical hierarchy process (AHP) has also been used in recent years in its conventional and advanced forms (e.g., AHP fuzzy approach) in order to select the best wastewater treatment technologies. For instance, Ouyang et. al [18] employed the integrated fuzzy analytical hierarchy process for selecting natural wastewater treatment alternatives. However, the fuzzy-Delphi method for ranking the influencing criteria and to prioritize the most suitable technologies to deal with industrial effluents loaded with recalcitrant biowastes has not been studied hitherto considering both conventional and novel treatment technologies.

2. Methodology

2.1. Problem description and criteria identification

Some efforts have been made to identify the most appropriate sewage treatment technologies using the multi criteria decision making processes [2]. However, there is a need to understand the current opinion of scientific community on the most important parameters to identify the most suitable technologies to deal with the industrial effluents as per sustainable development goals. In this investigation, seventeen criteria including technical (i.e., treatment efficiency, ease of implementation, combination possibility, process stability, and health and safety risks), environmental (i.e., biowaste waste generation, release of chemical substances, CO₂ emission, water reuse potential, and recovery of by-products), economic (i.e., initial investments, maintenance costs, and operating costs), and social (odor impact, noise impact, visual impact, and public acceptance) were considered in order to rank the studied methods for the treatment of industrial effluents. Table 1 presents a list of the studied sub-criteria and their descriptions have been provided in the supplementary information.

Table 1. The descriptions of the studied criteria

Criteria	Description
Treatment Efficiency	Effectiveness of the method for the treatment of recalcitrant biowastes from industrial effluents, considering all treatment parameters such as operating conditions, additives, etc..
Ease of Implementation	The level of complexity of the method in terms of required equipment, expertise needed, etc.
Combination Possibility	The combination potential of the method with other treatment methods.
Process Stability	Overall reliability of the treatment method against failures and possibility of re-establishment after probable failures.
Health and Safety risks	The health and occupational risks associated with the implementation of the method.
Solid Wastes Generation	Solid wastes (e.g., biowaste generation when the treatment method is applied.
Release of Chemical Substances	The release of additives (mainly chemicals) used for the treatment process into the effluents content.
CO ₂ Emission	The amount of CO ₂ emitted either directly from the treatment process or indirectly from the treatment facilities, etc.
Water Reuse Potential	Quality of the treated water.

Potential to Recover By-Products	Possibility of the by-products recovery i.e., energy and materials.
Initial Investments	Required initial investments (including land area, equipment, infrastructures, certificates, etc.).
Maintenance Cost	Overall costs required for the maintenance of the facilities.
Operating Costs	Overall costs required for energy, materials, labor, etc.
Odor Impact	Odor generated from the treatment process.
Noise Impact	Noise produced from the process.
Visual Impact	Impact of the treatment infrastructures on the local visual properties.
Public Acceptance	Overall public community's perception about the usefulness of the method for their routine life.

2.2. Study design

In this study, expert questionnaires were assisted to collect the current opinion of experts on the performance of the studied treatment methods. A careful protocol was followed to invite experts with excellent academic and/or industrial experience in the concerned area. Linguistic variables and triangular fuzzy numbers (Tables 2 and 3) were utilized in order to determine the importance of evaluation criteria and to rate the studied alternative methods. The questionnaire used to collect the opinion of the experts was provided in the supplementary information.

A fuzzy number is principally defined as a fuzzy set to elaborate a fuzzy interval in the real number, R. Fuzzy numbers are used to explain uncertain information in decision making process [19]. A triangular fuzzy number (as a type of fuzzy numbers) is defined as: $\tilde{A} = (a_1, a_2, a_3)$. Eq.1 is used to represent the triangular membership functions as [20]:

$$y = m(x) = \begin{cases} 0 & x < a_1 \\ \frac{x-a_1}{a_2-a_1} & a_1 \leq x \leq a_2 \\ \frac{a_3-x}{a_3-a_2} & a_2 \leq x \leq a_3 \\ 0 & x > a_3 \end{cases} \quad \text{Eq.1}$$

A vertex method was then used to estimate the consensus among the expert group (Eq. 2) by computing the distance, $d(\tilde{m}, \tilde{n})$, between the aggregated fuzzy numbers (m_L, m_M, m_U) computed from Eq. 3 and the triangular fuzzy numbers expressed by each expert in the form of linguistic terms.

$$d(\tilde{m}, \tilde{n}) = \sqrt{\frac{1}{3} (m_L - n_L^i)^2 + (m_M - n_M^i)^2 + (m_U - n_U^i)^2} \quad \text{Eq.2}$$

According to Cheng and Lin [21], a value of $d(\tilde{m}, \tilde{n}) < 0.2$ indicates consensus among the experts. The stability of results requires achieving of 75% of group consensus among the experts [22]. The geometric mean (Eq. 3) [23] was utilized to calculate the fuzzy weights of the criteria and relative efficiency of the industrial effluents treatment methods, in which L, M and U represent the fuzzy number components.

$$L_j = \text{Min}_i\{L_{ij}\}, \quad M_j = \frac{1}{n} \sum_{i=1}^n M_{ij}, \quad U_j = \text{Max}_i\{U_{ij}\} \quad \text{Eq. 3}$$

The above equation was used to calculate the relative importance of element number, j allocated by expert number, i . In order to perform defuzzification of the final triangular fuzzy numbers, Eq. 4 was utilized.

$$df = \frac{1}{4}(L + 2M + U) \quad \text{Eq. 4}$$

Table 2. Linguistic variables and the relevant fuzzy scales for the relative importance of the criteria

Linguistic variable	Fuzzy Scale (L, M, U)	$df = \frac{1}{4}(L + 2M + U)$
Extremely high	(0.9, 1.0, 1.0)	0.975
Very high	(0.7, 0.9, 1.0)	0.875
High	(0.5, 0.7, 0.9)	0.7
Fair	(0.3, 0.5, 0.7)	0.5
Low	(0.1, 0.3, 0.5)	0.3
Very Low	(0.0, 0.1, 0.3)	0.125
Extremely low	(0.0, 0.0, 0.1)	0.025

Table 3. Linguistic variables and the relevant fuzzy scales to evaluate the efficiency of the methods

Linguistic variable	Fuzzy Scale (L, M, U)	$df = \frac{1}{4}(L + 2M + U)$
Very good	(0.75, 1, 1)	0.937
Good	(0.5, 0.75, 1)	0.75
Fair	(0.25, 0.5, 0.75)	0.5
Bad	(0, 0.25, 0.5)	0.25
Very bad	(0, 0, 0.25)	0.0625

Aggregation of the fuzzy evaluations for each method was carried out according to Eq. 5.

$$\tilde{A} = \begin{bmatrix} \tilde{A}_1 \\ \tilde{A}_2 \\ \tilde{A}_3 \\ \vdots \\ \vdots \\ \tilde{A}_m \end{bmatrix}, \tilde{A} = \sum_{i,j=1}^n r_{ij} \times w_j \quad \text{Eq. 5}$$

where r_{ij} is the rating of alternative, i with respect to criteria, j and w_j is the j^{th} criterion weight.

3. Results and discussion

Due to inherent complexity in selecting the most appropriate industrial effluents treatment method, it is not feasible to rely only on a single aspect such as the technical characteristics. An international panel consisting of fifty high-quality experts from nineteen countries (all over the world with academic and/or industrial experience) contributed in this study to determine the importance of each criterion and to rate the treatment methods according to their previous experiences on the application of different methods for industrial effluent treatment.

3.1. Prioritization of the criteria

Figure 1 and Table 4 present fuzzy weights of the studied criteria and sub-criteria after achieving the consensus among experts in the second round of questioning. As per data obtained technical considerations received the highest importance among the studied criteria, with a high relevance allocated to “treatment efficiency” and “health and safety risks”. These results could reflect the fact that a technology to be chosen for mitigation of biowastes must be efficient and safe. Despite the fact that innumerable publications have been published on the performance of various industrial effluents treatments [21–24], the health and safety issues attributed to workers using those treatment technologies have not been well investigated. Exposure to biological agents (including bacteria, viruses, fungi (yeasts and mould) and parasites) is considered as one of the most important factors, which affect the safety and health of the workers. The entrance of the spores in human body via many ways such as respiratory tract, damaged skin, eye, etc. may cause severe health problems. In addition, environmental conditions such as humidity and temperature or their combination may provide a favorable environment for the growth of biological agents. Hence, the nature of treatment method applied, and the associated facilities could pose some risks.

Table 4. Linguistic variables and the relevant fuzzy scale for the relative importance of the criteria

Criteria	Sub-Criteria	Fuzzy values			De-fuzzy
		L	M	U	
Technical	Treatment Efficiency (TE)	0.50	0.90	1.00	0.83
	Ease of Implementation (EI)	0.30	0.79	1.00	0.72
	Combination Possibility (CP)	0.25	0.72	1.00	0.67
	Process Stability (PS)	0.30	0.83	1.00	0.74
	Health and Safety Risks (HSR)	0.50	0.90	1.00	0.83
Environmental	Solid Wastes (biowaste) Generation (SWG)	0.10	0.79	1.00	0.67
	Release of Chemical Substances (RCS)	0.00	0.90	1.00	0.70
	CO ₂ Emission (CE)	0.00	0.74	1.00	0.62
	Water Reuse Potential (WRP)	0.10	0.79	1.00	0.67
	Potential to Recover By-Products (PRB)	0.25	0.75	1.00	0.69
Economic	Initial Investments (II)	0.10	0.77	1.00	0.66
	Maintenance Cost (MC)	0.10	0.80	1.00	0.67
	Operating Costs (OC)	0.30	0.84	1.00	0.75
Social	Odor Impact (OI)	0.10	0.74	1.00	0.64
	Noise Impact (NI)	0.10	0.70	1.00	0.63
	Visual Impact (VI)	0.00	0.64	1.00	0.57
	Public Acceptance (PA)	0.00	0.75	1.00	0.63

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Fig. 1. Calculated weights of the criteria and sub-criteria, measured as the average of the respective sub-criteria. The results indicate that technical criteria are most important for selecting the most sustainable industrial effluents treatment technologies. Economic criteria are in the second position of importance followed by environmental and social parameters. In addition, treatment efficiency, health and safety risks are the most important parameters among the influencing criteria. The abbreviations are described in Table 4.

Economic criteria, i.e., initial investments, maintenance and operation costs have received the next importance among the studied criteria. For many industries, which need to comply with stringent environmental regulations, there is currently a declining and competitive market for their product [1]. This reflects the importance of economic considerations for the treatment of industrial effluents such as biowastes. However, the potential of water reuse as well as other environmental factors are of relatively high importance as also expressed by the participating experts. There are some factors determining the quality of treated water for reuse such as efficiency of the applied treatment methods and additives, which are normally added to the wastewater sources during the treatment process and extraction of by-products from the effluents.

The emission of CO₂ is an important environmental issue. The CO₂ emission (CE) has received the least importance among the studied environmental criteria. However, considering the values presented in Table 4, the weight of this criterion (0.62) is above the fair (see Table 2) and this should be considered in the decision-making process. In this regard, those methods with the ability to control emission of greenhouse gases and those in which the treatment process is conducted via mechanisms leading to the production of end products other than CO₂ (e.g., methane), can be more environmentally-friendly, and are better accepted.

The values of the most environmental sub-criteria are close to the economic sub-criteria, while the average weights of social criteria received the least importance among the main categories (table 4 and figure 1). This reflects the higher importance of both environmental and economic criteria in the experts' points of view as compared to social criteria. Furthermore, the fact that the importance of both environmental and economic criteria is similar indicates that nowadays pollution issues are as important as economic aspects related mainly to initial investment, operating and maintenance costs. Regarding the social criteria, their weights evidence the need to be considered when selecting the most appropriate treatment alternatives.

