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Pharmaceuticals as Emerging Contaminants in Aqueous Systems in Brazil: Consequences and Mitigation

Thais de Souza Neri[1], Alexandre do Nascimento[2], Marcos Batista Figueredo[1,*], Yusdel Diaz Hernández[1], Roberto Luiz Souza Monteiro[3]

[1]Postgraduate Program in Modeling and Simulations of Biosystems, Bahia State University, Alagoinhas, Bahia, Brazil.
[2]Postgraduate Program in Modeling and Simulations of Biosystems, State University of Santa Cruz, Ilhéus, Bahia, Brazil.
[3]Postgraduate Program in Computational Modeling, SENAI CIMATEC University Center, Salvador, Bahia, Brazil.

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ABSTRACT

The global commercialization and consumption of allopathic medicines have increased significantly. These pharmaceuticals, when ingested by organisms, generate by-products that, upon excretion, may interact with the environment, leading to numerous consequences that can alter flora and fauna at various levels. This study aims to consolidate findings from the past decade, highlighting the potential effects of pharmaceuticals and their by-products on biosystems when discharged into water systems without proper treatment, particularly within the Brazilian context. A review of articles from the last 10 years was conducted to identify regions in Brazil affected by pharmaceutical contamination. The review focused on the environmental impact and potential threats to aquatic biodiversity and public health. Many pharmaceuticals and their by-products have shown adverse effects even at low concentrations (μgL^{-1} or ngL^{-1}). However, a wide range of drugs whose effects and impacts on various biosystems are not yet fully understood. In Brazil, where freshwater availability is abundant, pharmaceutical contamination poses a growing threat to aquatic biodiversity and public health. The study underscores the importance of implementing effective mitigation policies and actions to address this problem.

1. Introduction

Anthropogenic activities have contributed significantly to changes in aquatic environments, which are increasingly exposed to a growing variety of chemical compounds that were previously absent or present at low concentrations. In this context, emerging contaminants (ECs) have gained prominence because of their occurrence in different ecosystems, raising environmental concerns that have become a warning for humanity.

ECs are chemical compounds of anthropogenic or natural origin, classified as such due to the lack of or ongoing environmental regulation, as their guidelines and legal frameworks are not yet well defined. Their anthropogenic occurrence is primarily a result of domestic, industrial, hospital, agricultural, and livestock activities [1, 2]. ECs include substances such as pharmaceuticals, compounds used in personal care products, hormones, illicit drugs, food additives, pesticides, by-products from water disinfection processes, nanomaterials, stimulants, microplastics, among others [3, 4].

Among the various types of ECs, pharmaceuticals can be defined as a group of chemical substances with medicinal properties, including organic and inorganic compounds [5]. These substances currently originate from the pharmaceutical industries, and in Brazil, their regulation, registration, authorization, inspection, and monitoring are the responsibility of the Brazilian Health Regulatory Agency (ANVISA).

Consumers obtain medications through medical prescriptions, pharmacists, and/or self-medication. It should be noted that in Brazil, the sale of pharmaceutical products is influenced by marketing [6]. After being metabolized, pharmaceuticals can generate by-products that are discharged into sewage systems, reach wastewater treatment plants, but are not removed. This is due to the lack of effective methods for removing ECs, which result in their discharge into receiving water bodies. Contamination can also occur through improper disposal of expired or unused medications directly into domestic wastewater. Industries, including pharmaceuticals, can also release substances containing these ECs into the environment [7]. Consequently, bioactive compounds from these substances can reach the surface and groundwater. Fig. 1 illustrates the possible routes through which pharmaceuticals can reach water bodies.

Studies on the presence of these compounds in the environment are driven by concerns about their potential risk to ecosystems as a result of adverse effects, which can affect human health, undergo bioaccumulation, and be transported to different environments. Even when released in extremely low concentrations, the effects can be adverse both to the aquatic environment and public health, leading to issues such as bacterial resistance, carcinogenicity, toxicity, endocrine disruption, and the complexity of risk assessment in affected regions [8-10].

