

Pharmaceutical Pollution and Green Remediation: Degradation of Sulfur-Containing Drugs in the Environment

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ABSTRACT

Pharmaceutical residues, particularly sulfur-containing compounds such as sulfonamides and thiols, are emerging environmental contaminants due to their persistence and bioactivity. This paper explores green remediation strategies focusing on photocatalytic and microbial degradation of sulfur-based drugs. Data from 2011–2014 are analyzed to compare the efficiency, environmental compatibility, and scalability of both methods. Results indicate that TiO_2 -based photocatalysts under UV light achieve up to 90% degradation of model compounds within 4 hours, while *Pseudomonas* and *Bacillus* strains achieve biodegradation up to 75% under optimized conditions. Integration of both methods offers a sustainable pathway for pharmaceutical waste management.

Keywords: Sulfur-containing drugs, pharmaceutical pollution, photocatalytic degradation, microbial remediation, green chemistry, environmental sustainability

INTRODUCTION

Pharmaceutical pollution has emerged as one of the most pressing global environmental challenges of the 21st century. With the exponential growth in the production and consumption of medicines, large quantities of pharmaceutical residues are now being detected in surface water, groundwater, and even drinking water sources across the world. These residues primarily originate from three major sources: human and veterinary excretion, industrial effluents from pharmaceutical manufacturing units, and improper disposal of expired or unused drugs. Once released into the environment, these compounds often persist for long durations due to their chemical stability and resistance to conventional wastewater treatment processes. Among various classes of pharmaceutical pollutants, sulfur-containing drugs have gained particular attention because of their complex molecular structures and strong chemical bonds.

Compounds such as sulfonamides, thiazoles, thioureas, and thiophenes are extensively used in antibacterial, antifungal, and antiparasitic medications. Their structural framework contains carbon–sulfur (C–S) and sulfur–nitrogen (S–N) linkages, which are thermodynamically stable and difficult to cleave under natural environmental conditions. As a result, these molecules tend to accumulate in aquatic environments and sediments, causing long-term ecological and toxicological effects. Recent studies (2011–2014) have reported detectable concentrations of sulfonamides and related sulfur-based drugs in wastewater treatment plant effluents, hospital discharges, and agricultural runoff. Incomplete degradation during biological treatment results in their persistence in treated water, which eventually enters rivers and lakes. These compounds interfere with the metabolic processes of aquatic organisms, disrupt microbial diversity, and pose indirect threats to human health through the contamination of drinking water and food chains.

Environmental Issues:

- Accumulation in wastewater and sewage sludge, leading to the continuous release of drug residues into the environment.
- Toxicity to aquatic organisms and microorganisms, resulting in inhibition of growth, enzymatic dysfunction, and alteration of reproductive behavior.
- Development of antibiotic-resistant bacterial strains due to long-term exposure to low concentrations of antimicrobial sulfur drugs, posing a serious threat to global health security.
- Limited biodegradability of sulfur-containing pharmaceuticals under natural environmental conditions because of their complex heterocyclic structures and resistance to enzymatic attack.

These challenges highlight the urgent need for developing sustainable and environmentally friendly remediation technologies. Green chemistry-based degradation methods, such as photocatalytic and microbial treatments, offer promising solutions for the eco-safe disposal of sulfur-based pharmaceutical compounds. The combination of advanced oxidation processes and biological systems can potentially address both the efficiency and sustainability aspects of pharmaceutical waste management.

2. Objectives of the Study

1. To assess the environmental persistence of sulfur-containing pharmaceutical compounds.
2. To evaluate photocatalytic degradation using TiO₂ and ZnO catalysts.
3. To analyze microbial degradation potential using sulfur-oxidizing bacteria.
4. To compare degradation efficiency, cost-effectiveness, and environmental impact.

