

Article

Removal of METH through Tertiary or Advanced Treatment in a WWTP

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Abstract: METHs are drugs that enter wastewater through the feces and urine of users. Conventional wastewater treatment plants are not capable of removing this type of emerging contaminant, but, in recent years, techniques have been developed to abate drugs of abuse. The present investigation focused on obtaining the technique that keeps the best balance between the comparison criteria considered: efficiency; costs; development stage; and waste generation. That is why a bibliographic review was carried out in the scientific databases of the last eight years, concluding that the six most popular techniques are: SBR, Fenton reaction, mixed-flow bioreactor, ozonation, photocatalysis, and UV disinfection. Subsequently, the Saaty and Modified Saaty methods were applied, obtaining a polynomial equation containing the four comparison criteria for the evaluation of the techniques. It is concluded that the UV disinfection method is the one with the best relationship between the analyzed criteria, reaching a score of 0.8591/1, followed by the Fenton method with a score of 0.6925/1. This research work constitutes a practical and easy-to-use tool for decision-makers, since it allows finding an optimal treatment for the abatement of METHs.



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Keywords: emerging pollutants; removal of drugs of abuse; Modified Saaty; WWTP; METHs

1. Introduction

Water is an essential natural resource for the development of the life of living beings. Currently, one of the challenges worldwide is to ensure the quality of water resources, since waste derived from human and productive activities is commonly thrown into different bodies of water through wastewater [1–3]. The presence of emerging contaminants (EC) in water is considered to be of high environmental concern, since it causes the deterioration of the quality of this resource, becoming one of the main problems that threatens the human population [4,5].

EC are compounds of different origin and chemical nature that are not commonly monitored because their presence in the environment and the damage they can cause are little studied and unnoticed [6]. In other words, there is little information about them, which further intensifies the problem. In addition, they are considered toxic and bioaccumulative substances, whose concentrations can reach water reservoirs for human consumption or migrate to groundwater sources [7,8].

Drugs of abuse are considered a type of EC, in the same way, recent research has indicated that these substances are able to cause carcinogenic as well as mutagenic problems and even alter hormonal functions [9]. The presence of drugs of abuse in raw water from wastewater treatment plants (WWTP) is becoming more frequent, in the same way, the existence of metabolites in bodies of water in lower concentrations compromises marine ecosystems and public health due to their toxicity and low biodegradability [10–14]. The evaluation of the consumption of drugs of abuse through WWTP has aroused the interest of different research groups, given that, being an environmental and social problem with little

information, it allows estimating the trends of consumers at the local and national level and, therefore, the potential effects within human health and environmental matrices [15–17].

Amine et al. (2014) performs a comparison of different types of water and wastewater treatment based on nanotechnology that do not depend on large infrastructures or centralized systems. The study of the revised techniques for the removal of viruses, inorganic solutes, heavy metals, metal ions, complex organic compounds, natural organic matter, nitrate, and other contaminants present in surface water, groundwater does not present values or provide the reader with tools for comparison and ranking [18].

So also, Asimakopoulos et al. (2016) in their review article compiled information on the presence of bioactive chemicals in wastewater treatment processes, concluding that neuropsychiatric pharmaceuticals and illicit drugs in wastewater are a growing environmental concern due to their persistence after a removal treatment. Their methodology consisted of performing a meta-analysis of recent literature and, subsequently, evaluating the concentrations and profiles of various drugs and their removal efficiencies in wastewater treatment plants. However, it does not indicate a comparison between drug abatement techniques, nor does it propose a balance of the compared criteria [19].

The concern of the presence of drugs of abuse such as amphetamine (AMP), cocaine (COC), cannabis (THC), methamphetamine (METH), heroin (HER), morphine (MOR), codeine (COD), methadone (MET), and caffeine (CAF), among others in the form of metabolites, could cause negative effects on the environment due to their continuous integration, even though the average concentrations are a few ng L^{-1} to more than $200 \mu\text{g L}^{-1}$, over time they bioaccumulate in living organisms or stay in the environment [20–22]. The main source of reception of these pollutants is through untreated wastewater (WW), however, in some studies it has been found that despite having passed through a WWTP, the compounds derived from METHs have not been completely eliminated [23].

METH is one of the most widely used illicit drugs in Europe and is closely related to synthetic amphetamine-type stimulants, usually made in clandestine laboratories as there are no commercial supplies available, as well as a natural source [24]. However, MET is also a drug approved by the US Food and Drug Administration to treat attention deficit hyperactivity disorder (ADHD) [25]. In addition, according to the United Nations Office on Drugs and Crime (UNODC), the world market for synthetic drugs continues to be dominated by MET. This drug enters wastewater through urine and feces after legal or illegal consumption and through clandestine laboratory manufacturing. On the other hand, the percentage of MET excreted as parent drug can range from 2% in alkaline urine (pH \sim 8.0) to 76% in acid urine (pH \sim 5.0) [25].

In this sense, WWTPs commonly use conventional primary and secondary purification treatments [26]. The primary one is in charge of the sedimentation and removal of solids, oils, fats, and rocky or sandy particles while the secondary destined to the degradation of the biological content of the WWTP. Therefore, the cleaning treatments of the EC, exclusively in METHs, must be incorporated into the systems of the WWTP in order to achieve their elimination [21,27].

