

## Environmental fate, toxicological impact, and advanced treatment approaches: Atrazine degradation and emphasises on circular economy strategy

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### ABSTRACT

The persistent nature of the herbicide atrazine in the environment and its possible negative impacts on ecosystems and human health has sparked growing concerns. The diverse effects of atrazine are explored in this review, which also discusses its environmental fate, toxicity mechanisms, and toxicological effects on aquatic ecosystems, soil, plants, humans, and animals. This review also discusses several physicochemical techniques for atrazine treatment, including adsorption on activated carbon and biochar, as well as chemical techniques like Fenton, ozone oxidation, sulfate radical oxidation, and photocatalytic methods. Biodegradation of atrazine is then analyzed, together with atrazine-degrading microorganisms, such as bacteria, fungi, other microbes, and genetically modified microorganisms. Knowledge of the molecular biology and microbial metabolic pathways of atrazine-degrading bacteria provides a deeper comprehension of the degradation process. The paper also looks at key factors that affect atrazine degradation and emphasises the circular economy strategy and life cycle analysis as effective options for long-term atrazine management. The paper concludes by outlining future prospects in the field, aiming to address the challenges posed by atrazine and pave the way for environmentally friendly alternatives and effective remediation techniques.

### 1. Introduction

The utilisation of atrazine herbicide in the agricultural sector is essential for the management of annual grasses, pests, and broad-leaved weeds, thereby ensuring the cultivation of crops of superior quality [1,2]. Since its invention in 1958, atrazine has been extensively utilised as a pre and post-emergent herbicide. This is attributed to its stable structure, efficacy against a wider spectrum of weeds, ability to mitigate soil erosion, and cost-effectiveness [3]. Despite the potential economic advantages associated with its utilisation, this chemical is widely regarded as posing a significant hazard due to its adverse effects on non-

target organisms [1]. The half-life of atrazine in soil ranges from approximately 4 to 57 weeks. Atrazine, a chemical compound, was subject to restriction in multiple nations during the year 1991, including Italy [4], Denmark [5], Finland, and Germany [6]. Additionally, the European Union implemented a ban on atrazine in 1992 [7]. This attributed to the capacity of metabolites and residues of atrazine to endure in agricultural fields and surface water for long time [8,9], yielding in the contamination of surface and water bodies. Several studies have reported the presence of atrazine levels exceeding 3 and 0.1  $\mu\text{g L}^{-1}$  in the drinking water of both the United States and Europe [10]. The maximum acceptable concentration (MAC) of atrazine in drinking water is

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$5 \mu\text{g L}^{-1}$ . The determination of the acceptable daily intake (ADI) is established by dividing a no-observed-adverse-effect level (NOAEL) by suitable uncertainty factors [11]. The herbicide in question is extensively utilized and has been frequently documented to exceed permissible thresholds in aquatic environments [12]. It has been recognised as a persistent organic pollutant within the natural environment [13,14]. The extensive investigation of atrazine's impact on soil organisms has been driven by concerns over its potential toxicity and persistent effects [15]. Nevertheless, the extended duration of atrazine exposure has led to an amplification of both environmental and health hazards on a global scale [16]. Atrazine possesses inherent characteristics such as an extended half-life in soil, significant mobility, limited volatilisation, resistance to aerobic degradation, and resistance to abiotic hydrolysis. These attributes contribute to its heightened capacity to accumulate in soil and subsequently contaminate both ground and surface water sources [17]. Because of the way this pesticide affects cholinesterase activity as well as central nervous system disorders, it poses a serious risk to human health. Moreover, intense cramping in the abdomen, tightness in the chest, dizziness, headache, Symptoms regarding pesticide poisoning have included diarrhea, hypotension, and psychological complications, peripheral nerve damage, decreased performance in psychological tests, as well as sensitivity to specific chemicals [18]. Various methods have been employed for the removal of atrazine from wastewater, encompassing both conventional and advanced techniques, due to the evident detrimental effects it poses. Among these techniques, adsorption, treatment, chemical degradation, and others are frequently employed [19]. These techniques involve intricate procedures that come with a hefty price tag and increased chemical usage. Researchers have discovered that the adsorption process has become more effective in this respect and that utilizing a variety of synthetic and natural adsorbents may decrease the process's operating costs [20–22].

However, it is important to note that many of these methods are accompanied by certain limitations, such as incomplete degradation, generation of secondary pollutants, and high energy consumption [23]. The Environmental Protection Agency (EPA) classifies atrazine as belonging to toxicity class III on a scale ranging from I to IV, with I representing the highest level of toxicity [24]. The central insecticides board and registration committee in India have granted registration for only two crops, namely apple and sugar [25]. US Environmental Protection Agency has classified atrazine as herbicide that disrupts the endocrine system [26]. Atrazine has been classified as a carcinogenic herbicide by the International Agency for Research on Cancer (IARC) [27].

Soil microbes hold significant significance due to their pivotal contributions to maintaining soil functions, ecosystem products, and services, particularly in relation to nutrient cycling and carbon sequestration in agricultural systems [28]. Consequently, alterations in soil microorganisms can potentially impact soil fertility and crop yield. A considerable body of research has been dedicated to examining the impact of atrazine, a persistent herbicide, on soil microorganisms [29]. Nevertheless, the findings from these studies have exhibited a lack of consistency. Multiple studies have documented both favourable and unfavourable impacts of atrazine on various aspects of soil microbial ecology, including soil microbial biomass (SMB), soil microbial respiration (SMR), soil microbial diversity (SMD), and soil enzyme activity (SEA). It is plausible that certain variables were not standardised across prior experiments, leading to variations in outcomes among distinct studies [29–31]. These variables may have encompassed dissimilarities in the dosage of herbicide application, characteristics of the soil employed in the experiment, and prevailing environmental conditions [32,33].

Based on the aforementioned information, our attention was drawn to two focal points: firstly, the potential impacts of atrazine on soil microorganisms, and secondly, the determinants that influence the effects of atrazine on soil microorganisms. This review provides an

examination of the toxicity of atrazine on humans, plants and other microbial organisms. Additionally, it explores the various aspects of atrazine degradation, including chemical, photochemical, and microbial processes.

## 2. Toxicity mechanisms of atrazine and its toxicological impact

The common pathway of herbicide appearance into the ecosystem is through agronomic applications. Inadequate production system management or uncontrolled releases of pesticides can also cause them to reach the ecosystem [34]. Herbicide production in factories and their use at large scales in metropolitan cities is another major source of pollution. Atrazine's effects on aquatic ecosystems, soil and plants, animals and humans are discussed as follows.

### 2.1. In aquatic ecosystems

Living organisms such as freshwater algae are badly impacted by the existence of atrazine in aqueous ecosystems [35–39]. The content of chlorophyll a and carbohydrates was improved by Atrazine, while the *Chlamydomonas mexicana* development was suppressed [40,41]. Ralston et al. [42] reported that atrazine caused major toxicity to *Hyaella azteca* and *Diporeia* sp., and *Pseudokirchneriella subcapitata*, algae as compared to its metabolites desethylatrazine and deisopropylatrazine. Atrazine exposure to a green alga, *Raphidocelis subcapitata* resulted in alterations at a cellular level and influenced metabolism and photosynthetic activity [43,44]. Photo-phosphorylation was found to be inhibited by atrazine, but there was no lethality or permanent cell disruption [45]. Atrazine caused raised lesion index levels in *Rhamdia quelen* by causing histopathological changes in the liver and altering osmoregulation activity [46]. Disruptions in DNA were noticed in *Corbicula fluminea* when herbicides atrazine and roundup were used in combination [47,48].

Exposure of atrazine to zooplankton, *Daphnia* resulted in improved content of carbonyl protein, thiols and activity of superoxide dismutase (SOD) enzyme and reduced lipid peroxidation and protein thiol levels [49]. Growth rate, other morphological attributes and reproduction activity were found to be reduced in zooplankton, *Daphnia pulex* [50]. It was shown that augmentation in the functioning of superoxide dismutase, glutathione peroxidase and reductase enzymes and diminished the glutathione S-transferase (GST) and catalase enzyme activities in atrazine-exposed zebra fish [51]. Transcriptional changes were noticed in the functioning of GST isoenzymes within and among organs of common carp by the use of atrazine [52,53]. Atrazine-exposed *Plotosus lineatus* showed genotoxic damage to various cell types, disrupted liver metabolism and suppressed the activities of different enzymes [54]. Atrazine treatment showed a decrease in the protein levels in *Ctenopharyngodon idella* [55]. Damaging effects were found in the *Crassostrea gigas* under atrazine exposure via causing alterations in the major molecular and biochemical aspects [56]. Atrazine induced a decline in the contents of glycogen and vitellogenin proteins in the ovary of estuarine crab, *Neohelice granulata* [57]. Fingerlings, *Caspian kutum* exhibited disruption of its tissue, pavement cells and ionocytes as the atrazine exposure negatively affected its gill respiration and ion regulation [58]. Atrazine treatment to *Phaeodactylum tricornutum* alga caused a decline in the photosynthetic pigments, suppression of photosynthesis efficiency, and disturbed the electron transport chain [38] (Fig. 1).

### 2.2. In soil and plants

Plants are known to be adversely affected by atrazine toxicity as it inhibits their growth and influences the translocation process, morphological alterations, physiological processes and overall development of plants [60–64]. There are different parameters affect the atrazine uptake in target and non-target plants, and the outcomes of its uptake may be related to the usage of various genotypes. Xylem is the major

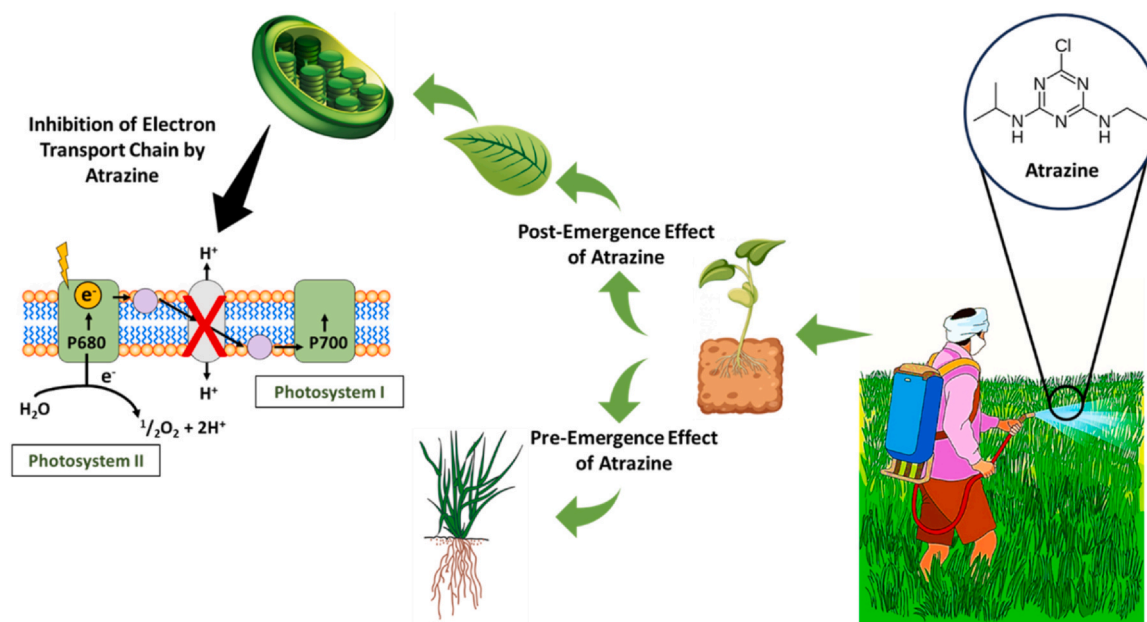


Fig. 1. Impacts of atrazine on plants by effecting transport chain and photosystems.

pathway through which atrazine is transported after its uptake by roots and shoots [65,66]. Atrazine exhibits both pre- and post-emergence activity in weeds as it is absorbed by leaves and roots [67–69]. Activities of peroxidase and ascorbate peroxidase enzymes were altered by atrazine treatment, which further resulted in lipid peroxidation [70]. Atrazine causes oxidative stress in plants by causing over accumulation of reactive oxygen species (ROS) such as  $O_2^-$ , OH, and  $^1O_2$  radicals and  $H_2O_2$ . However, plants possess ROS scavenging mechanisms to cope with atrazine-induced oxidative stress [71,61,60,65]. The antioxidant mechanism of *Zea mays* was altered by atrazine and resulted in oxidative stress [72]. A decrease in fresh weight and total chlorophyll content was noticed in *Zostera marina* seedlings under atrazine toxicity [73,74].

Atrazine treatment to three genotypes, i.e., *Lythrum salicaria*, *Acorus calamus*, and *Scirpus tabernaemontani* caused significant changes in their growth and physiological attributes [75]. Photosynthetic efficiency was inhibited, and alterations in the C:N ratio, nitrogen and sugars content were noticed in atrazine-exposed *Zostera marina* [76]. Maize plants exposed to atrazine exhibited a decrease in net  $CO_2$  assimilation rate and PS II efficiency [77]. Atrazine exposure resulted in augmented functioning of SOD, POD, CAT and APX enzymes in *Pennisetum americanum* seedlings to protect them from oxidative damage [78]. *Chrysopogon zizanioides* L. (vetiver) growth was inhibited in atrazine contaminated soil [79].

### 2.2.1. In humans and animals

The neuro-endocrine system is affected by atrazine toxicity by changing levels of pituitary hormones, as shown by several previous studies [80]. The abortion rate was increased in male Wistar rats due to the suppression of gonadotropin-releasing hormone (GnRH) release, which consequently declined the discharge of luteinizing hormone [81]. Male rats exposed to atrazine showed decreased sperm counts and the motility of their germ cells [82,83]. Levels of serum lipids and liver enzymes were diminished by atrazine treatment in adult mammals [53,84]. Aromatization of testosterone to estrogen was accelerated as the activity of phosphodiesterase was suppressed by atrazine treatment to augment the functioning of the aromatase enzyme [85,86,87]. Reproduction and development were affected, and cancers and the development of pre-existing estrogen-dependent cancers were also induced by the elevation in the estrogen level, which further resulted in alterations in the amount of relative sex hormone [88]. Cardiovascular function, the central nervous system and the immune system were also

affected by the metabolites of atrazine [89,90]. Atrazine exposure to adult humans causes non-Hodgkin's lymphoma [91,92].