The main improvement opportunities of some of the most widely used treatment alternatives for industrial effluents will be discussed here.

3.2. Ranking the treatment technologies

Various physico-chemical treatment methods have so far been applied for the treatment of industrial effluents such as biowastes. Coagulation and precipitation, membrane technologies, adsorption and oxidation processes are among the most effective and widely accepted techniques. Biological treatments, used as a single treatment or in combination with other physico-chemical methods have also been widely explored for the treatment of industrial effluents [13]. Although they have a number of advantages, including being eco-friendly and cost-effective, they exhibit lack of efficiency for the removal of recalcitrant biowastes due to their low biodegradability in highly polluted industrial effluents. However, scientific efforts are in progress to promote their efficiency for such applications. Pond systems, aerated lagoons, activated sludge and anaerobic sludge blanket technologies are the main biological treatments that have been applied so far for the treatment of industrial effluents around the world. The results of fuzzy-Delphi method for the investigated physico-chemical and biological treatment methods are summarized in Tables 5 and 6, respectively. As summarized in Table 5, the most sustainable physico-chemical method for the treatment of industrial effluents is membrane-based technologies followed by adsorption, oxidation with nanomaterials and Fenton process. Membrane-based technologies have gained the highest scores in technical criteria (2.79) as well as in environmental criteria (2.27), while those of adsorption-based technologies are the most efficient in terms of economic and social criteria. However, considering all criteria, membrane-based technologies have been identified as the most sustainable technology to deal with industrial effluents. Therefore, it can be highly correlated with the advances in the fabrication of novel membrane structures, mainly inorganic membranes (ceramic) and those decorated with engineered nanomaterials to increase the treatment efficiency and to decrease the fouling properties of the membranes [28].

Main	Sub- Criteria	Physico-chemical methods																							
		Fuzzy weights of sub-criteria				Electrocoagulation				Membrane				Adsorption				Fenton Process				Oxidation with			
						Technologies																nanomaterials			
		Fuzzy values				De-	Fuzzy values				De-	Fuzzy values				De-	Fuzzy values				De-	Fuzzy values			
L	M	U	fuzzy		L	M	U	fuzzy		L	M	U	fuzzy		L	M	U	fuzzy		L	M	U	fuzzy		
Technical	TE	0.50	0.90	1.00	0.83	0.50	0.85	1.00	0.80	0.50	0.90	1.00	0.83	0.25	0.79	1.00	0.71	0.25	0.76	1.00	0.69	0.25	0.78	1.00	0.70
	EI	0.30	0.79	1.00	0.72	0.00	0.64	1.00	0.57	0.25	0.69	1.00	0.66	0.25	0.75	1.00	0.69	0.00	0.73	1.00	0.62	0.25	0.63	1.00	0.63
	CP	0.25	0.72	1.00	0.67	0.25	0.75	1.00	0.69	0.25	0.78	1.00	0.70	0.25	0.82	1.00	0.72	0.00	0.79	1.00	0.64	0.25	0.74	1.00	0.68
	PS	0.30	0.83	1.00	0.74	0.25	0.76	1.00	0.69	0.25	0.71	1.00	0.67	0.25	0.78	1.00	0.70	0.00	0.72	1.00	0.61	0.25	0.68	1.00	0.65
	HSR	0.50	0.90	1.00	0.83	0.25	0.72	1.00	0.67	0.50	0.89	1.00	0.82	0.50	0.83	1.00	0.79	0.25	0.76	1.00	0.69	0.25	0.73	1.00	0.68
Environmental	SWG	0.10	0.79	1.00	0.67	0.00	0.63	1.00	0.56	0.00	0.70	1.00	0.60	0.00	0.57	1.00	0.53	0.00	0.58	1.00	0.54	0.25	0.75	1.00	0.69
	RCS	0.00	0.90	1.00	0.70	0.00	0.64	1.00	0.57	0.00	0.80	1.00	0.65	0.00	0.82	1.00	0.66	0.00	0.64	1.00	0.57	0.25	0.69	1.00	0.66
	CE	0.00	0.74	1.00	0.62	0.25	0.73	1.00	0.68	0.25	0.74	1.00	0.68	0.25	0.79	1.00	0.71	0.25	0.74	1.00	0.68	0.25	0.71	1.00	0.67
	WRP	0.10	0.79	1.00	0.67	0.49	0.80	1.00	0.77	0.25	0.95	1.00	0.79	0.00	0.75	1.00	0.63	0.25	0.73	1.00	0.68	0.25	0.79	1.00	0.71
	PRB	0.25	0.75	1.00	0.69	0.00	0.55	1.00	0.52	0.25	0.74	1.00	0.68	0.25	0.64	1.00	0.63	0.00	0.63	1.00	0.56	0.25	0.59	1.00	0.61
Economic	II	0.10	0.77	1.00	0.66	0.25	0.63	1.00	0.63	0.00	0.57	1.00	0.53	0.00	0.67	1.00	0.58	0.00	0.64	1.00	0.57	0.00	0.55	1.00	0.52
	MC	0.10	0.80	1.00	0.67	0.00	0.56	1.00	0.53	0.00	0.54	1.00	0.52	0.00	0.63	1.00	0.57	0.00	0.60	1.00	0.55	0.00	0.55	1.00	0.53
	OC	0.30	0.84	1.00	0.75	0.00	0.58	1.00	0.54	0.00	0.57	1.00	0.54	0.00	0.62	1.00	0.56	0.00	0.56	1.00	0.53	0.00	0.51	1.00	0.51
Social	OI	0.10	0.74	1.00	0.64	0.25	0.73	1.00	0.68	0.25	0.83	1.00	0.73	0.25	0.82	1.00	0.72	0.00	0.75	1.00	0.63	0.25	0.81	1.00	0.72
	NI	0.10	0.70	1.00	0.63	0.00	0.70	1.00	0.60	0.25	0.83	1.00	0.73	0.25	0.83	1.00	0.73	0.25	0.77	1.00	0.70	0.25	0.76	1.00	0.69
	VI	0.00	0.64	1.00	0.57	0.25	0.75	1.00	0.69	0.25	0.86	1.00	0.74	0.25	0.85	1.00	0.74	0.25	0.77	1.00	0.70	0.25	0.78	1.00	0.70

	PA	0.00	0.75	1.00	0.63	0.25	0.76	1.00	0.69	0.25	0.89	1.00	0.76	0.50	0.86	1.00	0.81	0.25	0.73	1.00	0.68	0.25	0.73	1.00	0.68
Sum									10.88				11.63				11.48				10.64				11.02
									Biological methods																
Fuzzy Value									7.48				7.99				7.87				7.29				7.55
									Fuzzy weights of sub-																
									Pond Systems				Aerated Lagoons				Activated Sludge				Anaerobic sludge blanket				
Rank									(4)				(1)				(2)				(5)				(3)

Table 5. The average fuzzy values and de-fuzzy numbers allocated by the experts to the studied physico-chemical methods.

Table 6. The average fuzzy values and de-fuzzy numbers allocated by the experts to the studied biological methods

Main Criteria	Sub-Criteria	criteria										technologies									
		Fuzzy values			De-fuzzy	Fuzzy values			De-fuzzy	Fuzzy values			De-fuzzy	Fuzzy values			De-fuzzy	Fuzzy values			De-fuzzy
		L	M	U		L	M	U		L	M	U		L	M	U		L	M	U	
Technical	TE	0.50	0.90	1.00	0.83	0.00	0.58	1.00	0.54	0.25	0.68	1.00	0.65	0.25	0.76	1.00	0.69	0.25	0.63	1.00	0.63
	EI	0.30	0.79	1.00	0.72	0.25	0.75	1.00	0.69	0.25	0.72	1.00	0.67	0.25	0.73	1.00	0.68	0.00	0.60	1.00	0.55
	CP	0.25	0.72	1.00	0.67	0.25	0.63	1.00	0.63	0.25	0.66	1.00	0.64	0.25	0.73	1.00	0.68	0.00	0.65	1.00	0.58
	PS	0.30	0.83	1.00	0.74	0.00	0.64	1.00	0.57	0.25	0.70	1.00	0.66	0.25	0.68	1.00	0.65	0.00	0.60	1.00	0.55
	HSR	0.50	0.90	1.00	0.83	0.00	0.65	1.00	0.58	0.00	0.63	1.00	0.57	0.00	0.64	1.00	0.57	0.25	0.60	1.00	0.61
Environmental	SWG	0.10	0.79	1.00	0.67	0.00	0.53	1.00	0.52	0.00	0.55	1.00	0.52	0.00	0.51	1.00	0.51	0.00	0.63	1.00	0.57
	RCS	0.00	0.90	1.00	0.70	0.00	0.59	1.00	0.55	0.25	0.58	1.00	0.60	0.00	0.65	1.00	0.58	0.00	0.63	1.00	0.57
	CE	0.00	0.74	1.00	0.62	0.00	0.47	1.00	0.48	0.00	0.45	1.00	0.47	0.00	0.54	1.00	0.52	0.00	0.58	1.00	0.54
	WRP	0.10	0.79	1.00	0.67	0.00	0.52	1.00	0.51	0.00	0.59	1.00	0.54	0.00	0.68	1.00	0.59	0.00	0.55	1.00	0.53
	PRB	0.25	0.75	1.00	0.69	0.00	0.47	1.00	0.48	0.00	0.44	1.00	0.47	0.00	0.59	1.00	0.54	0.00	0.79	1.00	0.64
Economic	II	0.10	0.77	1.00	0.66	0.00	0.67	1.00	0.58	0.00	0.68	1.00	0.59	0.00	0.64	1.00	0.57	0.00	0.57	1.00	0.54
	MC	0.10	0.80	1.00	0.67	0.00	0.72	1.00	0.61	0.00	0.59	1.00	0.55	0.00	0.62	1.00	0.56	0.00	0.68	1.00	0.59
	OC	0.30	0.84	1.00	0.75	0.25	0.79	1.00	0.71	0.00	0.58	1.00	0.54	0.00	0.58	1.00	0.54	0.25	0.74	1.00	0.68
Social	OI	0.10	0.74	1.00	0.64	0.00	0.37	1.00	0.43	0.00	0.38	1.00	0.44	0.00	0.44	1.00	0.47	0.00	0.48	1.00	0.49
	NI	0.10	0.70	1.00	0.63	0.00	0.61	1.00	0.56	0.00	0.46	1.00	0.48	0.00	0.54	1.00	0.52	0.25	0.69	1.00	0.66
	VI	0.00	0.64	1.00	0.57	0.00	0.44	1.00	0.47	0.00	0.49	1.00	0.49	0.00	0.52	1.00	0.51	0.00	0.65	1.00	0.57
	PA	0.00	0.75	1.00	0.63	0.00	0.52	1.00	0.51	0.00	0.61	1.00	0.55	0.00	0.65	1.00	0.57	0.25	0.63	1.00	0.63
Sum									9.42				9.43				9.75				9.93

Fuzzy Value	6.50	6.54	6.78	6.82
Rank	(4)	(3)	(2)	(1)

Novel technologies such as advanced oxidation with nanomaterials are also moderately sustainable due to some existing barriers, which needs to be overcome[29]. Due to high potential of such technologies to satisfy the future needs for clean water resources, more efforts are required, especially in terms of economic (i.e., operating cost) and social considerations (i.e., social acceptance) to push these technologies for commercialization. In this sense, development of cheaper nanomaterials with the ability to be recovered and re-used and with also low toxic effects [30], are highly welcome as a strategic key for the development of advanced oxidation technologies with nanomaterials. Fenton-based technologies have also suffered from the weaknesses such as the release of relatively high amounts of chemicals. Development of facilities to recover chemicals before discarding would push up this technology by reducing both subsequent environmental risks and operating costs.

