

JEBS-25-13

Harnessing Advanced Photocatalytic Oxidation for the Degradation of Drug Waste in Wastewater

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Received date: November 30, 2025; Accepted date: December 12, 2025; Published date: December 31, 2025

Citation: Chauhan G (2025) Harnessing Advanced Photocatalytic Oxidation for the Degradation of Drug Waste in Wastewater. J Environ Biol Sci. Vol.1 No.2: 13.

Abstract

Pharmaceutical Drugs (PDs), while essential for human healthcare, have emerged as significant environmental contaminants due to their widespread use, incomplete metabolism and improper disposal. These compounds persist in aquatic and terrestrial ecosystems, entering the environment through human excretion, hospital, waste water treatment plants, industrial effluents and agriculture runoff. Even at trace concentrations, pharmaceuticals exhibit bioactive properties that disrupt ecosystem functions, bioaccumulate in organisms and contribute to the development of antimicrobial resistance, posing ecological and human health risks. Conventional treatment methods are largely ineffective at eliminating pharmaceutical contaminants (PCs), whereas Advanced oxidation processes provide a sustainable and efficient approach by degrading these PCs into non-toxic byproducts. This review critically discusses the sources, environmental fate and impacts of PCs, evaluate the limitations of existing treatment methods and highlights the recent advances in AOPs based technologies for pollutant removal. This review also discusses future research directions, emphasizing the development of cost-effective, stable and highly efficient photocatalysts, alongside enhanced environmental monitoring to achieve long-term mitigation of pharmaceutical pollution and safeguard both human and ecosystem health.

Keywords: Pharmaceutical drugs; Advanced oxidation processes; Conventional methods; Wastewater treatment plants; Photocatalytic degradation

Introduction

Pharmaceutical Drugs (PDs) are the pillars of the modern medicine and play a significant role in the prevention, diagnosis and treatment of diseases. This is due to the fact that they have transformed the treatment of chronic diseases such as hypertension, diabetes and many others through agents like antihypertensives, insulin, statins and others, significantly improving patient outcomes and quality of life [1]. PDs have also revolutionized the control of infectious disease, with the help of antibiotics, antivirals and antifungals drastically reducing mortality and morbidity [1]. In the same way, advancements in oncology including chemotherapeutics, targeted therapies and immunotherapies have enhanced cancer survival rates, exemplified by drugs such as trastuzumab and imatinib [2,3]. Preventive PDs have benefited immensely from vaccines and prophylactic drugs like aspirin and statins, reducing disease incidence and long-term risks. Emerging fields such as

pharmacogenomics and biologics have enabled personalized and molecularly targeted treatments, while novel drug delivery systems like nanoparticles and liposomal carriers have improved precision and reduced side effects [4,5]. Despite these remarkable achievements, PDs have become emerging environmental pollutants, posing serious ecological and human health concerns.

Environmental contaminations from PDs waste are now recognized as a growing global issue, though often underestimated compared to other environmental challenges. Despite PDs essential role in disease treatment, they are not completely absorbed or metabolized resulting in excretion of unchanged compounds [6,7]. The release of these residues poses serious risks to aquatic and terrestrial ecosystems, biodiversity and human health [8]. Over the last two decades increasing PDs consumption have raised global concern [9,10]. PDs have been widely detected in soils,

sediments, wastewater sludge, surface and ground water and in marine systems [11,12]. Concentrations are generally very low, yet even trace levels of bioactive compounds adversely affect ecosystem and human health [13,14]. Major sources include effluents from Wastewater Treatment Plants (WWTPs), pharmaceutical industries, healthcare facilities, households and runoff from agriculture [14-16]. Conventional treatment methods are often ineffective in removing this waste, leading to their discharge into lakes and marine systems [14,17,18]. Additional inputs arise from food processing and improper disposal of expired and unused drugs [19]. For instance, Estuaries located at the freshwater-marine interface, host diverse habitats such as mudflats, mangroves and seagrass beds and are among the most productive ecosystems globally. They support fisheries, aquaculture and biodiversity. However, their exposure to these pollutants from both land and sea, combined with intense human activity, makes them highly vulnerable to PDs contamination. Therefore, there is an urgent need for sustainable approaches to degrade these PDs waste and minimize their environmental impacts. Also, stakeholders must adopt innovative waste reduction strategies to promote a cleaner and greener environment [14,20].

Sources and Pathway of PDS Waste to Environment

The growing detection of pharmaceuticals increases, while their distribution depends on physiochemical properties [21,22]. Major sources include human excretion, improper disposal, hospital and industrial waste and effluents from WWTPs [21-23].