Conventional treatments are not capable of removing EC; therefore, it has aroused the interest of researchers in the application of advanced treatments that demonstrate adequate removal efficiency considering economic and environmental aspects [3]. Advanced technologies such as oxidation, photocatalytic degradation, bioreactor filtration, Fenton reactor, and UV disinfection have been investigated for WW treatment [28–31]. However, although some of these technologies are showing promising results in the reduction of drugs of abuse and their metabolites, they are expensive and their large-scale application is not economically or sustainably viable [3,32,33].

Based on the context, it is observed that there are several alternatives for the abatement of drugs of abuse, so it is important to know the optimal method to apply. One of the methods used to evaluate several alternatives based on common parameters is the hierarchical analysis method (AHP) proposed by Tomas Saaty in 2008 [34,35]. This is a pairwise comparison procedure that allows establishing the importance of each of the criteria in

relation to with the others, through the assignment of weights. The process of weighing variables must be performed by a panel of experts. In order to have an adequate decision system, the method has allowed the inclusion of new criteria in order to balance coherence with the judgment of the experts, so that a decision is obtained based on the knowledge they provide and analytical consistency [36].

The Modified Saaty method uses the weights obtained from the AHP and additionally standardizes the variables through a membership function that depends on the relationship between the analyzed variable and the probability of occurrence of the event, having as limits of the function the maximum and minimum values of each criteria [37–39]. If the relationship is directly proportional, the sine function will be used and, if it is inversely proportional, the cosine function will be used in a range between 0° and 90°. Through a weighted linear sum (WLC), the weight is obtained for each of the study techniques, which is the result of the analysis and evaluation of the behavior of each variable with its respective weight obtained from the AHP [40].

The present work conducts a bibliographic review about the treatments used for the removal of METHs and their metabolites in wastewater. In addition, it indicates a comparison of the removal efficiency, construction cost, waste generation, and development stage of each of the exposed treatments to subsequently recommend its application in a WWTP through an evaluation by the Modified Saaty method.

2. Materials and Methods

Within the framework of this research, a bibliographic review was performed in different high-impact scientific databases, such as Scopus, SpringerOpen, ScieELO, and ScienceDirect, among others, in order to obtain significant information regarding drug abuse removal treatments in sewage water. During the compilation of the investigations, we worked with those of the last eight years of publication. Subsequently, the most relevant criteria of each treatment were extracted, such as construction cost efficiency, waste generation, and development stage (laboratory, pilot, and large-scale).

Subsequently, the Modified Saaty method was applied, six techniques studied, and their behavior against the parameters of interest were known. The parameters of efficiency, implementation cost, development stage, and waste generation were compared with respect to METH abatement. During the weighting of each parameter, an importance value was assigned on a scale of 1 to 9, with 1 being the worst and 9 being the best behavior, according to a panel of experts. Various categories were also considered for the study variables, such as poor, fair, good, very good, and excellent, according to the values and data, as shown in Table 1.

Then, the normalization of the variables was conducted considering the dependency relationship between the variable and the method, that is, for a direct relationship, the sine function was applied and for an inverse relationship, the cosine function was applied [41,42]. Once the normalization was obtained, the AHP was realized using Saaty. For this, the experts considered that the construction cost is more important in relation to the other variables, assigning it a value of 9. This is because within the economic sphere, those techniques that present a lower construction cost are more desirable of the efficiency that they are able to provide. Followed by this is the development stage variable with a weight of 7, consecutively the removal efficiency is placed with a value of 5 and, finally, the waste generation with a value of 3 [34–36,41,42]. Subsequently, the coefficients of each parameter were generated using the pairwise comparison matrix presented in Table 2.

With the factors w_i of each parameter, the WLC was proposed, obtaining Equation (1) for the evaluation of each drug abuse reduction technique.

$$\text{Technique} = 0.2083 \times \text{Efficiency} + 0.3750 \times \text{Cost} + 0.2917 \times \text{Stage} + 0.1250 \times \text{Waste} \quad (1)$$

With the application of Equation (1), the most adequate, profitable and economically viable technique was obtained in the removal of METHs for its application in a WWTP.

Table 1. Weighting of the study variables.

| Variable | Category | Values/Data | Weighting |
|--------------------|------------------------|--|-----------|
| Removal efficiency | Evil | 70–75% | 1 |
| | Regular | 76–80% | 3 |
| | Well | 81–85% | 5 |
| | Very good | 86–92% | 7 |
| | Excellent | 93–100% | 9 |
| Construction cost | Down | - | 9 |
| | Medium | - | 5 |
| | High | - | 1 |
| Development stage | Regular | Laboratory | 1 |
| | Well | Pilot Planpt | 5 |
| | Very good | Big scale | 9 |
| Waste generation | Does not generate | Does not imply | 9 |
| | Low detrimental | Carbon Diocid | 7 |
| | Slightly detrimental | Sludge and microbial biomass | 5 |
| | Moderately detrimental | Transformation products resulting from incomplete mineralization | 3 |
| | Highly detrimental | Free radicals, peroxides, and oxidative by products | 1 |

Table 2. Pairwise Comparison Matrix and WLC Coefficients.

| | Removal Efficiency | Construction Cost | Development Stage | Waste Generation | = | wi |
|--------------------|--------------------|-------------------|-------------------|------------------|---|--------|
| Removal efficiency | 1 | 0.5555 | 0.7142 | 1.6666 | = | 0.2083 |
| Construction cost | 1.8 | 1 | 1.2857 | 3 | = | 0.3750 |
| Development stage | 1.4 | 0.7777 | 1 | 2.3333 | = | 0.2917 |
| Waste generation | 0.6 | 0.3333 | 0.4285 | 1 | = | 0.1250 |

3. Results

The extraction of drugs of abuse in a WWTP depends on the nature of the contaminant, which is why it is necessary to identify its properties [43–45]. To determine the drugs of abuse present in WWTP, the high-performance liquid chromatography method coupled to low energy and high collision energy mass spectrometry can be used [46]. Once its presence in wastewater has been determined, it is essential to analyze a treatment that allows the removal of this type of EC. These removal treatments can be physical, chemical, or biological [47]. The following are drug abatement techniques with their respective advantages and disadvantages.