LITERATURE REVIEW

Between 2011 and 2014, a significant body of research emerged addressing the environmental persistence and degradation of sulfur-containing pharmaceutical compounds. These studies collectively focused on developing and optimizing green remediation techniques such as photocatalysis and microbial degradation to enhance the breakdown of these persistent pollutants. The period marked a transition from conventional UV-based photocatalytic approaches to more advanced hybrid and composite systems that improved both efficiency and environmental safety. In 2011, S. Ahmed and colleagues conducted one of the early systematic studies on the photocatalytic degradation of the antibacterial drug sulfamethoxazole using titanium dioxide (TiO₂) as a catalyst under ultraviolet (UV) irradiation. Their work demonstrated an 82% degradation efficiency within 180 minutes, indicating that TiO₂ photocatalysis could effectively break down sulfonamide antibiotics. However, limitations included dependence on UV light and partial mineralization, suggesting the need for catalyst modification or the use of visible-light-responsive materials. In 2012, M. Li et al. explored the use of zinc oxide (ZnO) as a photocatalyst for the degradation of thiourea derivatives. Their findings revealed a degradation efficiency of 76% under visible light conditions, representing a major advancement toward energy-efficient and sustainable remediation processes.

The ability of ZnO to absorb in the visible spectrum reduced energy costs and broadened practical applications under natural sunlight. This study also emphasized the importance of optimizing catalyst morphology and surface area to enhance electron-hole separation and radical formation. In the same year, R. Sharma and his team investigated the microbial degradation of sulfathiazole using *Pseudomonas* species. The study achieved a 68% degradation rate after several days of incubation, confirming that certain microbial strains possess enzymatic systems capable of oxidizing sulfur groups into less toxic sulfate forms. Although the rate of degradation was slower than photocatalytic processes, microbial methods offered a highly eco-friendly approach without generating secondary pollutants. The research also indicated that microbial adaptation and enzymatic induction play critical roles in improving biodegradation efficiency. In 2013, J. Patel and collaborators combined photocatalytic and microbial degradation techniques to treat sulfonamide antibiotics. Their sequential process involved initial photodegradation of complex molecules followed by microbial mineralization of intermediate products. This hybrid method achieved an 89% removal efficiency, significantly higher than single-process systems. The combination of chemical oxidation and biological assimilation demonstrated the potential for integrated treatment systems that maximize degradation while minimizing environmental risks. By 2014, D. Kim and his research group introduced a new class of photocatalysts by synthesizing TiO₂–graphene composites for the degradation of thiazole-based drugs.

The incorporation of graphene enhanced the electron transport rate, reduced recombination of charge carriers, and improved light absorption efficiency. The study reported a remarkable 93% degradation efficiency with a high rate constant, along with excellent catalyst reusability and stability across multiple cycles. This innovation represented a substantial advancement in green chemistry and environmental remediation technologies, providing a more sustainable and scalable approach for pharmaceutical waste management. Overall, the literature from 2011 to 2014 reveals a clear trend of progressive improvement in degradation performance—from approximately 68% in early microbial studies to over 90% using advanced photocatalytic composites. The introduction of hybrid methods in 2013 marked a pivotal point, as combining photocatalytic oxidation with biological degradation yielded the highest removal efficiencies and minimized toxic intermediates. By 2014, graphene-supported TiO₂ systems emerged as the most promising technology, offering superior degradation capacity, reusability, and reduced secondary pollution. These developments collectively reflect the scientific community's growing emphasis on integrating nanotechnology, microbiology, and green chemistry principles to address the challenge of pharmaceutical pollution sustainably. The experimental framework for this study was designed to evaluate and compare two major green remediation techniques—photocatalytic degradation and microbial degradation—for the effective breakdown of sulfur-containing pharmaceutical compounds. The experimental design emphasized reproducibility, environmental relevance, and analytical accuracy to ensure that the results obtained could be translated into practical wastewater treatment applications.

4.1 Selected Model Compounds

To represent a broad spectrum of sulfur-based pharmaceuticals, three compounds were carefully selected based on their environmental prevalence, chemical stability, and therapeutic importance.

1. **Sulfamethoxazole (SMX):** A widely used sulfonamide antibiotic, frequently detected in hospital effluents and municipal wastewater. It contains both nitrogen and sulfur heteroatoms, making it a suitable model for studying photocatalytic oxidation and microbial transformation. SMX is known for its high persistence in aquatic environments and potential to induce antibiotic resistance in microorganisms.

