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Integration of methodologies for the sanitation and treatment of wastewater contaminated by pharmaceuticals

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ABSTRACT

Introduction. The research aims to analyse the present physical, chemical, and combined methods and practices used to extract pharmaceuticals (PC) from wastewater (WW) starting from different sources, such as municipal waste and hospital release, emphasizing PC manufacturing companies. PC contaminants are primarily persistent organic chemicals not readily eliminated by standard WW treatment (WWT) procedures.

Materials and methods. The research examined suggests that enhanced oxidation methods can destroy these persistent medicines. The oxidation introduces harmful oxidation products if these procedures are not carefully controlled. Physical processes, including adsorption of carbon and membrane filtering, can give an obstacle that inhibits both parent substances and harmful products from flowing into treated effluent.

Results. A combination of multiple procedures can be an appropriate treatment plan for the persistence and degrading of both parent and conversion chemicals. The benefits of the procedures are integrated through combined technology, resulting in a maximization of pollutant cancellation. Sophisticated oxidation manipulation, either pre-treatment or post-treatment, paired with a natural adsorption or filtering method, is a promising approach.

Limitations. However, the best procedures for PCs-containing WW depend on the quality and amount of WW, the PC compound leftovers, and their dangerous consequences.

Conclusion. This research underscores the importance of combining enhanced oxidation methods with physical processes like adsorption and membrane filtration to effectively extract PC from WW. While these integrated approaches show promise in degrading contaminants, their success depends on the specific characteristics of the WW and the PC present. Continued exploration and refinement of these methods are essential for addressing PC pollution comprehensively. Future studies should focus on optimizing these strategies across varied WW contexts.

Keywords: Sanitation; Wastewater; pharmaceuticals; Review

Compliance with Ethical Standards. The research adheres to ethical guidelines as set forth by the relevant authorities. All procedures involving human or animal subjects were approved by the appropriate ethics committee, and all necessary consent forms were obtained.

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Внедрение методов очистки сточных вод при их загрязнении фармацевтическими отходами

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РЕЗЮМЕ

Введение. Целью исследования является анализ современных физических, химических и комбинированных методов и способов очистки сточных вод (СВ) от фармацевтических отходов (ФО), преимущественно коммунальных и медицинских, с уточнением компаний, производящих фармацевтические препараты. Фармацевтические отходы представляют собой преимущественно стойкие органические соединения, которые не поддаются удалению стандартными способами.

Материалы и методы. Предполагается, что разложение стойких фармацевтических отходов возможно с помощью высокоэффективного окисления. Однако при отсутствии направленного контроля возникает риск образования вредных продуктов окисления. Физические методы, в том числе, адсорбция углеродом и мембранная фильтрация, могут предотвращать загрязнение обработанных сточных вод ФО и вредными продуктами их окисления.

Результаты. При очистке от стойких ФО и разложении исходных и подвергнутых окислению соединений может быть эффективным комбинирование различных методов. В этом случае взаимное дополнение преимуществ разных методов позволяет добиться максимальной очистки воды от загрязняющих веществ. Перспективным подходом можно считать сочетание эффективных способов предварительного и последующего окисления и методов естественной адсорбции или фильтрации.

Ограничения исследования. Эффективность процедуры очистки сточных вод, загрязнённых ФО, зависит от качества и количества сточных вод, остаточного количества ФО и их вредных производных.

Заключение. Проведённое исследование подтверждает, что для надёжной очистки сточных вод от ФО необходимо применение методов эффективного окисления в сочетании с такими физическими процессами, как адсорбция и мембранная фильтрация. Несмотря на многообещающие результаты комплексных подходов к разложению загрязняющих веществ, успех реализации зависит от конкретных характеристик сточных вод и присутствующих в них ФО. Продолжение изучения и совершенствование этих методов имеет большое значение для комплексного решения задачи

очистки вод от загрязнения ФО. Будущие исследования должны быть сосредоточены на оптимизации этих стратегий применительно к разным источникам сточных вод.