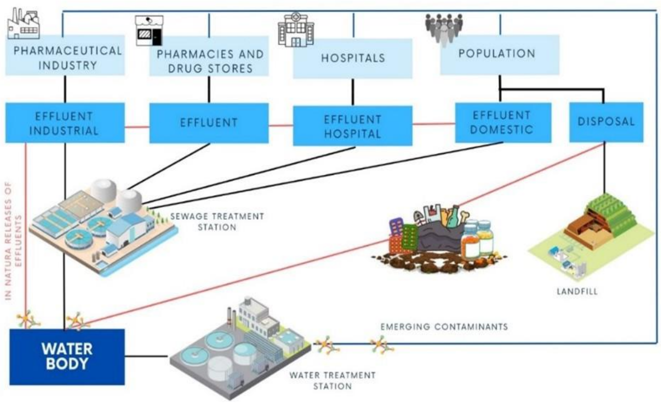

Fig. 1 Flow diagram of the potential routes of pharmaceutical compounds and their residues in the environment

In Sewage Treatment Plants (STPs), the responsible operators use the methods and go through a series of stages to treat these effluents according to the standard regulations of each country. However, in Brazil, there is no standardized method regulated for the removal of emerging contaminants (ECs). As a result, knowing that the final destination of

*Corresponding Author:mbfigueredo@uneb.br(Marcos Batista Figueredo)

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these ECs is natural water resources, these compounds can cause environmental imbalances and, in the long term, lead to significant changes in ecosystems, such as the feminization of fish [11], reduction in fertilization, mortality, growth inhibition of various aquatic organisms [12]. These short- and long-term interferences promote issues that also have an impact on humans, as biosystems have an interconnected network within the environment. Thus, the aim of this article is to gather data from previous studies (over the past 10 years) to identify pharmaceuticals.

2. Experimental Methods

Data were obtained from the databases Scopus, Springer, PubMed, Google Scholar, Web of Science, Science-Direct, EPA (Environmental Protection Agency), PubChem and Research-Gate between 1 May 2024 and 12 July 2024. The records were collected by a systematic search for documents corresponding to the search terms in the articles titles, abstracts, and keywords fields. The search was refined to include only articles and reviews as document types.

The initial search was conducted using the following search syntax: Emerging and Contaminants and Brazil, to include the most common terms referring to emerging contaminants (EC), such as emerging pollutant(s), emerging contaminant(s) of concern, emerging pollutant(s) of concern, emerging organic contaminant(s), emerging organic pollutant(s), emerging environ- mental contaminant(s) of concern, and emerging environmental pollutant(s) of concern. This search was limited to the period between 2014 and 2024 to obtain additional information on publishing activity during these years. The search included several keywords, the main results of which are shown in Fig. 2.

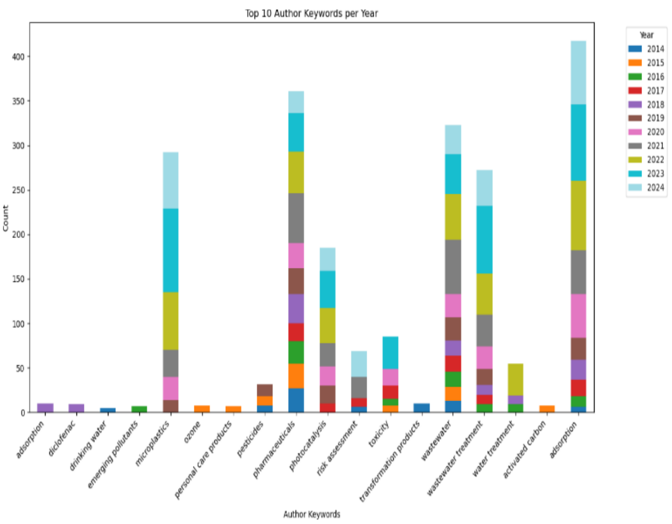

Fig. 2 Keywords and its search output

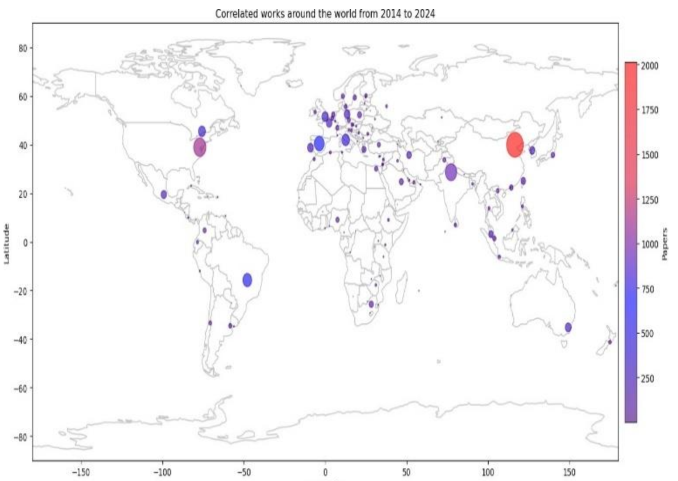

Fig. 3 Distribution of targetted articles worldwide

Additional and more specific searches were conducted to assess production in specific re- search areas related to ECs. In these cases, searches were performed within the results obtained from the initial search mentioned above, with the aim of determining research trends and topics of greater relevance.