Among the biological treatment methods (Table 6) activated sludge has been identified as the most promising technology in terms of technical criteria (score: 2.47). However, considering all criteria (i.e. technical, environmental, economic and social), anaerobic sludge blanket was identified as the most sustainable technology (overall score: 6.82). This technology has gained the highest score in environmental criteria (1.90) compared to other studied biological technologies. It means that anaerobic sludge blanket is a green biological technique to deal with industrial effluents. It has also achieved the highest score (1.45) in social criteria, which would indicate that this technology may be selected as a socially acceptable technique. In addition, while pond systems are the most economic biological technique (score: 1.32), it cannot be selected as the most sustainable technology for the treatment of industrial effluents. It can clearly reflect the fact that considering all the sustainability criteria can highly affect the decision-making process to identify the most sustainable technology to deal with biowaste mitigation.

Furthermore, biological treatment methods are not generally sufficient enough to deal with recalcitrant biowastes such as adsorbable organic halides (AOXs). The effluents with high loads of such environmental contaminants may decrease the performance of such systems and even causing their failure. In order to overcome this issue, a combination of biological methods, as post-treatment, with oxidation with nanomaterials may be an alternative approach. However, published reports are quite rare on such combinations, which need more efforts to make the

treatment methods more sustainable to deal with biowaste containing recalcitrant and toxic compounds.

3.3. Development opportunities

According to the results achieved, the main potentials for improvement of each method are briefly presented and discussed in the following sections.

3.3.1. Physico-chemical methods

Currently, membrane technologies are still the leading wastewater treatment methods to overcome global water pollution challenges[28]. Besides the high treatment efficiency of these technologies [31], they can contribute to the recovery of organics [32] and inorganic compounds [30,31] present in the effluents. However, as it can be seen in Table 5, the operating cost (0.54) is still the main obstacle of the wider application of this method. Recent studies have mainly focused on the development of polymeric and inorganic (ceramic) membranes. Goh and Ismail [28] reviewed the latest progress in the fabrication of a new class of inorganic nanostructures, i.e. ceramic membranes (such as metal oxide membranes, zeolite membranes, metal organic framework membranes), and carbon-based membranes (such as carbon nanotubes membranes, and graphene membranes) as the promising materials for industrial treatment purposes. They concluded that the operating cost is still the main barrier for rapid commercialization of new branches of membrane technologies. In this regard, reduction of production costs through the development and use of cheap raw materials and development of efficient and cost-effective fabrication methods for membranes can be considered to be interesting to study. There are some evidences for such approaches in the literature. For instance, Scheibler et al. [35] developed a ultrafiltration process composed of a low-cost multilayer $\gamma\text{-Al}_2\text{O}_3$ ceramic membrane for the pre-treatment of oily wastewater. Zhu et al. [36] prepared a titanium dioxide membrane supported onto mullite hollow fiber synthesized from industrial solid waste coal fly ash as a low-cost alternative for the treatment of oily effluent.

In addition to the operating costs associated with the membrane preparation, fouling is considered another bottleneck on the application of these technologies. Bagheri and Ahmad [37] reviewed fouling mitigation technologies to conclude that application of nanomaterials, electrical and mechanical methods, ultrasonic irradiation, and their combination with biological

treatments (Fig. 2) can be considered as effective strategies to deal with this problem. However, none of these methods has yet been used in full-scale technologies to prove their efficiency with real industrial effluents. Table 7 represents the main advantages and disadvantages of various physico-chemical treatment methods for the treatment of recalcitrant biowastes and mitigation of emerging pollutants from the industrial effluents.

Table 7. Some literature reports on the advantages and disadvantages of the studied physico-chemical methods

Method	Advantages	Disadvantages	Reference
Electrocoagulation	No need for chemical reagents.	Maintenance is required.	[38]
	Relatively low operating costs.	Electrode passivation occurs over	
	Low secondary pollution.	time.	
	Low sludge generation.	High water conductivity is required.	
	No moving parts.	The lack of reactor systematic design	
Membrane technologies	High treatment efficiency.	Process stability due to membrane	[26,39–41]
	Small footprint.	fouling.	
	High potential of the treated water to be re-used.	Relatively high operating costs.	
	Ease of implementation.		
	Fast start-up.		
Fenton Process	High treatment efficiency.	Secondary pollution which requires	[42–44]
	Non-selectivity.	additional treatments.	
	No need for especial reactor configuration (especially in case of non-UV irradiated systems.	Relatively high operating costs.	
Oxidation with engineered nanomaterials	High treatment efficiency.	Probable secondary health and	[45–47]
	Non-selectivity.	safety risks.	
	Low operating costs for some types of ENMs.	Availability of some efficient nanomaterials and nano-composites	
	No need for complicated reactor configurations.	in the current market.	
	Ease of implementation.		
	Stability of the process.		
Adsorption	High treatment efficiency.	Relatively high treatment costs.	[48–50]
	The potential of material recovery.	Selective removal of the	
	Ease of implementation.	contaminants.	
	Process stability.	High sludge generation.

Fig. 2. A schematic of an in-situ integration of microbial fuel cell with hollow-fiber membrane bioreactor in order for treatment of wastewaters. Such a combination is also very effective for the mitigation of the membrane fouling due to the biological decomposition of organic compounds responsible for the membrane fouling, reprinted with permission from Tian et al., 2015 [51].

Adsorption has also been widely studied as one of the most efficient and effective treatment technologies [52], easy to operate and well-suited for materials and nutrients recycling [53]. However, some environmental drawbacks related to solid waste generation (see Table 5) can hinder the promotion of adsorption to deal with the industrial effluents. Activated carbon is a widely used material for this process. However, such a conventional material is very expensive, especially for high quality products [54]. Hence, finding low-cost alternatives has been the subject of a number of recent studies. For instance, Castro et al. [55] indicated that application of cork granules is a cheap material for the treatment of textile effluents. Especially, the operating costs will significantly increase when a highly polluted effluent is subjected to treatment. For instance, Wang et al. [56] used combination of adsorption (wooden activated carbon) at a dosage of 10 g/L, and 1500 mg/L of polymeric magnesium ferric sulfate for subsequent coagulation process. This combination increased significantly the efficiency of the system to treat highly polluted effluents. This possibility derived from activated carbon (AC) can be economically beneficial compared to conventional AC processes.

Agriculture wastes have also been studied as the economic alternative material to be used in the treatment of industrial effluents. As an example, acid-washed coconut shell-based activated carbon (CSAC) has been used successfully for the treatment of palm oil mill effluents [56]. Bello et al. [54] used banana pseudo-stem, a plant waste of banana, as a cheap source of cellulose for the removal of

dyes from industrial effluents. In recent years, several nanostructured materials have also been developed for the adsorption of complex organic pollutants in the context of effluents treatment. For a nanomaterial to be used for adsorption applications, specific surface area is considered to be the determinant factor [47]. For instance, Heydartaemeh [52] prepared a nanocomposite ($\text{Ni}_x\text{Zn}_{1-x}\text{-X Fe}_2\text{O}_4$) with a specific surface area of $120 \text{ m}^2/\text{g}$ having a maximum of 90% of green malachite adsorption after 120 min. Being low-cost, this feature is a main advantage for these materials in terms of their application for industrial treatment purposes.

Advanced oxidation processes have been identified in recent years as efficient methods (see Table 5) to deal with recalcitrant biowastes. The generation of powerful hydroxyl radicals in the medium are the basis of advanced oxidation of recalcitrant pollutants. Fenton reactions and oxidation with engineered nanomaterials are the main processes based on the generation of hydroxyl radicals. The reaction between iron salts and hydrogen peroxide is the basis of Fenton process [57] and short reaction time is the main advantage of this process [58]. Photo-Fenton process using ultraviolet irradiation (UVI) offers higher removal efficiency compared to conventional Fenton process, although it may bring some safety issues and higher operating costs [59]. The main drawback of Fenton processes, however, is the generation of secondary pollutants by releasing ferric ion to the effluents [60]. As a result, additional treatment is required, for instance, by precipitation of the ferric ions through the pH increase of the effluents [61]. This additional treatment may result in generating iron-containing sludge, whose treatment would be expensive and needs a high amount of chemicals [62,63].

In order to overcome these drawbacks, heterogeneous Fenton reactions based on the application of iron oxide nanomaterials or other types of inorganic nanomaterials such as titanium dioxide, copper-based nanomaterials, etc., have gained a huge attention in recent years [27, 62, 63]. However, the reaction kinetics of these processes are slower than those with Fenton process [60]. The properties of nanomaterials such as high specific surface area and agglomeration state may also play a critical role in nano-catalytic processes. The main mechanisms involved in the removal or degradation of the pollutants using nanomaterials consist on the chemisorption of the pollutants on the surface of the materials, transformation of hydrogen peroxide to hydroxyl radicals on the surface of the materials followed by the decomposition of the adsorbed pollutants [66]. UVI can also assist in the generation and decomposition of hydrogen peroxide to hydroxyl radicals onto the surface of the nanomaterials.

As can be seen in Table 5, the economic considerations (i.e., operating costs) are still the main fields for improvements with the application of advanced oxidation processes. In recent years, a trend

can be seen in the literature for the synthesis of low-cost catalytic materials with the ability to work under visible light irradiation, for instance by introducing nitrogen into the structure of titanium dioxide nanomaterials, as one of the most widely applied nanomaterials for wastewater treatment [67]. In spite of the inherent advantages on the application of engineered nanomaterials for the treatment of industrial effluents such as biowastes, the subsequent health problems arising from the possible toxic effects of nanomaterials are now under much debate [66–69]. Although some *in vitro* and *in vivo* studies have shown that some types of the nanomaterials such copper oxide nanoparticles [47] can cause toxic effects on terrestrial and aquatic organisms, the probable health problems associated with the use of nanomaterials have not yet been widely investigated and understood.

Electrocoagulation has also been successfully applied in recent years for the treatment of a variety of pollutants in order to reduce parameters such as chemical oxygen demand (COD), color, recalcitrant compounds, etc., from the industrial effluents. The basis of coagulation process is to introduce metal salts to the stream in order to generate larger flocs from small particles. In the electrocoagulation process, metal cations are dissolved from the anode, resulting in simultaneous formation of hydroxide ions and hydrogen in the form of gas at the cathode (Eqs. 5, 6).

While in the conventional coagulation methods, coagulants such as aluminium chloride [72] are used, in the electrocoagulation process there is no need for any coagulant to be added to the effluents. Hence, the main advantage of this process is to mitigate the release of chemical substances into the treated effluent [73]. This method has been considered as an efficient solution for a sustainable industrial wastewater treatment. However, several parameters such as current density, operating pH, electrolyte type and passivation, reactor design, etc., can determine its efficiency for the treatment of highly polluted industrial effluents. These factors can determine electrical energy requirement and operating costs including the costs of electrodes, electrode replacement, chemicals used for pH and electric conductivity adjustment, etc. Recently, some measures have been applied in order to reduce the operating costs of this process such as the use of Fe electrodes instead of other metals such as aluminium [13].