Ключевые слова: методы очистки; сточные воды; фармацевтические отходы; обзор

Соблюдение этических стандартов. Исследование соответствует этическим нормам, установленным соответствующими органами. Все процедуры с участием людей или животных были одобрены соответствующим этическим комитетом, получены все необходимые формы согласия.

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Конфликт интересов. Авторы декларируют отсутствие явных и потенциальных конфликтов интересов в связи с публикацией данной статьи.

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Introduction

Pollution of the aquatic ecosystem with drugs is a critical developing problem. Pharmaceutical (PC) [1] contaminants can make it into this ecosystem through sewage discharged from residences, farmlands, clinics, and industry, with the inability of wastewater (WW) [2] treatments (WWT) [3] to eradicate them cited as a critical contributory cause. PCs can penetrate the natural setting owing to human activity. More than 600 reactive PC compounds have been detected globally in numerous water bodies, including water for consumption. While their occurrence happens in low quantities, various studies have revealed that PC-active compounds – can pose a substantial danger to the natural world and negatively impact organisms.

The PC sector can damage lakes and rivers with many dangerous contaminants and has a considerable water footprint. The water purification techniques necessary to remove the leftover PCs from WW released to acceptable limits can end in a higher remediation price [4]. This can be a concern in bulk medicine production plants in underdeveloped nations. For instance, research indicated a significant number of antimicrobial agents in drinking water emanating from production centres of PCs in nearby areas. The amount of fluoroquinolone in Musi River exceeded 5000 mg/L, whereas the amounts detected in WW samples across the planet were less than one mg/L [5].

The production of medications releases toxic, physiologically active, and possibly permanent organic chemicals into water solutions. These might involve antimicrobial agents, medicines, painkillers, and hormone-disrupting chemicals. Some can be easily eliminated from the water stage using traditional procedures, such as the method involving activated waste [6].

This study's fundamental goal is to collect and analyse the PC elimination treatment options, including techniques for identifying and eliminating PC compounds from water sources. The data gathered in this study offered a simply understandable notion in only one release, enabling swift knowledge of the existing position of the drug detection domains. This review outlines recent advancements in natural and sophisticated oxidation approaches for removing PCs from lakes and rivers. The PC adsorption characteristics of natural and sophisticated oxidation methods and their value in PC concentration elimination are explored in this paper.

Materials and methods

PC items are essential for enhancing human health. It is typically designated as PCs or narcotics. Humans and animals utilize drugs to improve health and address illnesses and disorders. The presence of PC goods has emerged as a global concern due to inadequate waste disposal practices [7].

Figure 1 depicts the mechanisms via which PC compounds infiltrate the environment. In more than 70 nations worldwide, about 630 PC drugs or their derivatives have been identified. PC materials reach the environment through media, including WWT plants, landfills, hospitals, cemeteries, and PC manufacturing factories. The amalgamation of many elements complicates the removal of drugs from aquatic environments. Humanity will perpetually suffer the consequences of its activities if a resolution to PC waste in the water is not achieved [8]. Figure 2 illustrates the origin of PC pollution in the environment.