In male mice, the production of spermatozoa was affected by atrazine via interfering with their meiotic process [93]. Immunotoxicity was induced in mice by atrazine, causing oxidative stress and alterations in  $Ca^{2+}$  homeostasis [94]. Moreover, it adversely affects the immune functioning in different organisms and exerts immune-toxic effects on murine lymphocytes [95]. *Drosophila melanogaster* showed altered gene expression, protein production, decline in mating rate, competitive fertilization ability and progeny number by their exposure to atrazine [96]. *D. melanogaster*, when confronted with atrazine, showed a decrease in emergence rate, pupation rate and longevity [97]. Alterations in bioreactivity, induction of necrosis and inflammation in human lungs were caused by atrazine [98]. The harmful effects of atrazine of different human body organs are presented in Fig. 2.

### 2.3. Toxicity of the degradation products of Atrazine

It is important to assess the toxicity of the degradation products of atrazine where these intermediate products may be more toxic than atrazine itself. Several studies have been highlighted the development of metabolic products resulted from the metabolism of atrazine including three common products such as desethylatrazine (DEA), desisopropylatrazine (DIA), [99] and deethyldeisopropylatrazine (DEDIA) [100,101], in addition to other products such as acetamidoatrazine, [102] 4'-demethyl HT, cephalotaxine, and dehydrated HT, [103] atrazine amide [104] and hydroxyatrazine [105]. However, the upcoming studies on atrazine degradation should focus in the degradation of these metabolic products owing to their persistence and toxicity. For example, DIA and DEA have been detected in surface waters, at up to 7.4 and 7.5  $mg L^{-1}$ , respectively and up to 1.1 and 0.6  $mg L^{-1}$  in ground waters, respectively owing to their stability [106]. Additionally, DEA and DIA show an equal toxicity to atrazine. For example, the United States Environmental Protection Agency (USEPA) has reported that atrazine and its three degradation products, i.e., DEDIA, DIA, and DEA, share a mechanism of toxicity concerning endocrine disruption [106]. Atrazine, a herbicide, has been found to have adverse effects on various aquatic organisms. Its presence in aquatic ecosystems can lead to cellular alterations, histopathological changes, and disruptions in metabolism and photosynthetic activity. Atrazine exposure has caused genotoxic damage and suppressed enzyme activities in different organisms.

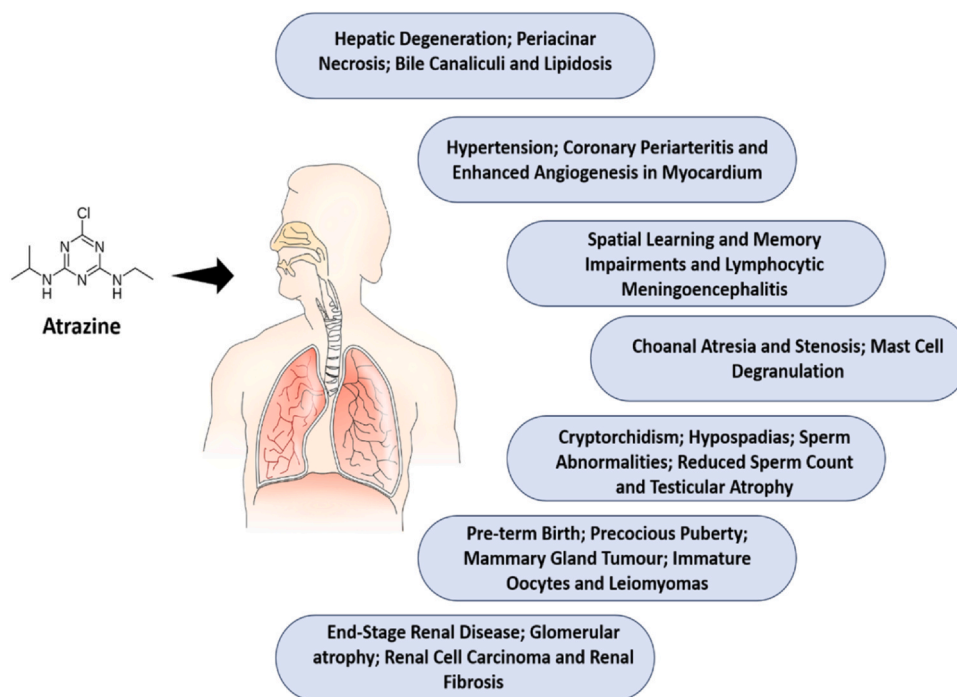


Fig. 2. Detrimental effects of atrazine on different organs of humans.

Although it has shown to improve certain protein levels, it has also resulted in a decrease in protein and pigment contents, and negatively impacted growth rate, morphological attributes, and reproduction activity. Atrazine exposure has also caused ion regulation disruption, gill respiration disruption, and altered major molecular and biochemical aspects. An unorthodox method of treating water, electrical discharge in contact with water produces extremely reactive transitory oxidative species ( $\text{OH}\cdot$ ,  $\text{H}_2\text{O}_2$ ,  $\text{O}_3$ , atomic  $\text{O}$ , singlet  $\text{O}_2$ , and UV/VUV photons) close to the water-gas interface [107].

### 3. Physicochemical methods for the treatment of atrazine

Atrazine is utilized for weed control because of its effective weed treatment and affordable price [108]. Because of its stable structure, difficult degradation, prolonged environmental residence duration, and toxicity on both humans and other living things and other factors, the research on atrazine elimination from the environment is extremely important. As a result, many processing technologies, including adsorption, photochemical catalysis, biodegradation, and others, have been developed and are frequently used for atrazine degradation. The four key features of adsorption and chemical methods (like Fenton/Fenton-like method, ozone oxidation, sulfate radical ( $\text{SO}_4^{\cdot-}$ ) oxidation and photocatalysis), as well as various parameters regulating atrazine degradation aspects of atrazine treatment technology are thoroughly examined. The benefits and drawbacks of these various approaches and the mechanisms of deterioration will be discussed in detail in the following sections. However, future studies must also concentrate on functional materials' great atrazine removal efficacy and minimal environmental impact, as well as on coordinating the removal of atrazine using two or more different technologies.

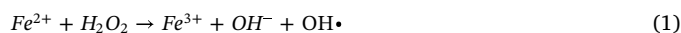
#### 3.1. Chemical methods for atrazine treatment

When it comes to chemical treatments, the oxidation-reduction reaction is primarily used to remove organic contaminants. The best options for treating hazardous wastes come from the oxidative breakdown of substances. Organic contaminants can be oxidatively destroyed in a variety of ways, including biological, chemical, and physical

(thermal). The fundamental mechanism of energy dissipation, which is inherent to the oxidative degradation of biological materials, is substantially the same in all of these systems, despite variances in their modes of functioning. This reaction generally produces a number of chemical compounds with potent oxidizing characteristics, such as hydroxyl and sulfate radicals which can breakdown as well as mineralize atrazine throughout the environment [109].

##### 3.1.1. Fenton/Fenton-like reactions

A tested and practical method for getting rid of several dangerous contaminants in wastewater is oxidation via Fenton reactions. A significant number of hazardous and organic contaminants can be destroyed using the Fenton reaction, which relies on ferrous ions plus hydrogen peroxide. Fenton made the initial discovery of the potent oxidizing abilities of a mixture of  $\text{H}_2\text{O}_2$  and  $\text{Fe}^{2+}$  salts around the turn of the 20th century [110]. Haber and Weiss identified the oxidizing species as the  $\text{OH}\cdot$  forty years later. By using electron spin resonance spectroscopy, the  $\text{OH}\cdot$  has recently been seen directly. According to literature, this Fenton reagent might oxidize organic molecules of almost any kind [111]. It has been proven that a variety of refractory organic compounds can be effectively decomposed through the Fenton reaction without releasing any harmful chemicals in the aquatic environment. The following reactions make up the majority of the mechanism for the Fenton reaction [112].



The ferric ion, the hydroxyl radical ( $\text{OH}\cdot$ ), plus the hydroxide ion are the reaction's byproducts. Compared to several sophisticated oxidation techniques, the Fenton approach had the best chances of success [113].  $\text{Ta}(\text{O})\text{N}$ , a novel type of catalyst, was combined with  $\text{H}_2\text{O}_2$  and  $\text{Fe}^{3+}$  to create a Fenton-like system.  $\text{Ta}(\text{O})\text{N}$  specimens remained made via nitriding  $\text{Ta}_2\text{O}_5$  nanomaterial that exhibited high absorption in the range 300–600 nm wavelength region. In addition, the detectable  $\text{Ta}(\text{O})\text{N}$ -Fenton-like system's intermediate generation,  $\text{H}_2\text{O}_2$  degradation, and Atrazine degradation were compared in both presence or without DO. Purifying using  $\text{N}_2$  and air, led to dissolved oxygen (DO) amounts of 0.4 and 8.4  $\text{mg L}^{-1}$  throughout the solution. It was discovered that DO has

a detrimental impact on atrazine degradation [114]. At 10 min, In both the absence or with DO the remaining atrazine concentrations were 60% or 74%; after 60 min, the amounts were 10% and 15%. With longer reaction times, the variation in degradation of atrazine between the existence and absence of DO diminish. Similar amounts of atrazine were still present in both systems after 120 min, and by 180 min, practically all of it had been broken down. The decomposition of  $H_2O_2$  was also prevented by the existence of DO. Afterward, 10 min and 180 min, correspondingly, 10% to 60% amount of  $H_2O_2$  decayed in the occurrence of DO. At 10 min and 180 min, individually,  $H_2O_2$  elimination ratios in the presence of  $N_2$  were 15% and 70%.

Multiwall carbon nanotubes have been functionalized and added to the Fenton system [115]. Atrazine adsorption on multi-walled carbon nanotubes (FCNT-H) and  $H_2O_2$  alone's ability to degrade it were both insignificant (1.8%). Fe(III)/ $H_2O_2$  eliminated around 15% of atrazine within 30 min. However, FCNT-H alone did not demonstrate any catalytic ability for  $H_2O_2$  to degrade atrazine (5%). Surprisingly, the elimination of atrazine by Fe(III)/ $H_2O_2$  was greatly improved by the addition of  $20\text{ mg L}^{-1}$  FCNT-H (92% within 30 min). Previous research suggested that increased  $H_2O_2$  concentrations greatly promote the oxidation-related Fenton reagent-related breakdown efficiency of organic contaminants [116]. Higher  $H_2O_2$  concentrations, however, have been shown to impede the removal of contaminants. Also, the dosage strategy affected the elimination of atrazine by the Fenton reagent [117]. They discovered that the segmented addition of  $H_2O_2$  was more effective than the conventional one-time addition, while the dosage was also reduced. As 2%  $H_2O_2$  was added, the atrazine elimination did not seem to improve, which is likely due to the organic matter in the soil quenching most of the  $H_2O$  [117]. According to the findings, adding more  $H_2O_2$  can considerably boost atrazine elimination. Atrazine concentrations decreased from  $617.5$  to  $554.5\text{ mg kg}^{-1}$  (10.2% atrazine elimination) following a 20 h reaction in the experiment with 5%  $H_2O_2$  concentrations. Additionally, atrazine was eliminated in trials with  $H_2O_2$  concentrations of 10% and 15%, respectively, by about 25.6% and 36.0%.

This implies that atrazine oxidation can be accelerated in the presence of greater  $H_2O_2$  concentrations. However, when  $H_2O_2$  concentration increased from 15% to 30%, the increase in atrazine elimination was not immediately apparent. Similar patterns have been seen in a number of other Fenton- and Fenton-like systems. It has been reported that the oxidation efficiency rises significantly with an  $H_2O_2$  dose up to a certain concentration but does not rise significantly after that [117]. The extremely high  $H_2O_2$  concentrations may even lower the oxidation efficiency [118,119]. As shown by the following reactions (Eqs. 3 and 4), this is caused by the scavenging effects of excess  $H_2O_2$  on OH and the subsequent creation of the less reactive  $HO_2$ .



Reaction (3) will be more prominent when  $H_2O_2$  is present in high amounts [117]. Fenton/Fenton-like procedures have the benefits of straightforward operation, quick reaction, wide application, and high removal efficiency. However, the range of pH values in the application is narrow, and huge sludge is produced during the reaction; hence, it is limited in practical applications.