For electrocoagulation process to be more economic there is a need to increase the efficiency of the energy used in the system. For instance, Cheng-ChunHe et al. [74] indicated that chloride addition and ultrasonic processing can increase the energy efficiency of the system by removing the passivation layer on the electrodes used for electrocoagulation. Reducing the sludge generation in coagulation processes is another potential for improvement in the application of this technology. Considering that the cost of sludge disposal is remarkable [75], the related costs should be included in the design of the treatment facilities using electrocoagulation techniques. Hence, there is a greater need for more studies on the reduction of sludge volume in this technology.

3.3.2. Biological treatments

Stabilization ponds, as an artificial ecosystem, consist on the co-existence of different biological communities such as bacteria, protozoa, alga, fungi, and crustacean larvae. Generally, stabilization ponds are partially aerobic and partially anaerobic in a basin with a depth of about 1 to 2 meters. These systems have been used for the treatment of a variety of industrial effluents, especially in under developed countries. For instance, in Malaysia, stabilization ponds are the most widely used methods for the treatment of palm oil production mill effluents[76].In fact, they are good options when large land areas are available for such treatment installations. Although they have shown to some extent as the acceptable treatment efficiency, there are still some drawbacks in the application of these systems.

Large amounts of greenhouse gases such as carbon dioxide and methane are released from the open ponds into the atmosphere. In addition to the effects of these systems on the global warming, the recovery of energy in the form of methane gas is highly limited from such systems [75,76].Sia et al. [76] stated that by 2020 ponding systems in Malaysia will release 7.2 million tonnes of CO₂into the atmosphere. Although some measures such as application of photosynthetic bacteria to recover CO₂ in the form of bacterial cell [79] have been adopted for capturing carbon dioxide, this problem is still considered as the major deficiency of these systems. The odor impact is another issue in pond systems though some studies are available in the literature to mitigate this problem. Truppel et al. [80] achieved a reduction of odor of a treatment plant located near a populated area by recirculation of the effluent followed by aeration of the pond. The combination of pond systems with other treatment facilities may also be considered as an effective solution to deal with this problem and also to increase the quality

of the treated effluents. For instance, Liu et al. [81] used a combination of a pond system and a wetland to increase the overall efficiency of the system for the treatment of refractory organic pollutants in petrochemical industrial wastewater.

Aerated lagoons have also been extensively used for the treatment of industrial effluents such as kraft mill effluents [80, 81]. These systems present acceptable performance for the removal of biological oxygen demand (BOD) from industrial effluents[13]. However, some studies have shown that their performance for the removal of color from the effluents is considered as one of the drawbacks of these systems. Also, energy input and daily maintenance costs are the main challenges of using these systems, which require the development of efficient aeration and mixing technologies with acceptable energy efficiency. However, by increasing the efficiency of the systems in order to promote biodegradation, the sludge accumulation will occur as one of the main disadvantages of such systems. These may lead to the increase of suspended biomass, which needs a final treatment such as filtration with an increase of further treatment cost.

Over the past decade, many industries have started upgrading their aerated lagoons to activated sludge systems due to the advantages of such systems [83]. Xavier et al. [83] compared the aerated lagoon with activated sludge processes to conclude that activated sludge system might present better efficiency for the treatment of kraft pulp and paper mill effluents, except for phenolic compounds.

In addition to the activated sludge processes, anaerobic sludge blanket processes are the most widely used anaerobic treatment methods in many industries[82, 83]. From Table 5, it is evident that although the treatment efficiency of activated sludge process has received a higher value (0.69) compared to that of the anaerobic sludge blanket technology (0.63), the latter technology was identified as the more sustainable one to deal with industrial effluents. It demonstrates the importance of other sustainability criteria to achieve a final decision to be adopted for an industrial wastewater treatment method. However, in spite of the acceptable performance of both the mentioned biological systems, these methods are now struggling with some certain disadvantages such as relatively high sludge production, especially in the case of activated sludge.

Although the degradation mechanism of the recalcitrant biowaste pollutants in anaerobic digestion processes leads to the formation of biogas (mainly methane), and thereby reducing the

emission of greenhouse gases, they are sensitive to some inhibitory elements such as sulfide and toxic substances [86]. Considering that methane is an important greenhouse gas with potential 34-times higher than carbon dioxide, the production of this gas can significantly contribute to the emission of greenhouse gases. However, by using the produced gas as a source of energy, the anaerobic process can be more cost-effective when compared to activated sludge processes. However, the need for the supply of alkalinity may increase the energy consumption and greenhouse gases (GHGs) generation in anaerobic treatment process. The average alkalinity required in anaerobic processes is in the range of 2000-4000 mg CaCO₃/l to maintain the neutral pH. Hence, production and transportation of the required alkalinity agents can be considered as significant up-stream sources of CO₂ emission [24].

4. Conclusions

The main objective of this study is to provide a framework for making sustainable decisions for selecting suitable wastewater treatment technologies considering the integration of multi-criteria (technical, environmental, economic and social). The application of fuzzy-Delphi method indicated that technical criteria, especially treatment efficiency and health and safety risks in the treatment plant are the most important parameters among the studied criteria. The results also revealed that there are other criteria (i.e., technical, economic, environmental and social) that have enough significance to be also considered when deciding on the most suitable industrial effluent treatment technologies.

Overall, the results of this study demonstrate that selection of the treatment methods to deal with industrial effluents cannot strictly be based only on treatment efficiency to satisfy the requirements of sustainable development. Among the treatment methods, membrane-based technologies are identified as the most viable physico-chemical approaches for recalcitrant biowastes mitigation, whereas anaerobic sludge blanket technology can be the most sustainable biological method to deal with highly polluted industrial effluents containing biowastes. The opportunities for improvements of each treatment method have also been discussed while providing a general perspective for the future. In any case, biowastes mitigation is a formidable issue and membrane technologies combined with biological treatment processes appear to be the solution to this problem.

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Supplementary Information

The format and structure of the questionnaires

A Fuzzy-Delphi approach for ranking the industrial effluents treatments to deal with recalcitrant compounds

Linguistic variables for ranking the criteria

Please identify the importance of each criteria for the treatment of highly polluted industrial effluents.

	Extremely unimportant (Extremely low)	Not important (Very low)	Not very important (Low)	Fair (Moderate)	Important (High)	Very important (Very high)	Extremely important (Extreme high)
Solid Wastes Generation (SWG)	☐	☐	☐	☐	☐	☐	☐
Release of Chemical Substances (RCS)	☐	☐	☐	☐	☐	☐	☐
CO2 Emission (CE)	☐	☐	☐	☐	☐	☐	☐
Water Reuse Potential (WRP)	☐	☐	☐	☐	☐	☐	☐
Treatment Efficiency (TE)	☐	☐	☐	☐	☐	☐	☐
Ease of Implementation (EI)	☐	☐	☐	☐	☐	☐	☐
Combination Possibility (CP)	☐	☐	☐	☐	☐	☐	☐
Process Stability (PS)	☐	☐	☐	☐	☐	☐	☐
Initial Investments (II)	☐	☐	☐	☐	☐	☐	☐
Maintenance Cost (MC)	☐	☐	☐	☐	☐	☐	☐
Operating Costs (OC) (Energy, Materials, Labour, etc.)	☐	☐	☐	☐	☐	☐	☐
Potential to Recover By-Products (PRB)	☐	☐	☐	☐	☐	☐	☐
Odor Impact (OI)	☐	☐	☐	☐	☐	☐	☐
Noise Impact (NI)	☐	☐	☐	☐	☐	☐	☐
Visual Impact (VI)	☐	☐	☐	☐	☐	☐	☐
Public Acceptance (PA)	☐	☐	☐	☐	☐	☐	☐
Health and Safety risks (HSR)	☐	☐	☐	☐	☐	☐	☐

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A Fuzzy-Delphi approach for ranking the industrial effluents treatments to deal with recalcitrant comounds

Electrocoagulation

Linguistic variables for rating the method.
Please NOTE:
Please fill this section if you have previous experience using this method otherwise you can proceed to next section by clicking NEXT button.

Please rate the method in terms of the studied criteria.

	Very Good	Good	Fair	Bad	Very Bad
Solid Wastes Generation (SWG)	☐	☐	☐	☐	☐
Release of Chemical Substances (RCS)	☐	☐	☐	☐	☐
CO2 Emission (CE)	☐	☐	☐	☐	☐
Water Reuse Potential (WRP)	☐	☐	☐	☐	☐
Treatment Efficiency (TE)	☐	☐	☐	☐	☐
Ease of Implementation (EI)	☐	☐	☐	☐	☐
Combination Possibility (CP)	☐	☐	☐	☐	☐
Process Stability (PS)	☐	☐	☐	☐	☐
Initial Investments (II)	☐	☐	☐	☐	☐
Maintanance Cost (MC)	☐	☐	☐	☐	☐
Operating Costs (OC) (Energy, Materials, Labour, etc.)	☐	☐	☐	☐	☐
Potential to Recover By-Products (PRB)	☐	☐	☐	☐	☐
Odor Impact (OI)	☐	☐	☐	☐	☐
Noise Impact (NI)	☐	☐	☐	☐	☐
Visual Impact (VI)	☐	☐	☐	☐	☐
Public Acceptance (PA)	☐	☐	☐	☐	☐
Health and Safety Considerations (HSC)	☐	☐	☐	☐	☐

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Sustainability of Treatment Technologies for Industrial Biowastes Effluents

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Abstract

Despite the huge efforts to develop efficient technologies for the treatment of recalcitrant biowastes and other emerging pollutants, selecting the most sustainable method among the possible alternatives is still a formidable task. This is mainly because of the integration of technical, economic, environmental, and social criteria in decision-making process. Traditionally, various multi-criteria decision-making approaches have been adopted to integrate innumerable criteria for environmental applications. In this study, we have examined the fuzzy-Delphi approach to evaluate seventeen parameters for integrating technical, economic, environmental and social criteria in order to rank the nine treatment technologies divided in two categories (physico-chemical and biological processes). The results of this study indicated that although efficiency of treatment methods is the most important criterion, but contribution of other sustainability criteria should also be considered because they are of high importance for the selection of sustainable wastewater treatment methods. As per our proposed framework on membrane technologies (among the many other physico-chemical methods) and anaerobic sludge blanket technology (among the biological treatment methods) are the most promising approaches for the treatment of highly polluted emerging industrial pollutants. The findings of this study are fully supported by the consensus achieved by a group of fifty experts from nineteen different countries. Opportunities for the improvement of the methods as per data generated are discussed.

Keywords: Sustainability, Fuzzy-Delphi methodology, Biowastes, Physico-chemical methods, Membrane technologies, Biological methods

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1. Introduction

Quality of the final discharged emerging effluents from innumerable industrial activities has been the subject of much debate over the past decades for improving the performance of the methods used either by physico-chemical, biological or a combination of these processes. On the other hand, industries around the world, particularly textile or pulp and paper industries are struggling for their economic profitability [1]. In this situation, stringent environmental regulations have forced the industries, especially those releasing the recalcitrant biowastes compounds, to adopt more efficient treatment methods. Therefore, it is vital to consider both the importance of technical and economic factors while selecting the most appropriate treatment technologies [2]. In addition, long-term sustainability considerations enforce the industries to include environmental and health issues [3] along with the social criteria in decision-making process for selecting the best treatment strategies [4]. Integration of all these aspects while selecting the most appropriate techniques for the treatment of recalcitrant biowastes in industrial effluents is a complex task requiring the need for a multi-criteria analysis framework to identify the most suitable technology.