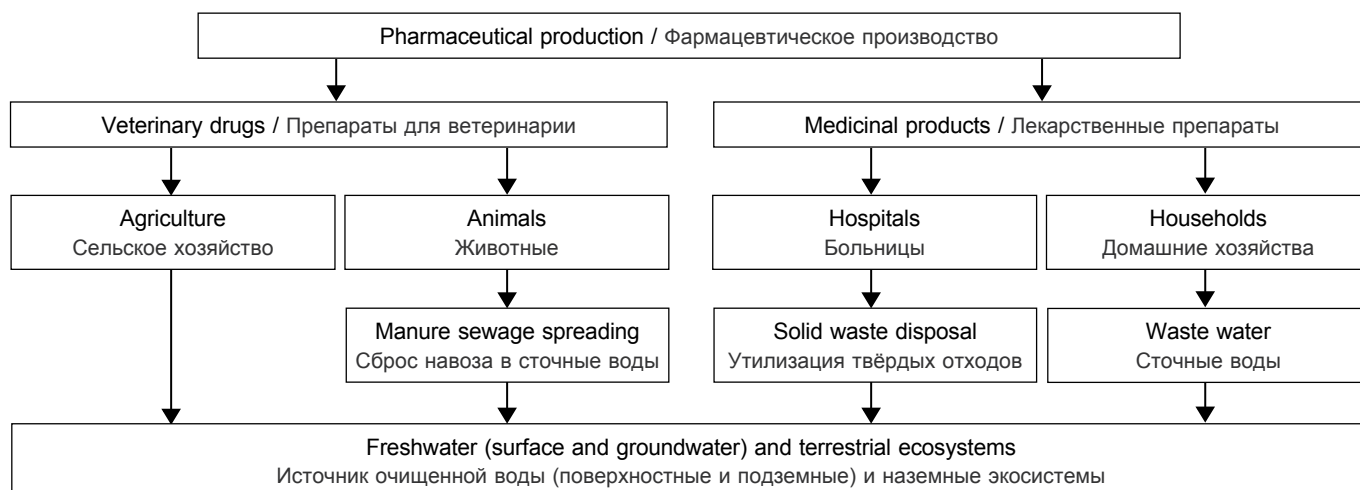


Fig. 1. Sources of PC elements.

Рис. 1. Источники фармацевтических отходов (ФО).

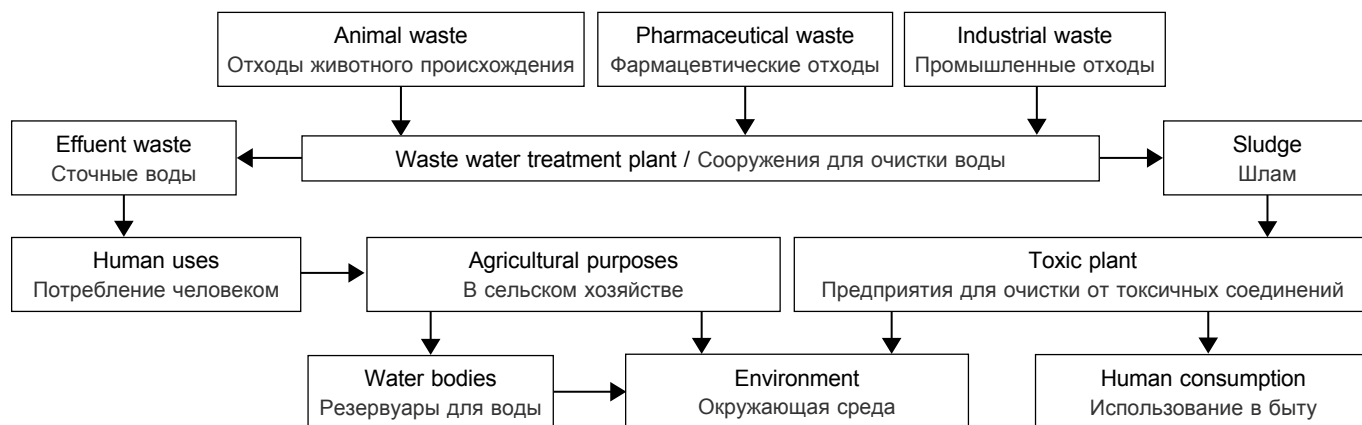


Fig. 2. Sources of C Pollution.

Рис. 2. Источники загрязнения ФО.

Sources of human PCs. Human PCs in the atmosphere and water pose a significant threat to creatures of all kinds. These PCs are mainly obtained from residences, hospitals, clinics, and medical establishments, and they are released into the water cycle, posing a threat to aquatic organisms and people. The status of this source of income fluctuates over the year or remains consistent for households with elderly individuals, who are more prone to use many prescriptions or over-the-counter drugs due to their socioeconomic status or living arrangements [9]. Humans can release medications or trash into the environment via urine or excrement. Expired or unused PCs discarded in household waste ultimately contribute to leachate from landfills at WWT plants before entering the environment. The role of various medical compounds in aquatic environments indicates that human waste products and urine account for a substantial portion, perhaps reaching 90%. The production and management of medication waste are rigorously controlled among producers, sellers, retailers, and healthcare facilities.