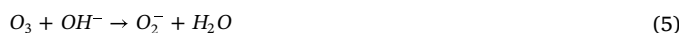
### 3.1.2. Electrochemical degradation

The oxidation of an organic substance is hard to manage, and ferrous iron regeneration is restricted, despite the simplicity of the Fenton approach. Lately, there has been a merger of electrochemical systems that enable the electrochemical production of hydrogen peroxide and/or ferrous iron, which in turn permits the creation of OH. It was demonstrated that Fenton's reagent may be electrochemically generated in aqueous solutions, resulting in a restricted number of competitive reactions and an effective, continuous OH generation. By reducing

ferric iron and simultaneously reducing oxygen to hydrogen peroxide, this straightforward method creates ferrous iron in situ. Hydroxyl radicals are created by the subsequent reaction between these two chemicals. Atrazine has been effectively degraded by this method; compared to the traditional Fenton's reagent, a quicker and maybe more thorough oxidation has been reported with the electrochemical system. It was suggested that an electro-Fenton system, in which hydrogen peroxide and ferrous iron are both progressively produced in the solution (avoidance of reagent use by competing reactions), may be used to produce OH under control. Other potential processes, such as the compounds' adsorption onto iron hydroxide precipitate or by-product reduction onto the electrode, could also be at play [120,121]. Four electrochemical advanced oxidation routes such as electro Fenton (EF), anodic-oxidation with simultaneous  $H_2O_2$  production and photoelectron Fenton (PEF), have been compared for the degradation of atrazine and its by-products. The most efficient method for eliminating atrazine and its by-products from synthetic solution (initial concn. =  $100\text{ }\mu\text{g L}^{-1}$ ) and actual agricultural surface water enriched with atrazine (initial concn. =  $10\text{ }\mu\text{g L}^{-1}$ ) was discovered to be the PEF process. With a boron-doped diamond (BDD) anode and a graphite cathode, various operational factors, such as light wavelength, pH, current density, and the presence of naturally occurring organic matter (humic acids), were studied for the PEF process. The two most significant factors affecting the atrazine degrading efficiency were the current density and the light's wavelength. For the synthetic materials, the ideal working parameters were found to be  $18.2\text{ mA cm}^{-2}$ , pH 3.0, and 45 min of treatment. After 15 min of treatment, more than 99% of atrazine oxidation was observed, and after 45 min of treatment, all significant by-product concentrations were below the limit of detection. Additionally, the PEF procedure was evaluated on actual atrazine-contaminated surface water. Even with the presence of iron complexation and radical scavenger, the PEF method removed atrazine from actual surface water more successfully when the pH was adjusted to be close to 3.0. With 0.01 mM of iron ( $k_a = 0.13\text{ min}^{-1}$ ) and 0.1 mM of iron ( $k_a = 0.17\text{ min}^{-1}$ ), the atrazine removal was 96.0% and 100%, respectively [122]. Moreover, a current-controlled electrolysis at a platinum electrode was studied to analyse the breakdown of atrazine in aqueous solution. The impacts of pH, the direction and strength of the current and temperature were discussed. It was shown that atrazine partly degrades to cyanuric acid in acidic environments at  $25\text{ }^\circ\text{C}$ , forming persistent intermediate molecules; however, atrazine totally degrades to cyanuric acid at  $60\text{ }^\circ\text{C}$ . According to the TOC data, no mineralization or electrochemical combustion was detected. [122].

### 3.1.3. Ozone oxidation

To destroy different organic compounds, ozone is considered one of the powerful oxidants ( $E_0$  14 2.07V) and has been used extensively [123]. Ozone oxidation, an efficient advanced sewage treatment method, has recently become a focus of research in the field of treatment of wastewater and is frequently utilized in the degradation of organic wastewater. During the reaction process, the technology may produce significant quantities of OH, and it oxidizes the majority of refractory organic compounds into tiny molecules. This approach's guiding premise is as follows [124]:



Since only ozone is present, the oxidation activity usually is not particularly high. Since this is the case, researchers' top priority is to improve the oxidation efficiency of ozone, and one key approach to do this is by introducing a catalyst [115]. An arranged mesoporous  $Fe_3O_4$  catalyst was developed and utilized to ozonize atrazine in their study [125]. Fig. 3 depicts the atrazine removal during the process. After 10 min, the atrazine elimination with the ordered mesoporous  $Fe_3O_4$  (om- $Fe_3O_4$ ) catalyst reached 82.0%, compared to 25.0% with nano-

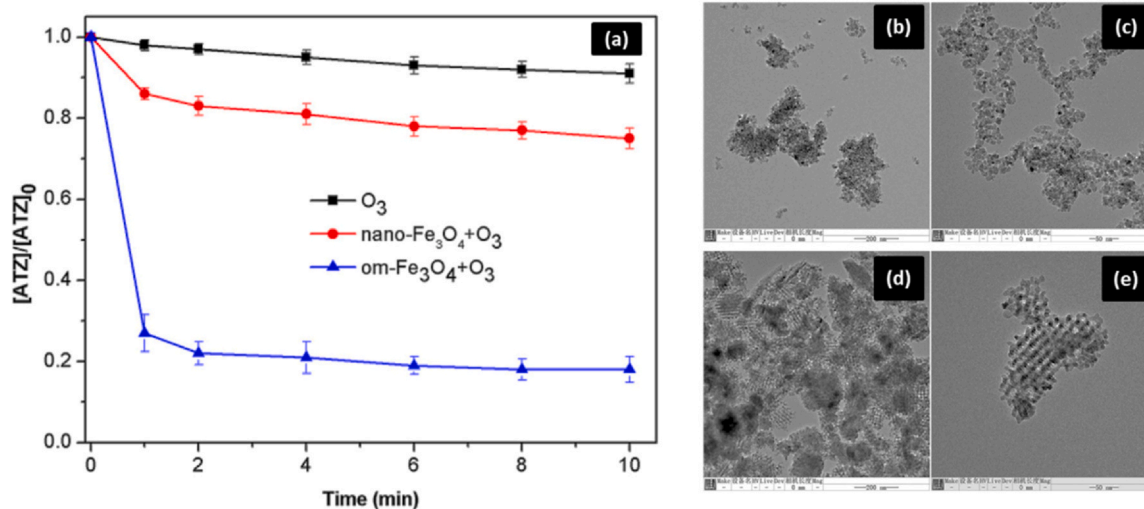


Fig. 3. The removal efficiency of atrazine with different catalysts (a), and TEM images of nano-Fe<sub>3</sub>O<sub>4</sub> (b& c) and om-Fe<sub>3</sub>O<sub>4</sub> (d& e) [130].

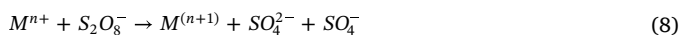
Fe<sub>3</sub>O<sub>4</sub> and just 9.1% with ozone alone. According to the findings, ordered mesoporous catalysts have better catalytic activity than non-mesoporous catalysts when it comes to catalytic ozonation of atrazine [126]. The huge surface area, as well as the pore size of om-Fe<sub>3</sub>O<sub>4</sub>, may be the cause of its exceptional catalytic activity [127]. The former can help increase the number of active sites on the ozone catalyst, and the latter is useful for speeding up intraparticle mass transfer and promoting heterogeneous catalytic reactions [128]. Using graphitic carbon and nitride, a superior catalytic composite was developed [128]. Broad leaf as well as grassy weeds are effectively controlled both pre- and post-emergence with the commonly utilized s-triazine herbicide atrazine (ATZ) in a variety of crops [129].

The findings of the evaluation and comparison of atrazine deterioration in various procedures is shown in Fig. (3b). After 5 min of reaction time at pH 6.0, the removal efficiencies of atrazine with the O<sub>3</sub> alone, O<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> and O<sub>3</sub>/O@g-C<sub>3</sub>N<sub>4</sub> procedures were 63.15%, 56.83%, as well as 92.91%, respectively. The findings unmistakably showed that O@g-C<sub>3</sub>N<sub>4</sub> greatly accelerated atrazine degradation and deterioration.

The combined process had a removal efficiency that was 4.78 times greater than the total of the two individual procedures. The effects of ozone oxidation, electrolysis, as well as the combination of the two technologies on the removal of atrazine were studied. The refractory organics in the environment can be effectively oxidized and mineralized using the ozone oxidation method [131,132], which does not result in secondary pollution. In addition, it does deodorization, disinfecting, and decolorization [133]. The poor actual ozone consumption and high expense of ozone generation limit its practical implementation, nevertheless. The creation of efficient and affordable catalysts should be the main emphasis of current research to improve ozone's oxidation performance [134].

### 3.1.4. Sulfate radical (SO<sub>4</sub><sup>•-</sup>) oxidation

The sulfate radical oxidation method is a recently developed advanced oxidation technology. The oxidation mechanism takes place when persulfate (PS) or peroxymonosulfate (PMS) gets activated by heat light or metal ions (M<sup>n+</sup>) to produce super oxidative sulfate radicals (SO<sub>4</sub><sup>•-</sup>), which oxidize and decompose the refractory organic pollutants in the environment [135]. Here is how the activated principle works [136].



It was looked into whether it would be possible to use soil-based s-triazine herbicides like terbuthylazine (TEA), atrazine, coupled with ametryn (AMT) to successfully breakdown persulfate (PS) catalyzed by Fe<sup>2+</sup> [137] to determine how PS and Fe<sup>2+</sup> dosages (3.3, 8.3, et 16.6 mM) affected the decomposition to atrazine via the PS/Fe<sup>2+</sup> mechanism. After 10 h, atrazine was slightly degraded (12%) with the addition of 16.6 mM PS, indicating that PS could have been catalyzed to some extent by organic species found in soil A1, such as Fe or Mn-containing minerals. A1 soil has 33.5 mg kg<sup>-1</sup> of total iron, of which 14.5 mg kg<sup>-1</sup> is readily available. The limited level of atrazine breakdown in soil A1 under the influence of PS lacking Fe<sup>2+</sup> injection could be related to a quick reaction time considering iron mineral-activated PS oxidation of contaminants normally occurs over long durations intervals, including several weeks due to soils' lower concentration of metal oxides than that of separate mineral systems. Atrazine degradation became worse with higher PS-Fe<sup>2+</sup> dosages when Fe<sup>2+</sup> was present. After 10 h, the concentration of PS-Fe<sup>2+</sup> went up from 3.3 to 16.6 mM, and at the same time, the rate of elimination of atrazine rose from 39% to 80%.

The effectiveness and mechanism of atrazine degradation under various conditions have been investigated where PS was activated using a self-made composite of graphene plus nanoscale zero-valent iron (nZVI/GR). The concentrations of PS had a substantial impact on the breakdown of atrazine. The rate of atrazine breakdown rose significantly from 34.2% to 79.5% when PS concentration increased from 0.2 to 0.4 mM. Atrazine was entirely destroyed at 1.0 mM after 41 min although the degradation efficiency was lower at 1.0 mM than at 0.8 mM after 21 min. The degradation efficiency decreased to 86.8% after 61 min at a dose of 2.5 mM PS, and the degradation change was similar to that at 0.4 mM PS. The outcomes demonstrated that the mass ratio of nZVI/GR at which this oxidation system was most effective at removing atrazine was 5:1. The ferrate (Fe(VI))/PMS system was also used to treat atrazine, when it was discovered that this combination was significantly more effective at degrading atrazine than either Fe(VI) or PMS alone. Additionally, Fe(VI)/PMS system was effective in removing atrazine across a wide pH range [137,138]. Organic contaminants that are resistive to other methods of removal can be effectively removed using the sulfate radical oxidation approach [139,140]. The use of sulfate radical oxidation technique is superior and has the potential to be used for environmental treatment [141], but more research is needed to determine how to activate sulfate radicals to enhance their oxidation performance [142,143].

### 3.1.5. Photocatalysis

The photocatalytic approach involves adding catalysts to a reaction system and exposing it to UV or visible light, which creates powerful

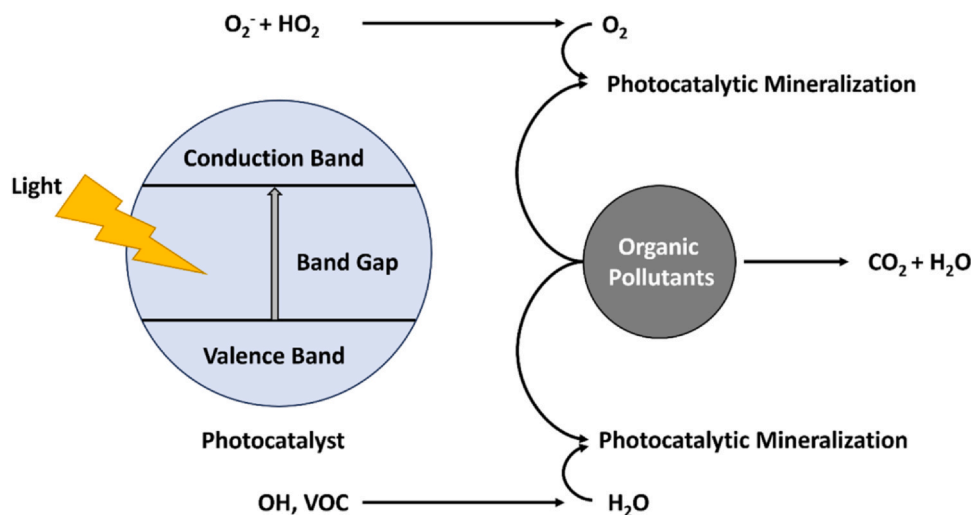


Fig. 4. A schematic diagram of the photocatalytic degradation [145].

OH $\cdot$  that oxidize and break down organic contaminants (Fig. 4). As a result, the materials as well as their oxides based on titanium, iron, and carbon are the most common catalysts [144].

