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Literature Review

A review of approaches to atrazine treatment employing advanced oxidation processes technologies

Uma revisão das abordagens de tratamento da atrazina empregando tecnologias de processos oxidativos avançados

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ABSTRACT

Atrazine is a synthetic pesticide widely used in many crops. It is considered a contaminant to soil and water, and due to its leaching and recalcitrant capacities, new treatment technologies have been developed for its removal, with an emphasis on advanced oxidation processes (AOPs), since conventional wastewater treatments show reduced capacity to remove persistent organic pollutants. This article provides a literature review of the main AOP approaches, such as photolysis, ozonation, photoperoxidation, fenton and photo-fenton, photocatalysis, and electrochemical processes, for the atrazine treatment and the fundamentals behind each process. In addition, the innovations and applications of hybrid AOP systems were documented. It is worth mentioning that despite the high efficiency obtained by AOPs in the degradation of ATZ, it is important to evaluate the resulting toxicity and by-products formed, as well as the costs associated with the application of these processes.

Keywords: oxidative process; degradation pathways; removal efficiency, contaminated environment, atrazine by-products.

RESUMO

A atrazina é um agrotóxico sintético amplamente utilizado em diversas culturas. É considerado um contaminante do solo e da água, e, devido à sua capacidade lixiviante e recalcitrante, novas tecnologias de tratamento têm sido desenvolvidas para sua remoção, com destaque para os processos oxidativos avançados (POAs), uma vez que os tratamentos convencionais de efluentes apresentam capacidade reduzida de remover poluentes orgânicos persistentes. Este artigo fornece uma revisão da literatura sobre as principais abordagens de POAs, como fotólise, ozonização, fotoperoxidação, fenton e fotofenton, fotocatálise e processos eletroquímicos, para tratamento da atrazina e os fundamentos reacionais por trás de cada processo. Em adição, as inovações e as aplicações de sistemas POAs híbridos foram documentadas. Vale ressaltar que apesar da alta eficiência obtida pelos POAs na degradação da ATZ, é importante avaliar a toxicidade resultante e os subprodutos formados, além dos custos associados à aplicação desses processos.

Palavras-chave: processo oxidativo; vias de degradação; eficiência de remoção; ambiente contaminado; subprodutos da atrazina.

INTRODUCTION

Among many organochlorine pesticides, atrazine (ATZ), as a s-triazine pesticide, has been used primarily to control broadleaf weeds and grasses in agriculture crops (Xu *et al.*, 2019). The extensive use and persistence of ATZ cause contamination of soil and water (Hanson *et al.*, 2019). Its presence in the environment is alarming, due to its potential toxicity in animals as a reproductive corruptor and immunomodulator (Albuquerque *et al.*, 2020; Jiang, C. *et al.*, 2020) and in humans as a potential endocrine deregulator and probably carcinogenic (Komtchou *et al.*, 2020; Zhu *et al.*, 2021).

From around 20 different classes of existing pesticides, the class of triazine substances is among the most consumed throughout the world, and the focus of this study is on ATZ, which remains among the most frequently monitored and studied compounds in groundwater, surface water, and soil (Lee, 2003; Hong *et al.*, 2019; Jiang, C. *et al.*, 2020; Esquerdo *et al.*, 2021). Due to its inevitable aquatic contamination, ATZ was banned in the European Union (EU) in 2003 (Yue *et al.*, 2017). Yet most countries, including the USA, India, China, and Brazil, still have large-scale consumption (Chandra; Usha, 2021). And even in territories where it has already been forbidden, ATZ is still found in aquatic

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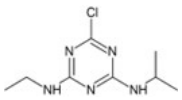
environments. While the hydrological cycle is the main cause of contamination due to leaching, the water-contaminated influx by runoff is continuous (Pascal-Lorber; Laurent, 2010). The study by Vizioli *et al.* (2023) reports the detection of ATZ and three of its by-products, at concentrations ranging from 2.0 to 2.7 ng L^{-1}, in the surface waters of the Capivari and Atibaia rivers, as well as detection in treated water (Campinas, Brazil). In addition, ATZ has also been detected in the Yangtze and Huangpu rivers in China, from 8 to 180 µg L^{-1} (Sun *et al.*, 2018), and in surface and groundwater in the United States, from 1 to 25 µg L^{-1} (Tillitt *et al.*, 2010; Wang *et al.*, 2022).

The physical–chemical properties of ATZ (Table 1) match its characteristics of high hydrophobicity (high log K_{ow} value), moderate polar nature, which is linked to its moderate solubility in water, high dipole moment, and complex molecular structure (Oliveira Jr; Koskinen; A Ferreira, 2001). Due to its half-life between 41 and 231 days (Karlsson *et al.*, 2016), low adsorption in soils, and moderate aqueous solubility, it has the potential to contaminate not only agricultural fields but also groundwater, mainly in a well-structured soil profile with macropores (Dias *et al.*, 2019).

