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Emerging Pharmaceutical Pollutants in Colombian Watersheds and Environmental Management Measures

María Eugenia Buitrago-González*, Jesús David Sánchez-Pinto, Mónica Martínez-Martina, Alejandra María Rodríguez-Correa, María José Virviescas-Ospina, Sandra Santacoloma-Londoño

(Environmental Engineering, Unidad Central del Valle del Cauca, Tuluá, Colombia)

* Corresponding author: mbuitrago@uceva.edu.co

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Abstract: This study presents the geographic distribution of emerging pharmaceutical contaminants in Colombian rivers and the most appropriate technology for their removal. The aim of this research was to study the presence of emerging pharmaceutical contaminants in the watersheds of Colombia, and to identify efficient environmental management options prior to their discharge into water bodies. For this purpose, a systematic review was carried out by searching for scientific articles published between 2017 and 2021 in the Gale, Science Direct, Springer Protocols, Taylor and Francis Online, and Google Academic databases. The search terms used were “emerging pollutants”, “emerging contaminants”, and “pharmaceutical emerging contaminants”, followed by Colombia. Research articles in English and Spanish that reported pharmaceutical-type compounds and/or affected watersheds and/or human health effects and/or treatment were included. This was supplemented with information from the country’s Regional Autonomous Corporations, the Ministry of Environment and Sustainable Development, and governmental environmental institutions. Subsequently, water sources in Colombia containing emerging contaminants were identified. Finally, a phase was conducted to identify treatment proposals for the management of emerging pharmaceutical contaminants in Colombia. A dynamic database was created with this information and



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managed using ArcGIS and R software. In Colombia, 117 emerging pharmaceutical contaminants have been reported in the watersheds. The most common drug was carbamazepine, followed by clarithromycin, erythromycin, and diclofenac. The watersheds with the highest concentrations of emerging pharmaceutical contaminants were Antioquia, Caquetá, Cundinamarca, Nariño, and Valle del Cauca. Reverse has been identified as the most effective treatment for the removal of emerging pharmaceuticals.

Keywords: emerging contaminants, chemical water pollution, watersheds, water treatment, environmental management.

哥倫比亞流域新出現的藥物污染物及環境管理措施

摘要: 本研究介绍了哥伦比亚河流中新兴药物污染物的地理分布以及最合适的去除技术。本研究的目的是研究哥伦比亚流域中新兴药物污染物的存在情况,并在其排入水体之前确定有效的环境管理方案。为此,通过搜索 2017 年至 2021 年期间在 Gale、Science Direct、Springer Protocols、Taylor and Francis Online 和 Google Academic 数据库中发表的科学文章进行了系统审查。使用的搜索词是“新兴污染物”、“新兴污染物”和“药物新兴污染物”,其次是哥伦比亚。包括报告药物型化合物和/或受影响流域和/或人类健康影响和/或治疗的英文和西班牙文研究文章。补充了该国区域自治公司、环境和可持续发展部以及政府环境机构的信息。随后,确定了哥伦比亚含有新兴污染物的水源。最后,开展了一个阶段,以确定哥伦比亚新兴药物污染物管理的治疗方案。利用此信息创建了一个动态数据库,并使用 ArcGIS 和 R 软件进行管理。在哥伦比亚,流域中已报告了 117 种新兴药物污染物。最常见的药物是卡马西平,其次是克拉霉素、红霉素和双氯芬酸。新兴药物污染物浓度最高的流域是安蒂奥基亚、卡克塔、昆迪纳马卡、纳里尼奥和考卡山谷省。逆转被认为是去除新兴药物的最有效治疗方法。

關鍵字: 新興污染物、化學水污染、流域、水處理、環境管理。

1. Introduction

Emerging contaminants (EC) are contaminants of a very different origin and chemical nature from the contaminants typically treated in wastewater treatment systems (WWTS), which are not regulated [1]. These contaminants need not be present continuously in the environment to be harmful; however, many WWTPs are not designed to remove them, and EC may even be present in WWTP sludge and leachate [2].

The EC group includes a wide variety of products, such as pesticides, pharmaceuticals (analgesics, antihypertensives, antibiotics), industrial products, personal care products, steroids, hormones, and even nicotine [1]. These contaminants cause serious damage to human health and aquatic organisms, including endocrine disruption, thyroid and other hormone disruptions, and reproductive damage [3]. ECs can also be carcinogenic and neurotoxic, causing problems in the immune system and kidneys. However, this depends on the frequency of exposure of the individual, whether sporadic or continuous, causing problems that can even be chronic. In addition, ECs can be bioaccumulative and can be distributed through the different trophic chains present in the ecosystem [4].

As demonstrated by [5], the harmful effects of ECs on aquatic life are so widespread that they affect everything from algae, owing to the toxicity of pharmaceuticals such as ciprofloxacin, to fish such as

rainbow trout, zebrafish, and even mussels, altering their various metabolic processes, reproductive cycles, and reducing fertility. Changes have also been found in humans, such as immune and reproductive system disorders caused by the pharmaceutical compound diethylstilbestrol, which is used to treat prostate cancer and prevent abortion. Bisphenol A, which is used industrially in the manufacture of sterilized pouches and can coatings, has hormonal effects that increase the risk of cancer and act as anti-androgens.

In Spain, in the Guadarrama, Jarama and Manzanares river basins, EC have been found as gastrointestinal pharmaceuticals at concentrations of (1.94 $\mu\text{g/L}$), loratadine at concentrations of 0.02 $\mu\text{g/L}$, causing various lesions. Histopathological effects in fish include effects on the reproductive system and organs such as the liver, kidneys, heart, and other parts of the body [6].

Despite their various adverse effects on aquatic life, these are not fully understood because of the lack of specific information on Pharmaceutical Emerging Contaminants (PECs). There are some known risks to aquatic biota that may be caused by persistence in the environment, such as diclofenac, which affects fish gills and kidney tissues; ibuprofen, which inhibits polyp regeneration and reduces reproduction; the mixture of carbamazepine, ranitidine, salbutamol, and sulfamethoxazole, among others, inhibits the growth of

embryonic kidney cells in humans; diltiazem, which is very toxic at a concentration of 8.2 mg/L is lethal to organisms such as *Daphnia magna*; and chloramphenicol, florfenicol, and thiamphenicol affect the growth of algae and microalgae [7].

In Brazil, although monitoring to detect ECs in different water bodies began in 2000, there is still an information gap, which means that discharges continue without any regulation, with high impacts on aquatic biota and damage to human health [8].

In 2018, the Water Environment Federation reported that different types of pollutants, such as pharmaceutical compounds, flame retardants, and steroid hormones, have been found in rivers such as the Danube in Europe, the Pirai Creek and Jundiai rivers in Brazil, and other watersheds in countries such as South Africa, Ukraine, and Serbia [9].

The Yangtze and Han rivers in Asia are sources of water used for purification, but a total of 33 pharmaceutical and metabolic contaminants have been found [9], posing a risk to the public health of consumers of the resource. Even if a purification process is carried out to remove impurities, these pharmaceutical contaminants may still be present.

Different PECs have been found in the Baltic Sea under the jurisdiction of each of the countries surrounding it, such as Denmark, Estonia, Finland, Germany, Poland, Russia, and Sweden. The long list of reported PECs includes diclofenac, naproxen, paracetamol, iopamidol, clarithromycin, metoprolol, sulfadiazine, and trimethoprim. In addition, the concentrations of some compounds are too high, affecting the health of aquatic life and human populations using the Baltic Sea ecosystem service [10].

Various wastewater treatment processes are becoming more innovative over time; therefore, it is necessary to implement guidelines for their design [11], which vary according to the characteristics of the wastewater. Many WWTPs are not specifically designed to treat pharmaceuticals present in water, but there are methods that can help reduce their concentrations.

Some conventional and advanced methods of wastewater treatment against PEC have been evaluated in different parts of the world in terms of their removal efficiency. According to [7], these treatments vary according to the characteristics of the water body and their implementation costs. However, not all compounds to be treated are documented because of their high costs at the time of treatment; therefore, there is a knowledge gap on this topic.

In Colombia, the cities of Medellín and Bogotá found more than 20 pharmaceutical products in their wastewater, from painkillers to antibiotics. Wastewater from a hospital in the city of Tumaco, which does not have a treatment process, was also assessed and found to contain concentrations of up to 5 µg/L of substances

such as paracetamol, azithromycin, losartan and other types of medicines, which is a problem for everyone [12].

In the department of Córdoba, in the Mocari and La Doctrina irrigation districts, also in this country, bisphenol was found at an average concentration of 0.036 µg/L; progesterone at 35.17 µg/L and carbamazepine at 23.46 µg/L; these substances are bio accumulative and affect the food security of the region's inhabitants, causing hormonal disorders, brain disorders and other diseases [13].

At the II International Forum for the Recovery of the Cauca River, it was reported that in the department of Valle del Cauca, Colombia, the Cauca River is polluted by various pollutants, such as pesticides, personal care products, hormones, illegal drugs and also pharmaceutical-type compounds, which alter the reproductive cycles of fish [14].

In Colombian legislation, the control of domestic and non-domestic wastewater discharges is regulated by Resolution 0631 of 2015; however, this resolution does not consider the permissible limits for PECs, which means that high concentrations of these compounds can be found in discharges not only from pharmaceutical industries but also from hospitals and households, and even in the treatment plants themselves, which can have an impact on human health and damage to aquatic biota.