The results of these searches are limited to work related to compounds classified as ECs and identified as such by the authors. We found 48,135 articles with worldwide distribution shown in Fig. 3. This number was reduced to 16,388 documents that specifically addressed the topic. Semantic filters were then applied to reduce this number to articles that exclusively dealt with the quantification of contaminants found in Brazil. Following this search, we studied the mitigation aspects without restricting the search to Brazil due to the low number of returns for this type of work, which allowed for a more in-depth analysis of the topic. We found a total of 224 studies related to Brazil. These studies cover a variety of topics, with a significant portion focused on environmental issues, such as the presence of emerging contaminants in water sources.

The information retrieved from the databases in each search included: (i) citation information, (ii) bibliographic details, (iii) abstract and keywords, (iv) funding details, and (v) other relevant information. Python and the Pandas package were used for data visualization and analysis.

3. The Scenario in Brazil

In Brazil, regulation for ECs, monitoring (quantifying and identifying) water quality in Sewage Treatment Plants (STPs) and Water Treatment Plants (WTPs) is still not a current reality, as there are no effective methods for routine analysis in these environments. It is essential to have close cooperation between researchers and regulatory authorities. However, it is not a mental priority for governments as there are no standard methods for the removal of ECs in STPs and WTPs. This has significant consequences for public health and the environment [13]. Recent studies show the presence of ECs in concentrations ranging from ng/L to μg/L in wastewater, surface water, groundwater, and drinking water in various regions of Brazil. These ECs may be associated with high consumption of pharmaceuticals, which has been increasing considerably in Brazil.

Fig. 4 illustrates the ten best-selling drugs in Brazil in 2023, providing a clear view of the high consumption of certain pharmaceuticals, such as paracetamol, losartan, and simvastatin. The significant demand for these drugs, as evidenced by the substantial number of units sold, suggests a potential increase in the load of ECs in aquatic environments, especially given that a significant fraction of these compounds is excreted and reaches STPs.

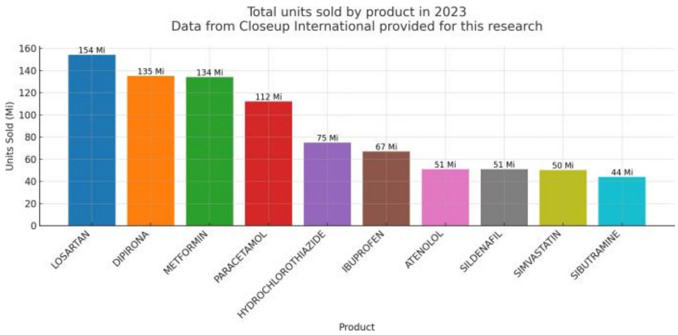

Fig. 4 Best-selling substances in the country by the main pharmaceutical distributor

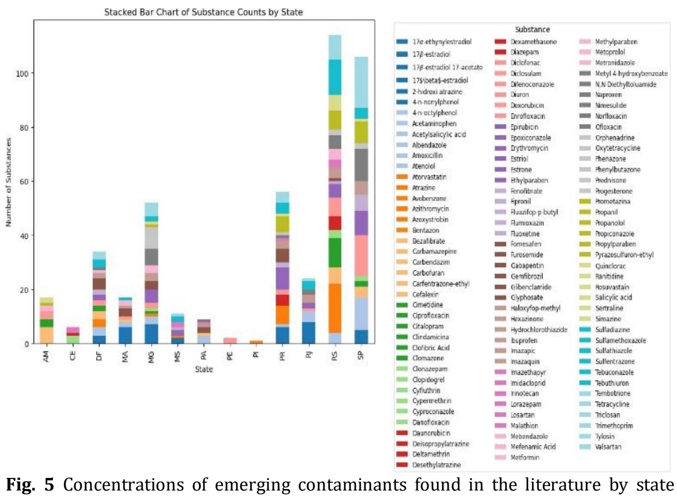

Fig. 5 Concentrations of emerging contaminants found in the literature by state evaluated in various environmental samples