It is known that the presence of ATZ in the aquatic environment directly interferes with the food chain of aquatic animals, as the pesticide can significantly inhibit algae growth and photosynthesis (Zhu *et al.*, 2016). Singh *et al.* (2018) reviewed studies in which ATZ manifested acute toxicity to some species of amphibians and fish, interfering mainly in biotransformation, generating a significant increase in genotoxic damage and oxidative stress, inducing changes in enzymatic activities, and damaging the endocrine and liver systems of these animals. Albuquerque *et al.* (2020) also reviewed studies that analyzed the toxic effect on mollusks, crustaceans, and insects, disrupting biomarkers, hormones, morphology, the reproductive system, and genetics, and also on reptiles, damaging the immune system and their development. In mammals, the main action of ATZ is to disrupt the endocrine system (La Casa-Resino *et al.*, 2012; Jin *et al.*, 2014; Cook *et al.*, 2020; Galbiati *et al.*, 2021), followed by oxidative stress (Jestadi *et al.*, 2014; Saalfeld *et al.*, 2018; Semren; Žunec; Pizent, 2018; He *et al.*, 2022), sexual hormone imbalance (Bohn *et al.*, 2011; Govers *et al.*, 2020; Abarikwu *et al.*, 2021), DNA damage (Yang *et al.*, 2010; Sánchez *et al.*, 2020; Xie *et al.*, 2021), which can even affect cardiovascular functioning (Lin *et al.*, 2016; Li *et al.*, 2017; Li *et al.*, 2018), as well as liver damage (Campos-Pereira *et al.*, 2012; Foulds *et al.*, 2017; Harper; Finger; Green, 2020). Proven evidence of ATZ's toxicity culminated in its classification as an endocrine-disrupting pesticide by the United States Environmental Protection Agency (USEPA, 2007) and also its entry into the International Agency for Research on Cancer's list of carcinogenic pesticides (IARC, 1999).

Table 1 – Physicochemical properties of atrazine.

Atrazine (ATZ)	
Molecular structure[a]	
Formula[a]	$C_8H_{14}N_5Cl$
IUPAC name[b]	2-Chloro-4-ethylamino-6-isopropylamino-1,3,5-triazine
Molecular weight[c]	215.685 g mol^{-1}
Melting point[a]	171–174 °C
Solubility[a] in water at 20–25 °C	33 mg L^{-1}
pKa[a]	1.68
Log K_{ow} [c]	2.61
Dipole moment[c]	4.6 D
Polluted biotopes[d]	Fields, Water networks
Geo-localization[d]	Watersheds
Persistence[d]	15–100 days
Plant uptake[d]	High
Phytotoxicity[d]	High

Source: Adapted from [a]Lee (2003), [b]Zhu *et al.* (2016), [c]Chandra and Usha (2020), and [d]Pascal-Lober and Laurent (2010).

ATZ may be removed by conventional techniques such as coagulation–flocculation by the addition of ferrous iron coagulants or low-cost biosorbents (Cheng *et al.*, 2017; Liu *et al.*, 2020), adsorption on humic substances, clays, oxyhydroxides, and activated carbon (Hong *et al.*, 2019; Sun *et al.*, 2021), nanofiltration and microfiltration membranes (Ahmad; Tan; Shukor, 2008; Chandra; Usha, 2021), or reverse osmosis (Naidu *et al.*, 2017). However, these technologies only transfer the contaminant from one state to another (Romero *et al.*, 2020). Other traditional wastewater treatment methods, such as biodegradation (Derakhshan *et al.*, 2018; Singh *et al.*, 2018), may not efficiently and completely remove ATZ in a short period, making the degradation of ATZ even more challenging. To mitigate this, a great deal of research effort has been invested to remove ATZ and its by-products with different approaches, mainly methods based on chemical reactions, which can eliminate the contaminants through complete mineralization (Komtchou *et al.*, 2020).

The advanced oxidation processes (AOPs), which involve the formation of radicals with excellent reactivity, have emerged as an effective solution to eliminating persistent organic pollutants (Hong *et al.*, 2019). Since the 21st century, publications on ATZ degradation by AOPs have increased exponentially over traditional methods, including Fenton and photo-Fenton reactions (Cheng *et al.*, 2017), photolysis (Moreira *et al.*, 2017), ozonation (Yang *et al.*, 2016), peroxidation (Kida; Ziembowicz; Koszelnik, 2018), heterogeneous catalysis (Santacruz-Chávez *et al.*, 2015), and even applications of combined AOPs (Ahmed *et al.*, 2017). These processes stand out for their increased efficiency in removing ATZ and its byproducts in a shorter time (Dhangar; Kumar, 2020). Furthermore, currently, advanced oxidation processes based on sulfate radicals have drawn considerable attention due to their high efficiency over a wide pH range and multiple operation advantages (Wang *et al.*, 2019). This review article makes a unique contribution to the literature on the application of AOPs in the degradation of atrazine, aiming to critically evaluate the feasibility of AOPs in physical, chemical, and hybrid treatments as a means of removing ATZ from water. Specifically, this article provides a summary of the effectiveness of different AOPs in water treatment for ATZ removal combined with their degradation pathways and the formation of by-products. It is discussed beyond conventional AOPs and hybrid processes for the removal of ATZ, including the challenges and current knowledge gaps that limit the effectiveness of AOPs.

BIOLOGICAL AND PHYSICAL TREATMENT TECHNOLOGIES

Biological treatment technology is one of the leading techniques applied for atrazine degradation (He *et al.*, 2019). Biological nitrification or denitrification is

considered an alternative. Phan *et al.* (2014) observed this process in an anoxic–aerobic membrane bioreactor applied to a set of 30 compounds, including atrazine, but only 8% was removed. Kamaz *et al.* (2020) also applied a membrane bioreactor to remove ATZ detected at 0.02 ppm in a real effluent, resulting in approximately 20% removal, consequently; it could estimate the probability of ATZ biodegradation in that wastewater treatment plant. Microorganisms (such as bacteria, fungi, and microalgae) with great adaptability and mutability in a polluted environment were also studied for the biodegradation capacity of atrazine alone and/or in vitro, including *Pseudomonas* sp. (Zhao *et al.*, 2017; Fernandes *et al.*, 2018; Sharma *et al.*, 2019), *Achromobacter* sp. (Fernandes *et al.*, 2018; Cao *et al.*, 2020; Jiang, Z. *et al.*, 2020), *Aspergillus niger* (Herrera-Gallardo *et al.*, 2021), *Aspergillus oryzae* (Lu *et al.*, 2021), and microalgae of the Phylum *Chlorophyta* (Matamoros *et al.*, 2015).