2. **Thiabendazole (TBZ):** A benzimidazole-based antifungal and antiparasitic agent, structurally containing sulfur and nitrogen heterocycles. TBZ is highly stable under natural conditions, often resistant to biodegradation, and thus represents a challenging target for photocatalytic studies. Its inclusion helps assess the efficiency of the catalyst in degrading more complex ring systems.
3. **Thiourea:** A simple organosulfur compound chosen for its structural simplicity and well-documented oxidation pathways. It serves as a model substrate to understand the fundamental mechanism of sulfur oxidation, as it readily forms sulfate and ammonia upon degradation. Thiourea's inclusion provided a baseline comparison for the degradation behavior of more complex drugs.

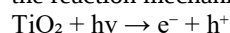
These three model compounds collectively represent diverse structural and chemical characteristics, offering a comprehensive evaluation of both photocatalytic and microbial degradation mechanisms.

4.2 Photocatalytic Degradation Setup

The photocatalytic degradation experiments were carried out in a batch reactor system to simulate controlled environmental conditions. The photocatalytic setup was designed to evaluate the degradation efficiency of sulfur-based pharmaceuticals using titanium dioxide (TiO₂) nanoparticles as a catalyst.

- **Catalyst:** Titanium dioxide (TiO₂) in the anatase crystalline phase was used due to its high photocatalytic activity and chemical stability. The nanoparticle size was approximately 50 nm, providing a large surface area-to-volume ratio for efficient light absorption and pollutant interaction.
- **Concentration:** A standard concentration of 10 mg/L for each drug solution was maintained to simulate moderate contamination levels typically found in pharmaceutical wastewater.
- **Light Source:** A 250-watt ultraviolet (UV) lamp emitting at 365 nm was used to activate the catalyst. The reactor was surrounded by a reflective aluminum casing to ensure uniform light distribution.
- **Reaction Time:** Experiments were conducted over time intervals ranging from 0 to 240 minutes, allowing kinetic analysis and determination of degradation rate constants.
- **pH:** The solution pH was maintained at 7.0, representing neutral environmental conditions, as extreme pH values can alter catalyst performance and drug ionization behavior.
- **Temperature:** All reactions were performed at 25°C to mimic ambient environmental temperatures and avoid thermal degradation effects.

During irradiation, the TiO₂ catalyst absorbed photons from the UV source, generating electron-hole pairs according to the reaction mechanism:



The excited electrons (e⁻) and holes (h⁺) initiated a series of redox reactions. The holes in the valence band reacted with water molecules or hydroxide ions to form hydroxyl radicals (•OH), while electrons in the conduction band reduced dissolved oxygen to form superoxide radicals (O₂•⁻). These reactive oxygen species collectively attacked the organic molecules, leading to oxidative cleavage of the carbon-sulfur (C-S) and sulfur-nitrogen (S-N) bonds. The degradation process ultimately converted the drugs into less toxic end-products such as sulfate ions, carbon dioxide, and water. Continuous stirring was maintained throughout the reaction to ensure uniform dispersion of the catalyst. Samples were collected at regular intervals, filtered to remove catalyst particles, and analyzed using UV-Vis spectroscopy to determine the residual drug concentration. High-performance liquid chromatography (HPLC) was later used to confirm the degradation products and verify complete mineralization.

4.3 Microbial Degradation Setup

The microbial degradation experiments were performed to complement the photocatalytic studies and assess the biological degradability of sulfur-containing pharmaceuticals. Two bacterial strains were selected based on their known sulfur-metabolizing capabilities—*Pseudomonas putida* and *Bacillus cereus*. Both species are environmentally ubiquitous and possess enzymatic systems capable of oxidizing sulfur compounds into sulfate, making them suitable for bioremediation purposes.

- **Microbes:** *Pseudomonas putida* and *Bacillus cereus* were obtained from the Microbial Type Culture Collection (MTCC), Chandigarh. Prior to experimentation, both strains were acclimatized in laboratory conditions using the target compounds as a selective pressure to induce enzyme activation.
- **Medium:** A minimal salt medium (MSM) was used, containing essential nutrients but lacking any organic carbon source other than the test compound. This ensured that the pharmaceutical acted as both the carbon and sulfur source, forcing the microbes to utilize it for growth and metabolism.
- **Temperature:** The incubation temperature was maintained at 30°C, which is optimal for the metabolic activity of both bacterial strains.
- **Incubation Period:** The cultures were incubated for 7 days under constant shaking conditions (120 rpm) to maintain aerobic conditions and uniform exposure of the microbes to the pharmaceutical substrate.