According to global and national evidence, environmental damage caused by pharmaceutical ECs is mainly documented in aquatic ecosystems, where they affect aquatic organisms, from fish to algae, altering reproductive cycles, and causing endocrine disruption. In addition, EC bioaccumulates in the food web, posing a serious risk because it can reach human consumption. Although the damage they can cause has been established, there is still a lack of knowledge about what FECs are and how they are produced, which aggravates the situation because these pollutants continue to be dumped in an uncontrolled manner into various water bodies, contributing to the deterioration of the environment. Therefore, studies are needed on different alternatives and/or technologies that help to reduce the concentrations of these compounds before they are discharged into surface waters. One of the main limitations in the study of ECs is the lack of knowledge about them, as high-tech equipment is required to determine their presence and carry out the appropriate treatment. The application of the results of this research on emerging pharmaceutical contaminants has a direct impact on both scientific and practical fields, influencing public policies, water treatment technologies, and environmental health. The objective of this research was to study the presence of emerging pharmaceutical contaminants in Colombian watersheds and identify efficient environmental management options before their discharge into water bodies.

2. Methods

A systematic review was conducted to identify the pharmaceutical compounds dumped in water bodies in Colombia. First, a search phase was conducted for information from scientific articles published between 2017 and 2021. The databases used were Gale, Science Direct, Springer Protocols, Taylor and Francis Online, and Google Academic. The search terms used were “emerging pollutants,” “emerging contaminants,” and “pharmaceutical emerging contaminants,” followed by Colombia, using the Boolean operator AND, so that the geographical matches were mainly in Colombia.

The following inclusion and exclusion criteria were considered in this review: The inclusion criteria were as follows: a) they presented at least one pharmaceutical compound, b) they related to at least one affected water basin, and c) they mentioned an impact on human health and presented a treatment for any type of emerging pharmaceutical contaminant. The following exclusion criteria were considered: a) review articles, b) duplicate articles, and c) full-text not available. These criteria were considered to reduce the risk of bias.

Once the search criteria were applied, the following criteria were used to organize and synthesize the information: author, year, title, search engine, language, pharmaceutical compound found, watershed affected,

related human impact, and mentioned treatment.

The information found was complemented with that available on the websites of the country’s Regional Autonomous Corporations (CAR’S), the Ministry of the Environment and Sustainable Development (MADS), the Institute of Hydrology, Meteorology and Environmental Studies (IDEAM), the Institute of Marine and Coastal Research (INVEMAR) and the Humboldt Institute, using keywords such as “emerging contaminants,” “emerging,” “medicines” and “pharmaceuticals” to find direct results on emerging contaminants, especially those of the pharmaceutical type. This information was organized according to criteria such as the year, document name, jurisdiction, specific information on emerging contaminants, and description.

The selection phase was then carried out, in which the water sources with the presence of emerging contaminants in Colombia were identified, and those most affected by emerging pharmaceutical contaminants in Colombia were selected, considering the five watersheds with the highest number of pharmaceutical compounds and the highest number of reported pharmaceutical compounds. In addition, the treatments were recorded. The search, screening, selection, and data extraction were based on PRISMA methodology, as shown in Figure 1.

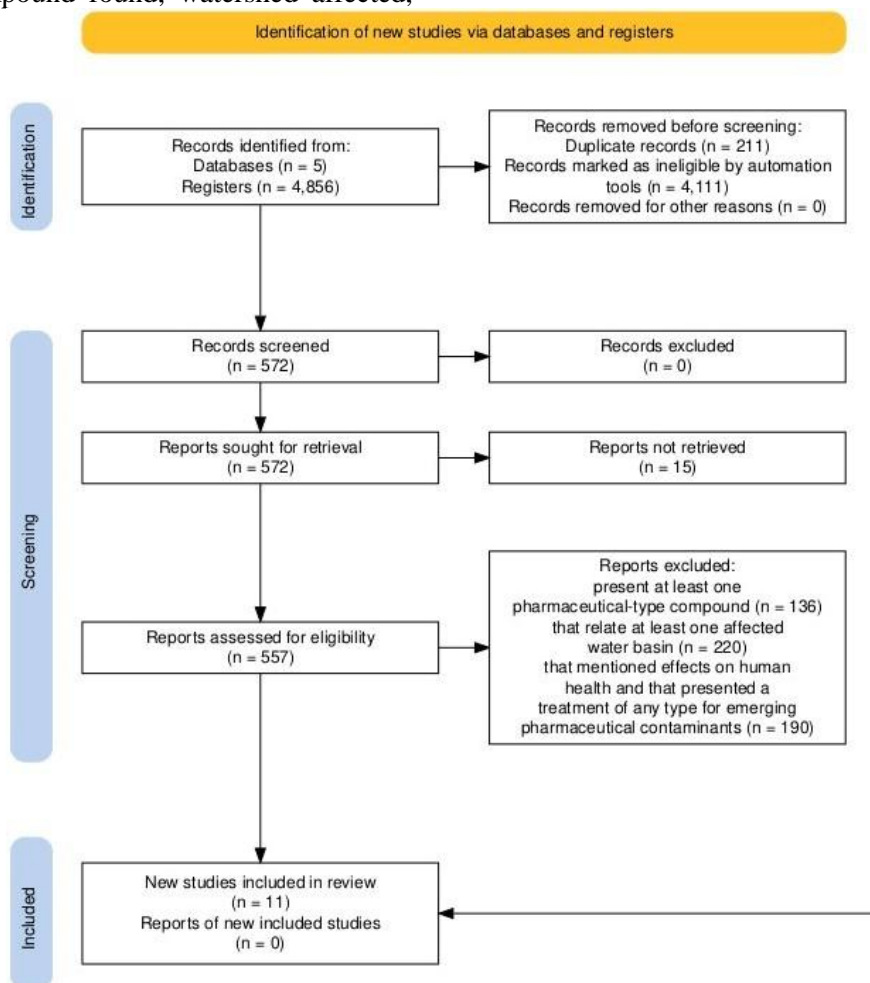


Figure 1. Information management diagram (Source: [15])

Finally, a phase of identification of the treatments proposed in Colombia for the management of pharmaceutical emerging contaminants was carried out, for which information was organized on the type of treatment (physical, chemical, or biological), the name of the treatment, and its description.

Table 1. Removal efficiency of some treatment systems for emerging contaminants (Source: adapted from [7])

| | Treatments | % efficiency |
|--------------|--|--------------|
| Conventional | Activated sludge | 7-99 |
| | Biological filter | 6-71 |
| | Primary sedimentation | 3-45 |
| | Coagulation, filtration and sedimentation | 5-36 |
| | Sand filter | 0-99 |
| | Ozonation | 1-99 |
| | Ozonation or ultrasound and photocatalysts | 23-45 |
| Advanced | Ozonation or catalytic ozonation | 9-100 |
| | UV radiation | ±29 |
| | Photolysis | 52-99 |
| | Fenton UV/TiO ₂ | 80->95 |
| | Biomembrane | 23-99 |
| | Microfiltration and reverse osmosis | 91-100 |
| | Reverse osmosis | 62-97 |
| Ultrasound | 24-99 | |

The efficiency of the identified treatments was recorded. For the treatments that did not report it, the theoretical efficiency was determined according to the method established by [7], as shown in Table 1. Using the above information, a dynamic database was created to compile and organize the information, which was then managed using ArcGIS and R software.

In addition, the reported environmental impacts of the treatment were considered. Its classification as “conventional” or “advanced” was also considered, along with the investment and operating costs.

To qualify the points of theoretical efficiency, environmental impact, treatment type, and cost, the following parameters were used according to the information in Table 2.

Table 2. Score theoretical efficiency, environmental impact, type of treatment and costs (compiled by the authors)

| Theoretical efficiency | Score | Environmental impact | Score | Type of treatment | Score | Investment and operation cost for one year | Score |
|------------------------|-------|-----------------------|-------|-------------------|-------|--|-------|
| <30% | 1 | One impact | 3 | Conventional | 1 | >100.000.000 | 1 |
| 30% - 70% | 2 | Two impacts | 2 | Advanced | 2 | 100.000.000 | 2 |
| >70% | 3 | Three or more impacts | 1 | | | 0< | |

The advanced treatment score is due to the fact that these treatments drive the growth and technological development of the sector where they are applied, being models of implementation of the latest technology equipment and machinery. To determine the investment and treatment costs, what was reported in the literature was used as a reference and an estimated value was taken.

Once the scores were obtained, the three treatments with the highest scores were chosen. For the selected treatments, models were constructed in which the conventional

The WWTS was outlined, and the treatment chosen to achieve better removal of emerging contaminants was added to a part of the process, as previously established. In addition, considering the selected watersheds, the management that should be done to the wastewater before it is discharged into the body of water that is affecting it was proposed.

3. Results

A total of 4856 results were obtained in the search of the databases listed above, of which 572 articles were chosen for a detailed review, as they were studies conducted in Colombia. Of the selected articles, 11 met all the established inclusion criteria, which date from 2017 to 2021. The results obtained from this search are shown in Figure 2.