Regarding physical methods, sedimentation and flocculation processes are not effective for removing pesticides such as ATZ, as they remain partitioned in the aqueous phase (Ahmed *et al.*, 2017). However, membrane filtration and adsorption technologies stand out. The membrane filtration process has been used in wastewater treatment plants, with the type of membrane varying according to the type of pollutant to be treated (Jatoi *et al.*, 2021). The study carried out by Ahmad, Tan, and Shukor (2008) evaluated the adsorption capacity of ATZ on four nanofiltration membranes: NF90, NF200, NF270, and DK, with the best result being obtained for NF90, with 95% retention of ATZ at 2 mg L^{-1}. According to Bodzek and Konieczny (2018), the type of membrane material and the molecular weight cut-off (MWCO) are the main parameters that influence ATZ removal in ultrafiltration processes, obtaining approximately 60% removal when using a membrane with an MWCO of 1–2 kDa. The adsorption process is characterized by the use of adsorbents such as activated carbon, biochar, zeolite, and bentonite, among others (He *et al.*, 2019). The efficiency of the adsorption process in the treatment of ATZ varies according to the surface area, the number of active sites available, and the type of interaction that occurs between the adsorbent and the adsorbate (Jatoi *et al.*, 2021).

The biological processes mentioned here have limitations, such as ambient temperature, salinity, pH, nutrient content, toxic substances, and other factors that will affect the efficiency of microorganism degradation. Among the advantages are the non-generation of toxic by-products, as occurs in some chemical treatments, low energy consumption, and low operating costs (Ahmed *et al.*, 2017). Therefore, there is still a search for microorganisms with better performance and environmental tolerance when applied to real conditions (He *et al.*, 2019). Physical processes such as membrane filtration and adsorption generally have a high efficiency in removing recalcitrant organic contaminants such as ATZ. In the case of nanomaterials, despite having excellent adsorption properties, they have a high preparation cost (He *et al.*, 2019), and the adsorption process involves the challenge of regenerating the adsorbent. The main disadvantage of physical processes is that most of them involve transferring the contaminant from the aqueous phase to a solid phase, leading to the problem of discarding or reusing this solid material.

ADVANCED OXIDATION PROCESSES (AOPS)

With the increasing detection frequency of contaminants in the water, new treatment technologies had to be developed. Currently, AOPs are recognized as one of the most effective alternatives for the degradation of compounds of environmental relevance (Lelario *et al.*, 2016). AOPs are based on the formation of reactive radicals, which have a high oxidizing power and can result in the degradation of different pollutant compounds in relatively short periods (Jiang, C. *et al.*, 2020). These radicals can be formed by several processes that can be classified into homogeneous or heterogeneous systems, depending on the absence or presence of catalysts in the solid state, in addition to whether it may be under irradiation (Oliveira *et al.*, 2020). The main AOPs can be identified as the processes of photolysis, ozonation, photoperoxidation, photo-Fenton, photo-catalysis, electro-Fenton, ultrasound, or gamma ray irradiation (Saleh *et al.*, 2021), and their reactions are shown in Table 2, besides combined approaches. These processes can be applied to ATZ removal from wastewater with high efficiency (Salimi *et al.*, 2017), as presented in Figure 1.

Photolysis

The initial studies involving atrazine photolysis were published in the 1990s, with emphasis on the study made by Bourgine *et al.* (1995), which achieved 30% degradation of ATZ when applied energy of 200 Wh m^{-3} and 70% with 700 Wh m^{-3}, promoting the statement that the degradation rate of atrazine is directly proportional to the photolytic energy supplied to the system. Chen *et al.* (2009) used a cylindrical reactor equipped with an immersible low-pressure mercury lamp (8 W), providing the system with an ultraviolet intensity of 0.96 mW cm^{-2}. The atrazine was mineralized at 90% after 50 min of reaction. In their work, it was observed that the pH of the reaction medium suffered a slight decrease, explained by the formation of acidic by-products such as cyanuric acid, ammelide, and ammeline. Hence, the possible reaction pathways for the photodecomposition of atrazine have been identified and quantified by liquid chromatography-mass spectrometry and the intermediates were determined: chloro-dealkylation, dechlorination-hydroxylation, alkylic-oxidation, dechlorination-hydrogenation, dechloro-dealkylation, deamination-hydroxylation, and olefination. And due to the formation of these byproducts that do not absorb the lower wavelengths of the spectrum of light, complete mineralization has been difficult to achieve. This same occurred with studies such as the one by Kong *et al.* (2016); they checked the direct photolysis of atrazine using a low-pressure UV lamp with λmax=254 nm and UV irradiance of 0.15 mW cm^{-2} after calibration. For [ATZ]=1.0 μM and pH=7.0, the process reached approximately 40% of the mineralization of ATZ. Similarly, Moreira *et al.* (2017) studied the atrazine degradation process (3 mg L^{-1}) and the formation of metabolites using the same kind of UV lamp, recording a 34% atrazine reduction after 300 s. These authors observed a substantial increase in the presence of atrazine-2-hydroxy (HAT), resulting from the hydroxylation of the halogenated carbon.