The manufacturing industry, which produces Active Pharmaceutical Ingredients (APIs) on a large scale, is primary source of contamination. Despite the presence of regulatory frameworks, many of them discharge effluents directly into wastewater systems. While waste water treatment processes are inefficient to remove these effluents, they enter natural water bodies, causing severe pollution. such discharged not only affect aquatic ecosystems but also pose long-term risks to human health through bioaccumulation and development of antibiotic-resistant microorganisms [24]. In addition to industrial sources, excretion serves as a substantial contributor to pharmaceutical pollution. A significant portion of ingested PDs is excreted unmetabolized in urine and feces and subsequently enters water bodies.

WWTPs are not specifically designed to eliminate these residues, resulting in their persistence in environment [23,25]. Similarly, improper disposal practices further amplify the environmental burden of Pharmaceutical

Contaminants (PCs) as there is direct disposal of unused or expired PDs into landfills or sewage systems. In landfills, these compounds can leach into surrounding soils and ground water, making it difficult for conventional methods to remove them [23,26]. Overtime, these persistent PCs accumulate in aquatic and terrestrial ecosystems, posing risks to environment and human health.

Understanding the various pathways through which PDs enter the environment is essential for addressing this multifaceted issue. Among the numerous routes WWTPs, direct release into water bodies, soil contamination, leaching and runoff play dominant role [23]. WWTPs although essential for wastewater management, are not specifically engineered to eliminate PCs. A significant proportion of PDs are excreted in unmetabolized forms, enabling these compounds to persist through conventional treatment methods and subsequently contaminate water bodies [27,28]. Furthermore, direct discharge from manufacturing units, untreated domestic sewage, storm water runoff exacerbates environmental contamination, particularly in regions with inadequate treatment infrastructure and regulatory oversight [23]. Soil contamination also represents a critical route, as the application of treated wastewater and biosolids in agriculture facilitates the accumulation of pharmaceutical residues in soils and leaching into groundwater. Within the soil matrix, these compounds may undergo complex physiochemical and biological transformations, generating metabolites with uncertain environmental behaviour and toxicities [23]. Leaching and surface runoff subsequently mobilize these residues into aquatic ecosystems, occasionally resulting in episodic increases in contaminant concentrations that pose acute risks to aquatic organisms [23]. A comprehensive evaluation is therefore essential for the formulation of advance and sustainable methods, effective monitoring systems and regulatory policies aimed at mitigating pharmaceutical pollution (**Figure 1**).

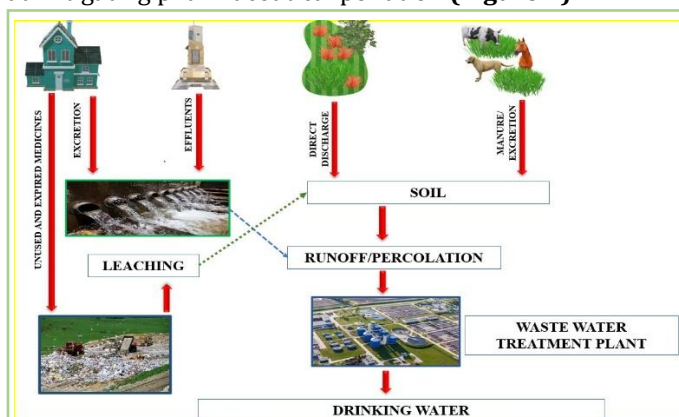


Figure 1: Sources and pathway of PDs waste to environment.

Environmental and Human Health Impacts

PDs waste contributes to persistent environmental contamination through multiple interconnected pathways that resist conventional remediation, creating long-term ecological and human health risks [29]. Environmental risks assessments reveal widespread contamination arising from inadequate disposal practices across healthcare and community settings. For instance, in Ghanaian municipalities, 96% of PDs are discarded via harmful routes, while 4% follow proper management, resulting in contamination across multiple media [30]. The persistence of these residues leads to sustained ecosystem exposure and potential bioaccumulation. Multiple contamination routes underscore the lasting environmental legacies of inadequate waste management [30]. Also, transport mechanisms facilitates the widespread movement of these PCs through interconnected systems, leading to complex exposure scenarios that impact multiple ecosystem components and tropic levels [31].

PCs enter aquatic systems through various sources, including untreated water, irrigation and domestic activities. Aquatic organisms, which are continually exposed to significant amounts of waste water residues, are highly vulnerable to a wide spectrum of chemicals present in these discharges. Due to their bioactive nature, pharmaceuticals can pose serious threats to both aquatic and terrestrial ecosystems [23].