Fig. 5, in turn, presents the geographical distribution of ECs in different regions of Brazil, showing that many of the most consumed pharmaceuticals are found in water bodies in the same areas of high

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population density. These data reinforce the correlation between high drug consumption and water resource contamination, which can lead to serious ecological imbalances. The presence of these ECs in aquatic environments, even after treatment in STPs, indicates the urgent need to develop and implement more effective methods for EC

Removal in treatment plants, to mitigate adverse impacts on aquatic biota and public health. Figs. 2 and 3 reveal the complexity of the challenge that Brazil faces regarding waste management, as current regulations lack methods for their removal.

In Fig. 4, Paracetamol stands out with 112 million units sold in the form of various pharma- ceutical products listed in the top 10, and its presence has been detected in water resources across different regions of Brazil, such as the North [14], South, and Central-West. In addition, other ECs such as Losartan and Simvastatin have also been reported. This occurs because, after being metabolized in the human body, a significant portion of pharmaceuticals and their metabolites are excreted through urine and feces, reaching sewage treatment plants (STPs) but may persist as there is no method to remove ECs. These effluents, after treatment in STPs, are discharged into surface waters, and the presence of ECs can interfere with various biosystems [15, 16] highlight that studies on the detection of ECs have been conducted in rivers in the southeast and south- ern regions due to the higher concentration of industries and the Brazilian population. Despite the higher levels of basic sanitation in these regions compared to others, the continued discharge of untreated effluents into low-flow rivers still compromises the quality of surface waters. In the southern region of the country, Pivetta et al. investigated the presence of ECs in two water bodies in the city of Santa Maria – RS [17]. The two urban watersheds analyzed were characterized by anthropogenic influence, particularly deficiencies in domestic sewage collection and treatment systems. The results indicated the presence of ibuprofen and paracetamol, whose concentrations can be observed in Fig. 4, suggesting a significant influence of population use on this drug.

In the Southeast region, in the state of São Paulo, a study conducted between 2005 and 2015 in the waters of the Atibaia, Capivari, and Jundiaí rivers, the Anhumas and Pinheiros streams, and the Salto Grande Dam also detected the presence of ECs [18]. A study by Martini et al. [19] analyzed samples from seven rivers and three reservoirs, detecting caffeine as a substance present in 95% of the samples with concentrations ranging from 5.5 to 69.9 ng L^{-1}.

In the Northeast region, a study by Santos et al. [20] detected the presence of several pharmaceuticals in the largest reservoir of the Caruaru Natural Park, which is intended for human consumption. This raises alarm, as these substances are considered endocrine disruptors and can affect an entire trophic chain.

In the North of the country, in Belém, samples from the Bolonha and Água Preta reser- voirs were analysed during both the rainy and dry seasons, detecting the presence of Loratadine (ranging from 15.7 to 45.0 ng L^{-1}) in both periods, and Gemfibrozil (ranging from 11.4 to 23.5 ng L^{-1}) which was only detected during the dry season, and Bezafibrate (ranging from 269.6 to 1364.9 ng L^{-1}) which was only identified during the rainy season [21].

The city of Cuiabá, in the state of Mato Grosso (MT), located in the central-west region, has three main water treatment systems responsible for providing 86% of the total population. A case study in these three systems revealed the presence of substances such as atenolol, caffeine, carbamazepine, diclofenac, estrone, ibuprofen, naproxen, paracetamol, propranolol, triclosan, and some hormones [22]. Among these, at least three pharmaceuticals can be seen in Figure 4, which are among the ten most consumed drugs by the population in Brazil.

Fig. 6 presents the spatial distribution of pharmaceuticals considered ECs found in surface water samples across the Brazilian territory. This figure complements Figs. 7 by providing a geographical view of EC contamination, demonstrating the extent and variability of the presence of these compounds in different regions of Brazil.

By comparing Fig. 7 with Figs. 4 and 6, it is possible to observe that the most sold pharmaceuticals in Brazil are also present in various regions, as indicated in Fig. 6, suggesting a direct relationship between high consumption and the detection of these compounds in the environment. The spatial distribution shown in Fig. 7 reinforces this correlation, highlighting that areas with a higher consumption of certain pharmaceuticals tend to have higher concentrations of these compounds in surface waters. Moreover, the joint analysis of the figures points to the importance of geography and regional characteristics in the behavior of ECs. Regions with higher population density and greater industrial activity, such as the Southeast, show higher concentrations of ECs, as highlighted in Fig. 7, which aligns with the information on consumption and the presence of pharmaceuticals in aquatic environments. This information is crucial for understanding the environmental impacts of these compounds and for developing mitigation strategies specific to each region.