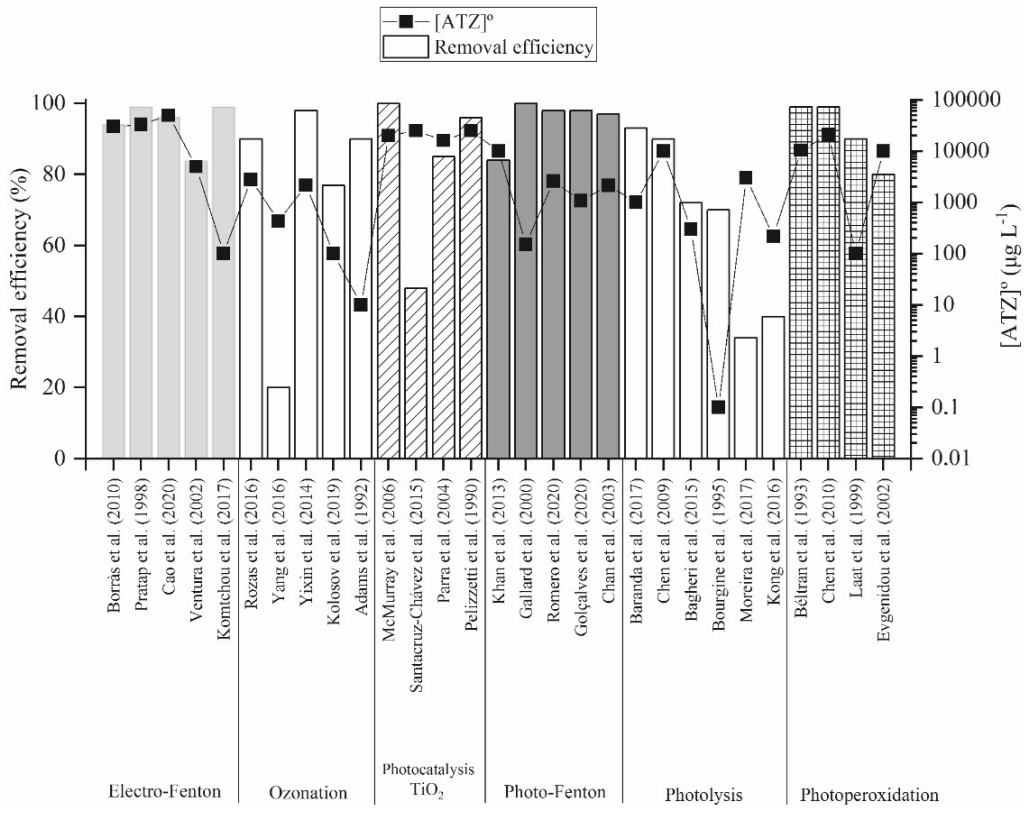
Ozonation

The ozonation process has been widely applied in water and wastewater treatment, as well as for disinfection and the degradation of toxic organic pollutants. Nevertheless, ATZ can be considered resistant to degradation by ozonation due to its low reactivity with ozone (Acero; Stemmler; von Gunten, 2000). In water, the ATZ and other ozone-refractory compounds were eliminated with lower efficiency; this was caused by the production phenomenon of OH• during aqueous ozonation. Yang *et al.* (2016) observed that only 20% of ATZ at an initial concentration of 2 μM was degraded by ozonation at a neutral pH. In an alkaline medium, Yixin *et al.* (2014) witnessed that the ATZ degradation

Table 2 – The generalized reactions of advanced oxidative processes.

AOP	Reactions	Light energy required
Photolysis	$H_2O + hv \rightarrow HO^{\bullet} + H^{\bullet}$	
Ozonation Photo-ozonation	$O_3 + OM \rightarrow O_2 + \text{oxidized OM}$ $O_3 + hv \rightarrow O_2 + O(^1D)$ $O(^1D) + H_2O \rightarrow 2HO^{\bullet}$	λ < 310 nm
Photoperoxidation	$H_2O_2 + hv \rightarrow 2HO^{\bullet}$	λ < 280 nm
Fenton Photo-Fenton	$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + HO^- + HO^{\bullet}$ $Fe^{3+} + H_2O_2 \leftrightarrow H^+ + FeOOH^{2+}$ $FeOOH^{2+} \rightarrow Fe^{2+} + HO_2^{\bullet}$ $Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + HO^- + HO^{\bullet}$ $Fe(OH)^{2+} + hv \rightarrow Fe^{2+} + HO^{\bullet}$	λ < 580 nm
Photocatalysis	$TiO_2 + hv \rightarrow TiO_2(e_{CB}^-) + TiO_2(h_{VB}^+)$ $TiO_2(e_{CB}^-) + OH^- \rightarrow TiO_2 + HO^{\bullet}$	λ < 280 nm
Electro-Fenton	$OM + H_2O \rightarrow OM(HO^{\bullet}) + H^+ + e^-$ $MO(HO^{\bullet}) + R \rightarrow MO + CO_2 + H_2O + H^+ + e^-$ $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$	

OM: organic matter; R: pollutant.
Source: author (2023).



Source: author (2023).

Figure 1 – Comparison of removal efficiencies of ATZ by oxidation processes.

rate significantly increased with ozone applications. The removal of ATZ with an initial concentration of 10 µM ATZ reached 98% when the solution was pH 10, five times greater than at pH 2. That was because the OH– ion is one of the main initiators of the decomposition of aqueous ozone into hydroxyl radicals. However, in addition to the high energy consumption and high cost, according to Wang and Chen (2020), some toxic disinfection by-products can be formed during the ATZ ozonation process, especially trihalomethanes and haloacetic acids, which have had a negative effect on human health. To mitigate this environmental impact, the catalytic ozonation process has earned increasing attention in recent years, as recent studies report a potentiate of the efficiency of ozone in the degradation of ATZ in the presence of other oxidants such as hydrogen peroxide, nitrite, and hydroxylamine, among others, as well as combined processes with microwaves, UV, and membranes (Kolosov; Yargeau, 2019; Rajah *et al*., 2019).

Photoperoxidation

Beltrán, Ovejero, and Acedo (1993) carried out one of the first studies involving the degradation of the pesticide atrazine in water as a function of the photoperoxidation process. In this study, the addition of different concentrations of H_2O_2 was analyzed with the aid of a mercury lamp (15 W), emitting radiation at 254 nm. For an H_2O_2 concentration of 0.01 M, approximately 99% of the ATZ oxidation rate was obtained just after 7 min of the process. When subjected to higher concentrations of H_2O_2, there was an abrupt decrease in efficiency, which was explained by the excess reagent added to the reaction. It is known that with excess peroxide and high concentrations of HO•, competitive reactions occur that slow the degradation rate (Ferreira; Maniero; Guimarães, 2015), but the conversion of H_2O_2 to hydroxyl radicals did not show results greater than 10% in any of the cases studied. These results were due to the amount of H_2O_2 added. However, it should be noted that there was a higher generation of hydrogen peroxide molecules because of the parallel reactions of the generation of the hydroperoxyl radical.