Water bodies such rivers, lakes and oceans often act as the final repositories for many PCs, posing considerable risks to aquatic life, including fish, amphibians, invertebrates and phytoplankton. These PCs can induce a range of toxic effects both acute and chronic affecting organisms at multiple biological levels, from molecular to population scales [23]. For examples, synthetic estrogens from contraceptives have been linked to the feminization of male fish, resulting in distorted sex ratios and reproductive dysfunction [32]. Acc. to Paut Kausturia et.al., the global human population releases approximately 30,000 kilograms of natural steroidal estrogens and about 700 kg of synthetic estrogens each year solely from contraceptive use. Furthermore, NSAIDs have been associated with near extinction of several vulture species in Asia due to renal failure [23, 33]. Similarly, antidepressants like fluoxetine can disrupt the behaviour of aquatic organisms, altering feeding patterns, growth rates and predator-prey dynamics [34]. Terrestrial wildlife, particularly species that depend on aquatic ecosystems or utilize irrigated agricultural land, are also at risk. Pharmaceuticals present in soil can be absorbed by plants,

creating a potential exposure route for herbivores. For instance, veterinary PDs including antibiotics and hormones administered in livestock production can persist in manure and when used as fertilizer, contaminate soils, adversely impacting soil organisms and plant health [35].

However, currently there is no conclusive research identifying long term human health effects resulting from prolonged exposure to low concentrations of PDs in the water supply. However, significant uncertainties and inconsistencies remain regarding the quantitative methodologies used to estimate both predicted and actual environmental concentrations of APIs [36]. Human health can be affected by environmental pharmaceuticals primarily through the ingestion of contaminated water and food sources. Both surface and groundwater used for drinking may contain PCs. Although WWTPs can eliminate a considerable portion of these compounds, certain PDs exhibit high persistence and can pass through treatment systems, ultimately being discharged back into the environment. Consequently, PDs have been detected in both raw and treated drinking water, albeit typically at low concentrations [23]. In addition to water, pharmaceuticals can enter the human food chain through contaminated plants and animals. For instance, crops irrigated with polluted water or grown in contaminated soils can absorb PDs, which may then be consumed by humans. Similarly, fish and other aquatic organisms can bioaccumulate pharmaceutical compounds in their tissues, presenting another potential exposure pathway [37]. While the concentrations of PDs detected in food and drinking water are generally low, the implications of chronic, long-term exposure to these low-level mixtures remain poorly understood. This poses a major challenge in accurately assessing potential human health risks. Mackul'ak et.al. further emphasized that evaluating whether elevated concentrations of certain PCs in agricultural crops could ultimately pose risks to humans is a complex and unresolved issue. They also noted that even the mere presence of pharmaceuticals or their metabolites in plants might adversely affect plant growth and development [38].

Moreover, Antimicrobial Resistance (AMR) has become a critical global health concern, particularly after the surge in antibiotic use during COVID-19 pandemic [39]. The extensive use of antibiotics in human medicine, veterinary practices and agriculture has been strongly associated with environmental contamination, thereby driving the emergence of antibiotic resistance and various ecotoxicological effects [40]. Additionally, the improper disposal of antibiotics such as the direct release of unused or expired medications into sewage systems intensifies

environmental pollution and presents an escalating risk to public health. Long-term exposure to antibiotic contaminants can also negatively impact human health, particularly among individuals with chronic conditions such as obesity, diabetes and asthma [33,41].

Antibiotics have been shown to trigger oxidative stress and growth inhibition in aquatic species, reduce nitrogen-fixing capacity in plants and cause toxic accumulation in humans, leading to gastrointestinal disturbances such as vomiting, rash and appetite loss [42-45]. Antidepressants, used to treat depression and other psychological disorders adversely affect aquatic and terrestrial organisms by reducing predatory efficiency in fish such as *Carassius*, inducing metabolic acidosis and hypocalcemia in mammals exposed in utero and impairing cognitive function in humans. They also cause hypo locomotion and delayed learning in *Danio rerio*, alter soil nutrient cycling affecting plant growth and may result in acute hepatocellular injury [34,46-49]. However, Analgesics, widely used as pain reliever, induce hepatic and gonadal oxidative stress in fish like *Rhamdia quelen* and cause renal tubular damage in vultures. In humans, they are linked to gastrointestinal, renal and cardiovascular disorders. Moreover, exposure to these drugs leads to kidney alterations and oxidative stress in *Oncorhynchus mykiss*, reduces root and shoot growth and leaf area in *Vigna unguiculata* and disrupts blood coagulation processes [50-56]. Furthermore, Anticancer drugs contribute to oxidative stress and genotoxicity in fish species such as *Oryzias latipes* and *Astyanax lacustris*, suppress photosynthetic efficiency and growth in *Chlorella vulgaris* and act as reproductive toxicants in humans, increasing the risk of lactic acidosis. They also induce metabolic dysfunction through enzyme downregulation in aquatic organisms, reduce plant growth and disrupt β -cell signalling and survival mechanisms [57-60]. Similarly, antifungal agents reduce swimming rate and feeding frequency while increasing mucus secretion in *Daphnia similis*; they also disrupt steroid hormone balance in mammals and cause hepatic, adrenal and gastrointestinal disorders in humans [61-65].