One of the most widely used nanomaterials, nano-TiO $_2$ , is used to make cosmetics, catalysts, and groundwater remediation products. Due to its huge specific area of surface, nanoscale size, photocatalysis, and chemical composition, nano-TiO $_2$  poses toxicological concerns to the ecosystem and to human health when it is discharged into the environment. When lights were present, reactive oxygen species produced by nano-TiO $_2$  could cause oxidative damage to bacteria, crustaceans, and other creatures [146]. To create an innovative nano-TiO $_2$  photocatalyst, the waste-reducing agent discharged by industrial processes was used. They effectively applied to the photocatalytic elimination of atrazine. Atrazine elimination appeared to follow Pseudo-first-order reaction kinetic, according to the kinetic investigations [147]. In suspensions containing 10 mg L $^{-1}$  of nano-TiO $_2$ , 1.0 mg L $^{-1}$  of atrazine, and various doses of CaCl $_2$  at pH 7.0, the impact of Ca $^{2+}$  on the removal of atrazine by nano-TiO $_2$  was assessed. Atrazine was primarily removed during the first six hours at dark by adsorption onto nano-TiO $_2$  surfaces, with photocatalytic destruction occurring during the final four hours of UV exposure. Atrazine was substantially less likely to bind with the nano-TiO $_2$  surface during the dark phase, thanks to the addition of Ca $^{2+}$ . After six hours, atrazine elimination via nano-TiO $_2$  was 49.2% without Ca $^{2+}$ . The adsorption was reduced to 17.6% with the inclusion of 1.0 mmol L $^{-1}$  CaCl $_2$ , and removal efficiency fell to 7.6% with an increase in CaCl $_2$  concentration to 100 mmol L $^{-1}$ . With the inclusion of Ca $^{2+}$ , atrazine's photocatalytic breakdown efficiency was similarly reduced under UV exposure. The photodegradation rate was 0.46 C/C $_0$ /h (from 6.5 h to 7.5 h) without the addition of CaCl $_2$ . However, the decrease in rate from 0.35 to 0.32 C/C $_0$ /h over the same time period was caused by the increase in CaCl $_2$  from 1.0 to 100 mmol L $^{-1}$ . In the absence of CaCl $_2$ , atrazine was entirely broken down within 2 h of UV exposure. However, after 2 h of the photocatalytic process, when the concentration of CaCl $_2$  was 100 mmol L $^{-1}$ , there was 38.9% of remained atrazine in the solution, and it wasn't totally destroyed until the experiment was over [148]. A straightforward sol-gel approach was used to create an aW-TiO $_2$ /clay photocatalyst that was capable of degrading atrazine with 90% efficiency after 4 h of reaction. However, other substances having photocatalytic characteristics were developed in addition to TiO $_2$  [149]. For example, a photocatalyst (VBO $_3$ ) was investigated to assess the impact of various crucial operating factors in a slurry system. The influence of initial atrazine concentration upon degradation efficiency which was carried out at 1 h, initial concentration (C $_0$ ) 1 g L $^{-1}$ , and pH 6. The degradation efficiency diminishes as atrazine content rises. Almost all atrazine is eliminated in 1 h at

5 mg L $^{-1}$  atrazine concentration. At 25 mg L $^{-1}$  atrazine concentration, the degradation efficiency drops significantly to about 36%. At high atrazine concentrations, the degradation efficiency was much lower due to a possible lack of active sites on the photocatalyst surface. These findings suggest that photocatalytic processes are often unfavourable at large levels of refractory organic contamination. At t = 1 h, C $_0$  5 mg L $^{-1}$ , and pH 6, the impact of photocatalyst concentration is then assessed. Practically speaking, choosing the right quantity of photocatalyst is essential for limiting excess and, thus, for lowering the costs related to this procedure [150]. The degree of atrazine degradation increases with the increase in photocatalyst concentration up to 0.8 g L $^{-1}$ . A screening effect of extra photocatalyst particles may occur at concentrations of more than 0.8 g L $^{-1}$  [151]. In this case, portions of the photocatalyst surface are covered up, which prevents incoming light from penetrating the system. Another factor that can lower the efficiency is the increased light reflection off the photocatalyst surface [152]. Amorphous Cu(I) nanoclusters were successfully deposited on the gC $_3$ N $_4$  surface using ultrasonication, and the material had a noticeable improvement in atrazine degradation and mineralization. It also had a comparatively high removal capacity for atrazine under visible light [153]. It is particularly effective to use materials' photosensitive properties to breakdown organic contaminants [154]. It has incomparable advantages over earlier adsorption, Fenton/Fenton-like approach, and other technologies, including high degrading performance and material reusability [155]. However, many materials have poor quantum yields, and some only exhibit photocatalytic characteristics when exposed to a particular type of light [156]. Therefore, it is required to examine the fabrication of new photocatalysts using appropriate materials to improve the catalytic efficiency in the future studies on photocatalytic deterioration. Also, Atrazine is a weed control herbicide that is effective and affordable, but it has a stable structure, which makes it difficult to degrade and can be toxic. Several processing technologies, including adsorption, photochemical catalysis, and biodegradation, have been developed to degrade atrazine. Future studies should focus on developing materials that can remove atrazine with minimal environmental impact and explore the coordination of using two or more different technologies for its removal.

### 3.2. Adsorptive removal of atrazine

Atrazine treatment typically uses a physicochemical approach that relies on the adsorption effect. The two groups of adsorbents that are most frequently utilized are activated carbon and biochar [157]. In addition, other materials like zeolite and bentonite are also widely employed.

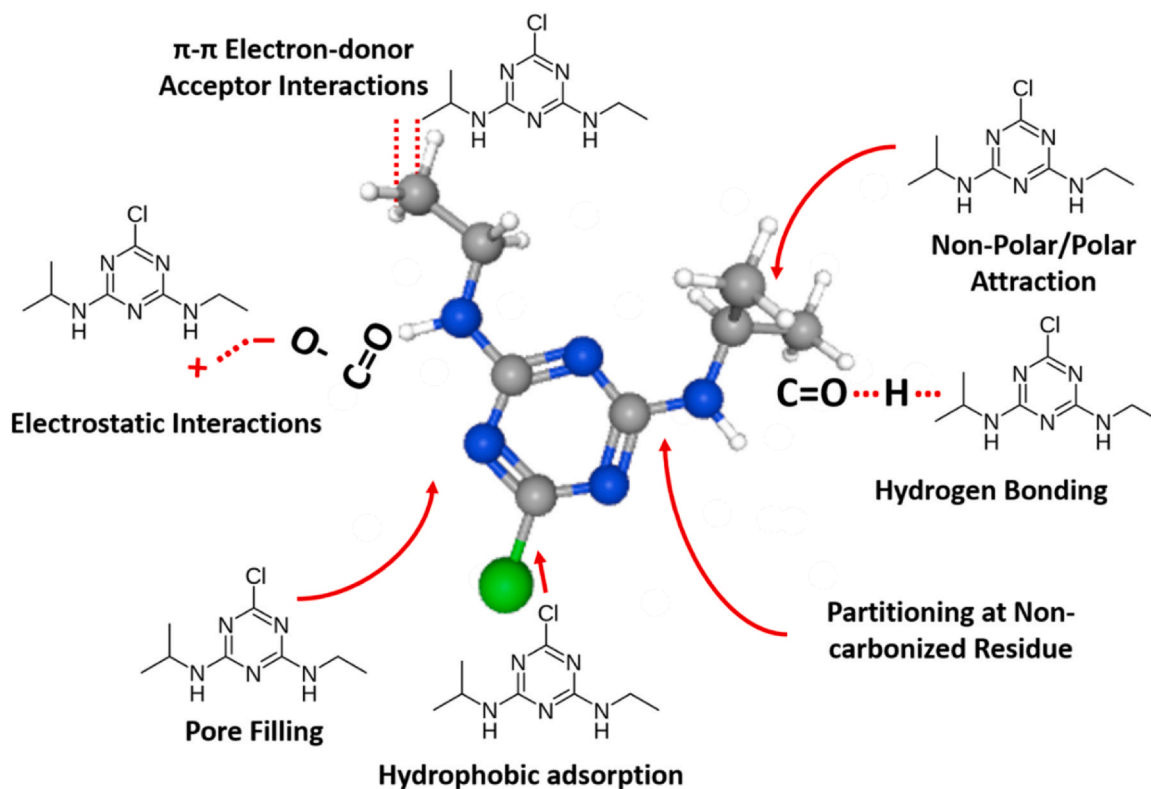


Fig. 5. Interaction mechanism for the removal of atrazine onto carbon-based materials.

### 3.2.1. Activated carbon adsorption

Activated carbon has significant adsorption characteristics for a range of pollutants because of its porous structure and large specific surface area [158,159]. To enhance the adsorption effect, researchers looked at the combined adsorption properties of different materials [160]. One of the most practical options for developing pollutants adsorption is porous carbon-based materials. These substances are produced utilizing a variety of synthesis methods having precursors containing a carbon source, and as a result, they have a large surface area, variable porosity, and a high capacity for interactions with adsorbed molecules (Fig. 5). The adsorptive removal of atrazine in aqueous solutions using porous based on carbon adsorbents has produced positive findings (> 90% in 1 h) for the development of novel environmental remediation technologies [161]. These substances include organic groups like -OH, -CH<sub>3</sub>, -CH<sub>2</sub>, as well as carbon chains, among others, that engage in physio-chemisorption processes with the adsorbate [162].

A graphitic-porous-carbon (GPC) which comprises mostly of carbon with graphite besides spheres of metal oxide such as iron which has a specified surface area of 358 m<sup>2</sup>/g, showed improvement in atrazine removal effectiveness up to 41% when the GPC dosage was increased as 200:500 mg L<sup>-1</sup>. Its increased effectiveness is the result of mechanisms controlled through physisorption interactions between GPC and atrazine, where the electrostatic interactions comprising carbon-hydrogen, the bonds of hydrogen, plus many more distinguish up [163]. Because ATZ acts as a refractory organic compound that has shown resistance to chemical reaction as well as microbial degradation, conventional methods such as coagulation, biodegradation, ultrasonic degradation, and chemical precipitation are ineffective. Fortunately, one of the most successful advanced oxidation process (AOP) techniques for the purification of resistant wastewater is photo-Fenton degradation technology [164,165]. There were no appreciable variations in the ultimate pH of the solution among pH = 5.9 on 9.2 besides PZC next to 7.6. This positive outcome demonstrates the substance's possible utility as an atrazine adsorbent at a neutral solution of pH. Nevertheless,

electrostatic interactions rather than effective bonding produced this surface charge [166]. The variability of the carbon groups may be the cause of this dynamic interaction with (+) or (-) charges. The applicability of GPC in the management of atrazine in real wastewater records promising results in some areas. The reusability related to GPC till four cycles show that the adsorption peaked in the first cycle at 100% and then steadily fell to 60 ± 7%, although desorption peaked at 100% at the 4th cycle at 39.6%.

In the attempt to develop an extensive surface area of active carbon that can efficiently eliminate atrazine in artificial solutions of water and rivers water, *Persimmon* seeds residual gathered by the fruit of *Diospyros kaki* pulp was employed as the precursor substance (Fig. 6a). The substitute activated carbon (Fig. 6b) shows superior qualities, including the large specific surface area (1067 m<sup>2</sup> g<sup>-1</sup>) as well as volume of pores (0.530 cm<sup>3</sup> g<sup>-1</sup>) values as well as some significant functional groupings on the outermost layer. Adsorption efficiency rose with temperature, from 194 ± 20 to 211 ± 51 mg g<sup>-1</sup>. The ideal classical to replicate equilibrium data was Freundlich model. The endothermic feature that characterizes the adsorption method was validated by thermodynamic data. Regardless of the starting concentration of atrazine, kinetic tests showed that equilibrium was not attained until 240 min [201].

As a mean for eliminating atrazine, the remainder of the husk that covers *Psidium cattleianum* fruit is carbonized by FeCl<sub>3</sub> to serve as an activated agent. Using an average pore size of 0.280 cm<sup>3</sup> g<sup>-1</sup> as well as an external area measuring 431 m<sup>2</sup> g<sup>-1</sup>, activated carbon displayed properties of microporous materials. The micrographs showed that the precursor had uniform-sized cavities with small cracks (Fig. 6c) while the developed activated carbon had fresh holes in the form of uniform-sized circles; the crack has been disappeared owing to the modification (Fig. 6d). It was revealed that certain bands had vanished and that the bands that were still there belonged to efficient groups that contained oxygen, carbon, plus hydrogen. Isotherm data were best suited by the Liu isotherms. As a result, activated carbon made from "araça" fruit leftovers showed the potential to eliminate atrazine from aqueous solutions. At 328 K, the adsorption capability was 35.67 mg g<sup>-1</sup> [167].



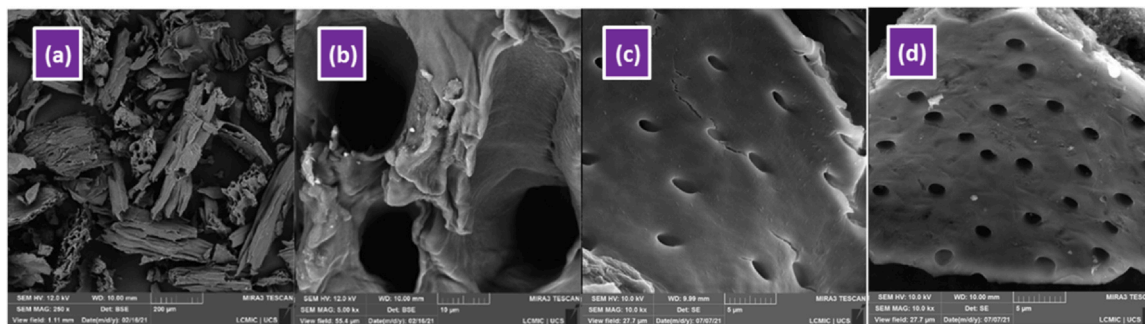


Fig. 6. SEM images of Persimmon seed residue (a) and the corresponding activated carbon (b) and psidium cattleyanum peel (c) and the corresponding activated carbon (d) [167].

Forthcoming research ought to pay attention to the enhancement related to the adsorption capacities by using developed activated carbons via functionalization and increasing the specific surface area and porosity. Much more attempts associated with the advancement of activated carbons from industrial wastes, such as microplastics in addition to wasted rubbers and agricultural wastes, should be increased within the concept of waste-treat-waste. The use of wastes as precursors is strongly encouraged and contributes to the circular economy via the valorization of numerous wastes.

### 3.2.2. Biochar adsorption

Biomass is pyrolyzed under oxygen-limited conditions to form biochar. Recently, interest in biochars has grown since their application in agriculture, where it has been promoted as a way to trap carbon and improve soil fertility [168]. Very potent adsorption affinities for pollutants are displayed by biochar [169]. Because of its adsorption capabilities, biochar can be used in different environmental treatments. Certain organic contaminants, including insecticides, synthetic biphenyls (PCBs), as well as polymers of aromatic hydrocarbons (PAHs), like carbaryl, acetochlor, as well as atrazine, can be effectively absorbed by it [170].