De Laat *et al*. (1999) performed a comparative kinetic study of processes using H_2O_2 in a cylindrical photochemical reactor with a low-pressure mercury-vapor lamp. The initial study conditions were [ATZ]=100 µg L^{-1}, and the pH was adjusted to 3.0 using solutions of perchloric acid and sodium perchlorate. At a dosage of 0.5–5 mM H_2O_2, a degradation of 90% of ATZ was achieved. Chen *et al*. (2011) investigated the oxidative decomposition of atrazine in water using a photochemical reactor and determined the ideal conditions for its decomposition. For [ATZ]=20.8 mg L^{-1}, total degradation was achieved with a dose of 300 mg L^{-1} of hydrogen peroxide. In this study, the experiments were restricted to a nearly neutral pH value, between 5.0 and 7.0. The oxidation rate of the atrazine molecule depended on three distinct contributions in this case: UV incidence, H_2O_2 addition, and HO• radicals generated. Thus, the action of hydrogen peroxide alone without combined photolysis did not result in a considerable oxidation potential for the degradation of this contaminant. In this case, two decomposition pathways of ATZ were highlighted: direct photolysis and HO• radical oxidation. The applied treatment was not able to reach complete mineralization of the molecule, but it had a great contribution in inducing the dealkylation of aromatic ring lateral chains, including its byproducts having less toxicity than the original compound.

Fenton and photo-Fenton

The study conducted by Gallard and de Laat (2000) was one of the pioneers in the kinetic evaluation of atrazine decomposition in the Fe^{III}:H_2O_2 process. The concentrations used were 0.2 mM<H_2O_2<1 mM and 1<H_2O_2:Fe^{III}<5.103 at pH 3.0, and the degradation rate reached approximately 100% in less than 30 min. Chan and Chu (2003) also studied ATZ removal by the Fenton reaction in multiple concentration ratios of ferrous ions (3<Fe^{II}:H_2O_2<0.33) and hydrogen peroxide (0.01 mM<H_2O_2<0.2 mM), obtaining a sharp removal curve in the first minutes and then gradually slower during the reaction. The concentrations of ferrous ion and hydrogen peroxide are significant in the oxidation rate of ATZ. When a high dose of iron was employed, the number of ferrous ions consumed by the process was not considerable, and the oxidation capacity was not affected. Conversely, when the Fe^{2+} dosage was low, the action of H_2O_2 was notable in the oxidation by the Fenton process.

The research conducted by Barreiro *et al*. (2007) evaluated the oxidative decomposition of ATZ at different pHs in an abiotic process using ferrihydrite and H_2O_2 to investigate the influence of parameters such as pH, ferrihydrite, and H_2O_2 concentrations. The applied process was the Fenton-like reaction, which is characterized by the use of native or synthesized iron oxide (FeO, Fe_2O_3, and Fe_3O_4) instead of free iron ions (Cheng *et al*., 2016). The contribution of pH was confirmed by detecting oxidation rates 10 times faster at pH 3.0, stimulated by the formation of Fe^{III} species. Therefore, when conducting experiments in a high pH range (6.5–8), the oxidation rate decreases, which can be explained by the reduction in the concentration of solubilized iron compounds in the system since ferrihydrite is better solubilized at acid pH.

Fenton-like reactions have an unconventional character for not using free iron ions, which means that they are replaced by iron-based industrial by-products or native iron compounds, thus making the Fenton-like reaction a sustainable alternative. However, this process is three times slower than conventional Fenton, as the regeneration of Fe^{II} is mainly responsible for the total reaction time. In this system, there is also the production of $HO_2^{\bullet}$, which is a radical with a lower oxidation potential compared to HO• (Romero *et al*., 2020). Therefore, there is a growing interest in designing working conditions more favorable to the Fenton-like reaction, expanding the time and pH range required (Zhang; Zhou, 2019). Rocha *et al*. (2024) used the extraction residue, basalt powder, as a natural photo-Fenton catalyst for ATZ degradation under UV-C (96%) and visible (56%) light under circumneutral pH. The authors evaluated the reusability of the catalyst, the degradation of by-products desethyl-atrazine (DEA), desisopropyl-atrazine (DIA), and hydroxyatrazine (HA), and toxicity tests. The research carried out by Gonçalves *et al*. (2020) evaluated a photo-Fenton process using organic iron ligands, biodegradable ethylenediamine-N and N′-disuccinic acid, to allow application in a wide pH range (3–9). This system achieved a 98% degradation of ATZ in only 15 min at pH 6.0, achieving greater mineralization than classic photo-Fenton at pH 2.7.

Photocatalysis

In the 1960s, photocatalysis emerged as an alternative to remove pesticides such as ATZ from contaminated water (Ameta *et al*., 2018; Long *et al*., 2020). The homogeneous catalytic degradation processes include transition metal ions (Mn^{2+}, Fe^{3+}, Co^{2+}, Cu^{2+}, and Zn^{2+}) as catalysts and result in the efficient degradation of organic pollutants (Xu *et al*., 2019; Wang; Chen, 2020). Wang *et al*. (2019) studied the degradation of ATZ at an initial concentration of

23 µM in the presence of activated peroxymonosulfate (PMS) by Cu-doped $LaFeO_3$ perovskite. In both heterogeneous and homogenous activation of PMS, ATZ was removed almost in its entirety; however, the homogeneous reaction process contributed limitedly to PMS activation. Cobalt-mediated activation of PMS was investigated by Chan and Chu (2009) with 95% ATZ removal by homogeneous process at an initial concentration of 0.1 mM, but four times slower than by heterogeneous process, in which PMS was activated by Co-TiO_2 particles in the presence of UV-Vis. Although homogeneous catalysis is an efficient process for removing organic pollutants because of the high solubility of transition metals, it is not technologically feasible to recover them, thus leading to secondary pollution caused by residual metal ions (Wang *et al*., 2019; Xu *et al*., 2019).