Due to the inherent advantages of multi-criteria decision making (MCDM) approaches [5, 6], such as their strong structure and logic [7], they have been effectively applied to support the decision makers to choose the most appropriate alternative to solve innumerable environmental problems [8]. Delphi, as a MCDM process, is basically conducted through a group of decision-making among the experts with different experiences and knowledge on the same application domain. Two main applications of this process are screening the criteria and forecasting (or evaluating) the performance of a method or technology [9]. Fuzzy-Delphi method (FDM) has thus been developed by the application of traditional Delphi methodology in a fuzzy environment. This technique has been previously employed in sustainable decisions in various fields [10–12]. In this study, we have employed FDM to assess the current opinion of experts in the field of various conventional and emerging technologies for the treatment of recalcitrant biowastes arising from pulp, paper and textile industries.

Selecting the most appropriate technologies to deal with the recalcitrant biowastes and other emerging pollutants in the context of industrial effluents to satisfy the stringent environmental standards while considering various technical, environmental, economic and social aspects is a

complex task [13]. Multi-criteria decision analysis, as a set of methods allows to identify the most important criteria to rank the available alternatives [14,15]. Even though these methods have been extensively used to rank the relevant criteria to select the most suitable alternatives in various scientific fields, yet only handful of reports are available on the application of MCDMs for selecting suitable wastewater treatment technologies. Arroyo and Molinos-Senante [2] used choosing-by-advantages (CBA) approach to select the most suitable municipal wastewater treatment technology among the widely used methods (constructed wetlands, pond systems, extended aeration, membrane bioreactor [16], rotating biological contactor, trickling filter and sequencing batch reactor) by considering several criteria including treatment efficiency, energy consumption, land area required, sewage sludge production, potential for water reuse, potential to recover by-products, reliability, odor impact, noise impact, visual impact, public acceptance, and complexity of operation. According to the opinions of nineteen participants in this research, odor impact was identified as the most important sustainability criterion and the extended aeration together with the sequencing batch reactor were ranked as the most promising treatment technologies.

A “technique for order of preference by similarity to ideal solution” (TOPSIS) approach was also used in some studies. For instance, Dursun [17] using this technique indicated that sustainability is the most important parameter among the studied factors (i.e., cost, global warming, eutrophication, land requirement, manpower requirement, reliability, sustainability and flexibility). However, the method of sequencing batch reactors (SBRs) is the best approach among of all the investigated technologies (i.e., activated sludge, up-flow anaerobic sludge blanket followed by a facultative aerated lagoon, sequential batch reactor, constructed wetlands). Analytical hierarchy process (AHP) has also been used in recent years in its conventional and advanced forms (e.g., AHP fuzzy approach) in order to select the best wastewater treatment technologies. For instance, Ouyang et. al [18] employed the integrated fuzzy analytical hierarchy process for selecting natural wastewater treatment alternatives. However, the fuzzy-Delphi method for ranking the influencing criteria and to prioritize the most suitable technologies to deal with industrial effluents loaded with recalcitrant biowastes has not been studied hitherto considering both conventional and novel treatment technologies.

2. Methodology

2.1. Problem description and criteria identification

Some efforts have been made to identify the most appropriate sewage treatment technologies using the multi criteria decision making processes [2]. However, there is a need to understand the current opinion of scientific community on the most important parameters to identify the most suitable technologies to deal with the industrial effluents as per sustainable development goals. In this investigation, seventeen criteria including technical (i.e., treatment efficiency, ease of implementation, combination possibility, process stability, and health and safety risks), environmental (i.e., biowaste waste generation, release of chemical substances, CO₂ emission, water reuse potential, and recovery of by-products), economic (i.e., initial investments, maintenance costs, and operating costs), and social (odor impact, noise impact, visual impact, and public acceptance) were considered in order to rank the studied methods for the treatment of industrial effluents. Table 1 presents a list of the studied sub-criteria and their descriptions have been provided in the supplementary information.

Table 1. The descriptions of the studied criteria

Criteria	Description
Treatment Efficiency	Effectiveness of the method for the treatment of recalcitrant biowastes from industrial effluents, considering all treatment parameters such as operating conditions, additives, etc..
Ease of Implementation	The level of complexity of the method in terms of required equipment, expertise needed, etc.
Combination Possibility	The combination potential of the method with other treatment methods.
Process Stability	Overall reliability of the treatment method against failures and possibility of re-establishment after probable failures.
Health and Safety risks	The health and occupational risks associated with the implementation of the method.
Solid Wastes Generation	Solid wastes (e.g., biowaste generation when the treatment method is applied.
Release of Chemical Substances	The release of additives (mainly chemicals) used for the treatment process into the effluents content.
CO ₂ Emission	The amount of CO ₂ emitted either directly from the treatment process or indirectly from the treatment facilities, etc.
Water Reuse Potential	Quality of the treated water.
Potential to Recover By-Products	Possibility of the by-products recovery i.e., energy and materials.
Initial Investments	Required initial investments (including land area, equipment, infrastructures, certificates, etc.).

Maintenance Cost	Overall costs required for the maintenance of the facilities.
Operating Costs	Overall costs required for energy, materials, labor, etc.
Odor Impact	Odor generated from the treatment process.
Noise Impact	Noise produced from the process.
Visual Impact	Impact of the treatment infrastructures on the local visual properties.
Public Acceptance	Overall public community's perception about the usefulness of the method for their routine life.

2.2. Study design

In this study, expert questionnaires were assisted to collect the current opinion of experts on the performance of the studied treatment methods. A careful protocol was followed to invite experts with excellent academic and/or industrial experience in the concerned area. Linguistic variables and triangular fuzzy numbers (Tables 2 and 3) were utilized in order to determine the importance of evaluation criteria and to rate the studied alternative methods. The questionnaire used to collect the opinion of the experts was provided in the supplementary information.

A fuzzy number is principally defined as a fuzzy set to elaborate a fuzzy interval in the real number, R . Fuzzy numbers are used to explain uncertain information in decision making process [19]. A triangular fuzzy number (as a type of fuzzy numbers) is defined as: $\tilde{A} = (a_1, a_2, a_3)$. Eq.1 is used to represent the triangular membership functions as [20]:

$$y = m(x) = \begin{cases} 0 & x < a_1 \\ \frac{x-a_1}{a_2-a_1} & a_1 \leq x \leq a_2 \\ \frac{a_3-x}{a_3-a_2} & a_2 \leq x \leq a_3 \\ 0 & x > a_3 \end{cases} \quad \text{Eq.1}$$

A vertex method was then used to estimate the consensus among the expert group (Eq. 2) by computing the distance, $d(\tilde{m}, \tilde{n})$, between the aggregated fuzzy numbers (m_L, m_M, m_U) computed from Eq. 3 and the triangular fuzzy numbers expressed by each expert in the form of linguistic terms.

$$d(\tilde{m}, \tilde{n}) = \sqrt{\frac{1}{3} (m_L - n_L^i)^2 + (m_M - n_M^i)^2 + (m_U - n_U^i)^2} \quad \text{Eq.2}$$

According to Cheng and Lin [21], a value of $d(\tilde{m}, \tilde{n}) < 0.2$ indicates consensus among the experts. The stability of results requires achieving of 75% of group consensus among the experts [22]. The geometric mean (Eq. 3) [23] was utilized to calculate the fuzzy weights of the criteria and relative

efficiency of the industrial effluents treatment methods, in which L, M and U represent the fuzzy number components.

$$L_j = \text{Min}_i\{L_{ij}\}, \quad M_j = \frac{1}{n} \sum_{i=1}^n M_{ij}, \quad U_j = \text{Max}_i\{U_{ij}\} \quad \text{Eq. 3}$$

The above equation was used to calculate the relative importance of element number, j allocated by expert number, i . In order to perform defuzzification of the final triangular fuzzy numbers, Eq. 4 was utilized.

$$df = \frac{1}{4}(L + 2M + U) \quad \text{Eq. 4}$$

Table 2. Linguistic variables and the relevant fuzzy scales for the relative importance of the criteria

Linguistic variable	Fuzzy Scale (L, M, U)	$df = \frac{1}{4}(L + 2M + U)$
Extremely high	(0.9, 1.0, 1.0)	0.975
Very high	(0.7, 0.9, 1.0)	0.875
High	(0.5, 0.7, 0.9)	0.7
Fair	(0.3, 0.5, 0.7)	0.5
Low	(0.1, 0.3, 0.5)	0.3
Very Low	(0.0, 0.1, 0.3)	0.125
Extremely low	(0.0, 0.0, 0.1)	0.025

Table 3. Linguistic variables and the relevant fuzzy scales to evaluate the efficiency of the methods

Linguistic variable	Fuzzy Scale (L, M, U)	$df = \frac{1}{4}(L + 2M + U)$
Very good	(0.75, 1, 1)	0.937
Good	(0.5, 0.75, 1)	0.75
Fair	(0.25, 0.5, 0.75)	0.5
Bad	(0, 0.25, 0.5)	0.25
Very bad	(0, 0, 0.25)	0.0625

Aggregation of the fuzzy evaluations for each method was carried out according to Eq. 5.

$$\tilde{A} = \begin{bmatrix} \tilde{A}_1 \\ \tilde{A}_2 \\ \tilde{A}_3 \\ \vdots \\ \vdots \\ \tilde{A}_m \end{bmatrix}, \tilde{A} = \sum_{i,j=1}^n r_{ij} \times w_j \quad \text{Eq. 5}$$

where r_{ij} is the rating of alternative, i with respect to criteria, j and w_j is the j^{th} criterion weight.

3. Results and discussion

Due to inherent complexity in selecting the most appropriate industrial effluents treatment method, it is not feasible to rely only on a single aspect such as the technical characteristics. An international panel consisting of fifty high-quality experts from nineteen countries (all over the world with academic and/or industrial experience) contributed in this study to determine the importance of each criterion and to rate the treatment methods according to their previous experiences on the application of different methods for industrial effluent treatment.

3.1. Prioritization of the criteria

Figure 1 and Table 4 present fuzzy weights of the studied criteria and sub-criteria after achieving the consensus among experts in the second round of questioning. As per data obtained technical considerations received the highest importance among the studied criteria, with a high relevance allocated to “treatment efficiency” and “health and safety risks”. These results could reflect the fact that a technology to be chosen for mitigation of biowastes must be efficient and safe. Despite the fact that innumerable publications have been published on the performance of various industrial effluents treatments [21–24], the health and safety issues attributed to workers using those treatment technologies have not been well investigated. Exposure to biological agents (including bacteria, viruses, fungi (yeasts and mould) and parasites) is considered as one of the most important factors, which affect the safety and health of the workers. The entrance of the spores in human body via many ways such as respiratory tract, damaged skin, eye, etc. may cause severe health problems. In addition, environmental conditions such as humidity and temperature or their combination may provide a favorable environment for the growth of biological agents. Hence, the nature of treatment method applied, and the associated facilities could pose some risks.