Although some accounts indicate that health institutions dispose of unneeded prescriptions and PC waste down drains, contamination from these sources is considered unlikely. The channels via which humans excrete drugs via urine and feces are more consequential. These drugs infiltrate aquatic ecosystems, water to consume, water on the surface, and groundwater due to inadequate disposal during WWT [10]. Disinfecting agents and antibiotics are thought to modify the WWT method and the microbe community of the water's surfaces. The suggested standard of care is for patients to give any unwanted medicine to the pharmacist, who would discard it; anyone is not responsible for the disposal of unneeded PCs.

Sources of animal PCs. Animal PCs reach the environment through numerous sources, including farm animals and cultivation. Veterinary antibiotics are employed globally to treat illnesses and maintain the wellness of animals [11]. Antibiotics are included in animal feed to enhance growth rates, improve feed effectiveness, and serve chemotherapeutic and preventive functions. Due to the inadequate absorption of antibiotics in the stomachs of animals, most of it is excreted unaltered in feces and urine. Considering the widespread use of animal feces as a fertilizer in several nations, there is increasing global apprehension over the potential environmental repercussions of antibiotic leftovers. Accurate information regarding the quantity of livestock PCs, particularly antimicrobials, that enter the soil must be included [12]. PCs published into the atmosphere from livestock farming activities are considered significant sources, primarily from leaks in anaerobic manure storage lakes, discharges from aquaculture activities, manure reproduction on farm fields, runoff from agricultural fields, and airborne dust. The predicted quantity of antibiotics farm animals generate annually totals thousands of tons.

Results

Traditional WWT techniques target micropollutants, comprising suspended particles and organic pollutants such as carbon, nitrogen, phosphorus, and viruses. They are inadequate for efficiently removing micropollutants, such as persistent PC chemicals. In conventional systems, the efficacy of eliminating these substances fluctuates significantly, ranging from ineffective to total removal, contingent upon the substances, their characteristics

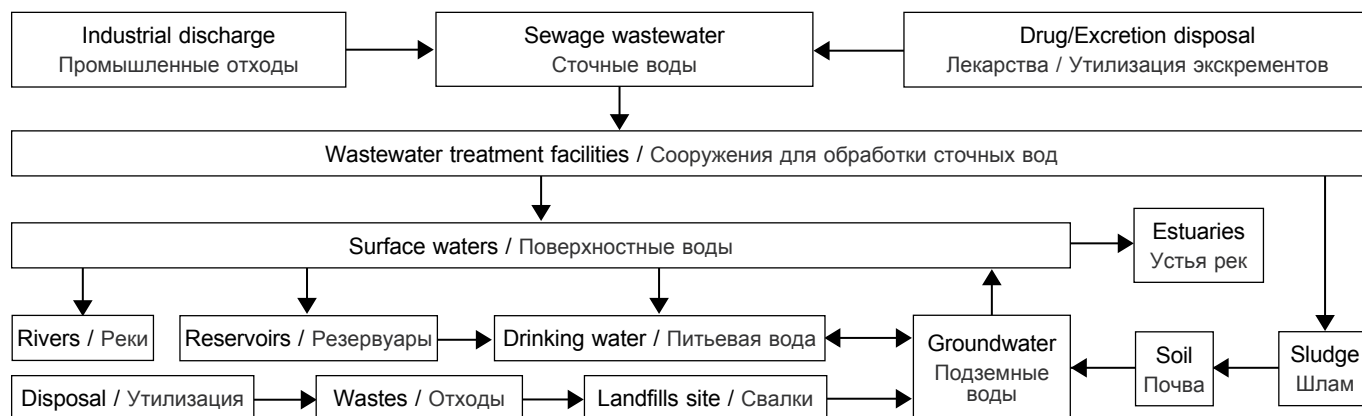


Fig. 3. Workflow of PC contamination into water sources.

Рис. 3. Источники загрязнения водных источников ФО.