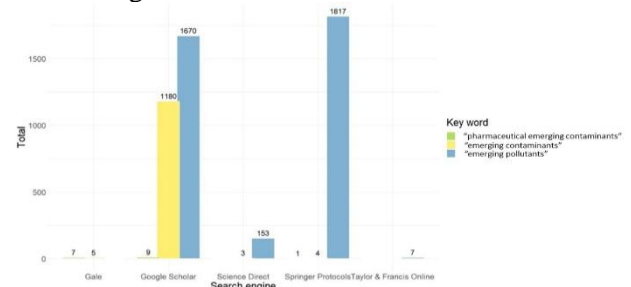


Figure 2. Data number obtained in the review of the databases (authors' estimation)

On the other hand, 7 of the 11 articles were found in English and published as research articles. In addition, only one of the selected articles was found directly in the Science Direct search engine with the keyword “Emerging pollutants,” and the other articles were found using the Google Scholar search engine. Although only 11 articles were chosen based on the applied criteria, the number of articles that were published in the different existing search engines is highlighted, which is why it is understood that there is a high interest in EC, mainly those of pharmaceuticals and pesticides. Table 3 lists the selected articles.

Table 3. Pharmaceutical emerging contaminants reported in Colombia (compiled by the authors)

| Authors | Year | Title | Location | Watershed | Compounds found |
|---------|------|---|---|--|--|
| [16] | 2020 | Evaluation of the removal efficiency of acetaminophen contained in water at a laboratory scale through the bioadsorption technique using cocoa and banana shells | Meta | Ocoa River | Acetaminophen |
| [12] | 2018 | An investigation into the occurrence and removal of pharmaceuticals in Colombian wastewater | -Antioquia -Nariño -Cundina marca -Caquetá | - Medellín River -Bogota River - Tumaco Bay - Orteguaza River | Acetaminophen, Azithromycin, Carbamazepine, Ciprofloxacin, Clarithromycin, Clindamycin, Diclofenac, Erythromycin, Irbesartan, Losartan, Metronidazole, Naproxen, Norfloxacin, Sulfamethoxazole, Tetracycline, Trimethoprim, Valsartan, Venlafaxine |
| [17] | 2019 | Effective elimination of fifteen relevant pharmaceuticals in hospital wastewater from Colombia by combination of a biological system with a sonochemical process | Nariño | Tumaco Bay | Acetaminophen, Diclofenac, Carbamazepine, Venlafaxine, Loratadine, Ciprofloxacin, Norfloxacin, Valsartan, Irbesartan, Sulfamethoxazole, Trimethoprim, Clarithromycin, Azithromycin, Erythromycin, Clindamycin |
| [18] | 2019 | Study of the presence, occurrence and final destination of a pharmaceutical compound (amoxicillin) in an educational institutional wastewater treatment plant | Santander | Mesuli Ravine | Amoxicillin |
| [19] | 2017 | Elimination of micropollutants by the Puerto Mallarino water purification plant, Cali, Colombia. | Valle del Cauca | -Cauca River - Cañaverelejo River - Cali River - Melendez River -Lili River - Aguacatal River -Pance River | Bezafibrate, Carbamazepine, Clofibrac acid, Diazepam, Diclofenac, Etofibrate, Fenofibrate, Atenolol, Betaxolol, Bisoprolol, Clenbuterol, Cyclophosphamide, Dimethyl amino phenazone, Aminotrizoic acid, Iohexol, Iomeprol, Amoxicillin, Chloramphenicol, Clarithromycin, Cloxacillin, Dapsone, of erythromycin A, Dicloxacillin, Azotomycin, Furazolidone, Chlortetracycline, Ciprofloxacin, Doxycycline, Enoxacin, Estrone, Estriol |
| [20] | 2020 | Multidrug resistance and diversity of resistance profiles in carbapenem-resistant Gram-negative bacilli throughout a wastewater treatment plant in Colombia | Antioquia | Medellin River | Carbamazepine |
| [21] | 2020 | Occurrence and removal of pharmaceutical and personal care products using subsurface horizontal flow constructed wetlands | Valle del Cauca | Cauca River | Carbamazepine, Sildenafil |
| [22] | 2019 | Degradation of seventeen contaminants of emerging concern in municipal wastewater effluents by sonochemical advanced oxidation processes | Cundinamarca | Bogota River | Diclofenac, Carbamazepine, Venlafaxine, Ciprofloxacin, Norfloxacin, Valsartan, Losartan, Irbesartan, Sulfamethoxazole, Azithromycin, Clarithromycin, Erythromycin, Metronidazole, Trimethoprim, Clindamycin |
| [23] | 2021 | Analysis of the occurrence of emerging contaminants (glyphosate, paraquat and ibuprofen) in surface sources and in drinking water of Cucuta Norte de Santander, and their removal using membrane technology | Norte de Santander | - Pamplonita River - Zulia River | Ibuprofen |
| [24] | 2019 | Pharmaceutical and personal hygiene products (PPcPs): A threat little studied in Colombian waters | -Cundina marca -Caquetá -Antioquia -Nariño -Risaralda | - Medellín river -Bogota River -Orteguaza River - Tumaco Bay - Otun River | Ibuprofen, Naproxen, Diclofenac, Aspirin, Ketoprofen, Caffeine, Hydrochloric acid, Lorazepam, Acetaminophen, Clarithromycin, Clindamycin, Doxycycline, Erythromycin, Carbamazepine, Irbesartan, Losartan, Metronidazole, Norfloxacin, Sulfamethoxazole, Trimethoprim, Valsartan, Venlafaxine |
| [13] | 2021 | Pharmaceutical and personal care products present in surface waters, for human consumption and wastewater in the department of Cordoba, Colombia | Córdoba | Sinu River | Naproxen, Ibuprofen, Gemfibrozil, Caffeine, Triclosan |

3.1 Reported Pharmaceutical Compounds

From the search carried out, a list of all the pharmaceutical compounds found in the watersheds of Colombia was prepared. The reported pharmaceutical compounds are listed in Table 4.

Table 4. Pharmaceutical compounds found in Colombian watersheds (compiled by the authors)

Mefenamic acid, aminotrizoic acid, clofibrac acid, oxolinic acid, nalidixic acid, salicylic acid, acetylsalicylic acid, nalidixic acid, fenofibrac acid, alpha-ethinyl estradiol, acetaminophen, alprazolam, atorvastatin, azithromycin, amoxicillin, atenolol, ampicillin, hydroxybenzoic acid, albendazole, aminoglycosides, anthracyclines, amikacin, aspirin, Betaxolol, bisoprolol, bezafibrate, benzodiazepines, bisphenol A, butylparaben, benzodiazepines, barbiturates

Caffeine, carbamazepine, ciprofloxacin, cloxacillin, clarithromycin, cumyl phenol, clindamycin, cephalosporin, chloramphenicol, ceftazidime, cephalixin, clenbuterol, cyclophosphamide, chlorphenamine, ceftriaxone, cyanine, codeine, cisplatin, cefadroxil, chloramphenicol, berberine hydrochloride

Diclofenac, dicloxacillin, doxycycline, diazepam,

dapsone, erythromycin A dihydrate, dimethyl amino phenazone, diatrizoate

Estrone, estriol, estradiol, enalapril, erythromycin, enrofloxacin, etofibrate, ethinyl estradiol, ethinyl estradiol, etoposide

Fluoxetine, flumequine, furaltadone, phenazone, fenofibrate, favipiravir, furosemide, fluoroquinolone, florfenicol, fenoprofen

Gemfibrozil, gabapentin, Iopromide, loratadine

Hydrocodeine, hydrochlorothiazide, doxycycline hyclate

Irbesartan, ivermectin, Iopromide

Ketoprofen, kanamycin

Levamisole, lincomycin, lorazepam, losartan, levonorgestrel

Metoprolol, metronidazole, methylparaben, monobactams, mitomycin, meropenem, macrolides

Naproxen, norfloxacin

Oxymorphone, omeprazole, ofloxacin, oxacillin, oxytetracycline hydrochloride, iohexol, oxicam, iopamidol

Primidone, piroxicam, progesterone, pantoprazole, primidone, propranolol, Propofol, penicillin

Roxithromycin, remdesivir

Salbutamol, sulfadiazine, sulfamethoxazole, sulfamethazine, sulfa pyridine, sulfamerazine,

| |
|---|
| sildenafil, sulfadimethoxine, sulfacetamide, sotalol |
| Tetracycline, triclosan, tramadol, trimethoprim, thiamphenicol, timolol |
| Valsartan, venlafaxine, vincristine |

Although the list of pharmaceutical compounds found was extensive, many were only reported in one study, which means that they are not very common compounds. Figure 3 shows the main pharmaceutical compounds reported in the selected articles, with frequencies greater than or equal to 3.

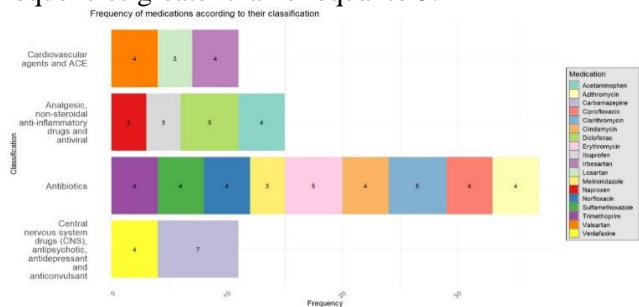


Figure 3. Main pharmaceutical compounds reported in Colombian watersheds (authors' estimation)

Carbamazepine is the medication that most frequently appears in rivers in Colombia. This pharmaceutical compound is mainly used in patients who suffer from seizures and epilepsy; however, it also helps stabilize the different moods of consumers, which is why its misuse makes people dependent on this compound [25].