Therefore, research has focused on heterogeneous catalysis and photocatalysis by insoluble metal oxides. Santacruz-Chávez *et al.* (2015) used TiO_2 in metallic nanoparticles, including Au, Ni, and Cu, by deposition–precipitation for the degradation and mineralization of the pesticide atrazine in solutions with an initial concentration of 25 ppm. The Au/TiO_2 catalyst was the most successful in approximately 80% of the degradation of ATZ, followed by Cu/TiO_2 and Ni/TiO_2. Aragay, Pino, and Merkoçi (2012) reported that photocatalysis of ATZ by ceramic modified with TiO_2 resulted in excellent catalyst performance concerning the degradation of ATZ with a removal efficiency of 96% of total organic carbon (TOC).

Moreover, recent studies on heterogeneous catalysis have been published that aim to remove ATZ, mainly regarding persulfate-based advanced oxidation processes (Li *et al*., 2019). Shen *et al*. (2020) applied bismuth molybdate nanosheets (Bi_2MoO_6) to activate PMS, resulting in a 99% ATZ removal efficiency at an initial concentration of 2.5 mg L^{-1} under visible light irradiation after 60 min of reaction. Xu *et al*. (2019) fabricated a new Fe_3O_4-sepiolite magnetic composite that was used as a catalyst to activate persulfate and resulted in the degradation of 71.6% of ATZ and 20.9% of solution TOC after 60 min with an initial concentration of 10 mM of ATZ. In short, heterogeneous catalysis, more specifically semiconductor-supported solar photocatalysis, has attracted attention in environmental remediation for removing atrazine owing to its unique characteristics of low cost and higher thermal and mechanical stability without residue formation. Although excellent pollutant removal efficiency exists, the catalyst properties, such as synthesis methods, energy-bandgap structure, crystallinity, surface features, and reusability, must be investigated.

Electrochemical processes

Malpass *et al.* (2006) published a study of the electrochemical oxidation of the pesticide ATZ at a Ti/$Ru_{0.3}Ti_{0.7}O_2$ dimensionally stable anode under the effect of using different supporting electrolytes, such as NaCl, NaOH, $NaNO_3$, $NaClO_4$, H_2SO_4, and Na_2SO_4. It was observed that the removal of ATZ was achieved only at considerable rates when NaCl was used as the supporting electrolyte because of the oxidizing species formed in this electrolyte. Therefore, the efficiency of these electrochemical processes would be directly related to the performance of fluid dynamics in the cells used. Barbosa *et al.* (2018) indicated factors that must be considered when designing an electrochemical reactor, such as size and geometry, fluid flow and electrode reaction kinetics, current intensity, potential difference and concentration distribution, heat transfer, costs, and operational simplicity.

Studies have supported the improvement of Electro-Fenton for the degradation of ATZ. Cao *et al.* (2020) improved the electro-Fenton process by designing a bifunctional catalyst with FeOx nanoparticles embedded into nitrogen-doped hierarchically porous carbon. The activity and selectivity for the catalytic production of H_2O_2 were improved. The catalyst exhibited excellent electro-Fenton performance for the degradation of sulfamethoxazole, atrazine, rhodamine B, and 2,4-dichlorophenol (all at 50 ppm) in a neutral reaction solution with 95%, 96%, 99%, and 99% in 90 min, respectively. In Electro-Fenton processes, there can be in situ generation of Fenton's reagents, and the formation of sludge is too low. But the problem is that energy is required for their installation and operational costs. However, the advantage of this process is that it can degrade pesticides such as atrazine in higher concentrations more successfully than by some conventional processes.

Emerging hybrid systems

Due to the poor performance of conventional treatment processes for the removal of several emerging contaminants, a variety of hybrid treatment systems have been reported in the literature, and significant improvements have been achieved in their application in wastewater treatment in the last few years (Dhangar; Kumar, 2020; Ahmed *et al*., 2017). The different aspects of AOPs have been used to improve the effectiveness of various physical and biological treatment systems through the exploration of hybrid systems. Hybrid systems using AOPs are summarized in Table 3.

Li *et al.* (2013), Sacco *et al.* (2015), Yola, Eren, and Atar (2014), Atarodi and Faghihian (2019), and Santacruz-Chávez *et al.* (2015) evaluated the heterogeneous photocatalysis process with modified TiO_2 and ZnO catalysts through the deposition–precipitation procedure on sol-gel, agitation, ultrasonic bath, and calcination, providing high efficiency of ATZ removal (80%–94%) under UV-Solar and UV-Vis exposure. The use of these catalysts is conventionally carried out under UV irradiation, considering the energy required for the activation of the photocatalysts. However, after co-doping by graphene-SiO_2, nitrogen and phosphorous (N/P), chitosan, and boron (B), respectively, showed high efficiency in the visible spectrum. Komtchou *et al.* (2020), in addition to modifying the catalyst by introducing N_2 reactive gas into TiO_2 and WO_3 targets and through the magnetron sputtering process on photoanodes for higher photoelectrocatalytic efficiency, performed a coagulation/flocculation process followed by nanofiltration combined with heterogeneous photocatalysis, resulting in a 99% removal of ATZ.