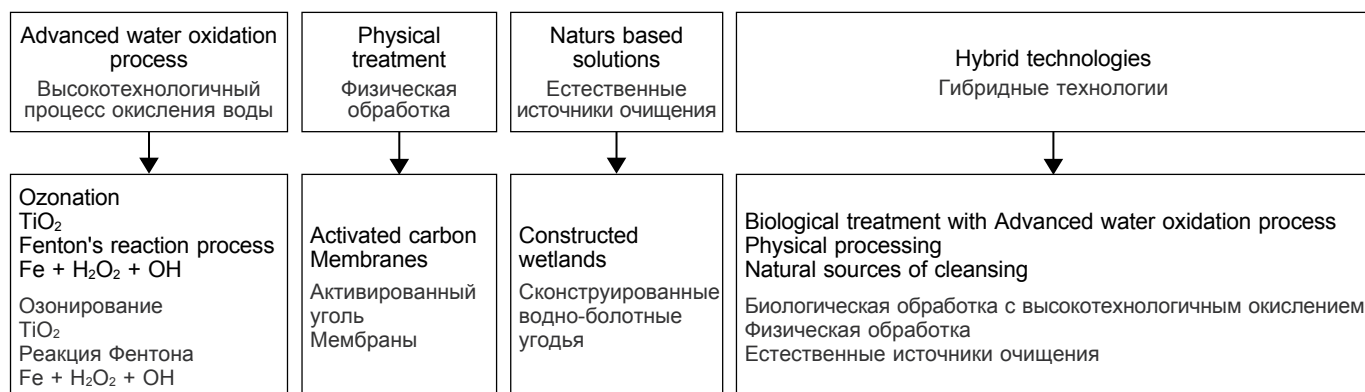


Fig. 4. Treatment system classifications.

Рис. 4. Классификация систем очистки.

(size, focus, category, polarity), and the surroundings and operational parameters of the remediation process. Traditional WW management exposes medicines to various physicochemical and biological methods, including mineralization, variation, diluting, sunlight, and vaporization [13]. The primary removal processes are breakdown, biotransformation by microbes, and absorption into biological solids.

Numerous PC compounds identified in locations, with the most significant amounts observed near WWT plant outfalls [14]. This suggests that the chemicals endure traditional treatment methods, and WWT plants represent a primary pathway for their introduction into the aquatic ecosystem. Figure 3 illustrates the potential pathways through which PMs infiltrate aquatic environments (both surface and underground) and soil, stemming from manufacturing and hospital effluents, the excretion of PCs and their byproducts from individuals or veterinary sources, and the elimination of unoccupied or expired drugs [15].

This study examines contemporary chemical, biological, and nature-based approaches employed in water purification to eliminate PC pollutants, either independently or as pre- or after other techniques. Other significant methods utilized in WWT, such as traditional biological processes, are beyond the purview of this analysis. The article examines prevalent Advanced Oxidation Procedures (AOPs) [16], encompassing the ozonation and photocatalysis facilitated by catalysts (TiO_2) [17]. It addresses therapies, such as absorbing and activating carbons, and separating techniques, such as reverse osmosis. It then explores nature-based remedies and concludes with a discussion on hybrid approaches that integrate traditional and sophisticated treatment methods (Figure 4).

AOP

Ozonation. It is a procedure that infuses ozone into water, often achieved by bubbling elements through the lower of a reservoir [18]. It exerts oxidative impacts by direct interactions with ozone or via radical responses in a series of oxidation processes. It is a potent oxidizing agent for specific chemical molecules. In the absence of liquid, it can combine with ionized to generate the hydroxyl radicals, a less discriminating but more potent agent.

A significant concern with using ozonation in PC effluent is the generation of by-products, which often exhibit more toxicity than the original chemicals. The processes involved in ozonation are influenced by pH, temperatures, and ozone dosage; hence, optimizing them is essential for generating end-products. The standard ozone concentrations utilized in managing WW often do not lead to mineralization, so the danger momentarily escalates during ozonation.