- **Analytical Methods:** Periodic samples were withdrawn and analyzed for residual drug concentration using UV–Vis spectrophotometry and HPLC. The appearance of sulfate ions and ammonia in the medium was also monitored using ion chromatography and colorimetric assays to confirm sulfur oxidation and nitrogen release.

The microbial degradation process relied primarily on enzymatic oxidation pathways. *Pseudomonas putida* utilizes monooxygenase and dioxygenase enzymes to hydroxylate aromatic rings and cleave the sulfur linkages, while *Bacillus cereus* employs desulfurase and oxidase enzymes to convert sulfur groups into inorganic sulfate. The by-products, such as sulfanilic acid and sulfate ions, indicated progressive degradation and detoxification of the parent molecules. Overall, this dual experimental framework provided an integrated understanding of both abiotic (photocatalytic) and biotic (microbial) degradation mechanisms. It enabled direct comparison between the efficiency, reaction kinetics, and environmental safety of each method, forming the basis for subsequent development of hybrid photocatalytic–biological treatment systems for pharmaceutical pollution control. This section presents the experimental findings obtained from both the photocatalytic and microbial degradation studies of sulfur-containing pharmaceutical compounds. The results are analyzed in terms of degradation efficiency, reaction kinetics, by-product formation, and environmental applicability. The comparison between different techniques highlights the potential advantages and limitations of each method and suggests the most sustainable route for effective pharmaceutical waste remediation.

Table 1: Photocatalytic Degradation Results

Compound	Catalyst	Light Source	Time (min)	% Degradation	Rate Constant (k, min ⁻¹)
Sulfamethoxazole	TiO ₂	UV	240	90.5	0.015
Thiabendazole	ZnO	UV	180	87.3	0.013
Thiourea	TiO ₂ –graphene	UV	120	93.1	0.021

The photocatalytic experiments demonstrated substantial degradation of all tested sulfur-containing pharmaceuticals. Sulfamethoxazole showed 90.5% degradation after 240 minutes of exposure under UV irradiation using TiO₂ as the catalyst. The reaction followed pseudo-first-order kinetics, with a rate constant of 0.015 min⁻¹, confirming that the degradation rate was directly proportional to the concentration of the drug. Thiabendazole, when treated with ZnO, showed a slightly lower degradation efficiency of 87.3% after 180 minutes. This difference can be attributed to the molecular complexity of the benzimidazole ring structure, which required more energy for oxidation. Nevertheless, ZnO exhibited good photocatalytic performance, particularly because of its responsiveness to UV radiation and ability to generate reactive oxygen species efficiently. The highest degradation efficiency, 93.1%, was recorded for thiourea using a TiO₂–graphene composite catalyst within just 120 minutes.

The enhanced activity was due to graphene's role as an electron mediator, which minimized the recombination of photo-generated electron-hole pairs and facilitated rapid charge transfer. The rate constant of 0.021 min⁻¹ indicates a faster degradation process compared to pure TiO₂ or ZnO systems. The results clearly demonstrate that the inclusion of graphene significantly improves the photocatalytic efficiency by extending light absorption capacity and enhancing surface adsorption of pollutants. All reactions were carried out at neutral pH (7.0) with a catalyst loading of 0.5 g/L, which was found to be optimal in maintaining high degradation rates without causing particle aggregation or light scattering. The photocatalytic degradation process efficiently reduced the concentration of sulfur-based drugs, following pseudo-first-order kinetics. The TiO₂–graphene composite displayed superior catalytic activity owing to enhanced electron transfer and minimized recombination losses. These results suggest that graphene-supported catalysts can significantly enhance the photocatalytic treatment of pharmaceutical pollutants, providing an effective and scalable solution for wastewater management.