It should be noted that the simultaneous application of microwave (MW) and UV irradiation leads to better results in photochemical processes because of its potential to accelerate the chemical reaction, therefore providing higher yields and selectivity in photochemical processes (Církva; Hájek, 1999). In this perspective, the study conducted by Chen *et al.* (2011) focused on combined AOP photoperoxidation with MW, resulting in the complete mineralization of ATZ. Zhanqi *et al.* (2007) proposed the addition of MW-assisted photocatalysis on conventional TiO_2 with 98.5% removal of ATZ. Ramasundaram *et al.* (2017), Rozas *et al.* (2017), Restivo *et al.* (2013), and Ahmed *et al.* (2017) reported that the ozonation process combined with activated carbon (AC) and biological activated carbon (BAC) promoted an efficiency of 70%–90% in ATZ degradation. When combined, ozonation promotes the generation of hydroxyl radicals that oxidize the contaminant adsorbed by activated carbon (Reungoat *et al*., 2012).

Table 3 – Removal efficiencies of ATZ by hybrid systems.

Treatment technology	Source	Catalyst/Adsorbent	Influent (μg L^{-1})	Removal (%)	References
UV-Solar+Cl	Simulated drinking W	-	0.5	75	Cheng *et al.* (2019)
UV-Solar+HomoCat	River W modified	Fe^{3+}-Montmorillonite	1077.5	100	Hong *et al.* (2019)
UV-Solar+HeteCat	Synthetic WW	TiO_2/SiO_2-Graphene	10,000	93.3	Li *et al.* (2013)
UV-Vis+HeteCat	Synthetic WW	TiO_2/N/P	2500	94	Sacco *et al.* (2015)
UV-Vis+HeteCat	Synthetic WW	ZnO-Chitosan	2,50,000	80.29	Atarodi and Faghihian (2019)
UV-Vis+HeteCat	Synthetic WW	TiO_2/B	60,000	94	Yola, Eren, and Atar (2014)
UV+HeteCat	Synthetic WW	TiO_2/Au	25,000	60	Santacruz-Chávez *et al.* (2015)
UV+HeteCat	Synthetic WW	TiO_2-Polyimide	2156.8	30	Ramasundaram *et al.* (2017)
CF+NF+UV+HeteCat	Synthetic/Real Agro WW	TiO_2/N/W	100	99	Komtchou *et al.* (2020)
MW+UV+HeteCat	Synthetic WW	TiO_2	20,000	98.5	Zhanqi *et al.* (2007)
MW+UV+H_2O_2	Synthetic WW	-	20,800	100	Chen *et al.* (2011)
O_3+HeteCat	Synthetic WW	Polonite®	100	99	Kolosov and Yargeau (2019)
O_3+HeteCat	Synthetic WW	Rutile TiO_2	2156.8	100	Yixin *et al.* (2014)
O_3+AD	Synthetic WW	AC	2800	90	Rozas *et al.* (2017)
O_3+AD	Ultrapure/Natural W	AC	1	75	Restivo *et al.* (2013)
UV+O_3	Synthetic WW	-	21,568	65	Bianchi *et al.* (2006)
NF+O_3+AD	Real WW	BAC	0.001	70	Ahmed *et al.* (2017)
NF+O_3+H_2O_2	River W	-	720	98	Saquib, Vinckier, and van der Bruggen (2010)

AC: activated carbon; AD: adsorption; AgroWW: Agriculture Wastewater; BAC: biological activated carbon; CF: coagulation/flocculation; Cl: chlorination; H_2O_2: peroxidation; HeteCat: heterogeneous catalysis; HomoCat: homogeneous catalysis; NF: nanofiltration; MW: microwave; O_3: ozonation; UV: photolysis by ultraviolet radiation; UV-Solar: photolysis by solar radiation; UV-Vis: photolysis by visible light radiation; W: water; WW: wastewater.
Source: author (2023).

Furthermore, there is the development of technologies to remove organic pollutants using the physicochemical properties of AOPs and the mineralization promoted by microorganisms (He *et al.*, 2019). Mahlalela *et al.* (2021) proposed a biological treatment based on an activated sludge system coupled with a photoreactor using a heterojunction of $BiVO_4$–Bi_2O_3 as the photocatalyst to remove ATZ at 0.0185 mM. The biological process was ineffective in the degradation of ATZ due to the possible toxicity of the compounds to the microorganisms present in the activated sludge, as the acclimatization time suggested by the authors (27–40 days) was not sufficient to increase the resistance of microorganisms to the pesticide, which generated a reduction in the efficiency of the treatment. However, with the application of the photocatalytic treatment, 70% degradation of ATZ was obtained. Hu *et al.* (2021) evaluated the removal and bioaccumulation capacity of ATZ by *Chlorella* sp. microalgae under UV-A irradiation (365 nm), and the photocatalytic degradation results showed that 31.4% of atrazine degradation occurred after 60 min, with detection of three degradation products, DIA, DEA, and desethyl-desisopropyl-atrazine (DEIA), and 83.0% removal of ATZ at 40 μg L^{-1} after 8 days. Similarly, Shin, Kim, and Park (2019) used microorganisms immobilized on catalytic materials to form a biomaterial with physicochemical and biological efficiency simultaneously. Yu *et al.* (2018) synthesized a magnetic bionanomaterial of *Penicillium* sp. yz11-22N2 doped with nano Fe_3O_4 entrapped in gel beads polyvinyl alcohol-sodium alginate (PVA-SA) (PFEPS) and promoted the degradation of 91.2% ATZ at 8 mg L^{-1}. According to the authors, nano Fe_3O_4 may promote atrazine degradation by reductive dichlorination. Moreover, nano Fe_3O_4 provided nutrients for the growth of the microorganism, which enabled atrazine degradation through its metabolism because it was the sole source of either carbon or nitrogen.