This integrated analysis underscores the need for regional public policies that take into account local specificities in combating EC contamination and highlights the importance of ongoing studies to monitor the presence of these contaminants and their effects on the environment and public health.

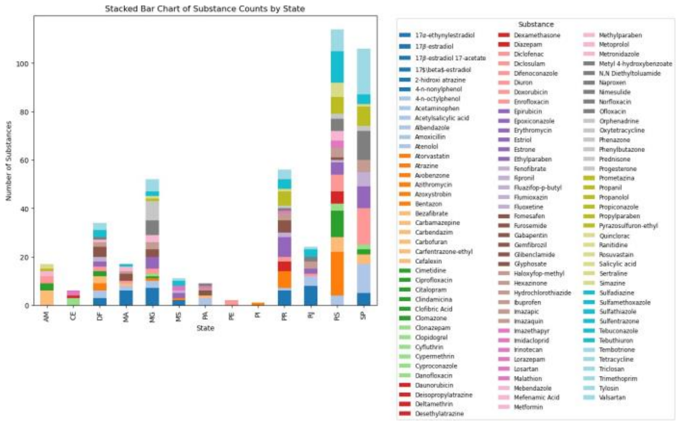

Fig. 6 Pharmaceuticals considered as ECs found in different regions of Brazil. Data obtained from Table S1

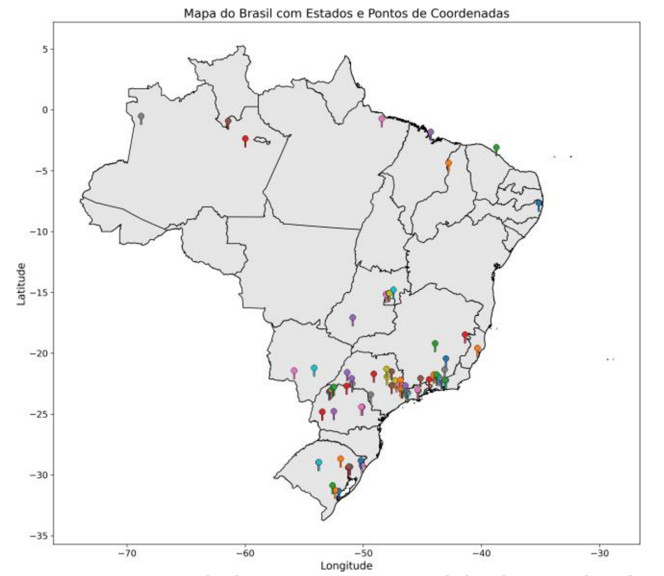

Fig. 7 Locations in Brazil where quantitative research has been conducted on emerging contaminants

4. Environmental and Public Health Impacts

A study conducted by Branco et al. [23] assessed the ecological risk of ECs in Brazilian river basins through the determination of the Risk Quotient (RQ), where RQ values greater than 1 indicate that the concentrations of the evaluated compounds pose a potential risk to the biota of the analysed aquatic environment. Among the various classes of ECs, pharmaceuticals were the sec- ond group with the highest presence of compounds with $RQ > 1$, with the most critical being: Atenolol, Bezafibrate, Caffeine, Carbamazepine, Diclofenac, Furosemide, Losartan, Sertraline, Triclosan, Trimethoprim, Valsartan and Venlafaxine.

According to a review of the literature conducted by Da Silva Escher, et al. [24], paracetamol, ibuprofen, and diclofenac can cause ecotoxicological effects in aquatic biota, such as algae, mollusks, fish and aquatic plants. The authors highlighted the feminization of fish and the reduction of species diversity as the main harmful impacts on these organisms.

In a study, an environmental risk analysis was performed for different pharmaceuticals [12]. The analysis was calculated on the basis of the concentrations of these compounds in treated sewage. As a result, Diclofenac, Bisphenol, Naproxen, Caffeine, Trimethoprim and Sulfamethoxazole were found to present a high environmental risk in all scenarios analyzed. It should be noted that the risk is associated with toxicological effects such as vitellogenin induction, fertilization, reduced fertility, mortality, growth inhibition, and morphological abnormalities in certain species of fish, crustaceans, microalgae, algae, and bacteria.