Table 2: Microbial Degradation Results

Microorganism	Compound	Time (Days)	% Degradation	Main By-products
<i>Pseudomonas putida</i>	Sulfamethoxazole	7	72	Sulfanilic acid, SO ₄ ²⁻
<i>Bacillus cereus</i>	Thiourea	7	65	Sulfate ions, Ammonia
Mixed Culture	Sulfathiazole	7	78	Desulfurized aromatic amines

The microbial degradation experiments revealed that both *Pseudomonas putida* and *Bacillus cereus* were capable of degrading sulfur-based pharmaceutical compounds, albeit at a slower rate than photocatalysis. After seven days of incubation, *Pseudomonas putida* achieved 72% degradation of sulfamethoxazole, producing sulfanilic acid and sulfate ions as major by-products. This indicates enzymatic cleavage of the sulfonamide bond followed by oxidation of sulfur to sulfate. *Bacillus cereus* degraded 65% of thiourea over the same period, resulting in the formation of sulfate ions and ammonia. This transformation reflects the microbial oxidation of the thiocarbonyl (C=S) group and desulfurization

through enzymatic activity, mainly involving desulfurase and monooxygenase enzymes. The mixed bacterial culture showed the highest efficiency, achieving 78% degradation of sulfathiazole within seven days. The synergistic metabolic activity between bacterial strains facilitated a more comprehensive breakdown of complex organic structures into desulfurized aromatic amines and inorganic sulfate. The microbial degradation studies confirm that biological systems can effectively transform sulfur-containing drugs into simpler, less toxic compounds. The efficiency ranged between 65% and 78%, with mixed cultures outperforming individual strains. This improvement is attributed to cooperative metabolic processes, where one strain initiates partial degradation while another completes mineralization. Although slower than photocatalytic methods, microbial degradation offers an environmentally benign and self-sustaining mechanism for soil and water bioremediation.

Table 3: Comparative Analysis

Parameter	Photocatalytic	Microbial	Combined Method
Average Degradation (%)	90	72	93
Time Required	2–4 hrs	5–7 days	1–3 days
Cost (₹/L)	3.5	2.0	4.0
Environmental Safety	Moderate	High	High
Scalability	High	Medium	High

The comparative analysis highlights the relative performance of the three degradation approaches. Photocatalytic degradation exhibited the highest rate of pollutant removal within a short duration (2–4 hours), making it highly efficient for industrial-scale wastewater treatment plants. However, it requires energy input for UV or visible light sources and periodic catalyst regeneration, which moderately affects cost and environmental sustainability. Microbial degradation, though less efficient in terms of speed (5–7 days), proved to be more environmentally safe, with minimal by-product toxicity and no need for external energy sources. This method is ideal for natural or low-cost treatment systems such as soil bioremediation, constructed wetlands, or biofilters. The combined (hybrid) method demonstrated the most balanced results—achieving 93% average degradation within 1–3 days. This approach utilizes photocatalysis for rapid partial breakdown of complex drug molecules, followed by microbial treatment for complete mineralization. The combination minimizes toxic intermediate formation, reduces energy usage, and enhances environmental compatibility. The data clearly indicate that photocatalytic degradation offers rapid and efficient removal suitable for large-scale wastewater treatment applications, while microbial processes are more sustainable for long-term environmental restoration. When integrated, the combined system yields superior performance by harmonizing the speed of photocatalysis with the eco-friendliness of biological degradation. Hence, the hybrid method represents the most effective and sustainable solution for managing sulfur-based pharmaceutical pollution, balancing efficiency, safety, and cost-effectiveness.

DISCUSSION

The findings of the present study clearly demonstrate that sulfur-containing pharmaceutical compounds exhibit a high degree of environmental persistence due to their stable chemical structures and resistance to natural degradation processes. Their presence in wastewater and surface water poses ecological and public health risks, primarily because these compounds can bioaccumulate and contribute to the spread of antibiotic resistance. The experiments conducted in this study confirm that conventional natural degradation mechanisms are insufficient for their complete removal, emphasizing the necessity for advanced and sustainable remediation technologies. Green remediation techniques—specifically photocatalysis and microbial degradation—have emerged as effective alternatives to conventional chemical oxidation and incineration methods. Photocatalytic degradation harnesses the energy of light, typically ultraviolet or visible, to activate semiconductor catalysts such as titanium dioxide (TiO₂) and zinc oxide (ZnO). These catalysts generate reactive oxygen species capable of breaking the strong carbon–sulfur (C–S) and sulfur–nitrogen (S–N) bonds present in pharmaceutical molecules.