TOXICITY ASSESSMENT

Several studies have focused on both identifying such substances and assessing their toxicity in comparison to ATZ, as the main goal in ATZ removal and degradation processes is to obtain by-products (described in detail in Supplementary Information) that are less toxic than the former pollutant to justify the application of the chosen treatment. In this regard, toxicity analysis has been implemented to enable a better evaluation of the effluent. Some of the main toxicity tests conducted in studies about the treatment of recalcitrant pollutants are bioassays with *Vibrio fischeri* luminescent bacteria, *Daphnia magna* crustacean, *Hyalella azteca* and *Diporeia* spp. amphipods, *Pseudokirchneriella subcapitata* unicellular algae, *Drosophila melanogaster* fly, and *Lactuca sativa* lettuce seeds (Choi; Kim; Lee, 2013; Gonçalves *et al.*, 2020; Tavares *et al.*, 2020).

ATZ is potentially toxic at different trophic levels, causing photosystem inhibition, changes in growth and enzymatic processes in microalgae, mutagenicity, genotoxicity, and endocrine disruption in aquatic organisms, oxidative stress and DNA damage in worms, enzyme inhibition, and changes in hepatic metabolism in fish, as well as affecting soil microbiota. ATZ contamination in humans can also cause carcinogenic effects as well as negative effects on the endocrine system, such as reduced testosterone production and sperm abnormalities (Rostami *et al.*, 2021). Regarding the ATZ toxicity by-products, Xu *et al.* (2019) stated that the dealkylation by-products (DEA and DIA) are less toxic than themselves, and the hydroxylated products do not show toxicity in aquatic organisms. According to the study conducted by Choi, Kim, and Lee (2013) about the by-products of the UV and UV/H_2O_2 processes in the degradation of ATZ, the toxicity analysis of the intermediates follows the order: HA>ATZ>DEA~DIA>DEIA. The hydroxydeethylatrazine and ammeline (DEHA and DEIHA) are two compounds that show no toxicity through the Daphnia magna toxicological method.

Table 4 – Advantages and challenges of different technologies in the removal of ATZ.

Treatment technology	Advantages	Challenges	References
Photolysis	High efficiency with UV-C light	Alone is frequently not adequate to degrade pollutants, so it is used in combination with other technologies	Jatoi *et al.* (2021)
Ozonation	High efficiency in the presence of H_2O_2; Disinfection and sterilization	High energy consumption; Formation of toxic by-products; Interference of radical scavengers	Ahmed *et al.* (2017); Jatoi *et al.* (2021)
Photoperoxidation	High efficiency with UV-C light	Excess peroxide causes a decrease in the rate of degradation; Formation of toxic by-products	Ferreira, Maniero, and Guimarães (2015)
Fenton and photo-Fenton	Degradation and mineralization; Degrades within short duration; Sunlight can be used by avoiding UV light	Decrease of $OH^{\bullet}$ forming chloro and sulfato-Fe(III) complexes or due to scavenge of $OH^{\bullet}$ forming $Cl_2^{\bullet}$ and $SO_4^{\bullet-}$ in the presence of chloride and sulfate ions; pH values are low in application; Massive sludge is generated during the reaction	Ahmed *et al.* (2017); Jatoi *et al.* (2021); Dhangar and Kumar (2020); He *et al.* (2019)
Heterogeneous photocatalysis	Sunlight can be used by avoiding UV light; High reaction rates upon using catalysts with low price, chemical stability, and easier recovery	Difficult to treat large volume of wastewater; Cost associated with artificial UV lamps and electricity; Separation and reuse of photocatalytic particles from slurry suspension; Some materials only possess photocatalytic properties under specific illumination	Ahmed *et al.* (2017); Ahmed *et al.* (2021); He *et al.* (2019)
Eletro-Fenton	Situ generation of the Fenton's reagents; Degradation and mineralization; Degrades within short duration	High energy consumption	Cao *et al.* (2020)

Source: author (2024).

FUTURE PERSPECTIVES

Despite the high efficiency in the degradation of ATZ and its intermediates, the AOPs hardly reach the total mineralization of this compound. One of the main challenges in the field of treatment of emerging pollutants is to promote a treatment in which these analytics are decomposed into inorganic compounds characteristic of the end of their degradation pathways. Accordingly, there is an incentive for research involving the study of hybrid treatments, whether between AOPs, with modification of heterogeneous catalysts, or even the addition of physical processes (see Table 3).

Whereas, when compared with other treatment technologies, the operating cost of AOP methods can be relatively high, the preparation of photocatalysts with excellent solar properties, reusability capacity, and low environmental impact is also the focus of future research. However, the view of the unique physicochemical properties of atrazine, it is difficult to achieve ideal removal efficiency without the generation of hazardous intermediates during the treatment in real matrices. In this sense, there is a lack of studies that emphasize the design of water treatment plants with economical approaches to remove several types of pesticides simultaneously. Furthermore, the identification of toxicity should be part of journal publishing requirements, as should a cost evaluation of the process with the finality of providing additional information to improve communication and transparency for the application of these AOPs.

CONCLUSIONS

A variety of different physical, chemical, and biological technologies have been used to remove or degrade atrazine from water in recent decades. Difficulty in biodegradation using physical and biological treatments has been reported. Chemical oxidation treatment technologies such as photolysis, ozonation, photoperoxidation, Fenton and photo-Fenton, photocatalysis, and electrochemical processes are considered the most efficient. The advantages and challenges of the different AOPs are summarized in Table 4. In conclusion, hybrid systems have recently been applied to improve the removal of this pollutant in real matrices.

AUTHORS' CONTRIBUTIONS

Dantas, Á.O.S.: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. Rocha, A.C.: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. Cardoso, V.: Funding acquisition, Project administration, Resources, Validation, Visualization, Writing – review & editing. Vieira, P.A.: Project administration, Resources, Supervision, Validation, Visualization, Writing – review & editing.

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