Among the other most frequent compounds, clarithromycin and erythromycin, which are used for respiratory infections, are widely used during the COVID-19 pandemic; in many cases, they are self-medicated [26], and diclofenac, which is mainly used

to treat inflammation and severe body pain; the latter is commonly used in hospital emergency rooms to treat patients with trauma due to blows. These pharmaceutical compounds are freely sold in the Colombian market, which makes them self-medicate and inappropriate for use. Table 5 presents the classification of reported PECs according to their medical use.

Additionally, the information reported by other sources such as government entities such as Regional autonomous corporations of Alto Magdalena/CAM; of Cundinamarca/CAR; of Risaralda/CARDER; of the Dique canal/CARDIQUE; regional of Sucre/CARSUCRE; of Santander/CAS; of the northern and eastern Amazon/CDA; Bucaramanga/CDMB; Choco/CODECHOCO; Antioquia/CORANTIOQUIA; the Macarena/CORMACARENA; the Magdalena/CORMAGDALENA; the Negro and Nare rivers/CORNARE; the south of the Amazon/CORPOAMAZONIA; Boyaca/CORPOBOYACA; Caldas/CORPOCALDAS; Cesar/CORPOCESAR; Chivor/CORPOCHIVOR; Guajira/CORPOGUAJIRA; Guavio/CORPOGUAVIO; Magdalena/CORPAMAG; Mojana and San Jorge/CORPOMOJANA; Nariño/CORPONARIÑO; northeastern border/CORPONOR; Orinoquia/CORPORINOQUIA; Uraba/CORPOURABA; Tolima/CORTOLIMA; Atlantico/CRA; Cauca/CRC; Quindio/CRQ; Bolivar/CSB; Valle del Cauca/CVC; Sinu and San Jorge Valleys/CVS; the archipelago of San Andrés, Providencia and Santa Catalina/CORALINA, responsible for environmental management in Colombia, showed low knowledge and management of PECs in the country.

Table 5 Classification of pharmaceutical emerging contaminants reported in water sources in Colombia, according to their medical use (compiled by the authors)

| Antibiotics | Cardiovascular agents and ACE | Hormones | Analgesic, non-steroidal anti-inflammatory drugs and antiviral | Central nervous system drugs (CNS), antipsychotic, antidepressant and anticonvulsant | Others (Opioid pain relievers, anti-cancer, contrast X-ray, β-agonists, others) | |
|------------------|-------------------------------|-------------|--|--|---|-------------------|
| Doxycycline | Chloramphenicol | Atenolol | Estrone | Acetaminophen | Caffeine | Tramadol |
| Oxytetracycline | Ceftazidime | Metoprolol | Estriol | Diclofenac | Carbamazepine | Codeine |
| Tetracycline | Cephalexin | Propranolol | Estradiol | Ketoprofen | Fluoxetine | Cyclophosphamide |
| Sulfadiazine | Ceftriaxone | Bezafibrate | Ethinyl estradiol | Naproxen | Venlafaxine | Cisplatin |
| Sulfamethoxazole | Cefadroxil | Gemfibrozil | Progesterone | Salicylic acid | Alprazolam | Etoposide |
| Azithromycin | Chloramphenicol | Betaxolol | | Mefenamic acid | Benzodiazepines | Vincristine |
| Clarithromycin | Dicloxacillin | Bisoprolol | | Acetylsalicylic acid | Barbiturates | Aminotrizoic acid |
| Erythromycin | Dapsone | Enalapril | | Atorvastatin | Lorazepam | Diatrizoate |
| Roxithromycin | Enrofloxacin | Irbesartan | | Aspirin | Diazepam | Iopromide |
| Amoxicillin | Flumequine | Salsartan | | Phenazone | Gabapentin | Iohecol |
| Ampicillin | Fluoroquinolone | Sotalol | | Fenoprofen | Primidone | Iopamidol |
| Meropenem | Doxycycline hyclate | | | Dihydrocodeine | | Fenofibric acid |
| Ciprofloxacin | Kanamycin | | | Piroxicam | | Albendazole |
| Enrofloxacin | Lincomycin | | | Oxymorphone | | Clenbuterol |

Continuation of Table 5

| | | | |
|-----------------|---------------|----------------|-------------------------|
| Norfloxacin | Mitomycin | Sulfa pyridine | Salbutamol |
| Ofloxacin | Macrolides | Remdesivir | Chlorpheniramine |
| Nalidixic acid | Oxacillin | Favipiravir | Loratadine |
| Aminoglycosides | Oxicam | | Berberine hydrochloride |
| Anthracyclines | Penicillin | | Etofibrate |
| Amikacin | Sulfacetamide | | Fenofibrate |
| Cloxacillin | Trimethoprim | | Ethinyl estradiol |
| Clindamycin | Thiamphenicol | | Levonorgestrel |
| Cephalosporin | Oxolinic acid | | Furosemide |
| Metronidazole | | | Hydrochlorothiazide |
| | | | Omeprazole |
| | | | Pantoprazole |
| | | | Propofol |
| | | | Sildenafil |
| | | | Timolol |

Currently, Colombia has 34 Regional Autonomous Corporations, of which only 22 mention issues relating to ECs in a general way, ranging from 2008 to 2021; however, in some cases, they do not use the term “emerging,” to describe compounds that are discharged directly into bodies of water because it is not a commonly mentioned term.

In the documents, the Regional Autonomous Corporations placed a general emphasis on the different emerging compounds that can be present in water, mainly pesticides, which are generated in the agricultural sector and have regulations regarding final disposal. of the containers where the substances are stored, but does not control what is applied to the soil or plant.

CARDER, CDMB, CORMAGDALENA, CORPOAMAZONIA, CORPOGUAVIO, and CRQ were the only regional Autonomous Corporations that mentioned problems regarding wastewater discharge from the health and pharmaceutical sectors.

However, they did not specifically mention that they

contain pharmaceutical compounds; therefore, there is no precise control or knowledge of what the PECs found in bodies of water are.

Compared to the review of scientific articles, the Regional Autonomous Corporations present a gap in knowledge about PECs, since they mainly choose to evaluate agricultural compounds such as pesticides and herbicides, because there is greater knowledge regarding the high negative impacts that they may have in the middle, because Colombia is a country that is characterized by being a food producer and requires compounds of this kind to improve the profitability of the agricultural sector [27]. In addition, Colombia has regulations regarding the use of this class of compounds, such as Resolution 1675 of 2013 of the Ministry of Environment and Sustainable Development, which regulates product return plans. post-consumption of pesticides [28].

After the 11 articles that were selected were examined, the following information was obtained 6 from the watersheds affected in Table 6.

Table 6. Watersheds where pharmaceutical compounds have been found in Colombia (compiled by the authors)

| Affected watershed | Pharmaceutical compounds found | Number of times pharmaceutical compounds were reported | Location | Treatments proposed | Author |
|--------------------|--|--|-----------|---|--------|
| Ocoa River | Acetaminophen | 1 | Meta | Bio adsorption with banana and cocoa peel | [16] |
| Medellin River | Acetaminophen, Azithromycin, Carbamazepine, Ciprofloxacin, Clarithromycin, Clindamycin, Diclofenac, Erythromycin, Irbesartan, Losartan, Metronidazole, Naproxen, Norfloxacin, Sulfamethoxazole, Tetracycline, Trimethoprim, Valsartan, Venlafaxine | 3 | Antioquia | Photo Fenton Electro Fenton Photo-electro Fenton Ultrasonic cavitation | [12] |
| Medellin | Ibuprofen, Naproxen, Diclofenac, Aspirin, Ketoprofen, Caffeine, Hydrochloric acid, Lorazepam, Acetaminophen, Clarithromycin, | | | Bioremediation Phytoremediation Aerobic/anaerobic lagoons | [24] |