3.1. Sequential Batch Reactor (SBR) Activated Sludge Process

According to Miao et al. (2014), it has been mentioned that a sequential batch reactor (SBR) represents an elementary form of activated sludge treatment process systems that is based on the sequence of filling and emptying [48–50]. That is, this process includes unit operations, such as mixing, treatment anaerobic, aerobic, anoxic, or the combination of them, which finally conclude with sedimentation, these processes will take place in the same reactor [51–54]. Drugs of abuse are removed in the primary settling tank, due to the relationship of treatment conditions, such as ambient/water temperature, wastewater composition, or biological population, as well as in the emptying stage, when the clarified supernatant is discharged from the reactor as effluent [17,55]. After discharging the treated effluent, all but a small part of the sludge, which is rich in microorganisms, is removed from the reactor [56–58] as indicated in Figure 1.

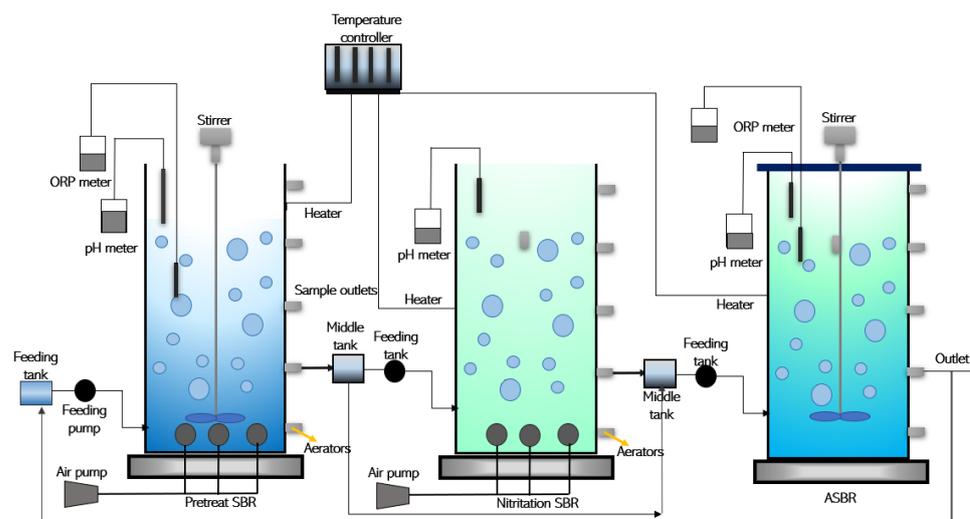


Figure 1. Stages of an operation cycle of an SBR reactor.

SBR technology is fundamental in wastewater treatment due to its high efficiency in decomposing organic pollutants [59–61]. For the abatement METH, a removal efficiency of more than 70% is achieved, likewise, the effectiveness of this technology has been demonstrated through the development of experiments in the laboratory and at the pilot plant level [17]. The main advantages of this process are the fast sedimentation speed, the high reaction speed, and the good degradation performance of the refractory organics [62,63]. In addition, it has potential construction cost savings due to not requiring the presence of an extended secondary sedimentation clarifier and return sludge pumping system, including minimal space requirement, ease of handling, and the ability to perform modifications in the treatment train [48].

3.2. Modified Fenton Reaction (FR)

The standard Fenton reaction (FR) abatement method is a type of chemical treatment that consists of an advanced oxidation process, which is based on the production of hydroxyl radicals ($\text{OH}\cdot$), considered one of the strongest oxidation agents [64–68]. The radicals that are formed during this process have low selectivity and are able to oxidize a wide spectrum of compounds corresponding to drugs of abuse [69,70]. Standard FR is a catalytic process based on the transfer of electrons between hydrogen peroxide (H_2O_2) and an iron salt as shown in Equation (2) [71].



Modified FR consists of the introduction of iron shavings as a source of Fe^{2+} cations in order to increase the efficiency of removal and degradation of contaminants such as drugs of abuse [72,73]. Figure 2 illustrates the general process of the modified Fenton reaction.

The modified FR is used in the treatment of WWTP in the effluents of the WWTP, in view of its simplicity and lower economic contribution compared to the standard FR. However, despite the fact that this technique has yielded good results in the efficiency of eliminating drugs of abuse, it is relatively expensive, and its implementation is not sustainable or economically viable [74]. Similarly, this treatment is in the laboratory stage to ensure a good performance of the Fenton process, constant control, and monitoring is maintained in the variation of concentration of the oxidizing and catalytic agent, pH, temperature and reaction time [71]. One of the negative aspects lies in the toxic effects on the environment due to the residues from the reactions involved in the Fenton process, that is, the environmental release of free radicals, peroxides, and oxidant by-products produced during the treatment of water, the same that can attack the genetic material of the ecosystem [75].