Table 4. Linguistic variables and the relevant fuzzy scale for the relative importance of the criteria

Criteria	Sub-Criteria	Fuzzy values			De-fuzzy
		L	M	U	
Technical	Treatment Efficiency (TE)	0.50	0.90	1.00	0.83
	Ease of Implementation (EI)	0.30	0.79	1.00	0.72
	Combination Possibility (CP)	0.25	0.72	1.00	0.67
	Process Stability (PS)	0.30	0.83	1.00	0.74
	Health and Safety Risks (HSR)	0.50	0.90	1.00	0.83
Environmental	Solid Wastes (biowaste) Generation (SWG)	0.10	0.79	1.00	0.67
	Release of Chemical Substances (RCS)	0.00	0.90	1.00	0.70
	CO ₂ Emission (CE)	0.00	0.74	1.00	0.62
	Water Reuse Potential (WRP)	0.10	0.79	1.00	0.67
	Potential to Recover By-Products (PRB)	0.25	0.75	1.00	0.69
Economic	Initial Investments (II)	0.10	0.77	1.00	0.66
	Maintenance Cost (MC)	0.10	0.80	1.00	0.67
	Operating Costs (OC)	0.30	0.84	1.00	0.75
Social	Odor Impact (OI)	0.10	0.74	1.00	0.64
	Noise Impact (NI)	0.10	0.70	1.00	0.63
	Visual Impact (VI)	0.00	0.64	1.00	0.57
	Public Acceptance (PA)	0.00	0.75	1.00	0.63

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Fig. 1. Calculated weights of the criteria and sub-criteria, measured as the average of the respective sub-criteria. The results indicate that technical criteria are most important for selecting the most sustainable industrial effluents treatment technologies. Economic criteria are in the second position of importance followed by environmental and social parameters. In addition, treatment efficiency, health and safety risks are the most important parameters among the influencing criteria. The abbreviations are described in Table 4.

Economic criteria, i.e., initial investments, maintenance and operation costs have received the next importance among the studied criteria. For many industries, which need to comply with stringent environmental regulations, there is currently a declining and competitive market for their product [1]. This reflects the importance of economic considerations for the treatment of industrial effluents such as biowastes. However, the potential of water reuse as well as other environmental factors are of relatively high importance as also expressed by the participating experts. There are some factors determining the quality of treated water for reuse such as efficiency of the applied treatment methods and additives, which are normally added to the wastewater sources during the treatment process and extraction of by-products from the effluents.

The emission of CO₂ is an important environmental issue. The CO₂ emission (CE) has received the least importance among the studied environmental criteria. However, considering the values presented in Table 4, the weight of this criterion (0.62) is above the fair (see Table 2) and this should be considered in the decision-making process. In this regard, those methods with the ability to control emission of greenhouse gases and those in which the treatment process is conducted via mechanisms leading to the production of end products other than CO₂ (e.g., methane), can be more environmentally-friendly, and are better accepted.

The values of the most environmental sub-criteria are close to the economic sub-criteria, while the average weights of social criteria received the least importance among the main categories (table 4 and figure 1). This reflects the higher importance of both environmental and economic criteria in the experts' points of view as compared to social criteria. Furthermore, the fact that the importance of both environmental and economic criteria is similar indicates that nowadays pollution issues are as important as economic aspects related mainly to initial investment, operating and maintenance costs. Regarding the social criteria, their weights evidence the need to be considered when selecting the most appropriate treatment alternatives.

The main improvement opportunities of some of the most widely used treatment alternatives for industrial effluents will be discussed here.

3.2. Ranking the treatment technologies

Various physico-chemical treatment methods have so far been applied for the treatment of industrial effluents such as biowastes. Coagulation and precipitation, membrane technologies, adsorption and oxidation processes are among the most effective and widely accepted techniques. Biological treatments, used as a single treatment or in combination with other physico-chemical methods have also been widely explored for the treatment of industrial effluents [13]. Although they have a number of advantages, including being eco-friendly and cost-effective, they exhibit lack of efficiency for the removal of recalcitrant biowastes due to their low biodegradability in highly polluted industrial effluents. However, scientific efforts are in progress to promote their efficiency for such applications. Pond systems, aerated lagoons, activated sludge and anaerobic sludge blanket technologies are the main biological treatments that have been applied so far for the treatment of industrial effluents around the world. The results of fuzzy-Delphi method for the investigated physico-chemical and biological treatment methods are summarized in Tables 5 and 6, respectively. As summarized in Table 5, the most sustainable physico-chemical method for the treatment of industrial effluents is membrane-based technologies followed by adsorption, oxidation with nanomaterials and Fenton process. Membrane-based technologies have gained the highest scores in technical criteria (2.79) as well as in environmental criteria (2.27), while those of adsorption-based technologies are the most efficient in terms of economic and social criteria. However, considering all criteria, membrane-based technologies have been identified as the most sustainable technology to deal with industrial effluents. Therefore, it can be highly correlated with the advances in the fabrication of novel membrane structures, mainly inorganic membranes (ceramic) and those decorated with engineered nanomaterials to increase the treatment efficiency and to decrease the fouling properties of the membranes [28].

Main Criteria	Sub- Criteria	Physico-chemical methods																							
		Fuzzy weights of sub-criteria				Electrocoagulation				Membrane				Adsorption				Fenton Process				Oxidation with			
						Technologies				nanomaterials															
		Fuzzy values				De-	Fuzzy values				De-	Fuzzy values				De-	Fuzzy values				De-	Fuzzy values			
L	M	U	fuzzy	L	M	U	fuzzy	L	M	U	fuzzy	L	M	U	fuzzy	L	M	U	fuzzy	L	M	U	fuzzy		
Technical	TE	0.50	0.90	1.00	0.83	0.50	0.85	1.00	0.80	0.50	0.90	1.00	0.83	0.25	0.79	1.00	0.71	0.25	0.76	1.00	0.69	0.25	0.78	1.00	0.70
	EI	0.30	0.79	1.00	0.72	0.00	0.64	1.00	0.57	0.25	0.69	1.00	0.66	0.25	0.75	1.00	0.69	0.00	0.73	1.00	0.62	0.25	0.63	1.00	0.63
	CP	0.25	0.72	1.00	0.67	0.25	0.75	1.00	0.69	0.25	0.78	1.00	0.70	0.25	0.82	1.00	0.72	0.00	0.79	1.00	0.64	0.25	0.74	1.00	0.68
	PS	0.30	0.83	1.00	0.74	0.25	0.76	1.00	0.69	0.25	0.71	1.00	0.67	0.25	0.78	1.00	0.70	0.00	0.72	1.00	0.61	0.25	0.68	1.00	0.65
	HSR	0.50	0.90	1.00	0.83	0.25	0.72	1.00	0.67	0.50	0.89	1.00	0.82	0.50	0.83	1.00	0.79	0.25	0.76	1.00	0.69	0.25	0.73	1.00	0.68
Environmental	SWG	0.10	0.79	1.00	0.67	0.00	0.63	1.00	0.56	0.00	0.70	1.00	0.60	0.00	0.57	1.00	0.53	0.00	0.58	1.00	0.54	0.25	0.75	1.00	0.69
	RCS	0.00	0.90	1.00	0.70	0.00	0.64	1.00	0.57	0.00	0.80	1.00	0.65	0.00	0.82	1.00	0.66	0.00	0.64	1.00	0.57	0.25	0.69	1.00	0.66
	CE	0.00	0.74	1.00	0.62	0.25	0.73	1.00	0.68	0.25	0.74	1.00	0.68	0.25	0.79	1.00	0.71	0.25	0.74	1.00	0.68	0.25	0.71	1.00	0.67
	WRP	0.10	0.79	1.00	0.67	0.49	0.80	1.00	0.77	0.25	0.95	1.00	0.79	0.00	0.75	1.00	0.63	0.25	0.73	1.00	0.68	0.25	0.79	1.00	0.71
	PRB	0.25	0.75	1.00	0.69	0.00	0.55	1.00	0.52	0.25	0.74	1.00	0.68	0.25	0.64	1.00	0.63	0.00	0.63	1.00	0.56	0.25	0.59	1.00	0.61
Economic	II	0.10	0.77	1.00	0.66	0.25	0.63	1.00	0.63	0.00	0.57	1.00	0.53	0.00	0.67	1.00	0.58	0.00	0.64	1.00	0.57	0.00	0.55	1.00	0.52
	MC	0.10	0.80	1.00	0.67	0.00	0.56	1.00	0.53	0.00	0.54	1.00	0.52	0.00	0.63	1.00	0.57	0.00	0.60	1.00	0.55	0.00	0.55	1.00	0.53
	OC	0.30	0.84	1.00	0.75	0.00	0.58	1.00	0.54	0.00	0.57	1.00	0.54	0.00	0.62	1.00	0.56	0.00	0.56	1.00	0.53	0.00	0.51	1.00	0.51
Social	OI	0.10	0.74	1.00	0.64	0.25	0.73	1.00	0.68	0.25	0.83	1.00	0.73	0.25	0.82	1.00	0.72	0.00	0.75	1.00	0.63	0.25	0.81	1.00	0.72
	NI	0.10	0.70	1.00	0.63	0.00	0.70	1.00	0.60	0.25	0.83	1.00	0.73	0.25	0.83	1.00	0.73	0.25	0.77	1.00	0.70	0.25	0.76	1.00	0.69
	VI	0.00	0.64	1.00	0.57	0.25	0.75	1.00	0.69	0.25	0.86	1.00	0.74	0.25	0.85	1.00	0.74	0.25	0.77	1.00	0.70	0.25	0.78	1.00	0.70

	PA	0.00	0.75	1.00	0.63	0.25	0.76	1.00	0.69	0.25	0.89	1.00	0.76	0.50	0.86	1.00	0.81	0.25	0.73	1.00	0.68	0.25	0.73	1.00	0.68
Sum									10.88				11.63				11.48				10.64				11.02
									Biological methods																
Fuzzy Value									7.48				7.99				7.87				7.29				7.55
									Fuzzy weights of sub-				Pond Systems	Aerated Lagoons				Activated Sludge	Anaerobic sludge blanket						
Rank									(4)				(1)				(2)				(5)				(3)

Table 5. The average fuzzy values and de-fuzzy numbers allocated by the experts to the studied physico-chemical methods.

Table 6. The average fuzzy values and de-fuzzy numbers allocated by the experts to the studied biological methods