| | | | | | | |
|-----------------|--|---|--|---------------|---|------|
| River | Clindamycin, Doxycycline, Erythromycin, Carbamazepine, Irbesartan, Losartan, Metronidazole, Norfloxacin, Sulfamethoxazole, Trimethoprim, Valsartan, Venlafaxine | | | Carbamazepine | Ultraviolet light | [20] |
| | Acetaminophen, Azithromycin, Carbamazepine, Ciprofloxacin, Clarithromycin, Clindamycin, Diclofenac, Erythromycin, Irbesartan, Losartan, Metronidazole, Naproxen, Norfloxacin, Sulfamethoxazole, Tetracycline, Trimethoprim, Valsartan, Venlafaxine | | | | Photo Fenton Electro Fenton Photo-electro Fenton Ultrasonic cavitation | [12] |
| Bogota River | Ibuprofen, Naproxen, Diclofenac, Aspirin, Ketoprofen, Caffeine, Hydrochloric acid, Lorazepam, Acetaminophen, Clarithromycin, Clindamycin, Doxycycline, Erythromycin, Carbamazepine, Irbesartan, Losartan, Metronidazole, Norfloxacin, Sulfamethoxazole, Trimethoprim, Valsartan, Venlafaxine | 3 | | | Bioremediation Phytoremediation Aerobic/anaerobic lagoons | [24] |
| | Diclofenac, Carbamazepine, Venlafaxine, Ciprofloxacin, Norfloxacin, Valsartan, Losartan, Irbesartan, Sulfamethoxazole, Azithromycin, Clarithromycin, Erythromycin, Metronidazole, Trimethoprim, Clindamycin | | | | Ultrasound Sono Fenton Sono-photo Fenton | [22] |
| | Acetaminophen, Azithromycin, Carbamazepine, Ciprofloxacin, Clarithromycin, Clindamycin, Diclofenac, Erythromycin, Irbesartan, Losartan, Metronidazole, Naproxen, Norfloxacin, Sulfamethoxazole, Tetracycline, Trimethoprim, Valsartan, Venlafaxine | | | | Photo Fenton Electro Fenton Photo-electro Fenton Ultrasonic cavitation | [12] |
| Tumaco Bay | Ibuprofen, Naproxen, Diclofenac, Aspirin, Ketoprofen, Caffeine, Hydrochloric acid, Lorazepam, Acetaminophen, Clarithromycin, Clindamycin, Doxycycline, Erythromycin, Carbamazepine, Irbesartan, Losartan, Metronidazole, Norfloxacin, Sulfamethoxazole, Trimethoprim, Valsartan, Venlafaxine | 3 | | | Bioremediation Phytoremediation Aerobic/anaerobic lagoons | [24] |
| | Acetaminophen, Diclofenac, Carbamazepine, Venlafaxine, Loratadine, Ciprofloxacin, Norfloxacin, Valsartan, Irbesartan, Sulfamethoxazole, Trimethoprim, Clarithromycin, Azithromycin, Erythromycin, Clindamycin | | | | Biological degradation Sono-photo Fenton | [17] |
| Orteguaza River | Acetaminophen, Azithromycin, Carbamazepine, Ciprofloxacin, Clarithromycin, Clindamycin, Diclofenac, Erythromycin, | 2 | | Caqueta | Photo Fenton Electro Fenton Photo-electro Fenton | [12] |

| | | | | | |
|-----------------------|---|---|--------------------|---|------|
| | Irbesartan, Losartan, Metronidazole, Naproxen, Norfloxacin, Sulfamethoxazole, Tetracycline, Trimethoprim, Valsartan, Venlafaxine | | | Ultrasonic cavitation | |
| | Ibuprofen, Naproxen, Diclofenac, Aspirin, Ketoprofen, Caffeine, Hydrochloric acid, Lorazepam, Acetaminophen, Clarithromycin, Clindamycin, Doxycycline, Erythromycin, Carbamazepine, Irbesartan, Losartan, Metronidazole, Norfloxacin, Sulfamethoxazole, Trimethoprim, Valsartan, Venlafaxine | | | Bioremediation Phytoremediation Aerobic/anaerobic lagoons | [24] |
| Mensuli Ravine | Amoxicillin | 1 | Santander | Phytoremediation | [18] |
| Cauca River | Bezafibrate, Carbamazepine, Clofibrac acid, Diazepam, Diclofenac, Etofibrate, Fenofibrate, Atenolol, Betaxolol, Bisoprolol, Clenbuterol, Cyclophosphamide, Dimethyl amino phenazone, Aminotrizoic acid, Iohexol, Iomeprol, Amoxicillin, Chloramphenicol, Clarithromycin, Cloxacillin, Dapsone, erythromycin A, Dicloxacillin, Azithromycin, Furazolidone, Chlortetracycline, Ciprofloxacin, Doxycycline, Enoxacin, Estrone, Estriol | 2 | Valle del Cauca | Activated carbon Ozonation UV oxidation Coagulation- flocculation- sedimentation Chlorination | [19] |
| | Carbamazepine, Sildenafil | | | Wetlands (Phytoremediation) | [21] |
| Cañaverelejo River | Bezafibrate, Carbamazepine, Clofibrac acid, Diazepam, Diclofenac, Etofibrate, Fenofibrate, Atenolol, Betaxolol, Bisoprolol, Clenbuterol, Cyclophosphamide, Dimethyl amino phenazone, Aminotrizoic acid, Iohexol, Iomeprol, Amoxicillin, Chloramphenicol, Clarithromycin, Cloxacillin, Dapsone, erythromycin A, Dicloxacillin, Azithromycin, Furazolidone, Chlortetracycline, Ciprofloxacin, Doxycycline, Enoxacin, Estrone, Estriol | 1 | Valle del Cauca | Activated carbon Ozonation UV oxidation Coagulation- flocculation- sedimentation Chlorination | [19] |
| Cali River | Bezafibrate, Carbamazepine, Clofibrac acid, Diazepam, Diclofenac, Etofibrate, Fenofibrate, Atenolol, Betaxolol, Bisoprolol, Clenbuterol, Cyclophosphamide, Dimethyl amino phenazone, Aminotrizoic acid, Iohexol, Iomeprol, Amoxicillin, Chloramphenicol, Clarithromycin, Cloxacillin, Dapsone, erythromycin A, Dicloxacillin, Azithromycin, Furazolidone, Chlortetracycline, Ciprofloxacin, Doxycycline, Enoxacin, Estrone, | 1 | Valle del Cauca | Activated carbon Ozonation UV oxidation Coagulation- flocculation- sedimentation Chlorination | [19] |

| Estriol | | | | | |
|------------------|---|---|--------------------|---|------|
| Melendez River | Bezafibrate, Carbamazepine, Clofibric acid, Diazepam, Diclofenac, Etofibrate, Fenofibrate, Atenolol, Betaxolol, Bisoprolol, Clenbuterol, Cyclophosphamide, Dimethyl amino phenazone, Aminotrizoic acid, Iohexol, Iomeprol, Amoxicillin, Chloramphenicol, Clarithromycin, Cloxacillin, Dapsone, erythromycin A, Dicloxacillin, Azithromycin, Furazolidone, Chlortetracycline, Ciprofloxacin, Doxycycline, Enoxacin, Estrone, Estriol | 1 | Valle del Cauca | Activated carbon Ozonation UV oxidation Coagulation-flocculation-sedimentation Chlorination | [19] |
| Lili River | Bezafibrate, Carbamazepine, Clofibric acid, Diazepam, Diclofenac, Etofibrate, Fenofibrate, Atenolol, Betaxolol, Bisoprolol, Clenbuterol, Cyclophosphamide, Dimethyl amino phenazone, Aminotrizoic acid, Iohexol, Iomeprol, Amoxicillin, Chloramphenicol, Clarithromycin, Cloxacillin, Dapsone, erythromycin A, Dicloxacillin, Azithromycin, Furazolidone, Chlortetracycline, Ciprofloxacin, Doxycycline, Enoxacin, Estrone, Estriol | 1 | Valle del Cauca | Activated carbon Ozonation UV oxidation Coagulation-flocculation-sedimentation Chlorination | [19] |
| Aguacatal River | Bezafibrate, Carbamazepine, Clofibric acid, Diazepam, Diclofenac, Etofibrate, Fenofibrate, Atenolol, Betaxolol, Bisoprolol, Clenbuterol, Cyclophosphamide, Dimethyl amino phenazone, Aminotrizoic acid, Iohexol, Iomeprol, Amoxicillin, Chloramphenicol, Clarithromycin, Cloxacillin, Dapsone, erythromycin A, Dicloxacillin, Azithromycin, Furazolidone, Chlortetracycline, Ciprofloxacin, Doxycycline, Enoxacin, Estrone, Estriol | 1 | Valle del Cauca | Activated carbon Ozonation UV oxidation Coagulation-flocculation-sedimentation Chlorination | [19] |
| Pance River | Bezafibrate, Carbamazepine, Clofibric acid, Diazepam, Diclofenac, Etofibrate, Fenofibrate, Atenolol, Betaxolol, Bisoprolol, Clenbuterol, Cyclophosphamide, Dimethyl amino phenazone, Aminotrizoic acid, Iohexol, Iomeprol, Amoxicillin, Chloramphenicol, Clarithromycin, Cloxacillin, Dapsone, erythromycin A, Dicloxacillin, Azithromycin, Furazolidone, Chlortetracycline, Ciprofloxacin, Doxycycline, Enoxacin, Estrone, Estriol | 1 | Valle del Cauca | Activated carbon Ozonation UV oxidation Coagulation-flocculation-sedimentation Chlorination | [19] |
| Pamplonita River | Ibuprofen | 1 | Norte de Santander | Reverse osmosis Nanofiltration | [23] |

Conitunation of Table 6

| | | | | | |
|-------------|--|---|--------------------|---|------|
| Zulia River | Ibuprofen | 1 | Norte de Santander | Reverse osmosis Nanofiltration | [23] |
| Otun River | Ibuprofen, Naproxen, Diclofenac, Aspirin, Ketoprofen, Caffeine, Hydrochloric acid, Lorazepam, Acetaminophen, Clarithromycin, Clindamycin, Doxycycline, Erythromycin, Carbamazepine, Irbesartan, Losartan, Metronidazole, Norfloxacin, Sulfamethoxazole, Trimethoprim, Valsartan, Venlafaxine | 1 | Risaralda | Bioremediation Phytoremediation Aerobic/anaerobic lagoons | [24] |
| Sinu River | Ibuprofen, Naproxen, Gemfibrozil, Caffeine, Triclosan | 1 | Cordoba | Facultative lagoons | [13] |

3.2. Affected Watersheds

Of the 17 affected watersheds, those with the highest frequency of PEC were the Medellín, Bogotá, Tumaco, Ortegua, and Cauca rivers. These are rivers of great importance in Colombia; In the case of the Medellín River, Bogotá River and Tumaco Bay, they are bodies of water that cross the aforementioned cities, in which domestic wastewater discharges are generated; It is noteworthy that Tumaco Bay presents a network of different watersheds.