Photocatalysis. It constitutes an additional sophisticated oxidation mechanism [19]. This behaves similarly to ozonation since photocatalysis works via the generation of hydroxyl radicals. In contrast to ozonation, hydroxyl radicals are generated by colliding water molecules with the lighted catalyst's surfaces.

Rutile TiO_2 is the prevalent catalyst employed in PC photocatalytic research owing to its biochemical and biological inertness, high photoactivity, cost-efficiency, and safety for both the natural world and people.

In addition to matrix consequences that restrict the use of photocatalysis, the efficacy of heterogeneous photocatalysis is contingent on the proportion of quantity to the starting amount of target molecules and ambient factors. A load below the optimal amount results in restricted production, thereby leading to inadequate elimination of the target chemicals. An elevated amount increases solution viscosity, reducing radiation penetration. The primary challenge of this method is the limited absorption spectrum and that of natural daylight.

Fenton reaction processes. It is widely employed to break down medicines [20]. The Fenton oxidation process entails the interaction of peroxide from hydrogen (H_2O_2) with the iron ions (Fe^{+2}) through a free radical chain response, generating hydroxyl agents.

Several operational factors, including the concentrations of H_2O_2 and Fe^{+2} , pH, and the composition of the WW substrate, influence the efficacy of the conventional procedures. The ideal beginning pH range for reactions is around 2 and 4, within which the maximal generation made from hydroxyl occurs, and iron deposition is prevented. Iron complexes mitigate this limitation, facilitating the process at near-neutral temperatures. Many treatment facilities, including PCs in WWT vegetation, generate acidic pollutants, which possess the optimal pH for Fenton processes without modification.

The primary disadvantages of the Fenton procedures are generating hazardous elements from the oxidization of dispersed material, pH sensitivity, development of iron WW, and using energy. Creating toxic substances is reduced by implementing ideal reagent dosage and reaction duration operating settings [21]. At the same time, complete oxidant reduction is essential to improve the removal of organic substances and prevent increased toxicity. The Fenton reagent possesses an oxidizing and coagulating function by creating ferric hydroxy combinations. It can replace the coagulation procedure in the initial process, resulting in diminished sludge development, and act as a polishing stage by eliminating residual pollutants following Fenton's oxidative stress.

Physical methods

Activated carbon removal. It is a transition method wherein chemicals are transported from a single stage (aqueous) to a different one (solid absorption) [22]. Activated Carbon (AC) is an appropriate substance and the most often utilized adsorbent, owing to its substantial ability to adsorb, ascribed to its elevated pores, extended region, and a significant degree of exterior contact. Additional materials, like carbon nanotubes and biochars, are widely utilized. The adsorption properties are contingent upon the origin of the ingredients utilized in their production and the procedure employed.

Research on concurrent adsorption has indicated that other medications, present as a combination in WW and various inorganic and organic compounds, negatively impact the procedure's efficacy. Regarding the elimination of PCs, drug adsorption onto activated carbon is an appropriate and economical alternative for PC in WW, contingent upon the particular substance or mixture targeted for elimination and the operational circumstances, provided that pre-treatment has occurred and particles reduced.

Membrane procedures. Reversed osmosis utilizes pressure to surpass the inherent osmotic pressure across the surface of a reversed membrane and the other end [23]. The main application of reversed osmosis is converting salt or salty water into potable water. It generates purified water from inferior-quality water supplies. The supply water has usually undergone many processing operations to enhance the performance and longevity of reverse osmosis membranes. Techniques such as quick or gradual sand filtering and powdery or AC adsorption eliminate chemicals contributing to membranes' fouling and degradation. These substances encompass microbes, extrinsic polymeric chemicals, substances, particles, and non-biological organic molecules.

As shown in the preceding section on photosynthesis, the WW generated by a PC manufacturing facility, contingent upon the goods generated, is likely to possess significantly lower concentrations of living matter compared to other types of WW, such as household WW. The application of reverse osmosis for treating PC WW is more advantageous than other kinds of WW, as the likelihood of membrane fouling from biological growth is diminished due to the lower number of organisms and additional polymer compounds they generate. Fouling is caused not only by biological factors but also by suspended and other non-biological chemicals. Implementing preliminary therapies before reverse osmosis is prudent to guarantee optimal efficiency.