Overall, the ecological consequences of pharmaceutical contaminants are intricate and far reaching, with the potential to disrupt entire ecosystems. Moreover, these compounds may interact with each other or with other environmental pollutants, resulting in additive, antagonistic or synergistic effects that further complicate risk prediction and assessment. Therefore, there is an urgent need to develop and implement sustainable strategies to mitigate pharmaceutical pollution and promote a greener, healthier environment.

Limitations of Current Treatment Methods

Although multiple strategies have been introduced to mitigate pharmaceutical pollution, several major challenges remain. The limited efficiency of current wastewater treatment technologies, inadequate regulatory enforcement and low levels of public awareness continue to restrict the overall effectiveness of existing management approaches. Conventional waste water treatment methods such as coagulation-flocculation, precipitation, evaporation, filtration and adsorption are commonly employed to eliminate colloidal solids, organic nutrients and metallic contaminants from industrial and municipal effluents. Despite their widespread applications, these techniques suffer from several inherent drawbacks that limit their overall efficiency and sustainability. More importantly, conventional methods are largely ineffective in completely degrading PCs, instead, they merely transform or partially remove these compounds without achieving full mineralization [66].

Adsorption and filtration though widely applied, involve high costs and suffer from rapid adsorbent saturation, reducing their long-term efficiency. Electrolysis processes require cost equipment, frequent maintenance limiting their practicality for large scale applications [66,67]. Evaporation on the other hand are energy-intensive and ineffective in removing volatile contaminants, while incineration methods, although capable of destroying organic matter, are expensive and constrained by strict emission control regulations [67-69]. Similarly, coagulation and flocculation processes involve high operational costs and generate large quantities of sludge that require further handling and disposal [66,67].

Furthermore, landfilling, often used for solid PDs waste, poses significant risks of soil and ground water contamination and microwaving, an emerging treatment method, also suffers from high operational costs and uncertain air emissions profiles, raising concerns about its environmental safety and sustainability [69].

All these limitations clearly demonstrate that existing treatment approaches are inadequate for the complete and safe elimination of PCs contaminants from the environment. Therefore, there is urgent need to explore cost-effective and environmentally sustainable solutions.

AOPs: A Sustainable Approach for Green Environment

Advanced Oxidation Processes (AOPs) have been gaining significant attention as potential alternatives for removing

PCs from environment. Initially introduced in 1980s for water purification, AOPs were defined as processes capable of generating hydroxyl radicals ($\bullet\text{OH}$) in sufficient quantities to purify water [70,71]. Over time, their scope has broadened and highly reactive oxidants effectively degrade a wide range of pollutants in air and water [72]. Unlike conventional treatment methods AOPs promote complete degradation and mineralization of contaminants. AOPs particularly function through mechanisms like photocatalysis, Fenton and Fenton like reactions, ozonation, sonochemistry each involving the generation of Reactive Oxygen Species (ROS) that enhances the degradation of PCs in environment [72].

The operating parameters of AOPs vary depending on the specific technique employed. Photocatalysis is influenced by factors such as catalyst type, light source, intensity, reaction time, pH and temperature [73]. Fenton and Fenton like processes depend on catalyst type, persulphate or hydrogen peroxide concentration, pH, temperature, reaction time and initial concentration of pollutant [74]. In ozone based AOPs key parameters include ozone concentration, reaction time, pH, temp and presence of additional oxidants [75]. Similarly, electrochemical oxidation is governed by electrode material, current density, voltage, electrolyte composition, pH, reaction time and initial concentration of pollutant [76]. Finally, sonolysis depends on ultrasound frequency and intensity, reaction time, medium composition, temp, pH and initial concentration [77].