Biochar made from pig dung was used to absorb atrazine. For example, pig dung stock was gathered in China, mid-air-dry after that crushed toward fit via a 2 mm strain. The fertilizer was after that cooked for 2 h in a ceramic pot with a tight-fitting lid at 350 °C /700 °C in a heated muffle furnace (where oxygen was quickly expelled). The created biochars were placed in amber glass bottles after being crushed to go through filters of 200:400 meshes and 0.038: 0.075 mm. The trials of the unprecedented biochars stayed exposed to acidic treatment to eliminate the ash from the biochars. Samples of the resultant biochar were restriction-dried furthermore kept in desiccators aimed at future use. Deashed biochar samples are known as DABC350 and DABC700, respectively, based on the initial heating [171]. The FTIR fingerprints (Fig. 7a) seem to be quite distinct in biochars produced at various temperatures. The adsorption data was fitted to Freundlich model (Fig. 7b). Except for atrazine onto BC350, all samples exhibited non-linear isotherms; the nonlinearity of the sorption isotherm increased with increasing pyrolyzing temperature. The corresponding affinity coefficient,  $K_f$ , values of BC700 was  $10^{2.77}$ , and was greater ( $P < 0.001$ ) compared to that on BC350, indicating that biochars could effectively sorb carbaryl and atrazine ( $10^{2.06}$  and  $10^{1.40}$ , respectively). Following deashing, biochar's adsorption ability significantly improved ( $P = 0.001$ ). On DABC350, the atrazine  $K_f$  rose to  $10^{3.35}$ , while at DABC700, it increased to  $10^{3.55}$ . Consequently, BC700 had higher two biochars' capability for sorption, and deashing increased sorption even more [172].

The outlines reflect the computed Freundlich model appropriate to the deashed biochar adsorption records fractionated by original biochar's organic matter concentration (BC350 has an unbroken line, while BC700 has a line with dots.). The two biochars had comparable atrazine adsorption capacities, with BC700 having a stronger adsorption effect

than BC350 due to the biochar's higher ash content at higher pyrolysis temperatures. The hydrolysis ranged from 23.5% to  $90 \pm 6\%$ , taking place higher at higher biochar dose up afterwards higher pyrolysis temperature (Fig. 7c). In addition to the dose and pyrolysis temperature of biochars, other adsorption parameter shows a significant the biochar's ability to bind atrazine such as pH (Fig. 7d) where Maize stalk was also pyrolyzed to develop biochar, which was then utilized to eliminate atrazine [173]. It was concluded that the developed biochar had a considerable capacity for atrazine adsorption and that its adsorption performance was superior following ammonium dihydrogen phosphate treatment. The  $q_e$  values of atrazine decreased significantly at higher pH values, pointing to biochar affinity for atrazine occurring encouraged at small pH. Compared to activated carbon, biochar synthesis requires fewer secondary pollutants because the raw components are far more common in the environment. As a result, biochar is an economical and sustainable material with a wide range of potential applications. The research on biochar is currently in a highly active development stage. The collection, in addition amount of natural substances, process optimisation, and then ensuing adsorption performance adjustment, all require additional studies [174].

Biochar is produced by heating biomass in the absence of oxygen. It is known for its strong adsorption properties and can effectively absorb certain organic pollutants. Pig dung can be used to produce biochar, which can absorb atrazine. The biochar's adsorption ability significantly improves after deashing. The corresponding affinity coefficient values for biochar were higher after deashing, indicating that biochars could effectively sorb carbaryl and atrazine.

### 3.2.3. Other functional materials

Alongside activated carbon and biochar, other functional materials like polycation-clay composites, zeolite, and nanoparticles can effectively adsorb atrazine [177]. For example, polycation-clay composites constructed on poly (4-vinylpyridine-co-styrene)-montmorillonite for removing atrazine from water. A popular polymer for water treatment is PDMAC. High levels of PDMAC adsorption on the clay led to a rate exchange of the mud with facilitated imazapyr combining. Based on the idea that "like attracts like" PVP-co-S clay composites have been developed to enable atrazine elimination from water [178]. Atrazine and PVP-co-S polymers are structurally compatible. The structural similarity between the s-triazine ring of atrazine and the pyridine rings connected to PVP, in addition to a styrene ring, helped atrazine adhere to PVP-co-S composites. This theory is supported by the significant adherence of atrazine to PVP-co-S composites compared to its brittle binding to PDADMAC composites, which are structurally incompatible with atrazine. While polymers with lower charge density could act as a loop as well as tails, polycations having a high charge density frequently adsorption as trains on a negatively charged surface. Such a design ought to increase the amount of atrazine-binding hydrophobic regions in the composite that are available for interactions. However, in comparison with its attachment with the smaller charged density polymer-clay composite (PVP/co/S45%) (0.2 g/g), the atrazine

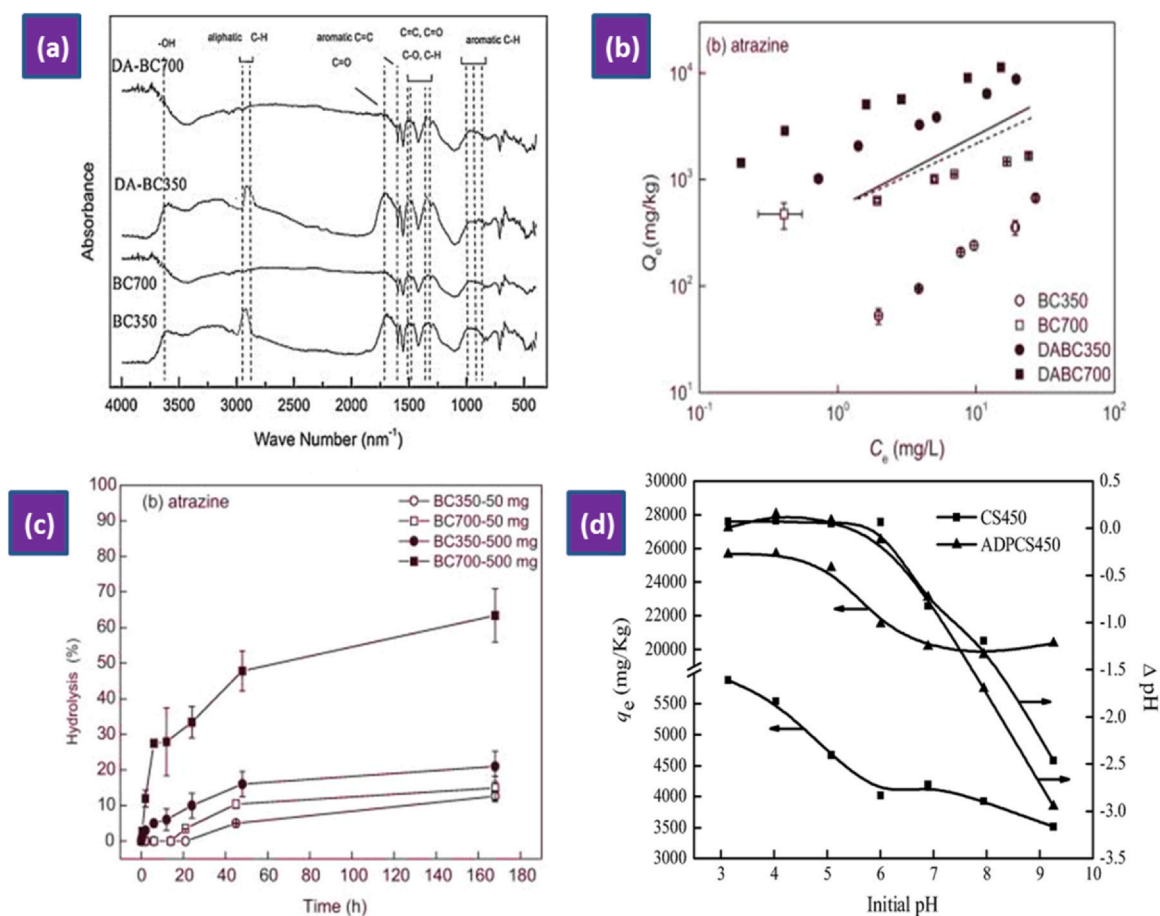


Fig. 7. FTIR spectra of the developed biochars (a), adsorption isotherms (b) and hydrolysis (c) of atrazine using the biochars [175]. The impact of initial solution pH on sorption capacities atrazine onto biochars at biochar dose = 200 mg L<sup>-1</sup> and initial atrazine concentration of 5 mg L<sup>-1</sup>. [176].

interaction to the higher charge densities polymer (PVP/co/S90%) adsorption onto montmorillonite (0.2 g/g) were significant (93%) while it only obtained 58% binding [179]. The decreased pH, in this case, is connected via the PVP-co-S90% compound's capacity to bind atrazine, though. The polymer loadings of 0.15 till 0.25 gm polymer per gm clay produced the maximum atrazine combining (95 ± 1%) for PVP/co/S90% over the clay composites. Even at high amounts of atrazine, 90–99% of it was removed from the water. After 20 to 40 min, the removal was at its maximum. For instance, after 18 h, a significantly higher concentration of dye-clay compounds (50 g L<sup>-1</sup>), 94–98% removal of the atrazine (24.5 mg L<sup>-1</sup>) was bound with dye-clay compounds [16].

Another frequent adsorbing substance is zeolite, but it needs to be modified to improve its adsorption effectiveness due to its low adsorption performance when left untreated. Two different forms of zeolites were developed from Egyptian kaolin, to investigate the atrazine adsorption behaviour. At 25 °C, the results of increasing the concentration of atrazine from 2 to 10 mg L<sup>-1</sup>, however, maintaining all other conditions fixed for atrazine adsorption onto zeolites A as well as zeolites X surfaces [180]. At low initial concentrations of atrazine, zeolite X demonstrated superior clearance than zeolite A. This might be explained by the fact that zeolite X contains more positive cations than zeolite A does. The electrical attraction between the negative end of the atrazine (chloride atom) and these cations, which behave as strong localized positive charges, accelerates quickly until equilibrium. Zeolite X has a surface area 688 m<sup>2</sup> g<sup>-1</sup>, while zeolite A has a specific surface area 333 m<sup>2</sup> g<sup>-1</sup>. Though, small holes in zeolite X account for the majority of its surface area since atrazine cannot penetrate them. At great primary concentrations of atrazine, the low removal percentage is caused by positive sites that are active in tiny pores of zeolite X having

no role in the process of adsorption. As a result, High clearance percentages are possible because atrazine may be absorbed or adsorbed into the inner surface of such holes. In contrast, Zeolite A gives a greater percentage of broad holes. Additionally, Atrazine adsorption increased with lowering pH, suggesting how the adsorption process had been molecular in pH-neutral settings and then ionic in pH-acidic conditions, demonstrating that pH has major effects on atrazine adsorption. Due to the somewhat acidic pH of the solution, Because of its molecule-like form of zeolite A, the atrazine has been absorbed in low quantities in addition to high levels onto zeolite X. In the course of the adsorption procedure, atrazine may pass into zeolite mass's porosity or may bind to zeolite's surface. The atrazine molecules travelled more slowly via the smaller diameter channels while diffusing more quickly through the pores. In this example, the adsorption mechanism and zeolite pores may have played a major role in atrazine uptake [181].

With the advancement of nanotechnology, nanoparticles are now being utilized to remove atrazine. Using precipitation and mini-emulsion polymerization, some researchers created highly specific molecularly imprinted polymer nanoparticles (MIP-NPs) [177]. They discovered that the MIP-NP-packed filtration unit could effectively remove atrazine at very low concentrations and that the MIP-NPs could be renewed and reused [16]. Creation of a magnetic molecularly imprinted polymer in mesoporous silica (mSiO<sub>2</sub>)-coated nanoparticles of Fe<sub>3</sub>O<sub>4</sub> for atrazine adsorption resulted in good atrazine adsorption performance, with no appreciable reduction in atrazine adsorption capacity after five cycles [182]. While polycation-clay composites and zeolite are accessible and use fewer resources, their adsorption capacities are poor, necessitating the need for improved adsorption performance in future studies [183]. The majority of nanomaterials have high adsorption characteristics. However, the preparation procedure is difficult, and the

**Table 1**  
The degradation of atrazine at relevant environmental conditions.

Mechanism of degradation	Initial conc. of atrazine	Time	Removal %	Main finding	References
Bioaugmentation (Pseudomonas and Arthrobacter)	5 mg L <sup>-1</sup>	44 d	91.8	The bioaugmentation could improve atrazine degradation in constructed wetland	[185]
bioremediation (strain TT3)	50 mg L <sup>-1</sup>	66 h	100	The high atrazine concentrations might inhibit the growth of the strain TT3	[186]
Photocatalytic degradation	1 µg mL <sup>-1</sup>	<ul style="list-style-type: none"> <li>• Pd CNT (120 min)</li> <li>• PdO-CNT (40 min)</li> <li>• Ag-Pd CNT (40 min)</li> </ul>	100	This architecture of catalyst provide a reusable and economic solution for the removal of atrazine from water	[187]
Photocatalytic degradation (ACF/CoFe <sub>2</sub> O <sub>4</sub> composites)	15 mg L <sup>-1</sup>	15 h	86.7	ACFs and H <sub>2</sub> O <sub>2</sub> improved the removal efficiency	[188]
Electrocatalytic degradation using CoFe@NC-CNTs/nickel foam	10 mg L <sup>-1</sup>	105 min	100	The free radical (O <sub>2</sub> <sup>•-</sup> ) and (•OH) and non-radical ( <sup>1</sup> O <sub>2</sub> ) contributed to atrazine removal	[189]
Electrochemical system	10 mg L <sup>-1</sup>	30 min	90.1	An efficient and stable (at pH 3-9) bifunctional cathode was developed <ul style="list-style-type: none"> <li>• OH is the main active free radical to remove atrazine, on the surface of the electrode.</li> </ul>	[190]
Fenton processes with nZVI/H <sub>2</sub> O <sub>2</sub> /UVA systems	10 mg L <sup>-1</sup>	120 min	80	The impact on the atrazine oxidation rate of water from different sources (pure, supply, simulated and secondary effluent real water) was studied. Cl <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> , HCO <sub>3</sub> <sup>-</sup> , and DOC, presented a moderate inhibitory impact on atrazine degradation rates, and their combination is considerably lower the removal efficiency at real secondary effluent water.	[191]
Fenton process using AFG@MIL-101(Fe) composite	30 mg L <sup>-1</sup>	105 min	81 ± 1%	At higher concentrations of H <sub>2</sub> O <sub>2</sub> (> 1.5 mL), it acts as an inhibitor and reduces the production of OH; and the removal efficiency decreased	[192]
photo-oxidation using montmorillonite and indole-3-acetic acid (IAA)	6 µM	24 h	38.7	These results demonstrated the strong dependence of the degradation process on the reaction pH and the exchangeable cations in the clay interlayer	[193]
Photocatalysis using W@TiO <sub>2</sub> @clay	2.5 mg L <sup>-1</sup>	240 min	100	W-TiO <sub>2</sub> /clay with 5% of W presents the highest photocatalytic activity under solar light	[194]
Bioaccumulation on Chlorella sp.	40 µg L <sup>-1</sup>	8 d	83	The degradation products of atrazine may inhibit algal growth as well as decrease removal percentage	[100]
Adsorption onto polyethylene	1-15 mg L <sup>-1</sup>	24 h	0.94 mg g <sup>-1</sup>	Adsorption capacities of atrazine on six target MPs were in the following order: aged PE > aged PP > aged PS > PS > PE > PP. Atrazine was easier to desorb from MPs in simulated gastric juice than in freshwater.	[195]
Adsorption	6.25 - 150 mg L <sup>-1</sup>	9 h	<ul style="list-style-type: none"> <li>• Graphene oxide (GO) (1.012 g/g)</li> <li>• Graphene nanoplatelets (1.006 g/g)</li> <li>• Reduced-GO (1.084 g/g)</li> <li>• BE350 (45%)</li> <li>• BE450 (51%)</li> <li>• BE550 (61%)</li> </ul>	The adsorption has a multi-mechanism feature and can be attributed to H-bonding and conjugated interaction between the adsorbents and atrazine	[196]
Adsorption onto biochar	2-10 mg L <sup>-1</sup>	240 h	60.9-84.3%	The temperature used to develop biochars plays a significant role in atrazine removal efficiency from water.	[197]
Goat manure amended treatment	10 mg kg <sup>-1</sup>	40 d		Goat manure has a dual effect on atrazine degradation in soil where it act as soil ameliorant and bacterial inoculum.	[198]