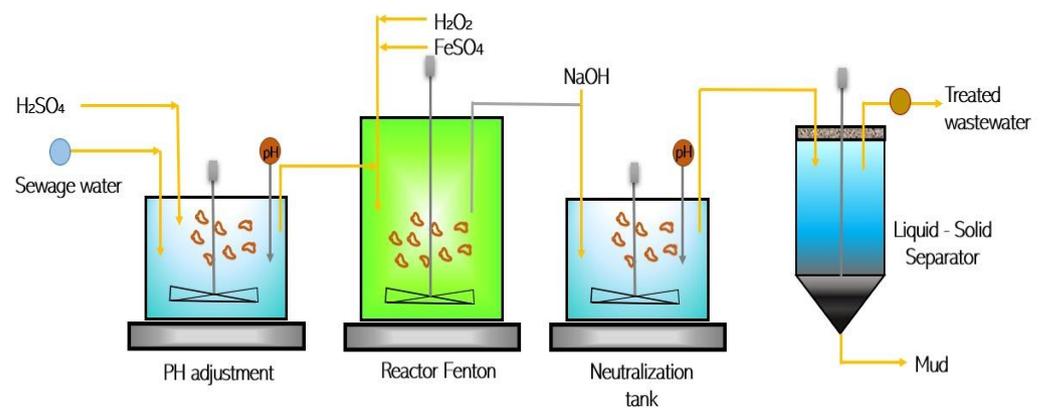


Figure 2. General scheme of the modified Fenton reaction (FR) process.

3.3. Mixed-Flow Bioreactor

Mixed-flow bioreactors are the combination of aerobic and anaerobic type bioreactors for the elimination of drugs of abuse present in WWTP [76,77]. The aerobic bioreactor uses dissolved oxygen as an electron retainer, which, in turn, converts organic material to its highly oxidized inorganic form, yielding carbon dioxide, nitrates, sulfides, and orthophosphates as byproducts [78]. The membrane aerobic biological bioreactor allows the detachment of sludge and RA containing drugs of abuse through the walls of filtration membranes. The filtered liquid is extracted into the system, meanwhile, the sludge and substances larger than the pore size of the membrane are detained in the reactor [78].

The anaerobic bioreactor joins the sedimentation of solids and their deposit in the base, with the floating material of the WWTP and the suspended active biomass. Both bottom mud and floating material provide adequate space for microbial growth [77,79]. This type of bioreactor generates sludge and microbial biomass as residual products, which are treated for reuse before leaving [80,81]. This method has indicated good efficiency in eliminating drugs of abuse at low concentrations, but in recent studies it has been accompanied by chlorination and maturation in ponds in order to allow better removal of drugs of abuse and their metabolites from WWTP [77,82,83]. Most of the processes that use this type of mixed bioreactor have been applied on a large scale for the treatment of water from industrial and domestic activities [78,84–86], as illustrated in Figure 3.

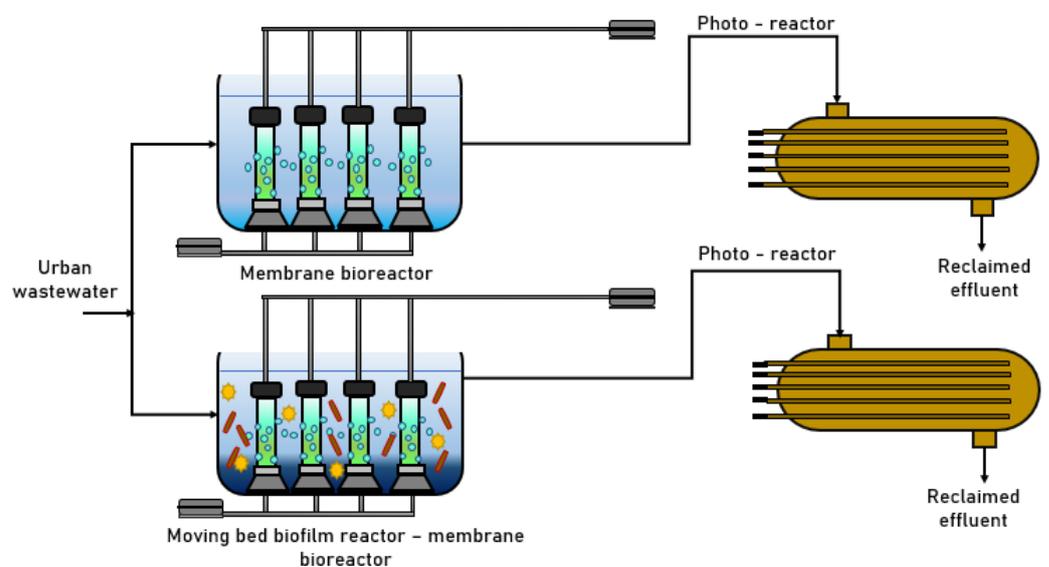


Figure 3. General scheme of the mixed-flow bioreactor.

Currently, this type of treatment constitutes an alternative with great potential, since it offers different advantages, such as low costs during its implementation for the treatment of a wide variety of WWTP, simple, and automated operation. Among the disadvantages is the constant cleaning of the membranes, which requires high maintenance costs. Likewise, one of the operational problems is the fouling of the membrane surface due to sludge, colloids, and metabolites that affect the capacity to remove contaminants [78].