The Cauca River is one of the most important rivers in the country and, in addition, the most important among the affected watersheds, as it crosses several departments of Colombia, including the Cauca, Valle del Cauca, Risaralda, Caldas, Antioquia, Sucre, and Bolívar, which can drag and leave pharmaceutical compounds in their sludge, which can affect nearby residents and aquatic ecosystems. It is noteworthy that the pharmaceutical compound that the selected water

basins present in common is carbamazepine, which, according to the state of the art consulted, generates estrogenic effects, successively altering the genetics of species and causing serious hormonal alterations. The species *Pseudoplatystoma magdaleniatum* or striped catfish was analyzed in the Cauca River and Magdalena River basins, and the concentrations of the pharmaceutical compound carbamazepine are presented. These substances can bioaccumulate, which magnifies the problem in the food chain. Based on the above, the watersheds that were most affected were selected, considering the frequency of the reported PEC presence, which is presented in Figure 4.

However, because Tumaco Bay has a network of different water basins, no specific basin with this network was chosen. Figure 5 shows the locations of the basins in the countries where PECs have been reported.

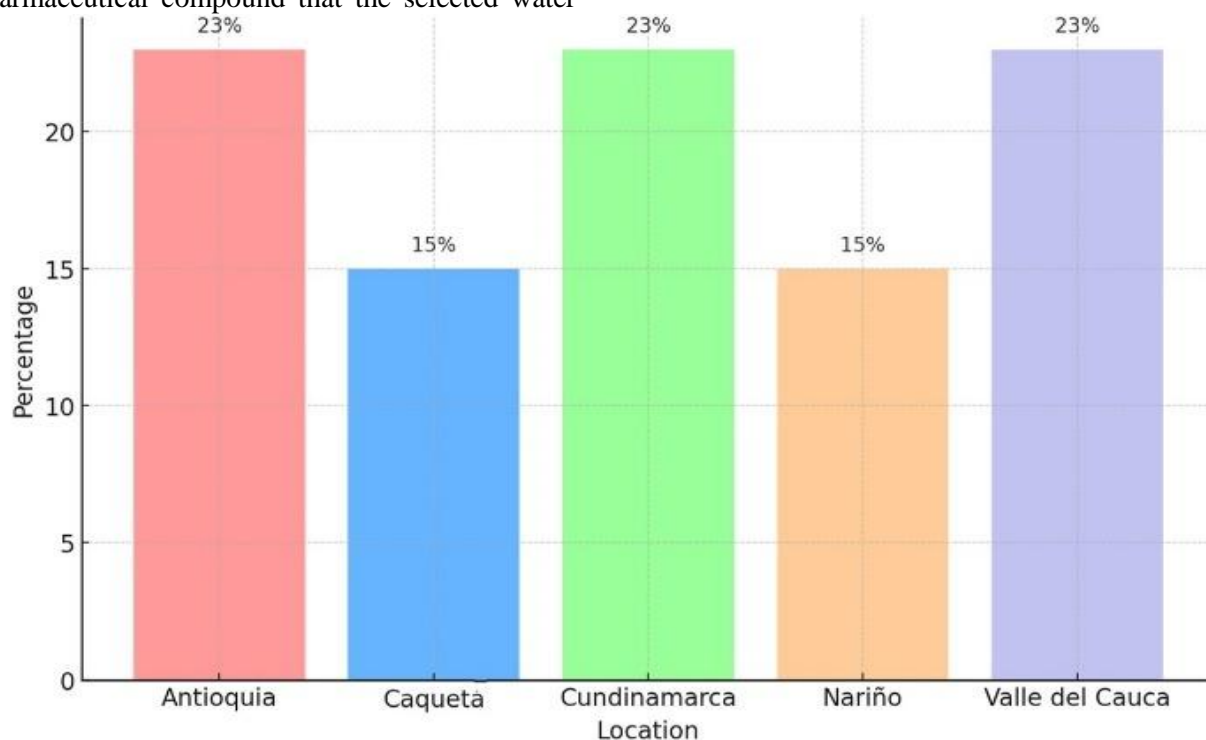


Figure 4. Main watersheds affected by pharmaceutical emerging contaminants in Colombia (authors' estimation)

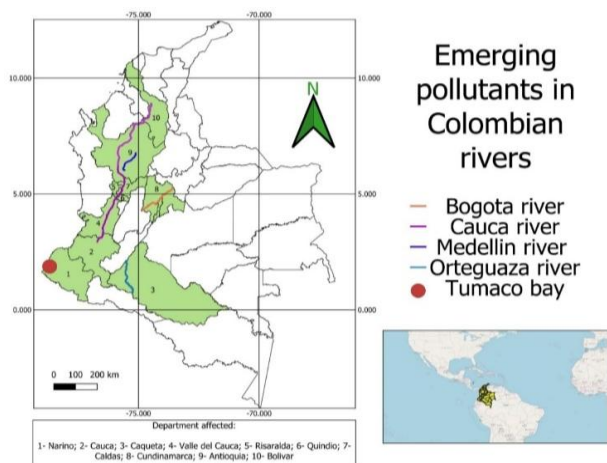


Figure 5. Watersheds in Colombia where pharmaceutical emerging contaminants have been reported (authors' estimation)

The five main bodies of water that have been most

affected and, at the same time, have been the greatest object of study in the country are four rivers: Orteguaza, Bogotá, Cauca, and Medellín, and a bay located in the city of Tumaco. Although Colombia has only five water bodies, 10 of the 32 departments in the country represent 31% of the national territory. It is necessary to mention that other bodies of water also present PEC, with the difference that they do not present a greater number of studies like the aforementioned watersheds.

3.3 Treatments for the removal of emerging contaminants raised for Colombia.

In studies carried out in the water bodies of Colombia, where PECs were present, different treatments were proposed for their removal, which are listed in Table 7.

Table 7. Suggested treatments for the removal of pharmaceutical emerging contaminants in the watersheds of Colombia (compiled by the authors)

| Type of treatment | Treatment | Treatment description | Author |
|-------------------|---------------------------|---|--------|
| Biological | Bio adsorption | It is a treatment which has the property that some biomolecules can concentrate and bind ions present in solutions, and that develops an affinity between the sorbate and the bio adsorbent. For this particular case, the authors mention the use of banana peel and cocoa | [16] |
| Chemical | Photo Fenton | It is a technology which uses Fe salts in conjunction with hydrogen peroxide that acts as a catalyst to form HO radicals, which in conjunction with solar radiation increases the oxidative power due to the photo reduction of Fe (III). | [12] |
| Chemical | Electro Fenton | It is a technology that uses Fe salts in conjunction with hydrogen peroxide that acts as a catalyst to form HO radicals, which using electrical energy significantly increases its efficiency because it favors the regeneration of Fe ²⁺ and increases the number of OH radicals | [12] |
| Chemical | Photo-electro Fenton | It is a technology that uses Fe salts in conjunction with hydrogen peroxide that acts as a catalyst to form HO radicals that, combining electrical energy and solar radiation, makes oxidation more efficient | [12] |
| Chemical | Ultrasonic cavitation | It is the formation of vapor bubbles in the liquid when considerable decreases in the sound wave front (rarefaction) occur. These waves increase in size and finally when the sound field is compressed, these cavities or bubbles collapse and lead to the release of energy. Under conditions of increased temperature and pressure, reactions occur that form free radicals H ⁺ and OH ⁻ | [12] |
| Biological | Bioremediation | It is a process by which living organisms are used that have the capacity to degrade and transform contaminants through fungi or bacteria | [24] |
| Biological | Phytoremediation | It is the process in which plants are used to remove contaminating loads | [24] |
| Biological | Aerobic/anaerobic lagoons | Anaerobic lagoons are those in which the degradation of organic matter occurs in the absence of oxygen. Aerobic lagoons degrade organic matter through the aerobic activities of bacteria and algae | [24] |
| Physical | Ultraviolet light | It is electromagnetic radiation with wavelengths between 200 and 400 nm for the elimination of microorganisms and pathogens | [20] |
| Physiochemical | Sono Fenton | It is a technology that uses Fe salts in conjunction with hydrogen peroxide that acts as a catalyst to form HO radicals, which with the help of ultrasound waves facilitate the regeneration of Fe (OOH) ²⁺ , giving rise to Fe ions. (II) | [22] |
| Physiochemical | Sono-photo Fenton | It is a technology that uses Fe salts in conjunction with hydrogen peroxide that acts as a catalyst to form HO radicals that, with the help of ultrasound waves and solar radiation, significantly increase oxidation | [22] |

Continuation of Table 7

| | | | |
|----------------|--|---|------|
| Physiochemical | Activated carbon | It is a method which has a high removal capacity due to the definition of the size of its pores, in addition to its chemical structure influencing the interaction with polar and non-polar adsorbates, which allow different chemical reactions that help to retain different pollutants | [19] |
| Chemical | Ozonation | It is a compound with a high oxidation potential at 25°C, which allows parameters such as COD to be significantly reduced by up to 90% | [19] |
| Chemical | Chlorination | It consists of the application of chlorine to eliminate organic matter and ammonia, which form chloramine compounds and organic chlorine compounds and, if more chlorine is added, completely eliminates these compounds. | [19] |
| Physiochemical | Coagulation-flocculation-sedimentation | Coagulation and flocculation is a chemical method. In coagulation, some compound is applied that neutralizes charges and can form a mass that has a good density. Flocculation causes the particles to form larger masses that, due to their weight, can easily settle | [19] |
| Physiochemical | Reverse osmosis | It is the treatment that consists of applying pressure to the solution that has the highest concentration, which reverses the natural flow and the water to be treated flows from the most concentrated to the least concentrated. | [23] |
| Physical | Nanofiltration | It is a technique which consists of using a membrane with pores smaller than 1nm, which require pressures between 10 and 50 bars. It is capable of retaining even minor species between 200 and 300g/mol | [23] |
| Biological | Facultative lagoons | They are reservoirs, into which wastewater enters, and are treated biologically by means of algae and bacteria, having anaerobic and aerobic oxidation zones for the stabilization of organic matter. | [13] |

Several advanced treatments have been presented for the elimination of PEC, including biological, chemical, physical, and physicochemical properties. These include bio-adsorption, photo-Fenton, electro-Fenton, ultrasonic cavitation, ozonation, chlorination, reverse osmosis, and nanofiltration. Coagulation, flocculation, sedimentation, and physicochemical treatments that are present in conventional wastewater treatment plants reduce PEC [29].