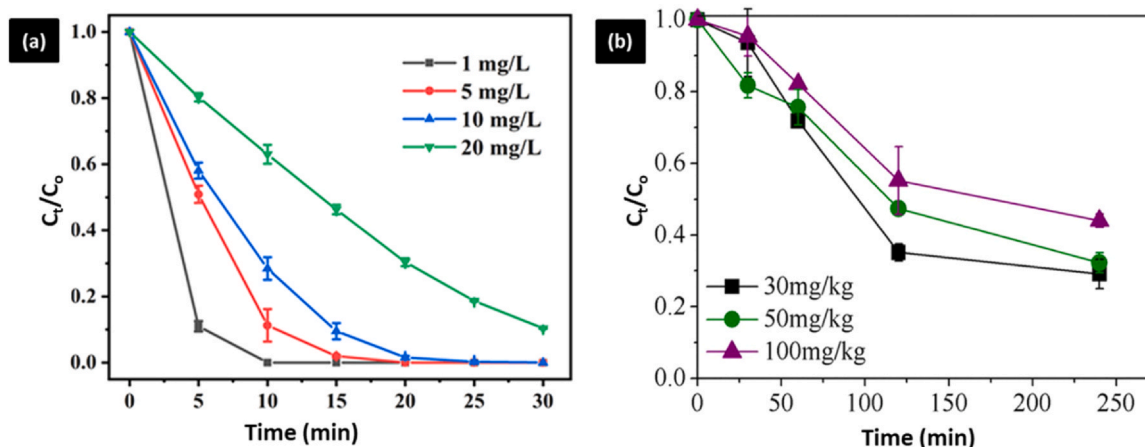


Fig. 8. Effect of initial ATZ concentration on degradation in soil by thermal activated PS process (a) [206] and (b) [207].

components are frequently pricey. As a result, the use of low-cost raw materials and straightforward preparation techniques is the way of the future [184]. Table 1 represents the degradation of Atrazine at environmental relevant conditions.

### 3.3. Various parameters regulating physicochemical degradation of atrazine

#### 3.3.1. Temperature effect on atrazine degradation

The impact of temperature change on atrazine deterioration with thermal-activated persulfate (PS) (20, 40, 50, and 60 °C) was carefully investigated. After 240 min of processing at 20 °C, no discernible atrazine degradation was found  $4.4 \pm 4.8\%$ . According to a report, PS is thermally inert in water at room temperature. On the other hand, the deterioration rate increased to 64% after 60 min when the temperature approached 60 °C and to 97% after 240 min. The increased PS breakdown rate with greater  $\text{SO}_4^-$  production at higher temperatures between 20 °C and 60 °C may be the cause of the improved atrazine degrading efficiency [199]. With a pseudo-first-order kinetics model, the degrading efficiency of atrazine under varied active temperatures may be fitted (Eq.10)

$$\ln\left(\frac{C_t}{C_o}\right) = -k_{\text{obs}}t \quad (10)$$

Where  $C_o$  and  $C_t$  stand for the atrazine levels during time zero (beginning concentration) and  $t$  (in  $\mu\text{M}$ ), accordingly. A linear regression analysis of a chart of  $\ln(C_t/C_o)$  versus reaction time might be used to determine the pseudo-first-order rates constant and  $K_{\text{obs}}$ .

It is demonstrated how temperature affects atrazine is adsorbed by activated carbon as an adsorbent. Since the system's temperature rose, all curves were in favour. The highest recorded  $q_e$  values jumped from  $21.71 \text{ mg g}^{-1}$  until  $35.67 \text{ mg g}^{-1}$  whenever the temperature went up from 298 K to 328 K. This increase in adsorption capability can be attributed to molecular mass transfer of the atrazine increasing and its solubility decreasing [200]. The remaining husks of *Psidium cattleianum*'s edible fruits were carbonised with  $\text{FeCl}_3$  as an activator and employed as an adsorbent to draw out atrazine. When the temperature fluctuated between 298 K and 328 K, via 26.39 up to  $35.67 \text{ mg g}^{-1}$ , the adsorption capability improved, indicating that temperature had a significant impact on equilibrium isotherms. The isotherm data were best suited by the Liu isotherms. The "araça" fruit's leftover activated carbon, which is highly effective in removing atrazine from aqueous solutions even at low temperatures, showed promise in this regard [201].

#### 3.3.2. pH effect on atrazine degradation

The impact of varying pH (3.0, 4.3, 7.0, 9.0, and 11.0) on atrazine degradation was also investigated by different researchers. It can be seen that with a  $\text{p}K_a$  of 1.7 and a tendency to get protonated when pH drops, atrazine is a weak base that may be more electrostatically attracted to

negatively charged  $\text{SO}_4^-$ . Lower pH values of 3 and 4.3 produced the most deterioration. Increasing pH may noticeably reduce atrazine's ability to degrade. Particularly, the degradation rate was just 13% at a pH of 11 [202]. The same finding was recorded in another study where the impact of pH on photo-Fenton degradation of atrazine was investigated via varying pH from 3.04 to 6.08, where  $\text{Fe@BiOBr}$  was applied as the catalysts in a visible light-based Fenton system. These results showed that  $\text{Fe@BiOBr}$  was able to greatly slow down the recombination of charge carriers and encourage the conversion of  $\text{Fe(II)}$  to  $\text{Fe(III)}$ , which allowed for the sustained development of  $\text{OH}$  from  $\text{H}_2\text{O}_2$ . In addition, the photoelectrons' prolonged  $\text{Fe(III)}$  to  $\text{Fe(II)}$  conversion restrained the interaction of  $\text{Fe(III)}$  with  $\text{H}_2\text{O}_2$  that creates  $\text{H}^+$ , resulting in a small pH shift (from 6.1 to around 5.7) [203]. The effect of pH change on atrazine degradation using peroxymonosulfate (PMS) activated with  $\text{MoS}_2$ . At elevated initial pH value, the degradation efficiency of atrazine is decreased owing to the electrostatic attraction or repulsion among atrazine,  $\text{MoS}_2$  and PMS at various pH values [204]. The impact of pH on the degradation of atrazine is important in some treatment approaches such as adsorption and advanced oxidation in contrast to other treatment approaches such as UV/microwave process [205].

#### 3.3.3. Concentration effect on atrazine degradation

Additionally, the effect of the initial atrazine concentration on deterioration was evaluated by various researchers. For example, a 3D-Co-MOF was hydrothermally prepared to activate PMS. the advanced oxidation processes showed that the degradation efficiency of atrazine is increased by decreasing the initial atrazine concentration from  $20 \text{ mg L}^{-1}$  to  $1 \text{ mg L}^{-1}$  (Fig. 8a), suggesting that a lower concentration of atrazine is preferable for its degradation [206]. At an initial soil atrazine concentration of  $30 \text{ mg kg}^{-1}$ , the decomposition efficiency of atrazine may reach 71% after 240 min (Fig. 8b). For  $50 \text{ mg kg}^{-1}$  as well as  $100 \text{ mg kg}^{-1}$  atrazine concentrations, the process of decomposition efficiency fell to 56% and 68%, accordingly. Whenever the PS dose remains constant, the higher concentration of atrazine molecules may cause competition as  $\text{SO}_4^-$  reacts with atrazine molecules, which might diminish the entire reaction's efficiency. Nevertheless, it should be noted that when using initial atrazine concentrations of 30, 50, and  $100 \text{ mg kg}^{-1}$ , correspondingly, the overall quantity of removed atrazine rose from 0.07 to 0.10 and 0.16 mg, respectively. This increase was caused by the greater effectiveness of interaction between  $\text{SO}_4^-$  and atrazine molecules [207].

## 4. Biodegradation of atrazine

### 4.1. Atrazine degrading bacterial strains

Removal of atrazine from contaminated areas by bacteria is one of the most widely reported microorganisms [136,208]. Several atrazine-

degrading bacteria strains have been characterized as potential bioremediation machines. Different catalytic enzymes such as 1-carboxybiuret amidohydrolase, cyanuric acid amidohydrolase, atrazine chlorohydrolase, allophanate amidohydrolase, hydroxyatrazine (HA) hydrolase etc., are produced by bacteria that break down atrazine. These enzymes perform different biochemical processes, such as dechlorination, dealkylation, hydroxylation, and ring cleavage, to diminish the half-life of atrazine deprivation [209]. *Pseudomonas* strain ADP uses three enzymatic processes to convert atrazine into cyanuric acid [210]. Atrazine was significantly degraded from atrazine-contaminated soil by using the composites of FeMBC (iron-modified biochar) and *Acinetobacter lwoffii* DNS32 (bFeMBC) [211]. *Acinetobacter lwoffii* DNS32 bacteria, combined with biochar, was observed to degrade atrazine from liquid and soil experiments efficiently and improved the soil's available phosphorus [212].

Atrazine is also converted into cyanuric acid, which is further metabolized to produce compounds that are rich sources of carbon and nitrogen by different types of bacteria from the genera *Alcaligenes*, *Klebsiella*, *Agrobacterium*, *Acinetobacter*, *Rhodococcus*, *Clavibacter*, *Streptomyces*, *Bacillus*, *Pseudomonas* and *Arthrobacter* [17]. But, only *Arthrobacter* sp., *Pseudomonas* sp., and *Bacillus* sp. have been reported in the last ten years to be fully capable of degrading atrazine into CO<sub>2</sub> and NH<sub>3</sub> among the most isolated atrazine-degrading bacteria [213]. When two strains of bacteria, i.e., *Bacillus licheniformis* and *B. megaterium* were combined after their isolation from soil, they exhibited faster atrazine degradation than their individual use after 7 d [214]. A 95.05% removal rate for atrazine was observed by the application of a novel bio-organic fertilizer named DNB10, which was prepared by mixing *Arthrobacter* sp. DNS10 strain, biochar, agricultural solid wastes, and poly-( $\gamma$ -glutamic acid) [215]. *Pseudomonas* and *Arthrobacter* were significantly noticed to be markedly involved in atrazine degradation in the constructed wetland [216]. Co-culture of *Arthrobacter* sp. DNS10 and *Enterobacter* sp. P1 bacterial strains caused 99.18% degradation of atrazine, indicating the benefits of this novel biostimulation technique to augment the atrazine biodegradation [217]. Zhang et al. (2023) found that degradation of atrazine in rhizosphere soil was majorly performed by different bacterial genera, i.e., *Arthrobacter*, *Bradyrhizobium*, *Nocardioides*, and *Rhodococcus* [79].

#### 4.2. Atrazine degrading fungal strains

Breakdown of atrazine is also performed by another major part of soil microflora, i.e., fungi after bacteria. They perform N-dealkylation of either alkylamino group to produce various metabolites, which are helpful in atrazine degradation at varying rates [218]. Fungi-mediated atrazine removal from polluted soil may be the most significant technique. Fungi produce various enzymes which help in catabolizing different steps during chemical degradation by using different carbon sources for metabolism. Therefore, they are considered most effective in bioremediation [219]. Atrazine degradation is also performed by basidiomycetes, wood-degrading fungi. The ligninolytic ability of white rot fungi makes them suitable for chemical degradation [220]. Different enzymes, which are of significant importance, are produced by basidiomycetes and ascomycetes for the deprivation of both herbicides and pesticides [221]. The highly tolerable ability of white-rot fungi under extreme ecological conditions makes it advantageous for atrazine degradation. For instance, atrazine was degraded under water deficit circumstances by basidiomycetes and *Trametes versicolor* in nonsterile soil [17,222]. Atrazine was successfully degraded by ectomycorrhizae fungi in association with plants [223,224]. Soil microorganism activity and soil enzyme activities were improved by arbuscular mycorrhizal fungi (AMF) for atrazine removal [218]. Atrazine was degraded in the near rhizosphere and bulk soils by *Glomus caledonium* and *G. etunicatum* [17,225]. *Bjerkandera adusta* showed efficient atrazine degradation with a removal efficiency of 92% and thus offers a novel and environmentally friendly method for the removal of pollutants without

affecting the environment [226]. Different basidiomycete strains exhibited a high potential for atrazine biodegradation by producing ligninolytic enzymes [227]. Fungus *Metarhizium robertsii* IM 6519 converted atrazine to various hydroxy-, dechlorinated or dealkylated metabolites to degrade atrazine with the help of cytochrome P450 monooxygenases [228]. Atrazine was significantly degraded by ligninolytic-producer fungi by transforming it into deethylatrazine [229].