3.4. Ozonation

Ozone (O_3) is a selective oxidant reactive against double bonds, aromatic systems, non-protonated secondary and tertiary amines, and reduced sulfur species. In addition, the hydroxyl radicals that are formed by the decomposition of O_3 also add to the oxidative potential of the ozonation process. For batch ozonation, a concentrated O_3 stock solution is used. A gas wash bottle filled with pure water and placed in an ice-filled bucket is then sprayed with a gaseous mixture of O_3 and oxygen gas [87]. Figure 4 illustrates a possible ozonation process.

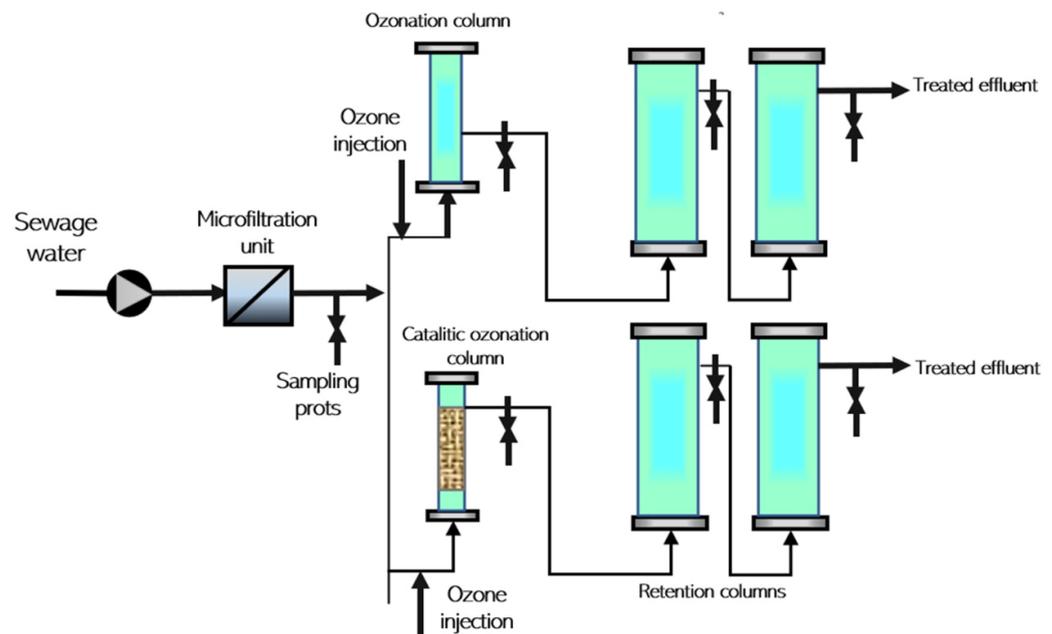


Figure 4. Ozonation process.

Oxidation with O_3 is considered as a removal technique in WWTP that favors the degradation of toxic metabolites from drugs of abuse, reaching an efficiency of at least 25% in the case of AMP and KET up to reach an efficiency of 96% in the case of BE. Although its development is on a laboratory scale, it is expected that through studies it will be possible to implement a pilot plant [88–90].

The implementation of this technique is quite feasible in terms of energy and cost, the estimated total price of ozonation is between €0.05 and €0.20 per m^3 of WWTP, which does not represent a large part of the total cost of maintaining a WWTP considering the size of the plant and the level of dissolved organic carbon remaining in the treated water. A problem that arises from the application of this method is the generation of possible transformation products that occur as a result of the incomplete mineralization of organic compounds in aqueous solution that often persist after the original compound has been completely removed and, in some cases, are more toxic to aquatic organisms than the parent compound [91].

3.5. Photocatalytic Degradation Using Illuminated TiO_2

Aziz et al. (2019) indicates that the photocatalytic treatment allows the degradation of pharmaceutical products, but it has a low mineralization rate, making it impractical

for the complete elimination of diclofenac (DCF) and ibuprofen (IBP), since it requires a long period of time for a high reaction [92]. However, it mentions that it is effective for the degradation of recalcitrant organic contaminants, such as pharmaceutical residues, or, at least, to transform them into biodegradable species [93–96]. The schematic diagram of the photocatalytic reactor is shown in Figure 5.

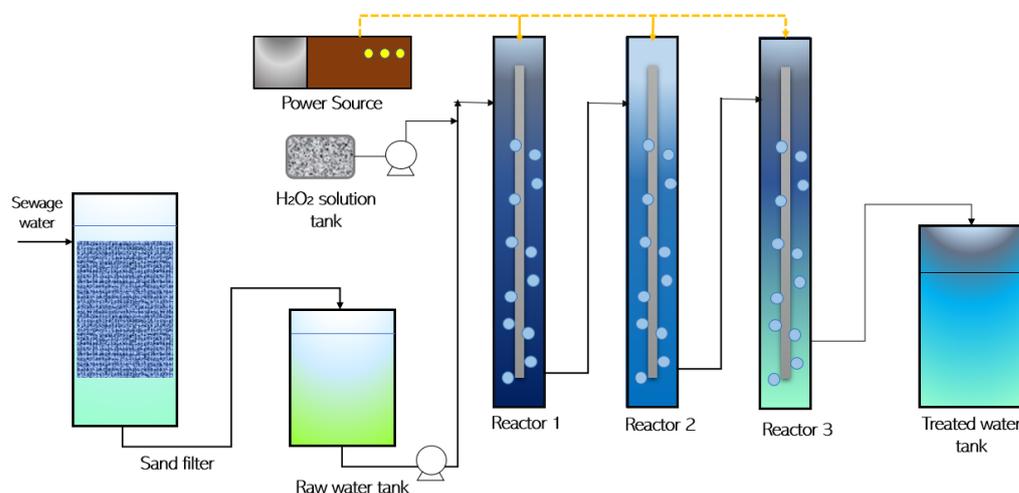


Figure 5. Scheme of a photocatalytic reactor.