Biological processes have been shown to remove many PECs [30]. PEC biodegradation studies have gained a lot of interest, as they are processes that allow for high PEC removal efficiency [31] and have also been reported, with a higher percentage of removal in hybrid treatments that combine biological and physicochemical treatments [29].

The treatments proposed for the removal of PEC in different water basins of Colombia have a theoretical efficiency that can be used for their qualification. The theoretical efficiencies of these treatments have been established by [7] and other authors such as [32-46].

It should be noted that the efficiencies of the

treatments depend on the characteristics of the wastewater; therefore, additional studies have reported efficiencies in the removal of both EC and heavy metals, as the latter presented greater information reported and had a relationship with pharmaceutical compounds [47].

The highest theoretical efficiencies for EC removal occur in advanced treatments, such as microfiltration and reverse osmosis, ozonation and catalytic ozonation, photolysis, bio-membrane, Fenton, UV/TiO₂, and ultrasound. For conventional treatments, the highest removal efficiencies occurred in chlorination, activated sludge, and sand filters. The best efficiency range is recorded in the hybrid treatment of microfiltration and reverse osmosis, with values ranging between 91 and 100%, which remove the majority of EC in the water [48].

The operating costs considered for the different water treatments with EC, listed in Table 8, were established according to what was reported in the consulted information.

Table 8. Operational costs of treatments for the removal of emerging contaminants (compiled by the authors)

| Treatment | Cost USD/m ³ * | Reference |
|-----------------------|---------------------------|-----------|
| Bio adsorption | 5.03 | [49] |
| Photo Fenton | 20.83 | [50] |
| Electro Fenton | 2.52 | [51] |
| Photo-electro Fenton | - | - |
| Ultrasonic cavitation | 20.13 | [52] |
| Bioremediation | - | - |
| Phytoremediation | 1.96 | [53] |
| Facultative lagoons | 0.34 | [45] |
| Ultraviolet light | - | - |
| Sono Fenton | - | - |

| Continuation of Table 8 | | |
|--|------|------|
| Sono-photo Fenton | - | - |
| Activated carbon | 0.19 | - |
| Ozonation | 0.20 | [54] |
| Chlorination | 0.32 | - |
| Coagulation-flocculation-sedimentation | 0.29 | - |
| Reverse osmosis | 0.28 | [55] |
| Nanofiltration | 0.76 | [55] |

Note: *Costs in Colombian pesos (COP) converted to USD.

The highest EC removal costs were recorded for the photo-Fenton and ultrasonic cavitation treatments, with a significant difference compared to the costs of the other treatments. The lowest EC removal costs occurred in activated carbon and ozonation treatments.

The high costs of the photo-Fenton treatment are associated with the incorporation of energy sources to optimize the process [56]. The treatments for EC removal were qualified according to the criteria of theoretical efficiency, amount of environmental impact, type of treatment, and costs. The rating matrix is presented in Table 9.

The best-evaluated wastewater treatments were activated carbon, ozonation, nanofiltration, ultrasonic cavitation, photo-Fenton, and reverse osmosis. The treatment with the highest rating was reverse osmosis.

Currently, wastewater treatment systems have a succession of steps, which generally begin with the implementation of a grease trap system, a system for the removal of suspended solids and finally a system for the decomposition of organic matter in the water. Once these steps are completed, the water is poured [46]. However, wastewater treatment systems vary in their characteristics.

Table 9. Treatment qualification matrix for the removal of emerging contaminants (compiled by the authors)

| Treatment | Theoretical efficiency | Environmental impact | Type of equipment | Cost | Score |
|--|------------------------|----------------------|-------------------|------|-------|
| Bio adsorption | 2 | 1 | 1 | 1 | 5 |
| Photo Fenton | 3 | 2 | 2 | 1 | 8 |
| Electro Fenton | 3 | 1 | 2 | 1 | 7 |
| Photo-electro Fenton | 3 | 1 | 2 | 1 | 7 |
| Ultrasonic cavitation | 2 | 3 | 2 | 1 | 8 |
| Bioremediation | 1 | 2 | 1 | 2 | 6 |
| Phytoremediation | 1 | 2 | 1 | 2 | 6 |
| Facultative lagoons | 1 | 2 | 1 | 2 | 6 |
| Ultraviolet light | 1 | 2 | 2 | 1 | 6 |
| Sono Fenton | 3 | 1 | 2 | 1 | 7 |
| Sono-photo Fenton | 3 | 1 | 2 | 1 | 7 |
| Activated carbon | 2 | | 2 | 2 | 8 |
| Ozonation | 1 | 3 | 2 | 2 | 8 |
| Chlorination | 1 | 3 | 1 | 2 | 7 |
| Coagulation-flocculation-sedimentation | 1 | 2 | 1 | 2 | 6 |
| Reverse osmosis | 3 | 3 | 2 | 2 | 10 |
| Nanofiltration | 3 | 3 | 2 | 2 | 9 |

Considering that these treatments remove such specific compounds, they are classified as tertiary treatments [57], which are the last treatments applied for the management of wastewater. Theoretically, these treatments have very high efficiencies, but only if they have the appropriate technology and the corresponding maintenance to preserve the systems. Figures. 6-11 present the design of the treatment systems for the control of PEC.

3.3.1. Nanofiltration

It is a physical methodure that involves the implementation of a membrane to remove pharmaceutical compounds. Nanofiltration is an alternative that shows great potential for advanced

water treatment, especially to eliminate EC, including pharmaceutical EC [58]. This treatment has a theoretical efficiency that ranges from 91% to 100%, which makes it an attractive method for removing not only pharmaceutical compounds, but other contaminants [59]. Regarding its operating and maintenance costs, it is only \$3.089 COP/m³, and its investment value can have values ranging from 1.06 million dollars to 1,77 million dollars, according to the volume to be treated (between 1000 m³, up to 3000 m³ daily) which requires a high initial investment [55]. Table 10 lists the advantages and disadvantages of nanofiltration.

Table 10. Advantages and disadvantages of nanofiltration (compiled by the authors)

| Advantages | Disadvantages |
|--|---|
| High removal efficiency | High content of suspended solids must be removed to avoid deterioration of the membrane |
| Removal of particles between 0.01 to 0.001 μm | Constant change of the membrane Specific conditions for correct operation |



Figure 6. Scheme of the nanofiltration wastewater treatment system (developed by the authors)

3.3.2. Ultrasonic Cavitation

This technique consists of the application of ultrasound waves ranging from 20 kHz to 500 kHz [60], generating, in this way, bubbles as a result of the tension caused by the sound, which burst and at the

same time, decomposing the various compounds that are present in the water. Different studies have suggested that the ultrasonic cavitation effect improves dispersion and mass transfer [61]. Table 11 shows the advantages and disadvantages of this treatment.

Table 11. Advantages and disadvantages of ultrasonic cavitation (compiled by the authors)

| Advantages | Disadvantages |
|-----------------|-------------------------------|
| Easy operation | High operating cost |
| Long durability | Constant inlays |
| | Specific operating conditions |

Although it is an innovative technique to implement in the wastewater treatment systems of Colombia, its efficiency range is quite variable, ranging from 24% to 99%, which is not a viable alternative because it does not guarantee that the dumping conditions are always optimal. In addition, it has high operating costs of approximately \$81.425 COP/m³ [52], which makes its application in a large population economically unviable.