#### 4.3. Other microbes degrading strains

A significant atrazine degrading ability was also shown by different yeast species. Yeast strain *Pichia kudriavzevii* strain Atz-EN-01 significantly degraded atrazine [230]. According to GC-MS study, it was found that atrazine degradation was performed by several metabolites which were produced from *Cryptococcus laurentii* that were taken from atrazine-polluted cultivated soil [231,232]. Zhu et al. (2018) reported atrazine degradation by *Saccharomyces cerevisiae* [233]. Atrazine was broken down into hydroxyisopropylatrazine, atraton (2-methoxy-4-ethylamino-6-isopropylamino-1,3,5-triazine), demethylated atrazine, HA [234], and hydroxy-dehydrogenated atrazine, and 2-OH-isopropyl-IPU [235] by *Pichia pastoris* strain X-33. A new Fe<sub>3</sub>O<sub>4</sub>-ECH-CS nanocomposite which was synthesized from Fe<sub>3</sub>O<sub>4</sub> nanoparticles, *Saccharomyces cerevisiae*, chitosan and epichlorohydrin, showed significant degradation of atrazine with a removal efficiency of [233].

#### 4.4. Genetically modified microorganisms

Different soil microorganisms transform atrazine into CO<sub>2</sub> and NH<sub>4</sub><sup>+</sup> [231]. The intricate catabolic route, which is modulated by a wide variety of enzymes encoded by a number of genes, allows microflora to degrade and utilize atrazine [213]. Atrazine conversion into its intermediate cyanuric acid involves various genes which are responsible for the degradation mechanism by associating with different steps [236]. Eight different genes, i.e., *atzA*, *atzB*, *atzC*, *atzD*, *atzE*, *atzF*, *trzN* and *trzD* have been characterized as a part of atrazine metabolic route [17]. *Pseudomonas* sp. strains ADP's *atzABC* genes were found to have homology to five microbial isolates that break down atrazine. Genes, namely *atzA/trzN*, encoding atrazine chlorohydrolase, were identified in *Arthrobacter agilis* and *Nocardioides nanhaiensis*, which ultimately produced HA by causing atrazine dechlorination [236]. Moreover, *atzD/trzD* genes encoding cyanuric acid amidohydrolase enzyme performed ring cleavage to transform cyanuric acid into biuret. In addition to this, hydrolysis of allophanate and biuret deamination was associated with *atzF/trzF* and *atzE* genes, respectively, whereas the dealkylation catabolic step was performed by *atzB/atzC* genes (Table 2).

Understanding different microbial biochemical processes through gene editing and system biology is imperative. For improving the bioremediation process, microbes with a desired gene can be designed for its breakdown using gene editing methods such as clustered regularly interspaced short palindromic repeats CRISPR-Cas, transcription activator-like effector nucleases, and zinc-finger nucleases [17]. This will result in improved and effective microbes-mediated herbicide removal by modulating the existing metabolic routes. By improving the capacity of microbes, genetic engineering can also create new opportunities for the breakdown of atrazine. Hence, an effective method for bioremediation could be the genetic transfer of degradation ability from one microorganism to another [17].

#### 4.5. Material-microbial combined technology

For the purpose of degrading various ecosystem's contaminants, different composite technologies have been in use in recent times. Necessary methods are required to create new combined biomaterials for cleaning the polluted ecosystem by using the mineralization of microbes and the physicochemical properties of materials. In this

**Table 2**  
Various atrazine degrading genes isolated from different microorganisms.

Genes involved	Enzyme encoded	Microbial species	Degradation product	Reference
<i>trzN</i> and <i>atzA</i>	Atrazine chlorohydrolyase	<i>Arthrobacter, Nocardioides</i> and <i>Pseudomonas</i> sp.	Cyanuric acid	[236]
<i>atzB</i> and <i>atzC</i>	Hydroxyatrazine ethylaminohydrolyase, and N-isopropylammelide isopropylaminohydrolyase	<i>Arthrobacter crystallopoietes</i>	Cyanuric acid	[243]
<i>atzA</i>	Atrazine chlorohydrolyase	<i>Arthrobacter</i> sp.	-	[244]
<i>atzA</i> , <i>atzB</i> , <i>atzC</i> , <i>atzD</i> , <i>atzE</i> and <i>atzF</i>	-	<i>Ensifer</i> sp	Cyanuric acid	[245]
<i>TrzN</i>	-	<i>Arthrobacter</i> strain DAT1	Hydroxyatrazine	[246]
<i>trzN</i> , <i>atzB</i> and <i>atzC</i>	-	<i>Arthrobacter</i> sp	Cyanuric acid	[247]
<i>trzN</i> , <i>atzB</i> and <i>atzC</i>	-	<i>Arthrobacter</i> sp.	Cyanuric acid	[248]
<i>atzA</i> , <i>atzB</i> and <i>atzC</i>	-	<i>Pseudomonas</i> sp. strain ADP	Cyanuric acid	[249]
<i>atzD</i> , <i>atzE</i> and <i>atzF</i>	Cyanuric acid amidohydrolyase, allophanate hydrolase,	Rhizospheres of <i>Inga striata</i> and <i>Caesalpinia ferrea</i>	NH <sub>3</sub> <sup>+</sup> and CO <sub>2</sub>	[250]
<i>trzNI</i> , <i>atzB</i> and <i>atzR</i>	Adenosine aminohydrolyase, amidohydrolyase	<i>Frankia alni</i> AClN14a and <i>Frankia</i> sp. <i>Eul1c</i>	-	[251]
<i>trzN</i> , <i>atzB</i> and <i>atzC</i>	Atrazine chlorohydrolyase, hydroxyatrazine ethylaminohydrolyase and N-isopropylammelide isopropylamidohydrolyase	<i>Citricoccus</i> sp. strain IT3	Cyanuric acid	[252]
<i>atzA</i> / <i>trzN</i> , <i>atzB</i> and <i>atzC</i>	-	<i>Arthrobacter</i> spp.	Cyanuric acid	[253]
<i>trzN</i> , <i>atzB</i> and <i>atzC</i>	Atrazine chlorohydrolyase, hydroxyatrazine ethylaminohydrolyase and N-isopropylammelide isopropylamidohydrolyase	<i>Arthrobacter aureescens</i> TC1	Isopropylamine	[254]
<i>trzN</i>	-	<i>Arthrobacter</i> sp. C3	Dechlorination	[255]
<i>atzB</i> and <i>atzD</i>	N-ethylaminohydrolyase and cyanuric acid hydrolase	<i>Puenaarthrobacter</i> sp. W11	NH <sub>4</sub> <sup>+</sup>	[256]
<i>trzN</i>	Atrazine chlorohydrolyase	<i>Nocardioides</i> sp.	Hydroxyatrazine	[257]

context, immobilization technology is the typical synthetic technique [145], for which different physical or chemical methods are used to fix microbes and materials together in a limited space which subsequently leads to the formation of biological material with both biological and physicochemical impacts. With regard to varying loading rates, these innovative biomaterials show effective action influence, greater microorganism count and large flexibility [237]. Atrazine was removed with a degradation rate of  $83.1 \pm 3.9\%$  when a reactor was prepared by encapsulating *Pseudomonas* sp. strain ADP in core-shell electrospun microtubes [238]. Almost 100% removal rate for atrazine was reported by a novel biomaterial prepared by encapsulating *Pichia kudriavzevii* Atz-EN-01 in a PVA-SA matrix [230]. A degradation rate of 94% for atrazine was reported by a biomaterial which was made through immobilization of *Pseudomonas* sp. ADP on thin silica layers [239]. A 95.53% removal rate for atrazine was achieved by using the microspheres biomaterial made from Fe<sub>3</sub>O<sub>4</sub> and *S. cerevisiae* [240]. Similarly, 91.2%, 88% and 99.67% removal rates for atrazine were reported by using Fe<sub>3</sub>O<sub>4</sub> -*Penicillium* sp. yz11-22N2, Fe<sub>3</sub>O<sub>4</sub> -chitosan- *S. cerevisiae* and sodium alginate- *Arthrobacter* sp. DNS10 biomaterial composites, respectively [233,241,242].

#### 4.6. Microbial metabolic pathway of atrazine bio-degradation

In recent years, research on atrazine degradation has yielded significant progress in identifying and isolating microbes that can degrade atrazine. Numerous studies have reported the enrichment and isolation of atrazine-degrading microbes from contaminated sites across different geographical locations [258,259-261]. Extensive research on atrazine degradation has revealed that atrazine breakdown by microbes occurs through dealkylation, hydrolytic dechlorination, and deamination reactions. Primarily, the researchers focused on the dealkylation of atrazine, which led to the identification of various dealkylated metabolites like DEA (deethylatrazine), DIA (deisopropylatrazine), and DEDIA (dealkylatrazine) [262-265]. These metabolites were generally found in the soil, which supported the fact that dealkylation was the chief pathway involved in atrazine breakdown by microbes. However, successive research revealed the presence of bacterial strains that have the ability to catalyze hydrolytic dechlorination, resulting in the formation of hydroxy-atrazine (HA), the first intermediate of the degradation pathway [264].

Generally, the mineralization of atrazine occurs in two stages. In the first stage, N-alkyl side chains and chlorine are removed, leading to the formation of cyanuric acid. Interestingly, the enzyme involved in this stage has the potential to perform diverse reactions on different substrates [266]. While in the second stage, the s-triazine ring is removed and converted into NH<sub>4</sub><sup>+</sup> and CO<sub>2</sub> via specific enzymes with a narrow substrate range. The extensive research on atrazine degradation has led to the identification of three distinct pathways for converting atrazine to cyanuric acid, one of which is purely hydrolytic. At the same time, the other two are mixed oxidative-hydrolytic according to enzymatic activities compiled at the University of Minnesota Biocatalysis and Biodegradation Database (<http://umbbd.msi.umn.edu/>) [267].

The hydrolytic pathway involved in the degradation of atrazine has been extensively studied in *Pseudomonas* sp. ADP, involves three enzymatic steps catalyzed via the gene products of *atzA*, *atzB*, and *atzC* [268]. The *atzA* gene has been found to be involved in the dechlorination of atrazine [269], whereas *atzB* and *atzC* genes catalyze the removal of N-alkyl substituents from the s-triazine ring to produce cyanuric acid [270,271]. These genes are prevalent and are present on plasmids of the multiple atrazine-degrading isolates from different regions [243,272,273,258]. In a few cases, Gram-positive strains of microbes, *trzN* substitutes for *atzA* [274].

On the other hand, oxidative-hydrolytic pathways involve the initial oxidative N-dealkylation of atrazine to deisopropylatrazine or deethylatrazine. These reactions can lead to the formation of deisopropylatrazine or deethylatrazine, which can undergo further dealkylation,

dehalogenation, deamination, and hydrolytic reactions to produce cyanuric acid. Different enzymes from various organisms are involved in these oxidative reactions, such as *Rhodococcus* sp. N186/21 cytochrome P450, AtrA from *Rhodococcus* sp. TE1, and TriA, which performs the deamination of atrazine derivatives [275–277]. However, no single organism has been found to perform all the necessary enzymatic reactions for complete atrazine degradation via oxidative-hydrolytic pathways. Instead, a combination of enzymes from different organisms, such as a mixed culture of *R. corallinus* NRRLB-15444R and *Pseudomonas huttiensis* NRRLB-12228, has been shown to be capable of degrading deisopropylatrazine [278,279,274].

The microbial degradation of atrazine typically involves a consortium of microorganisms rather than a single species. Different bacterial strains within the consortium have complementary enzymatic activities. For example, one bacterium may be responsible for dealkylation, while another carries out dechlorination or deamination reactions. The interactions between microorganisms in the mixed culture are crucial for the efficient breakdown of atrazine into simpler and less toxic compounds [280,281]. Through co-metabolism, where atrazine is used as a carbon or nitrogen source, enzymes that have evolved to break down similar compounds can be utilized for atrazine degradation [264]. The rates and pathways of atrazine degradation can be influenced by environmental conditions such as pH, temperature, oxygen availability, and the presence of other substances. Thus, progressive exploration needs to be continued to uncover new microbial strains and enzymes that play crucial roles in the degradation of atrazine, which will enhance our understanding of these microbes, enzymes and pathways involved in atrazine degradation.

#### 4.7. Molecular approaches for atrazine bioremediation

The progressive development in the field of genetic engineering has emerged as a promising solution since it allows the modification of microbes to show higher metabolic potential [282,283,284]. Researchers are now exploring various tools as well as techniques to enhance and optimize biocatalysts for the degradation of atrazine. Rational and semi-rational design, accompanied by directed evolution, are currently fast-growing fields that are now also employing machine learning and artificial intelligence to enhance the biocatalysts performance [285,286,287]. The primary aim of genetic modification is to improve the key characteristics of enzymes like solvent robustness, tolerance to varied temperatures and enantioselectivity. Besides enzyme optimization, genetic engineering has also opened new way for modifying biocatalytic sites to amend their functionalities and allow us to develop artificial multifunctional enzymes [288]. This will open new possibilities for developing tailored enzymes for bioremediation of atrazine.

Additionally, enzyme cascades have also emerged as a promising approach for the effective degradation of atrazine and have gained attention as means of creating "biological factories". This concept involves combining different enzymatic activities in a strategic manner to take advantage of the synergistic effects of multiple enzymes [289]. By tactically designing enzyme cascades, researchers now have the ability to improve the degradation process and attain more efficient removal of atrazine pollutants.