Advanced oxidative processes (AOP) use the production of oxidants, such as hydroxyl (OH^\cdot), peroxy (O_2^\cdot), and hydroperoxide (HO_2^\cdot) radicals, to degrade organic materials and mineralize the contaminant to carbon dioxide and water. The hydroxyl radical is a strong oxidizing agent, but it is not selective, so it can react with several classes of compounds. The presence of these radicals initiates a series of reactions capable of oxidizing organic matter, justifying their importance in advanced oxidative processes for environmental decontamination [93,94]. The photocatalytic removal of organic contaminants turns out to be an effective alternative to biological methods for the separation of organic contaminants. Titanium dioxide (TiO_2) is one of the best known photocatalysts due to its efficient photoactivity, electron hole recombination rate, high stability, low cost, and safety for the environment and humans [95–97].

The application of this method has certain advantages, such as the successful removal of a wide range of molecules from drugs of abuse, for instance, KET, METH, and MOR, which reduces reagent costs, and also avoids the presence of traces of iron in the water, which could be a threat to human health. Although it is worth mentioning that its implementation has not yet reached commercialization, mainly due to the difficulties in the separation and recovery of the catalysts and their transparency to visible light, which is why it is still under development, presenting itself as a pilot plant [98,99].

3.6. UV Disinfection

It is a physical process based on UV radiation; they transmit a high radiation energy at <400 nm. The UV dose refers to the energy that enters the water, multiplied by the time the WW containing drugs of abuse is exposed to such energy. UV disinfection when combined with membrane bioreactors (MBR) can achieve the elimination of pathogens and even the destruction of contaminants, such as drugs of abuse [100–102]. In a constant flow system, the UV lamp is on all the time. However, excessive downtime causes a reduction in its useful life. In addition, the efficiency of the method will depend on the characteristics of the WWTP, emphasizing the concentration of colloidal components and residual particles, in addition to the intensity of the radiation and the configuration of the reactor [23].

The chemical reactions that occur in UV treatment can be complex and could include both direct and indirect photolysis reactions (Figure 6). This technique has been used in different industries, since among the advantages it is mentioned that being a physical

process does not imply the need to generate, handle, transport, and store residual toxic chemical substances. In addition, it is easy to use for operators and the disinfection equipment occupies less space compared to other methods [103].

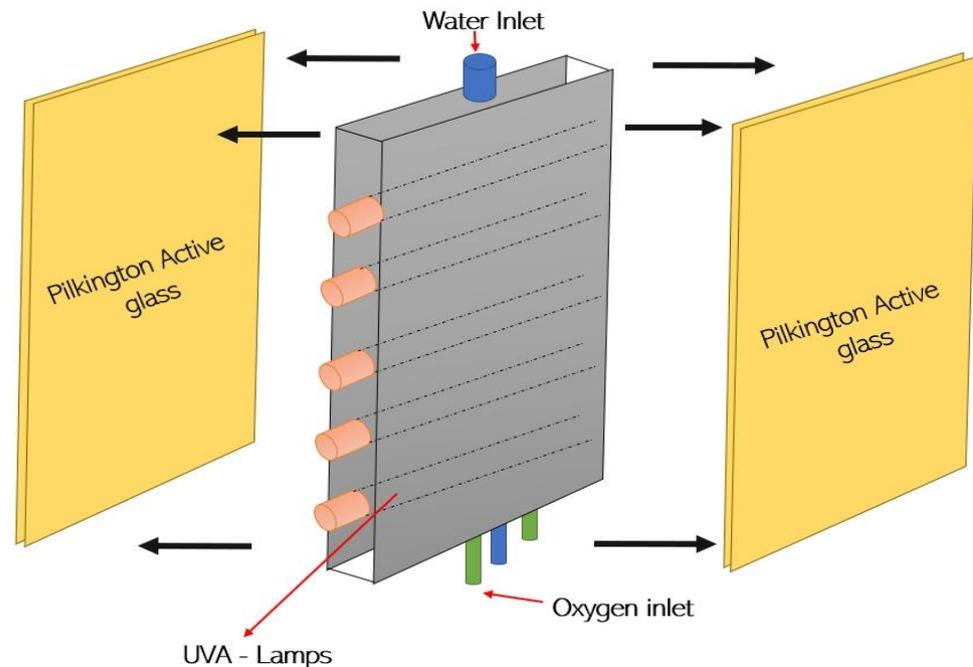


Figure 6. Scheme of UV disinfection process.

This treatment has been applied on a large scale in certain WWTPs, however, it has disadvantages such as constant and preventive maintenance in order to control the accumulation of solids on the outside of the light tubes. Similarly, its implementation is not economically viable since it requires a good investment for its operation [11]. Figure 6 demonstrates the wastewater treatment scheme using the UV disinfection technique.

A third treatment applied to the removal of EC, specifically drugs of abuse, is essential within the WWTP system, due to its better efficiency in the degradation of these compounds and their metabolites. Therefore, it is necessary that this measure be considered in order to avoid harmful damage to human health and environmental matrices in a prolonged time [78].