Nature-based answers

Constructed Wetlands (CWs) are a price-effective and environmentally sustainable method for treating WW [24]. A CW is a compact habitat that emulates natural wetland systems, facilitating the proliferation of creatures and native flora and enabling various physicochemical processes for WWT. CWs are categorized into three categories based on the characteristics of the flow dynamics inside the CW. The primary removal methods in constructed wetlands include biodegradation, sorption to organic materials, phytoremediation, hydrolysis, and photosynthesis.

Researchers revealed mixed results in evaluating the effectiveness of one underground flow CW and surface flow CW. The input WW of the two surface flow CW comprised a combination of household and industrial/hospital WW, having undergone pre-treatment via an activating sludge method with sand filtration and an oxidization ditch. The overall bacterial count rose in all examined constructed wetlands. Regarding the total microbes, most antibiotic resistance genes exhibited stability or a decline following constructed wetland treatment.

Other investigators have reported positive reductions in Antibiotic Resistance Genes (ARGs) [25]. Several studies indicate that CWs facilitate the dissemination of antibiotic-resistant genes, owing to prolonged usage of antibiotics that leads to the accumulation of abundant ARGs, which tend to increase with period. The findings underscore the variability of ARGs and indicate that the efficacy of elimination is significantly influenced by several aspects, including flow framework, age, category, Hydrodynamic Retention Terms (HRT), seasons, influent motion, and the state of the water.

Hybrid technologies

Hybrid techniques integrate two or more traditional or innovative approaches to treatment to provide optimal or total elimination of microorganisms or recalcitrant chemicals. The necessity for hybrid technologies stems from the inefficacy of individual treatment methods, whether biological or AOPs, in substantially eliminating all parent chemicals and their transformation byproducts. The breakdown and calcification of some persistent contaminants can be augmented by integrating several processes, leveraging mutually beneficial effects and the

benefits of different approaches. Several innovative and hybrid therapy protocols have been explored. Studies utilized a lab-scale modelling reactor to eliminate 18 antimicrobial agents and 10 ARGs from actual medical waste concurrently.

The prevalence of all ARGs escalated, suggesting that mechanisms facilitate the proliferation of opposition. ARGs were predominantly eliminated by AOPs, with the Fenton/UV mixture proving to be the most productive for eliminating both rRNA and ARGs. UV and ozonation exhibited only marginal clearance capabilities.

Statutory guidelines for PC wastes

Water is a vital resource for fundamental existence and the progression of national socioeconomic prosperity. Given the escalating water crisis, it is imperative to implement water-use regulations to address increasing water shortages. The EU Decision identified forty-five priority chemicals related to ecological quality requirements and water regulations. Ten chemicals were designated for tracking on a watch list within the realm of water policy. Health officials might enforce reducing waste by promoting awareness, providing guidance on waste reduction methods, and fostering collaboration and partnerships. Possibilities for waste reduction techniques differ significantly among nations. In many countries, regulatory rules or their absence hinder the extensive use of waste-reduction methods. In some countries, the re-dispensing of medicine is constrained by the lack of clinical standards or legislation.

Degradation of PC by AOP

The degradation route is crucial for comprehending the mechanism of PC breakdown by AOPs. AOPs are technical processes that produce hydroxyl radicals to effectively oxidize dangerous byproducts from industrial sources, organic pollution streams, healthcare WW, and trash from municipalities. The primary process produces free hydroxyl radicals and other reactive compounds, including peroxide from hydrogen and superoxide anions. The breakdown of goods, particularly in the PC sector, necessitates utilizing this AOP's energy to attain optimal reaction effectiveness. Mass Spectrometry (MS) was utilized to detect the breakdown of products. Regarding the recognition and measurement in WW, Tandem mass chromatography is the predominant method employed to analyse antimicrobial agents, anti-inflammatory medications, and antifungal agents, among others. The Electro-Fenton (EF) technique was used to examine the breakdown pathway of enoxacin, revealing four kinds of metabolites employing spectroscopy analysis.