Main Criteria	Sub-Criteria	criteria										technologies									
		Fuzzy values			De-fuzzy	Fuzzy values			De-fuzzy	Fuzzy values			De-fuzzy	Fuzzy values			De-fuzzy	Fuzzy values			De-fuzzy
		L	M	U		L	M	U		L	M	U		L	M	U		L	M	U	
Technical	TE	0.50	0.90	1.00	0.83	0.00	0.58	1.00	0.54	0.25	0.68	1.00	0.65	0.25	0.76	1.00	0.69	0.25	0.63	1.00	0.63
	EI	0.30	0.79	1.00	0.72	0.25	0.75	1.00	0.69	0.25	0.72	1.00	0.67	0.25	0.73	1.00	0.68	0.00	0.60	1.00	0.55
	CP	0.25	0.72	1.00	0.67	0.25	0.63	1.00	0.63	0.25	0.66	1.00	0.64	0.25	0.73	1.00	0.68	0.00	0.65	1.00	0.58
	PS	0.30	0.83	1.00	0.74	0.00	0.64	1.00	0.57	0.25	0.70	1.00	0.66	0.25	0.68	1.00	0.65	0.00	0.60	1.00	0.55
	HSR	0.50	0.90	1.00	0.83	0.00	0.65	1.00	0.58	0.00	0.63	1.00	0.57	0.00	0.64	1.00	0.57	0.25	0.60	1.00	0.61
Environmental	SWG	0.10	0.79	1.00	0.67	0.00	0.53	1.00	0.52	0.00	0.55	1.00	0.52	0.00	0.51	1.00	0.51	0.00	0.63	1.00	0.57
	RCS	0.00	0.90	1.00	0.70	0.00	0.59	1.00	0.55	0.25	0.58	1.00	0.60	0.00	0.65	1.00	0.58	0.00	0.63	1.00	0.57
	CE	0.00	0.74	1.00	0.62	0.00	0.47	1.00	0.48	0.00	0.45	1.00	0.47	0.00	0.54	1.00	0.52	0.00	0.58	1.00	0.54
	WRP	0.10	0.79	1.00	0.67	0.00	0.52	1.00	0.51	0.00	0.59	1.00	0.54	0.00	0.68	1.00	0.59	0.00	0.55	1.00	0.53
	PRB	0.25	0.75	1.00	0.69	0.00	0.47	1.00	0.48	0.00	0.44	1.00	0.47	0.00	0.59	1.00	0.54	0.00	0.79	1.00	0.64
Economic	II	0.10	0.77	1.00	0.66	0.00	0.67	1.00	0.58	0.00	0.68	1.00	0.59	0.00	0.64	1.00	0.57	0.00	0.57	1.00	0.54
	MC	0.10	0.80	1.00	0.67	0.00	0.72	1.00	0.61	0.00	0.59	1.00	0.55	0.00	0.62	1.00	0.56	0.00	0.68	1.00	0.59
	OC	0.30	0.84	1.00	0.75	0.25	0.79	1.00	0.71	0.00	0.58	1.00	0.54	0.00	0.58	1.00	0.54	0.25	0.74	1.00	0.68
Social	OI	0.10	0.74	1.00	0.64	0.00	0.37	1.00	0.43	0.00	0.38	1.00	0.44	0.00	0.44	1.00	0.47	0.00	0.48	1.00	0.49
	NI	0.10	0.70	1.00	0.63	0.00	0.61	1.00	0.56	0.00	0.46	1.00	0.48	0.00	0.54	1.00	0.52	0.25	0.69	1.00	0.66
	VI	0.00	0.64	1.00	0.57	0.00	0.44	1.00	0.47	0.00	0.49	1.00	0.49	0.00	0.52	1.00	0.51	0.00	0.65	1.00	0.57
	PA	0.00	0.75	1.00	0.63	0.00	0.52	1.00	0.51	0.00	0.61	1.00	0.55	0.00	0.65	1.00	0.57	0.25	0.63	1.00	0.63
Sum									9.42				9.43				9.75				9.93

Novel technologies such as advanced oxidation with nanomaterials are also moderately sustainable due to some existing barriers, which needs to be overcome[29]. Due to high potential of such technologies to satisfy the future needs for clean water resources, more efforts are required, especially in terms of economic (i.e., operating cost) and social considerations (i.e., social acceptance) to push these technologies for commercialization. In this sense, development of cheaper nanomaterials with the ability to be recovered and re-used and with also low toxic effects [30], are highly welcome as a strategic key for the development of advanced oxidation technologies with nanomaterials. Fenton-based technologies have also suffered from the weaknesses such as the release of relatively high amounts of chemicals. Development of facilities to recover chemicals before discarding would push up this technology by reducing both subsequent environmental risks and operating costs.

Among the biological treatment methods (Table 6) activated sludge has been identified as the most promising technology in terms of technical criteria (score: 2.47). However, considering all criteria (i.e. technical, environmental, economic and social), anaerobic sludge blanket was identified as the most sustainable technology (overall score: 6.82). This technology has gained the highest score in environmental criteria (1.90) compared to other studied biological technologies. It means that anaerobic sludge blanket is a green biological technique to deal with industrial effluents. It has also achieved the highest score (1.45) in social criteria, which would indicate that this technology may be selected as a socially acceptable technique. In addition, while pond systems are the most economic biological technique (score: 1.32), it cannot be selected as the most sustainable technology for the treatment of industrial effluents. It can clearly reflect the fact that considering all the sustainability criteria can highly affect the decision-making process to identify the most sustainable technology to deal with biowaste mitigation.

Furthermore, biological treatment methods are not generally sufficient enough to deal with recalcitrant biowastes such as adsorbable organic halides (AOXs). The effluents with high loads of such environmental contaminants may decrease the performance of such systems and even causing their failure. In order to overcome this issue, a combination of biological methods, as post-treatment, with oxidation with nanomaterials may be an alternative approach. However, published reports are quite rare on such combinations, which need more efforts to make the

treatment methods more sustainable to deal with biowaste containing recalcitrant and toxic compounds.

3.3. Development opportunities

According to the results achieved, the main potentials for improvement of each method are briefly presented and discussed in the following sections.

3.3.1. Physico-chemical methods

Currently, membrane technologies are still the leading wastewater treatment methods to overcome global water pollution challenges[28]. Besides the high treatment efficiency of these technologies [31], they can contribute to the recovery of organics [32] and inorganic compounds [30,31] present in the effluents. However, as it can be seen in Table 5, the operating cost (0.54) is still the main obstacle of the wider application of this method. Recent studies have mainly focused on the development of polymeric and inorganic (ceramic) membranes. Goh and Ismail [28] reviewed the latest progress in the fabrication of a new class of inorganic nanostructures, i.e. ceramic membranes (such as metal oxide membranes, zeolite membranes, metal organic framework membranes), and carbon-based membranes (such as carbon nanotubes membranes, and graphene membranes) as the promising materials for industrial treatment purposes. They concluded that the operating cost is still the main barrier for rapid commercialization of new branches of membrane technologies. In this regard, reduction of production costs through the development and use of cheap raw materials and development of efficient and cost-effective fabrication methods for membranes can be considered to be interesting to study. There are some evidences for such approaches in the literature. For instance, Scheibler et al. [35] developed a ultrafiltration process composed of a low-cost multilayer $\gamma\text{-Al}_2\text{O}_3$ ceramic membrane for the pre-treatment of oily wastewater. Zhu et al. [36] prepared a titanium dioxide membrane supported onto mullite hollow fiber synthesized from industrial solid waste coal fly ash as a low-cost alternative for the treatment of oily effluent.

In addition to the operating costs associated with the membrane preparation, fouling is considered another bottleneck on the application of these technologies. Bagheri and Ahmad [37] reviewed fouling mitigation technologies to conclude that application of nanomaterials, electrical and mechanical methods, ultrasonic irradiation, and their combination with biological

treatments (Fig. 2) can be considered as effective strategies to deal with this problem. However, none of these methods has yet been used in full-scale technologies to prove their efficiency with real industrial effluents. Table 7 represents the main advantages and disadvantages of various physico-chemical treatment methods for the treatment of recalcitrant biowastes and mitigation of emerging pollutants from the industrial effluents.

Table 7. Some literature reports on the advantages and disadvantages of the studied physico-chemical methods

Method	Advantages	Disadvantages	Reference
Electrocoagulation	No need for chemical reagents.	Maintenance is required.	[38]
	Relatively low operating costs.	Electrode passivation occurs over	
	Low secondary pollution.	time.	
	Low sludge generation.	High water conductivity is required.	
	No moving parts.	The lack of reactor systematic design	
Membrane technologies	High treatment efficiency.	Process stability due to membrane	[26,39–41]
	Small footprint.	fouling.	
	High potential of the treated water to be re-used.	Relatively high operating costs.	
	Ease of implementation.		
	Fast start-up.		
Fenton Process	High treatment efficiency.	Secondary pollution which requires	[42–44]
	Non-selectivity.	additional treatments.	
	No need for especial reactor configuration (especially in case of non-UV irradiated systems.	Relatively high operating costs.	
Oxidation with engineered nanomaterials	High treatment efficiency.	Probable secondary health and	[45–47]
	Non-selectivity.	safety risks.	
	Low operating costs for some types of ENMs.	Availability of some efficient nanomaterials and nano-composites	
	No need for complicated reactor configurations.	in the current market.	
	Ease of implementation.		
	Stability of the process.		
Adsorption	High treatment efficiency.	Relatively high treatment costs.	[48–50]
	The potential of material recovery.	Selective removal of the	
	Ease of implementation.	contaminants.	
	Process stability.	High sludge generation.

Fig. 2. A schematic of an in-situ integration of microbial fuel cell with hollow-fiber membrane bioreactor in order for treatment of wastewaters. Such a combination is also very effective for the mitigation of the membrane fouling due to the biological decomposition of organic compounds responsible for the membrane fouling, reprinted with permission from Tian et al., 2015 [51].

Adsorption has also been widely studied as one of the most efficient and effective treatment technologies [52], easy to operate and well-suited for materials and nutrients recycling [53]. However, some environmental drawbacks related to solid waste generation (see Table 5) can hinder the promotion of adsorption to deal with the industrial effluents. Activated carbon is a widely used material for this process. However, such a conventional material is very expensive, especially for high quality products [54]. Hence, finding low-cost alternatives has been the subject of a number of recent studies. For instance, Castro et al. [55] indicated that application of cork granules is a cheap material for the treatment of textile effluents. Especially, the operating costs will significantly increase when a highly polluted effluent is subjected to treatment. For instance, Wang et al. [56] used combination of adsorption (wooden activated carbon) at a dosage of 10 g/L, and 1500 mg/L of polymeric magnesium ferric sulfate for subsequent coagulation process. This combination increased significantly the efficiency of the system to treat highly polluted effluents. This possibility derived from activated carbon (AC) can be economically beneficial compared to conventional AC processes.

Agriculture wastes have also been studied as the economic alternative material to be used in the treatment of industrial effluents. As an example, acid-washed coconut shell-based activated carbon (CSAC) has been used successfully for the treatment of palm oil mill effluents [56]. Bello et al. [54] used banana pseudo-stem, a plant waste of banana, as a cheap source of cellulose for the removal of

dyes from industrial effluents. In recent years, several nanostructured materials have also been developed for the adsorption of complex organic pollutants in the context of effluents treatment. For a nanomaterial to be used for adsorption applications, specific surface area is considered to be the determinant factor [47]. For instance, Heydartaemeh [52] prepared a nanocomposite ($\text{Ni}_x\text{Zn}_{1-x}\text{-X-Fe}_2\text{O}_4$) with a specific surface area of $120 \text{ m}^2/\text{g}$ having a maximum of 90% of green malachite adsorption after 120 min. Being low-cost, this feature is a main advantage for these materials in terms of their application for industrial treatment purposes.

Advanced oxidation processes have been identified in recent years as efficient methods (see Table 5) to deal with recalcitrant biowastes. The generation of powerful hydroxyl radicals in the medium are the basis of advanced oxidation of recalcitrant pollutants. Fenton reactions and oxidation with engineered nanomaterials are the main processes based on the generation of hydroxyl radicals. The reaction between iron salts and hydrogen peroxide is the basis of Fenton process [57] and short reaction time is the main advantage of this process [58]. Photo-Fenton process using ultraviolet irradiation (UVI) offers higher removal efficiency compared to conventional Fenton process, although it may bring some safety issues and higher operating costs [59]. The main drawback of Fenton processes, however, is the generation of secondary pollutants by releasing ferric ion to the effluents [60]. As a result, additional treatment is required, for instance, by precipitation of the ferric ions through the pH increase of the effluents [61]. This additional treatment may result in generating iron-containing sludge, whose treatment would be expensive and needs a high amount of chemicals [62,63].

In order to overcome these drawbacks, heterogeneous Fenton reactions based on the application of iron oxide nanomaterials or other types of inorganic nanomaterials such as titanium dioxide, copper-based nanomaterials, etc., have gained a huge attention in recent years [27, 62, 63]. However, the reaction kinetics of these processes are slower than those with Fenton process [60]. The properties of nanomaterials such as high specific surface area and agglomeration state may also play a critical role in nano-catalytic processes. The main mechanisms involved in the removal or degradation of the pollutants using nanomaterials consist on the chemisorption of the pollutants on the surface of the materials, transformation of hydrogen peroxide to hydroxyl radicals on the surface of the materials followed by the decomposition of the adsorbed pollutants [66]. UVI can also assist in the generation and decomposition of hydrogen peroxide to hydroxyl radicals onto the surface of the nanomaterials.