The environmental and public health risks posed by the concentrations of pharmaceuticals found worldwide in surface waters have been reported

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by various researchers. Losartan, the best-selling drug in Brazil and detected in different surface water samples, has been found in effluents in regions such as Germany [25], Colombia [26], Spain [27], China[28] and Cuba [29], among others. Losartan by-products have been identified to exhibit even more toxic effects when degraded in the environment [30], and adverse effects on aquatic organisms, such as crustaceans, algae, and fish, have been documented [31]. Many of these organisms are consumed by humans in Brazil, especially by the riverside population, which can lead to ingestion, causing endocrine disruption. Among the main adverse effects caused by EC pharmaceuticals on human health are the increased incidence of cancers (breast, testicular, and pulmonary) prostate), infertility, spontaneous abortions, metabolic disorders, and endometriosis [32]. A study by Mello, et al. [33] analyzed the presence of pharmaceuticals in seafood from two areas of the Brazilian coast, Sepetiba Bay and the Parnaíba Delta. Among the results, ibuprofen and other nonsteroidal anti-inflammatory drugs (NSAIDs), as well as Simvastatin and carbamazepine, were the main bioaccumulated pharmaceuticals in the edible parts of the species analyzed. In particular, carnivorous species at a high trophic level, snook, were the most contaminated by NSAIDs.

The anthropogenic introduction of ECs into biosystems has been found to be significantly related to human health impacts. When a drug is absorbed by a species at a certain trophic level of the food chain, the substance accumulates in the tissue and this bioaccumulation tends to increase with time. Consequently, organisms at the top of the food chain (such as humans) are exposed to more concentrated levels of ECs, as there is a progressive increase along the food web due to the biomagnification process [34].

Table 1 List of various mitigation methods

Year	Mitigation Method	Ref.
	Adsorption and Pollutant Removal	
2024	SPE/GC/MS for the determination of organic UV filters	[38]
2024	Modified almond shells for pollutant adsorption	[42]
2023	Adsorption using mesoporous MgFe2O4 for organophosphate pesticides	[43]
2023	Functionalized porous polymers for adsorption and photocatalysis Analytical degradation	[44]
2023	Aerogels for solvent/oil cleanup, ion removal, and pollutant gas Adsorption	[45]
	Oxidation Processes	
2024	UV-Activated Persulfate Oxidation for Antibiotic Degradation	[46]
2024	Rehabilitation of the sewer infrastructure and advanced oxidation processes	[47]
2020	Integration of the membrane bioreactor with advanced oxidation processes	[48]
2023	Modification of processes and equipment, adsorption, and advanced oxidation	[49]
	Environmental Management and Monitoring	
2023	Regular monitoring for timely interventions and mitigation	[50]
2022	Containment measures to reduce flood-induced microplastic mobilization	[51]
2020	Integrated coastal zone management and environmental monitoring plans	[52]
2019	Real-time monitoring using biosensors	[18]
2018	Control strategies for pharmaceutical contaminants in water	[53]
	Soil and Water Improvement	
2024	Alternating irrigation to reduce CEC accumulation in crops	[54]
2022	Ozonation for pharmaceutical removal	[37]
2022	Preservation and contaminant control in drinking water sources	[55]
2023	Biochar for soil amendment and pollutant immobilization	[56]
2023	Soil amendment with Quicklime for Pathogen Disinfection	[57]
	Specific Pollutant Reduction	
2023	Restriction of toxic plastic additives and eco-friendly alternatives	[58]
2021	Source control and advanced technologies for PFAS reduction in Biosolids	[59]
	Effluent Treatment Technologies	
2016	Membrane filtration and combined physical/chemical processes	[39]
2014	Horizontal subsurface flow bed with activated sludge for micro pollutant removal	[60]

Similarly, when efforts are made to protect ecosystems from the harmful impacts of EC con- centrations, such as pharmaceuticals, not only is the biota of the environment in question pro- tected from potential adverse effects of this contamination, but the benefits also extend to human health. A study by Baran, et al. [12] concluded that establishing an environmental standard for the maximum concentration of the pharmaceuticals evaluated in their research, aimed at protecting aquatic biota, would also indirectly protect human health for 94% of the compounds analysed.