Analysis of propyphenazone using tandem MS demonstrated optimal Limits of Detections (LOD) and Quantifications (LOQ) at 0.04 and 0.06 ng/L, respectively. The sulfamethazine test utilized a high-performance liquids chromatography-tandem MS. The inherent characteristics of several antibiotics, such as sulfonamides, amide ethanol, lactams, tetracyclines, and chemicals, need ring cleavage for their degradation by electrochemical-enhanced oxidation processes, forming multiple intermediates.

Discussion

Scientific and technical breakthroughs have led to various illnesses that have significantly harmed both human and aquatic organisms. Releasing industrial effluents into the environment has led to several prevalent waterborne illnesses, necessitating urgent remedial action. Academic groups have addressed these difficulties by advancing WWT from traditional to contemporary techniques. All these strategies seek to diminish the concentration of PC pollutants in the surrounding area. Scientists have utilized an integrated approach to address water pollution, even though contaminants are converted from one form to another during treatments. The complete removal and breakdown of these pollutants have yet to be accomplished correctly and effectively. Developing a successful and effective adsorbent in conjunction with a separation approach is necessary to enhance results and address these shortcomings in the future.

Despite using adsorption and sophisticated oxidation procedures independently to eliminate PC contaminants in aquatic environments, these methods have yet to eradicate such substances. The following study should integrate sophisticated oxidation and adsorption mechanisms to eradicate medicinal compounds, even in minimal quantities. Future research on biological processes should focus on optimal conditions and microbial mechanisms that facilitate the concurrent elimination of medicinal chemicals and the impact of various habitats on removal efficiency. It is essential to examine the synergistic removal mechanisms of medications, genetic resistance, and traditional pollutants using innovative CW treatment methods. Future investigation is required regarding the elimination of gene resistance, the efficacy of functional materials infused with pharmacologic gene-resistance inhibiting agents, their superior adsorption properties, and their beneficial effects on microbial growth and activities. The current study primarily addresses the small test size; future research should concentrate on industrial size and evaluate operational circumstances alongside indoor simulations. A prediction model for various operational factors should be developed to assess and enhance artificial wetland circumstances in conjunction with actual requirements.

Conclusion

The presence of PCs in aquatic environments is a pervasive challenge that is receiving more scrutiny owing to its healthcare risks and harmful impacts. Pollutants arise through several pathways, with the PC sector being a significant source. Industrial WW exhibits variability in quality and quantity, influenced by diverse PC substances, contaminant concentrations, WW matrices, volumes, and the initial supplies and techniques employed in

manufacturing procedures. Providing a precise treatment protocol for such a varied business is challenging.

Following the treatment operations, although the elimination of PCs is substantial, total mineralization still needs to be achieved. Estimating the residual levels of PCs is crucial, as they have detrimental effects upon disposal or reuse. Alongside the quality of the effluent, treatment expenses must be considered in the construction of these treatment facilities since these methods must be feasible for pertinent businesses and economically accessible for low-income nations.

Due to the long-lasting effects of many medications and their limited biodegradability, more than a single treatment technique is required for total eradication. Physical procedures, such as desorption and membrane techniques, offer excellent effectiveness for removal and brief treatment time. These phase-changing methods generate focused retentate water, necessitating extra treatment before elimination due to elevated levels of persistent pollutants. One must consider the complexities of the procedure, the expenses associated with adsorbent planning, regrowth, destruction, and the operational costs and fouling of membranes.

The review identifies many predominant mechanisms essential for the degradation of medicines, including eliminating other functioning categories, ring cleavage, oxygen-containing functional categories, and acidification. Effective rules designed to ensure environmental adherence would significantly benefit both humanity and the ecosystem, serving as a substantial measure in mitigating water contamination. Integrating environmental regulations into the interests and aspirations of many stakeholders contributing to ecological degradation can enhance policy efficacy. The legislator must compel all relevant parties to assume responsibilities and collaborate to attain a sustainable environment.

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