Among all these photocatalysis is recognized as one of the most cost-effective and efficient techniques for degrading and mineralizing PCs in wastewater. This renewable and sustainable approach holds significant promise for addressing environmental remediation challenges [72,78,79]. In a photocatalytic system, the Valence Band (VB) and Conduction Band (CB) are responsible for generating hydroxyl radicals ($\bullet\text{OH}$) and superoxide anions (O_2^-), respectively, which play a vital role in pollutant degradation [72]. It is classified into homogeneous and heterogeneous

types, with the latter being more commonly applied due to its effectiveness. Various chalcogenides act as a key photocatalysts, including metal oxides, metal sulfides. Additionally, non-metal-based materials, particularly carbon-based, are frequently used in solar driven photocatalytic applications [80]. However, conventional metal oxide photocatalysts are limited by their large band gap energies, which restrict light absorption and slow down degradation. To overcome this, modification strategies such as doping in metal/ non-metal and incorporation of nanocomposites have been adopted [72]. These approaches reduce band gap energy and extended light absorption into visible range. However, smaller band gap often causes rapid electron-hole recombination, reducing efficiency [81]. Therefore, advanced modifications are used to enhance charge separation, boost ROS generation and improve overall degradation performance [72] (**Figure 2**). However, Photocatalysis occurs when light energy equal to or greater than semiconductor band gap excites electrons from VB to CB, leaving holes behind. These charge carriers initiate redox reactions where electrons react with oxygen to form superoxide radicals ($\bullet\text{O}_2^-$), which further produce hydroxyl radicals ($\bullet\text{OH}$). Contaminants adsorbed on to catalyst surface are then oxidized into harmless products [82] (**Table 1**).

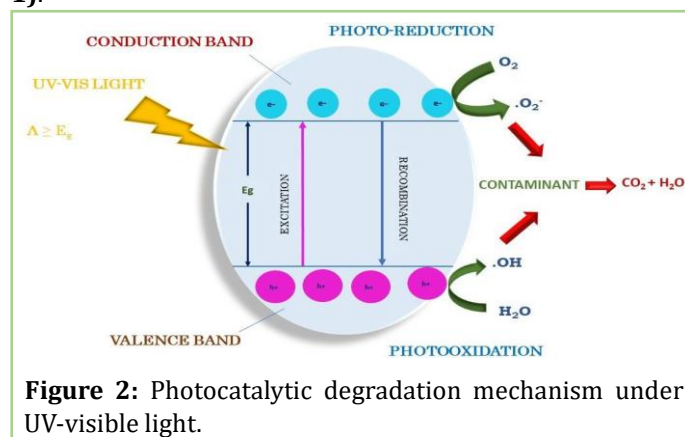


Figure 2: Photocatalytic degradation mechanism under UV-visible light.

Table 1: PDs and the composite materials used for their degradation.

Drug	Material Used	Degradation	References
Tetracycline hydrochloride	TiO ₂ /CdS photocatalyst	53.89%	[83]
Tetracycline hydrochloride	Mn-BPC and Ti-BPC composites	79% and 92%	[84]
Diclofenac	Nitrogen doped Carbon Quantum Dot-Graphitic Carbon Nitride (CNQD)	62%	[85]
Diclofenac	Mn-WO ₃ photocatalyst	100%	[86]
Ibuprofen	Er-doped ZnO nanoparticles	52.30%	[87]
Amoxicillin	TiO ₂ -N, Cu photocatalyst	95.76%	[88]
Ciprofloxacin	Ag-ZnO plasmonic catalyst	82%	[89]
Acetaminophen	g-C ₃ N ₄ /CQD/Ag nanocomposites	85%	[90]

Naproxen	TiO ₂ -La ₂ O ₃ photocatalysts	93.6 and 99.8%	[91]
paracetamol	Au-TiO ₂ nanomaterials	99.17%	[92]

Conclusion and Future Prospects

PCs are persistent and biologically active pollutants that pose significant risk to human health and ecosystem. Conventional wastewater treatment methods often fail to completely remove these compounds, highlighting the need for more effective remediation strategies.

AOPs, particularly photocatalytic degradation, offer a sustainable solution by efficiently mineralizing PCs through ROS while minimizing secondary pollution. However, Future efforts should prioritize the development of cost-effective, stable, photocatalysts, along with thorough assessment of the toxicity and fate of degradation of byproducts.

Combining AOPs with green technologies can enhance scalability and environmental sustainability. Strengthening regulatory frameworks, environmental monitoring and public awareness alongside interdisciplinary research across chemistry, environmental engineering is essential for long-term mitigation of pharmaceutical pollution. Ultimately, integrating innovative treatment technologies with proactive policies offers a promising path toward protecting ecosystems and human health from pharmaceutical contamination.

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