The use of a biological process such as bioreactors and SBR, contribute to reduce the space required for wastewater treatment. This is due to the fact that the four phases (filling, reaction, sedimentation, and emptying) are carried out in the same tank, with the advantage of a rapid speed in the reaction and sedimentation process, obtaining a high yield of degradation of these compounds. On the other hand, occupying little space within WWTPs helps make it economically viable and avoid incurring unnecessary expenses [17]. The incorporation of an additional treatment to those conventionally found in a WWTP (primary, secondary) for the cleaning of drugs of abuse in wastewater, attracts different advantages such as: achieving an effective removal of these contaminants since there are several investigations in those that demonstrate that physical, chemical, biological, or advanced processes have great potential to obtain treated water with greater biodegradability, and, in turn, hinder bioaccumulation in and around living organisms [47].

The bibliographic review used in this article allowed us to differentiate the efficiency, advantages and disadvantages of each drug of abuse removal treatment applied in different wastewater treatment plants, which is listed in Table 3.

Table 3. Advantages and disadvantages of treatments for the removal of drugs of abuse.

| Treatment/Type | Drug | Efficiency | Advantages | Disadvantages |
|---|-----------------------|------------|---|---|
| Sequential Batch Reactor (SBR)/ Biological Activated Sludge Process | METH | 70–95% | <ul style="list-style-type: none"> Fast sedimentation rate. High reaction speed. Good degradation performance of organic refractory materials. Low cost for its application. | <ul style="list-style-type: none"> It is not applicable to all types of organic effluent. The presence of toxic compounds affects the performance of this treatment, the toxic impacts being favored by punctual feeding. Requires a higher energy demand due to the discontinuous power supply of the system. |
| | Cocaine | 40–50% | | |
| | Benzoyllecgonine | >90% | | |
| | Amphetamine | >40% | | |
| | Methadone (MET) | >90% | | |
| | Codeine (COD) | >90% | | |
| | Ketamine (KET) | >75% | | |
| 3,4-Methylenedioxyamphetamine (MDA) | >60% | | | |
| Caffeine | 99% | | | |
| Fenton Reaction (FR)/Chemical | Amphetamine | 80% | <ul style="list-style-type: none"> Great applicability in the treatment of contaminated sludge. The reagents used in the reaction are easy to handle and abundant. Does not require energy sources for the generation of hydroxyl radicals (OH). | <ul style="list-style-type: none"> Large-scale application is not sustainable or economically viable. Requires adjustment and control of factors, such as: pH, concentration of the oxidizing and catalytic agent, and reaction time to ensure good performance. |
| | METH | 87% | | |
| | Benzoyllecgonine | 50–70% | | |
| | Cocaine | 50–70% | | |
| | THC-COOH | 80% | | |
| | Codeine | 50–70% | | |
| Methadone | 10% | | | |
| Mixed-flow bioreactor (anaerobic and aerobic)/Biological | Codeine | 95% | <ul style="list-style-type: none"> Simple and automated operation. Low costs for its implementation. A large space is not required to apply this treatment. Treatment applicable to different types of wastewaters. | <ul style="list-style-type: none"> Requires constant maintenance costs. Operational problems due to fouling of the membrane surface due to sludge, colloids, and metabolites. |
| | Caffeine | 91% | | |
| | Benzoyllecgonine | 85% | | |
| | Codeine | 90% | | |
| | METH | 85% | | |
| Methaqualone | 60% | | | |
| Ozonation/ Chemical | Benzoyllecgonine (BE) | 96% | <ul style="list-style-type: none"> It is quite feasible in terms of energy since it has a strong oxidizing power. The estimated total price is between € 0.05 and € 0.20 per m³ of wastewater. | <ul style="list-style-type: none"> Generation of possible transformation products that are more toxic to aquatic organisms than the original compound. |
| | Amphetamine (AMP) | >25% | | |
| | Ketamine (KET) | >25% | | |
| | Cocaine (COC) | 70% | | |
| | METH (METH) | 70% | | |
| Extasis (MDMA) | 50% | | | |
| Photocatalytic degradation using illuminated TiO ₂ /Chemical | Ketamine | 99.9% | <ul style="list-style-type: none"> High efficiency in the removal of organic and inorganic molecules due to its high reactivity, chemical and photocatalytic stability. Low acquisition cost and does not require meticulous care. | <ul style="list-style-type: none"> This area of research is still new. Little information on how to use it for operators. |
| | METH | 99.9% | | |
| | Morphine | 99.9% | | |
| UV Disinfection/ Physical | Benzoyllecgonine | 83% | <ul style="list-style-type: none"> There is no generation of toxic chemical substances. Simple use for operators. Disinfection equipment occupies less space in relation to other methods. | <ul style="list-style-type: none"> Constant and preventive maintenance in order to control the accumulation of solids in the external part of the light tubes. Inexpensive implementation. |
| | METH | 92% | | |
| | MDMA | 73% | | |
| | Codeine | 89% | | |
| | Morphine | 99% | | |

3.7. Modified Saaty Analysis

Once the principles of the six techniques studied and their behavior against the parameters of interest were known, the AHP was applied using Modified Saaty. Initially, the parameters of efficiency, implementation cost, development stage, and waste generation were compared with respect to METH abatement.

Applying Equation (1) for each of the abatement techniques, the values shown in Table 4 were obtained.