In the field of bioremediation, genetic engineering extends beyond individual microbes and focuses on whole microbial communities through in situ metagenomic engineering [290]. This approach involves insertions of transposons genes into native microbial populations via *Escherichia coli* as a carrier. The application of RNA-targeting CRISPR systems has accelerated this process, allowing researchers to effectively deliver genetic payloads to microbial communities in their natural environment [291]. This method, known as in situ metagenomic engineering, holds great promise for enhancing bioremediation processes by harnessing the collective power of microbial communities [282,292].

Specific genes and enzymes have been identified as crucial in the degradation of atrazine. For instance, the *atzA* gene, which encodes the atrazine chlorohydrolase, is an enzyme responsible for the effective degradation of atrazine [293]. Recombinant *Escherichia coli* expressing atrazine chlorohydrolase has been developed and used for successful field-scale bioremediation of atrazine-polluted soil [294]. In another study, a gene responsible for the deamination of de-ethylatrazine and de-isopropylatrazine was transferred from *Rhodococcus corallinus* to *Rhodococcus* TE1, resulting in a complete atrazine degradation to cyanuric acid in the recombinant *Rhodococcus* strain [295].

Genetic rearrangement events have been observed in the movement of atrazine-degrading genes from plasmids to the chromosomes of microbial strains, facilitated by transposons or insertion sequences [296]. Horizontal gene transfer of atrazine-degrading genes has also been demonstrated in soil bacteria through the conjugative transfer of plasmids [297]. These findings highlight the ability of indigenous microbial communities to acquire atrazine-degrading capabilities, expanding the potential effectiveness of bioremediation strategies.

In conclusion, molecular biology offers exciting opportunities for addressing the challenges posed by atrazine through genetic engineering methods like in situ metagenomic engineering. By utilizing the capabilities of atrazine-degrading genes and enzymes, researchers can optimize biocatalysts, construct enzyme cascades, and transfer desirable traits to microbial communities. These advancements hold great promise for developing efficient, sustainable, and customized bioremediation solutions that mitigate the environmental impact of atrazine and other persistent pollutants.

#### 5. Circular economy-driven atrazine treatment

Incorporating circular economy into the removal of atrazine is important for the sustainable management of pollutants. Due to its high specific surface area, micro-porosity, and insensitivity to toxic pollutants, the adsorption of atrazine with commercially activated carbon is effective in the removal of the contaminant from water [298]. However, commercial activated carbon is non-renewable. This means that once it has fulfilled its purpose, the by-product is considered waste, which promotes a linear economy. If the disposed waste is not managed properly, it can release adsorbed contaminants into the environment. Production of commercial activated carbon also depletes resources while contributing to greenhouse gas emissions during the process. However, cheap, renewable, and sustainable alternative adsorbent materials such as Persimmon seed residue obtained from *Diospyros kaki* fruit wastes are now being used as a precursor material to develop a high surface area activated carbon that effectively removes atrazine. Leftover husks from the edible fruits of *Psidium cattleianum* are carbonized with  $\text{FeCl}_3$  as an activating agent. The activated carbon made from arça fruit leftovers exhibited potential in eliminating atrazine from aqueous solutions. The use of agricultural waste as precursors promotes a circular economy via the valorization of numerous wastes. Biochar is an adsorbent produced from biomass rich in carbon and is a low-cost technology that has been applied in removing atrazine. The high surface area of the substance enables effective adsorption of atrazine and increases the lifecycle of waste while promoting a circular economy. Biochar offers an economical and sustainable alternative to non-renewable commercial activated carbon. Waste-based materials such as sediment, sand, clay, soil, dairy manure, agricultural and industrial waste, and rubber granules can also be used for the removal of atrazine [299]. Due to their availability, large specific surface area, low cost, and chemical and mechanical stability, clay minerals have been widely used to adsorb different molecules [300]. Sepiolite is one such clay that is abundantly available at a low cost and has a high specific surface area and porosity for adsorption [300]. Pine bark, due to its high potential sorption capacity by chelating processes and ion exchange, has been proposed as a biosorbent to remove pollutants from contaminated wastewater [300]. Cork has suitable properties like

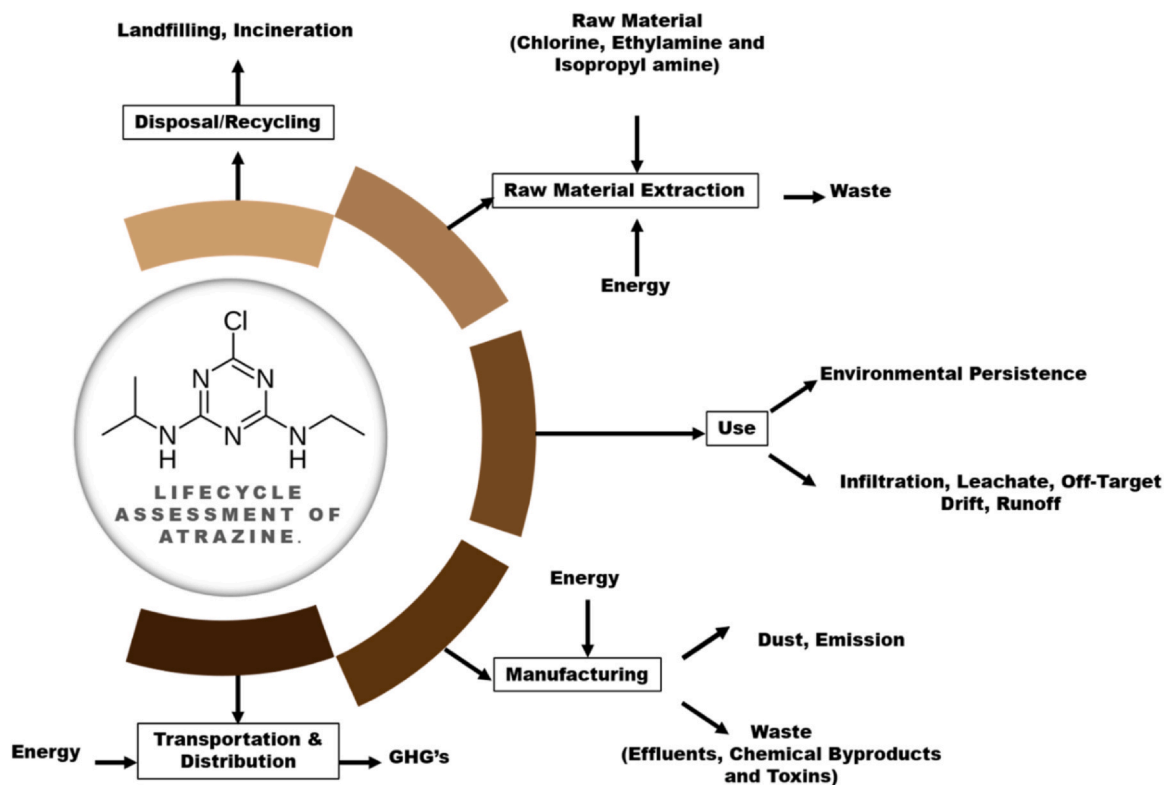


Fig. 9. Life cycle assessment of atrazine.

buoyancy, low density, and important binding sites on its structure that make it feasible for sorption [300]. Cork powder rejected from industries and used cork stoppers can be acquired cheaply and used as an adsorbent [301]. Spent tires can be turned into an alternative adsorbent by recycling the abundantly available used tires which increases the lifecycle of the waste and promotes a circular economy [301]. In a study conducted by reference [300], it was concluded that cork, sepiolite, and pine bark could be regenerated at least three times using a desorbing eluent and reused without the loss of adsorption efficiency. This enhances product value and lifecycle and promotes a circular economy with a reduced cost and a smaller environmental impact from final waste. To treat contaminated water, permeable reactive barriers are placed in the subsurface and filled perpendicular to the direction of groundwater flow. The target contaminants are then immobilized, adsorbed, and changed into less toxic and harmful forms because of various physical and biochemical processes [300]. Instead of replacing the adsorbent, solvents are used to regenerate adsorbents and recover their adsorption capacity, which reduces waste production [300]. Extraction or recovery of the target substance causes low mass loss of the adsorbent, which promotes multiple uses [302]. Recycling or repurposing atrazine by-products can be done if certain by-products have value in specific applications and the extraction and purification of these compounds can be done for use in other industries or chemical synthesis which can contribute to a circular economy.

Studying the degradation pathways of atrazine and its by-products can help explore new catalytic or enzymatic methods for degradation and identification of potential value-added products that can be derived from these compounds. Atrazine-degrading bacteria and fungi can aid in the biodegradation of the contaminant and their by-products by breaking down the complex compounds into simpler, less toxic substances over time [218]. Water treatment processes such as activated carbon filtration, advanced oxidation, and biofiltration can be used to remove or degrade residual compounds before the water is used for consumption. The previous experiments indicated that soybean, corn, and cotton plants were able to take up atrazine into their foliar tissue

within 7 h of application and gradually degraded over time. Genes that uptake and transport contaminants could be overexpressed to enhance the effectiveness of phytoremediation and once the contaminant is degraded to non-toxic metabolites, the plants could be disposed of safely. Bioremediation with the help of soil microorganisms can degrade atrazine through biochemical reactions productively and sustainability. In the case of inefficient degradation of pollutants by bacteria, fungal degradation can be more effective. Although these methods provide a natural solution to degradation and promote circularity, the degrading ability would depend on several environmental factors.

## 6. Life cycle assessment

Atrazine is a systemic herbicide that is a chlorinated triazine used to selectively control grasses and broadleaf weeds before they emerge [218]. It is synthesized from raw materials such as chlorine, ethylamine, and isopropyl amine (according to The National Library of Medicine). The extraction, production, and manufacturing process of these materials involves energy consumption and resource extraction that further depletes natural sources while posing significant environmental impacts from waste generation and the emission of pollutants and contaminants [303]. In the manufacturing process, chemical reactions and processes used to synthesize atrazine consume energy and produce chemical waste by-products and effluents. The manufacturing process contributes to the emission of smoke, dust, and greenhouse gases while also polluting water resources and generating toxic and hazardous waste if not properly managed [304,305]. Once produced, atrazine is distributed to farmers and applied to crops for weed control and the transportation of atrazine products to the users can result in greenhouse gas emissions and energy consumption [306]. Atrazine infiltrates the soil from agricultural fields through leaching and agricultural runoff and contaminates groundwater and ends up in surrounding water resources [307]. The potential off-target drift and atrazine runoff impact non-target organisms, especially aquatic life



[12]. It inhibits photosynthesis and decreased plant growth and may also lead to the death of aquatic plants when sufficiently concentrated [74]. Exposure can also lead to changes in sensitive species that may suppress or kill them, which can alter the structure and function of the aquatic ecosystem. It also affects reproductive capacity and hinders development [12]. Atrazine does not bioaccumulate, but it persists in the environment, and the by-products of degradation may vary in toxicity and persistence. The fate and behavior of atrazine and its degradation by-products in soil, water, and ecosystems are important considerations in assessing their environmental impacts [19]. The disposal of atrazine products, by-products, and the waste generated during the manufacturing process should be managed properly and safely to prevent environmental contamination. Incineration of atrazine is commonly followed. However, landfilling as well as other waste management methods, may also be associated with significant environmental impacts [177,308]. Atrazine has been regulated in many regions due to its potential environmental and health impacts and simultaneously the development and adoption of alternative renewable, eco-friendly herbicides and agricultural field additives need to be encouraged to mitigate associated environmental concerns [309,310]. An overall view of typical life cycle assessment of atrazine is represented in Fig. 9.

## 7. Conclusion and future prospective

The distinctive characteristics of atrazine pose significant difficulties in attaining optimal degradation efficiency with any single technique. Aforementioned treatment methods for atrazine have notable advantages, including its ease of operation and relatively short treatment duration. Nevertheless, the efficacy of the approach in eliminating organic contaminants is not comprehensive, hence limiting its practical implementation. The chemical approach exhibits a broad spectrum of applicability owing to its efficient breakdown of organic contaminants. In contrast to other treatment technologies, the chemical technique exhibits a comparatively higher operating cost and the potential generation of harmful intermediates during the treatment process. Therefore, its practical uses are limited. The biological approach is characterized by its cost-effectiveness and minimal environmental impact. However, it is important to note that the processing time associated with this method is very lengthy. As a result, the primary focus of research in this sector is on identifying and evaluating highly efficient strains and species. The novel biomaterial integrates the inherent benefits of both materials and microorganisms, exhibiting remarkable efficacy in the elimination of atrazine. This innovative approach represents a promising avenue for further exploration in the field of environmental remediation. Nevertheless, the drawbacks of this approach are readily apparent, necessitating additional investigation. Due to the distinctive physicochemical characteristics of atrazine, attaining optimal removal efficacy through the use of a singular technique for atrazine treatment poses challenges. Hence, the prioritization of research in the domain of organic pollution treatment is in the synergistic use of two or more methods. Hence, recent research has indicated that the concurrent implementation of multiple strategies and the synergistic utilization of two or three approaches concurrently hold promise as the forthcoming paradigm for soil herbicide treatment. Consequently, future investigations should prioritize the examination of the simultaneous application of a select few technologies in a synergistic manner. Furthermore, given that the majority of investigations pertaining to this topic have been conducted using synthetic soil and controlled laboratory settings, transferring the research to real-world environments would help bridge the disparity in efficacy between laboratory-based scenarios and practical implementation. This would also enhance the accuracy of prediction models. Furthermore, the focal point of future research lies in the development of functional materials that possess exceptional properties while maintaining a minimal environmental footprint.

## Compliance with ethical requirements

This article does not contain any studies with animals performed by any of the authors.

## CRediT authorship contribution statement

**Simranjeet Singh:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Original draft Writing - review & editing; **Nadeem A Khan, Rania Ramadan, Nabila Shehata, Dhriti Kapoor, Daljeet Singh Dhanjal, Nikhita Sivaram:** Writing and editing; Critical revision of the manuscript and final approval: **Praveen C Ramamurthy, D. Barcelo and Joginder Singh.**

## Data Availability

Data will be made available on request.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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