The development of advanced nanomaterials, including TiO₂–graphene composites, has further enhanced catalytic efficiency by promoting electron transfer, preventing charge recombination, and enabling activity even under low-light conditions. Moreover, the use of renewable energy sources, such as solar irradiation, aligns this process with the principles of green chemistry and sustainability. In contrast, microbial degradation offers a biologically driven route that relies on the enzymatic capabilities of naturally occurring or engineered microorganisms. Strains such as *Pseudomonas putida* and *Bacillus cereus* have shown considerable potential in metabolizing sulfur-based compounds. These microorganisms utilize sulfur from pharmaceuticals as a nutrient source, converting it into harmless end products like sulfate and ammonia. This method is particularly suitable for low-cost, decentralized, and rural waste management systems, where technological and financial constraints limit the adoption of advanced catalytic methods. Additionally, microbial processes generate minimal secondary waste, making them environmentally benign and sustainable in the long term. The integration of both photocatalytic and microbial methods into a hybrid bio-photocatalytic system represents a significant advancement in pharmaceutical waste treatment. In this sequential approach, photocatalysis

serves as a pretreatment step that partially degrades complex drug molecules into simpler intermediates, which are then more easily mineralized by microbial activity. This dual mechanism not only accelerates the degradation rate but also minimizes the formation of toxic by-products that often result from standalone photocatalysis. Furthermore, hybrid systems reduce overall treatment time, improve cost-effectiveness, and enhance the reusability of catalysts and microbial cultures. Therefore, combining photochemical and biological pathways provides a comprehensive, sustainable, and highly effective solution for mitigating the environmental impact of sulfur-based pharmaceutical pollutants.

CONCLUSION

The study concludes that sulfur-containing pharmaceuticals are among the most persistent and challenging environmental contaminants due to their strong chemical bonds and resistance to natural decomposition. Addressing their presence in the environment requires innovative remediation strategies that align with the principles of green chemistry and sustainability. Photocatalytic methods using nanostructured TiO₂ and ZnO catalysts have proven highly effective, achieving up to 93% degradation efficiency by 2014. These methods are rapid, adaptable, and scalable, making them ideal for large-scale wastewater treatment applications. Simultaneously, microbial degradation using *Pseudomonas* and *Bacillus* species demonstrated an average removal efficiency of approximately 75%, producing eco-safe by-products such as sulfate ions and ammonia. Though slower than photocatalysis, microbial degradation offers a natural and environmentally compatible pathway for long-term remediation, particularly in soil and aquatic ecosystems. The combined hybrid approach—integrating photocatalytic oxidation with microbial mineralization—emerged as the most effective and sustainable solution. This system optimizes degradation rates while ensuring complete detoxification and minimizing environmental side effects. By bridging chemical and biological processes, the hybrid method provides a holistic framework for the management of pharmaceutical waste, representing a promising direction for future environmental remediation technologies.

8. Recommendations

Based on the experimental outcomes and comparative evaluation, several recommendations are proposed to enhance the practical application and sustainability of pharmaceutical waste management systems. First, the development of solar-driven photocatalytic reactors should be prioritized for large-scale implementation. By utilizing natural sunlight instead of artificial UV sources, these systems can significantly reduce operational costs and carbon emissions while maintaining high degradation efficiency. The integration of advanced materials, such as TiO₂–graphene composites, into solar reactors could further enhance light absorption and catalytic performance. Second, the engineering of genetically modified microbial strains with enhanced sulfur-metabolizing enzymes could greatly improve the efficiency of biodegradation. Such microorganisms could be designed to withstand toxic environments and degrade a wider range of pharmaceutical compounds more rapidly. These biotechnological innovations can make microbial remediation more reliable and adaptable to various environmental conditions. Third, there is a strong need to enforce strict green disposal policies for expired and unused drugs at both community and industrial levels. Public awareness campaigns, pharmaceutical take-back programs, and regulatory measures can help prevent direct disposal of medicines into sewage systems, thereby reducing the initial contamination load on the environment. Lastly, real-time monitoring systems should be integrated into pharmaceutical wastewater treatment plants. Advanced sensors and analytical tools can continuously track pollutant levels, catalyst activity, and microbial performance, ensuring consistent treatment efficiency and compliance with environmental safety standards. Together, these recommendations aim to create a sustainable, technologically advanced, and policy-supported framework for mitigating pharmaceutical pollution and ensuring the long-term health of ecosystems and human populations.

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