5. Policies and Mitigation Actions

The Brazilian legislation on basic sanitation establishes a set of services that should be provided to the population to maintain public health, conserve natural resources, and protect the environment. These services include water supply, sewage collection, urban drainage, solid waste, and stormwater management [13]. It is known that the deficiencies in these services, considered essential for the population, are directly related to the contamination of water resources by pharmaceutical residues and other types of emerging contaminants (EC) [16].

According to data from the National Sanitation Information System – SNIS (2022), 56% of the Brazilian population is served by sewage collection services and around 52% of the country's wastewater undergoes treatment before discharged to receiving water bodies. However, even in places where sewage services are available, part of the population can still discharge its effluents irregularly into stormwater collection networks or directly into water bodies, as reported by Pivetta, et al. [17].

Even more aggravating, as previously reported in this article, although lower concentrations are identified in treated effluents, a considerable portion of these contaminants resist the treatment processes used in wastewater treatment plants (WWTP) [16]. Similarly, Cartaxo, et al. [35] pointed out that the conventional treatment used in most water treatment plants (WTPs) is in- effective in removing ECs, requiring the use of more advanced technologies to remove these contaminants. The main alternatives that can be associated with these systems are: adsorption with powdered activated carbon (PAC) and granular activated carbon (GAC), advanced oxidation processes (AOPs), and membrane filtration.

Furthermore, as pointed out by El-Kalliny, et al. [36], although Brazilian legislation has specific resolutions for water quality, drafted by the National Environment Council (CONAMA), such as Resolu- tion No. 357/2005, which deals with water body classification standards, and Resolution No. 430/2011, which establishes effluent discharge standards, the country still does not set limits for pharmaceutical ECs [13]. Therefore, it becomes more challenging to prevent these contaminants from reaching water bodies and harming the environment and public health.

Global research on policies and mitigation actions highlights several strategies and approaches to tackle environmental challenges. An analysis of available studies reveals a series of actions taken to minimize the impacts of emerging contaminants on the environment and public health. Many studies have focused on developing and improving water treatment technologies to remove emerging contaminants. Advanced methods such as ozonation, membrane filtration, and the combination of physical and chemical processes have been explored to enhance the efficiency of removing toxic compounds from water. These technologies are crucial in ensuring that drinking water is free of contaminants that can harm human health and the environment [37-39].

Implementing effective public policies is essential to mitigate the impacts of emerging contaminants. Several countries have adopted strict regulations for the disposal and treatment of industrial and pharmaceutical waste. In addition, policies have been implemented that encourage the research and development of new treatment technologies to promote innovation and the adoption of sustainable practices. The importance of strict pharmaceutical waste management policies to minimize the entry of contaminants into the environment is often highlighted [40, 41].

Continuous monitoring of contaminant levels in different environmental matrices is crucial to risk mitigation. Studies have demonstrated the importance of environmental monitoring per gram using advanced technologies to detect and quantify emerging contaminants. These grams help identify pollution sources and assess the effectiveness of the mitigation measures implemented. In addition, the use of biosensors and other real-time monitoring technologies has been explored to provide a rapid and effective response to contamination problems [41, 18, 38]. Tables 1 summarizes the various mitigation methods found in recent studies, categorizing them by approach type and year of publication. The analysis of these data provides a comprehensive overview of the strategies used to combat environmental contamination by various types of emerging pollutants.

6. Conclusion

This study highlighted the growing concern regarding pharmaceuticals as ECs in Brazilian water systems, emphasizing the urgent need for effective mitigation strategies. Analysis of data collected over the past ten years revealed a significant presence of pharmaceuticals in various regions of the country, correlating the high consumption of these compounds with their detection in surface waters. These results indicate

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that despite efforts at wastewater treatment plants (STPs), current methods are insufficient to remove these contaminants, which pose potential risks to both aquatic ecosystems and public health.

The adverse impacts of these contaminants, such as bacterial resistance, toxicity, and endocrine disruption, represent a real threat, especially considering the persistence and bio-accuracy of these compounds in the environment. Furthermore, the geographical distribution of pharmaceuticals points to the need for regional public policies that take into account local specificities in waste management.

In conclusion, this study reinforces the importance of stricter regulations and the importance of advanced effluent treatment technologies for the removal of pharmaceuticals from water systems. Cooperation between researchers, regulatory authorities, and civil society is essential to address the challenges posed by emerging contaminants and to protect human health and aquatic biodiversity in Brazil.

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