Table 4. Results applying the Modified Saaty method.

| Technique = | 0.2083 x Efficiency | + | 0.3750 x Cost | + | 0.2917 x Stage | + | 0.1250 * Waste | = | Results |
|--------------------------|------------------------|---|------------------|---|-------------------|---|-------------------|---|---------------|
| SBR= | 0.0000 | + | 0.2652 | + | 0.2062 | + | 0.0884 | = | 0.5598 |
| Ozonation = | 0.0000 | + | 0.0000 | + | 0.0000 | + | 0.1155 | = | 0.1155 |
| Fenton = | 0.1925 | + | 0.3750 | + | 0.0000 | + | 0.1250 | = | 0.6925 |
| TiO ₂ = | 0.2083 | + | 0.0000 | + | 0.2062 | + | 0.0478 | = | 0.4624 |
| Bioreactor = | 0.1473 | + | 0.0000 | + | 0.2917 | + | 0.0884 | = | 0.5274 |
| UV disinfection = | 0.1925 | + | 0.3750 | + | 0.2917 | + | 0.0000 | = | 0.8591 |

4. Discussion

The removal of drugs of abuse depends on the method and the characteristics of the compound. On the other hand, the method of photocatalysis with TiO₂ is proposed as a tertiary treatment process of a WWTP, this is due to the fact that it has a low acquisition cost and good efficiency compared to the rest of the techniques mentioned, additionally, it does not require careful treatment [104–107].

To implement a third treatment, it is necessary to know the type of EC that reaches the WWTP from wastewater, given that they belong to drugs of abuse such as METH, BE, COC, and KET, among others. The remediation for each of these compounds will depend on their properties, since in some cases they persisted, which did not allow a satisfactory removal, that is, a common treatment cannot be deduced for each of these compounds and their metabolites. However, the application of advanced biological processes (SBR and bioreactor) emerges as an optimal alternative for large-scale application [78]. Such is the case of the compound called BE, which has a removal efficiency of 96% by ozonation. However, when treating this metabolite by Fenton reaction, it has a removal rate that can vary between 50–70% [71].

Once the Modified Saaty method was applied, it was established that the best technique for abatement of METH is UV disinfection, since it has the highest weighting compared to the other techniques (0.8591). One of the factors that allows the highest weighting to be achieved is that the value corresponding to the generation of waste as stated by the authors Sun et al. (2022) and Afonso-Olivares et al. (2016) [101,103] is 0, that is, it is the only one of the six techniques studied that does not present residues after its application in a WWTP. Although this criterion is considered of lesser importance by the panel of experts. Likewise, the coefficients for implementation cost and efficiency represent the highest values, as indicated in Equation (1). While the ozonation technique is presented as the least viable option in the implementation of a WWTP for abatement METH, this is due to the fact that the values that accompany each of the weights of the WLC are zero, specifically in three (efficiency, cost of implementation, and stage of development) of the four criteria analyzed.

The modified Fenton technique is the second-best alternative for implementation in a WWTP, given that it has a good weighting with respect to the evaluation of the criteria analyzed. According to the authors Mackul'ak et al. (2015) and Catalá et al. (2015) [71,75], the elimination efficiency for the abatement of METHs reaches up to 87%, so the weighting

of this technique is high. In the same way, Fenton affects the generation of highly harmful waste, however, this criterion has less weight compared to the rest.

Biological-type techniques, such as SBR, and mixed-flow bioreactor are in a range of less than 0.6 regarding the final weighting. This occurs despite the fact that biological techniques always imply less impact on the environment, as stated by Osman Awaleh et al. (2014) [59]. However, the applied ranking techniques (Saaty and Modified Saaty) indicate the best one that maintains a balance between the analyzed variables.

In the case of the photocatalytic degradation technique using TiO_2 , it indicates a weighting of less than 0.5. This result is due to the fact that its implementation was realized in a pilot plant, which puts it at a disadvantage with some of those studied that are already on a large scale. The difficulty of this technique in moving to the next stage of development is commercialization as indicated by Al-Abduljabbar et al. (2021) [98], affecting the weighting variables. In addition, the generation of waste is relatively unprofitable which gives it a score that can be deduced that this technique is within a moderate range.

The Saaty and Modified Saaty ranking methods allowed the generation of a tool that is easy and quick to apply for decision-makers, through a mathematical analysis whose result is to find the abatement method with the best characteristics, that is, the one that saves a better balance with the study variables. In summary, each treatment technique previously analyzed has its advantages and disadvantages which is reflected in the final weighting through the prioritization techniques previously studied in this scientific document with which it was obtained that the optimal technique for the abatement of METH in WW is UV disinfection.

5. Conclusions

In recent years, knowledge about the presence of different ECs in aquatic environments has intensified. However, it is necessary to increase the information regarding the removal of drugs of abuse in a WWTP. The present work conducted a bibliographic review with relevant scientific articles of the last eight years, finding that the SBR, Fenton reaction, mixed-flow bioreactor, ozonation, photocatalysis, and UV disinfection techniques are the most popular to abate METHs. Subsequently, the Saaty and Modified Saaty methodology was applied with the aim of finding the technique that maintains a good relationship between efficiency, implementation costs, development stage, and waste generation, resulting in the UV Disinfection technique presenting the highest weighted value of 0.8591, while the ozonation technique presents the lowest value 0.1155, which means that the UV technique has a higher balance (score) between the parameters presented.

The work performed in this research is presented as a useful tool for decision-making based on the knowledge of the panel of experts and analytical consistency, since it allows to ensure maximum efficiency with the lowest implementation costs and the lowest generation of waste with proven techniques on a large scale.

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