As can be seen in Table 5, the economic considerations (i.e., operating costs) are still the main fields for improvements with the application of advanced oxidation processes. In recent years, a trend

can be seen in the literature for the synthesis of low-cost catalytic materials with the ability to work under visible light irradiation, for instance by introducing nitrogen into the structure of titanium dioxide nanomaterials, as one of the most widely applied nanomaterials for wastewater treatment [67]. In spite of the inherent advantages on the application of engineered nanomaterials for the treatment of industrial effluents such as biowastes, the subsequent health problems arising from the possible toxic effects of nanomaterials are now under much debate [66–69]. Although some *in vitro* and *in vivo* studies have shown that some types of the nanomaterials such copper oxide nanoparticles [47] can cause toxic effects on terrestrial and aquatic organisms, the probable health problems associated with the use of nanomaterials have not yet been widely investigated and understood.

Electrocoagulation has also been successfully applied in recent years for the treatment of a variety of pollutants in order to reduce parameters such as chemical oxygen demand (COD), color, recalcitrant compounds, etc., from the industrial effluents. The basis of coagulation process is to introduce metal salts to the stream in order to generate larger flocs from small particles. In the electrocoagulation process, metal cations are dissolved from the anode, resulting in simultaneous formation of hydroxide ions and hydrogen in the form of gas at the cathode (Eqs. 5, 6).

While in the conventional coagulation methods, coagulants such as aluminium chloride [72] are used, in the electrocoagulation process there is no need for any coagulant to be added to the effluents. Hence, the main advantage of this process is to mitigate the release of chemical substances into the treated effluent [73]. This method has been considered as an efficient solution for a sustainable industrial wastewater treatment. However, several parameters such as current density, operating pH, electrolyte type and passivation, reactor design, etc., can determine its efficiency for the treatment of highly polluted industrial effluents. These factors can determine electrical energy requirement and operating costs including the costs of electrodes, electrode replacement, chemicals used for pH and electric conductivity adjustment, etc. Recently, some measures have been applied in order to reduce the operating costs of this process such as the use of Fe electrodes instead of other metals such as aluminium [13].

For electrocoagulation process to be more economic there is a need to increase the efficiency of the energy used in the system. For instance, Cheng-ChunHe et al. [74] indicated that chloride addition and ultrasonic processing can increase the energy efficiency of the system by removing the passivation layer on the electrodes used for electrocoagulation. Reducing the sludge generation in coagulation processes is another potential for improvement in the application of this technology. Considering that the cost of sludge disposal is remarkable [75], the related costs should be included in the design of the treatment facilities using electrocoagulation techniques. Hence, there is a greater need for more studies on the reduction of sludge volume in this technology.

3.3.2. Biological treatments

Stabilization ponds, as an artificial ecosystem, consist on the co-existence of different biological communities such as bacteria, protozoa, alga, fungi, and crustacean larvae. Generally, stabilization ponds are partially aerobic and partially anaerobic in a basin with a depth of about 1 to 2 meters. These systems have been used for the treatment of a variety of industrial effluents, especially in under developed countries. For instance, in Malaysia, stabilization ponds are the most widely used methods for the treatment of palm oil production mill effluents[76].In fact, they are good options when large land areas are available for such treatment installations. Although they have shown to some extent as the acceptable treatment efficiency, there are still some drawbacks in the application of these systems.

Large amounts of greenhouse gases such as carbon dioxide and methane are released from the open ponds into the atmosphere. In addition to the effects of these systems on the global warming, the recovery of energy in the form of methane gas is highly limited from such systems [75,76].Sia et al. [76] stated that by 2020 ponding systems in Malaysia will release 7.2 million tonnes of CO₂into the atmosphere. Although some measures such as application of photosynthetic bacteria to recover CO₂ in the form of bacterial cell [79] have been adopted for capturing carbon dioxide, this problem is still considered as the major deficiency of these systems. The odor impact is another issue in pond systems though some studies are available in the literature to mitigate this problem. Truppel et al. [80] achieved a reduction of odor of a treatment plant located near a populated area by recirculation of the effluent followed by aeration of the pond. The combination of pond systems with other treatment facilities may also be considered as an effective solution to deal with this problem and also to increase the quality

of the treated effluents. For instance, Liu et al. [81] used a combination of a pond system and a wetland to increase the overall efficiency of the system for the treatment of refractory organic pollutants in petrochemical industrial wastewater.

Aerated lagoons have also been extensively used for the treatment of industrial effluents such as kraft mill effluents [80, 81]. These systems present acceptable performance for the removal of biological oxygen demand (BOD) from industrial effluents[13]. However, some studies have shown that their performance for the removal of color from the effluents is considered as one of the drawbacks of these systems. Also, energy input and daily maintenance costs are the main challenges of using these systems, which require the development of efficient aeration and mixing technologies with acceptable energy efficiency. However, by increasing the efficiency of the systems in order to promote biodegradation, the sludge accumulation will occur as one of the main disadvantages of such systems. These may lead to the increase of suspended biomass, which needs a final treatment such as filtration with an increase of further treatment cost.

Over the past decade, many industries have started upgrading their aerated lagoons to activated sludge systems due to the advantages of such systems [83]. Xavier et al. [83] compared the aerated lagoon with activated sludge processes to conclude that activated sludge system might present better efficiency for the treatment of kraft pulp and paper mill effluents, except for phenolic compounds.

In addition to the activated sludge processes, anaerobic sludge blanket processes are the most widely used anaerobic treatment methods in many industries[82, 83]. From Table 5, it is evident that although the treatment efficiency of activated sludge process has received a higher value (0.69) compared to that of the anaerobic sludge blanket technology (0.63), the latter technology was identified as the more sustainable one to deal with industrial effluents. It demonstrates the importance of other sustainability criteria to achieve a final decision to be adopted for an industrial wastewater treatment method. However, in spite of the acceptable performance of both the mentioned biological systems, these methods are now struggling with some certain disadvantages such as relatively high sludge production, especially in the case of activated sludge.

Although the degradation mechanism of the recalcitrant biowaste pollutants in anaerobic digestion processes leads to the formation of biogas (mainly methane), and thereby reducing the

emission of greenhouse gases, they are sensitive to some inhibitory elements such as sulfide and toxic substances [86]. Considering that methane is an important greenhouse gas with potential 34-times higher than carbon dioxide, the production of this gas can significantly contribute to the emission of greenhouse gases. However, by using the produced gas as a source of energy, the anaerobic process can be more cost-effective when compared to activated sludge processes. However, the need for the supply of alkalinity may increase the energy consumption and greenhouse gases (GHGs) generation in anaerobic treatment process. The average alkalinity required in anaerobic processes is in the range of 2000-4000 mg CaCO₃/l to maintain the neutral pH. Hence, production and transportation of the required alkalinity agents can be considered as significant up-stream sources of CO₂ emission [24].

4. Conclusions

The main objective of this study is to provide a framework for making sustainable decisions for selecting suitable wastewater treatment technologies considering the integration of multi-criteria (technical, environmental, economic and social). The application of fuzzy-Delphi method indicated that technical criteria, especially treatment efficiency and health and safety risks in the treatment plant are the most important parameters among the studied criteria. The results also revealed that there are other criteria (i.e., technical, economic, environmental and social) that have enough significance to be also considered when deciding on the most suitable industrial effluent treatment technologies.

Overall, the results of this study demonstrate that selection of the treatment methods to deal with industrial effluents cannot strictly be based only on treatment efficiency to satisfy the requirements of sustainable development. Among the treatment methods, membrane-based technologies are identified as the most viable physico-chemical approaches for recalcitrant biowastes mitigation, whereas anaerobic sludge blanket technology can be the most sustainable biological method to deal with highly polluted industrial effluents containing biowastes. The opportunities for improvements of each treatment method have also been discussed while providing a general perspective for the future. In any case, biowastes mitigation is a formidable issue and membrane technologies combined with biological treatment processes appear to be the solution to this problem.

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Supplementary Information

The format and structure of the questionnaires

A Fuzzy-Delphi approach for ranking the industrial effluents treatments to deal with recalcitrant compounds

Linguistic variables for ranking the criteria

Please identify the importance of each criteria for the treatment of highly polluted industrial effluents.

	Extremely unimportant (Extremely low)	Not important (Very low)	Not very important (Low)	Fair (Moderate)	Important (High)	Very important (Very high)	Extremely important (Extreme high)
Solid Wastes Generation (SWG)	☐	☐	☐	☐	☐	☐	☐
Release of Chemical Substances (RCS)	☐	☐	☐	☐	☐	☐	☐
CO2 Emission (CE)	☐	☐	☐	☐	☐	☐	☐
Water Reuse Potential (WRP)	☐	☐	☐	☐	☐	☐	☐
Treatment Efficiency (TE)	☐	☐	☐	☐	☐	☐	☐
Ease of Implementation (EI)	☐	☐	☐	☐	☐	☐	☐
Combination Possibility (CP)	☐	☐	☐	☐	☐	☐	☐
Process Stability (PS)	☐	☐	☐	☐	☐	☐	☐
Initial Investments (II)	☐	☐	☐	☐	☐	☐	☐
Maintenance Cost (MC)	☐	☐	☐	☐	☐	☐	☐
Operating Costs (OC) (Energy, Materials, Labour, etc.)	☐	☐	☐	☐	☐	☐	☐
Potential to Recover By-Products (PRB)	☐	☐	☐	☐	☐	☐	☐
Odor Impact (OI)	☐	☐	☐	☐	☐	☐	☐
Noise Impact (NI)	☐	☐	☐	☐	☐	☐	☐
Visual Impact (VI)	☐	☐	☐	☐	☐	☐	☐
Public Acceptance (PA)	☐	☐	☐	☐	☐	☐	☐
Health and Safety risks (HSR)	☐	☐	☐	☐	☐	☐	☐

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Google Forms

A Fuzzy-Delphi approach for ranking the industrial effluents treatments to deal with recalcitrant comounds

Electrocoagulation

Linguistic variables for rating the method.
Please NOTE:
Please fill this section if you have previous experience using this method otherwise you can proceed to next section by clicking NEXT button.

Please rate the method in terms of the studied criteria.

	Very Good	Good	Fair	Bad	Very Bad
Solid Wastes Generation (SWG)	☐	☐	☐	☐	☐
Release of Chemical Substances (RCS)	☐	☐	☐	☐	☐
CO2 Emission (CE)	☐	☐	☐	☐	☐
Water Reuse Potential (WRP)	☐	☐	☐	☐	☐
Treatment Efficiency (TE)	☐	☐	☐	☐	☐
Ease of Implementation (EI)	☐	☐	☐	☐	☐
Combination Possibility (CP)	☐	☐	☐	☐	☐
Process Stability (PS)	☐	☐	☐	☐	☐
Initial Investments (II)	☐	☐	☐	☐	☐
Maintanance Cost (MC)	☐	☐	☐	☐	☐
Operating Costs (OC) (Energy, Materials, Labour, etc.)	☐	☐	☐	☐	☐
Potential to Recover By-Products (PRB)	☐	☐	☐	☐	☐
Odor Impact (OI)	☐	☐	☐	☐	☐
Noise Impact (NI)	☐	☐	☐	☐	☐
Visual Impact (VI)	☐	☐	☐	☐	☐
Public Acceptance (PA)	☐	☐	☐	☐	☐
Health and Safety Considerations (HSC)	☐	☐	☐	☐	☐

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