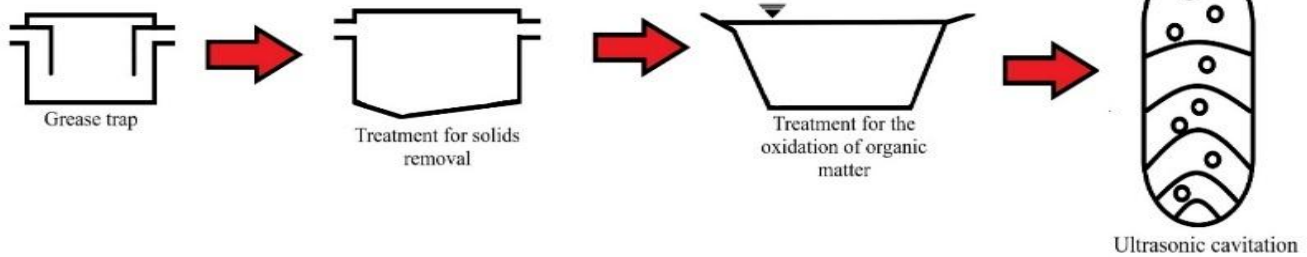


Figure 7. Scheme of a wastewater treatment system with ultrasonic cavitation (developed by the authors)

3.3.3 Photo Fenton

It consists of the application of hydrogen peroxide together with an iron (II) salt, which forms OH-radicals, where the different compounds remain attached to the free radicals, which helps remove the various contaminant loads that the water can present. Fenton oxidation (based on H₂O₂) has shown good potential for the degradation of pharmaceuticals [62]. In addition to the application of these compounds, solar

radiation can be used as a catalyst for the decomposition of hydrogen peroxide, thereby accelerating the process. Although it has efficiencies ranging from 80% to more than 95%, its cost of \$84.267 COP/m³ makes it a high-cost treatment; thus, its application to large volumes of wastewater may be economically unviable [50]. Table 12 lists the advantages and disadvantages of applying this treatment.

Table 12. Advantages and disadvantages of photo Fenton (compiled by the authors)

| Advantages | Disadvantages |
|--|---|
| It is carried out at room pressure and temperature | Does not achieve complete mineralization of the contaminants present in the water |
| Iron is an abundant element | Strict pH control is required Generation of byproducts and residual sludge |
| The use of solar radiation makes it more efficient and economical to apply | Constant investment in inputs |
| Easy handling of hydrogen peroxide | |

Although the application of photo Fenton as a wastewater treatment is a viable alternative because of its high efficiency (between 80 and 95%) and its broad advantages, the generation of sludge and byproducts is

a highly important aspect, because the sludge generated in the primary treatment must also be added, which, in Colombia, continues to be a problem due to the poor final disposal that may be given.

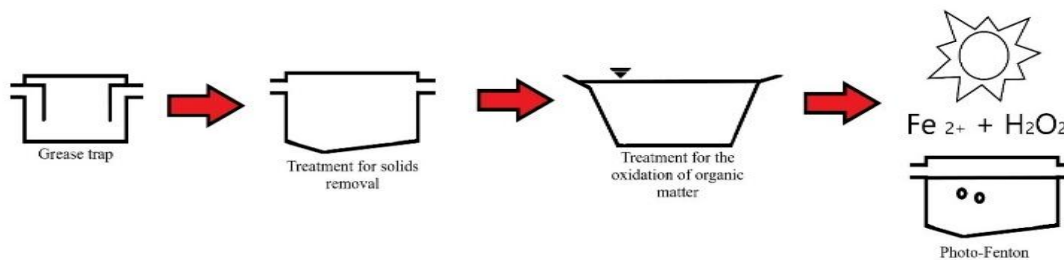


Figure 8. Schematic of a wastewater treatment system with photo-Fenton (developed by the authors)

3.3.4. Reverse Osmosis

This technique uses a semipermeable membrane through which the fluid is decontaminated, where it passes from a region that has a high concentration to another where it has a lower concentration. To perform reverse osmosis, a pressure greater than osmotic pressure must be applied. Although it has an approximate cost of \$1.133 COP/m³ [63], it may require high initial investment. Reverse osmosis is excellent for efficiently removing low-molecular-weight contaminants, making it a promising treatment for the removal of PECs, such as antibiotics and estrogens [64]. Table 13 shows the advantages and disadvantages of reverse osmosis.

Table 13. Advantages and disadvantages of reverse osmosis (compiled by the authors)

| Advantages | Disadvantages |
|----------------------------------|--------------------------------------|
| High removal efficiency (75-97%) | Generation of solid waste due to the |
| Removal of heavy metals | constant change of membranes |
| Low cost in terms of labor | High investment cost |

According to the articles consulted, osmosis is classified as a high-cost treatment; at the same time, it has very high efficiencies for removing compounds that can cause significant impacts to the environment and is considered an optimal treatment for wastewater.

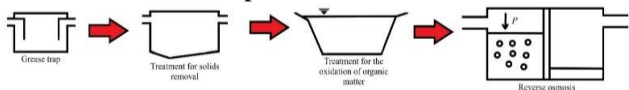


Fig. 9 Scheme of a reverse osmosis wastewater treatment system.

Although the membranes have to be constantly changed, which has a useful life of at least three months according to their use, they can be compensated again in the high efficiency of removing compounds, in this case, compound pharmacists.

3.3.5. Activated Carbon

It is a physicochemical treatment, which consists of the removal of mainly organic compounds, which helps to improve organoleptic properties, thanks to the fact that its chemical structure interacts with polar and non-polar adsorbates, which helps to retain different contaminants. Although its operating cost is low, approximately \$789 COP/m³, it has a fairly wide

efficiency range, ranging from 61% to 83%, which does not ensure the complete removal of PEC. However, it has been established that, in water treatment, activated carbon adsorption is suitable for the removal of pharmaceutical compounds such as diclofenac and carbamazepine [65]. Table 14 presents the advantages and disadvantages of the activated carbon.

Table 14. Advantages and disadvantages of activated carbon (compiled by the authors)

| Advantages | Disadvantages |
|--------------------|---------------------------------------|
| Low operating cost | Generation of waste water by backwash |
| | By-product generation |

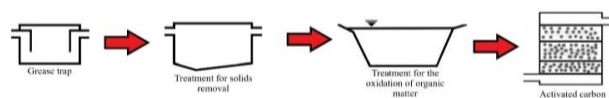


Figure 10. Scheme of a wastewater treatment system with activated carbon (developed by the authors)

3.3.6. Ozonation

This technique involves applying ozone gas to water to eliminate microorganisms, leaving the water disinfected. This treatment makes it possible to significantly reduce COD parameters by up to 90% at an operating cost of approximately \$824 COP/m³, making it an economically viable treatment. However, its wide range of efficiencies makes

It is not a reliable treatment, as it ranges from 1% to 99%. The removal of EC with ozone has been documented for 23 pharmaceutical compounds, with efficiencies of up to 80% [66]. Table 15 lists the advantages and disadvantages of ozonation.

Table 15. Advantages and disadvantages of ozonation (compiled by the authors)

| Advantages | Disadvantages |
|--------------------|----------------------------|
| Low operating cost | Generation of by-products |
| | Very wide efficiency range |

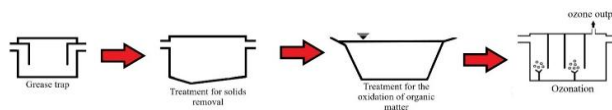


Figure 11. Scheme of a wastewater treatment system with ozonation (developed by the authors)

Of the six selected treatments, nanofiltration, reverse osmosis, and activated carbon presented the greatest viability in terms of operating costs, mainly activated carbon, which has a lower value than reverse osmosis and nanofiltration; however, its wide range of efficiency makes it an unreliable treatment compared to reverse osmosis and nanofiltration, in terms of removal efficiency. In this case, reverse osmosis is the most feasible treatment because its cost is lower than that of nanofiltration, and it has good removal percentages, similar to nanofiltration. These treatments can be applied in treatment systems such as the El Salitre wastewater treatment plant (WWTP) (Río Bogotá), Aguas Claras EPM WWTP, Bello WWTP (Río Medellín), Cañavalejo WWTP, Yumbo WWTP (Río Cauca), and Heliconias WWTP (Río Orteguzaza), which are water basins that have a greater presence of these compounds.

At the institutional level in Colombia, IDEAM and the Alexander Von Humboldt Biological Resources Research Institute, according to the information consulted, do not refer to ECs of any kind, reflecting a gap in knowledge on the part of these entities targets have been set at the country level for sustainable management of water resources [67]. Nevertheless, INVEMAR [68] evaluated the presence of microplastics and pesticides in the coastal areas of the country, which are considered EC. It is important to note that they are not only present in freshwater bodies but also in the seas and oceans, altering the marine ecosystem and magnifying the problem.

4. Conclusion

More than 100 emerging pharmaceutical contaminants have been reported in watersheds in Colombia. The most common drug was carbamazepine, followed by clarithromycin, erythromycin, and diclofenac. The largest number of PECs related to research belonged to the antibiotic category. It is noteworthy that the selected articles were contemporary with the COVID 19 pandemic, which may be a product of the self-medication generated during the contingency, even increasing the sales of antibiotics such as clarithromycin and erythromycin, which are used for lung infections.

The Colombian watersheds identified in this research with a greater presence of PEC have been large cities, such as Bogotá and Medellín, and the Cauca River basin. However, it is possible that they do not have a higher content of pharmaceutical compounds, but rather have been the subject of further study, so there is more information regarding their PECs. These PECs can occur in small city basins, which can even generate discharges with a higher concentration of these pollutants because some do not have wastewater treatment systems. Therefore, it is

necessary to conduct research that reports the presence of PEC in small watersheds in the country.

Only the Ministry of Environment and Sustainable Development has a plan for the control of emerging contaminants in its National Strategic Plan for Environmental Research between 2021 and 2030. However, no other government entity has implemented or planned actions to control emerging contaminants of any type, which is why it is necessary to reinforce the current regulations, thus leading to an improvement in quality of the environment and human health, which are exposed because emerging contaminants still do not have control in Colombia.

Among the treatments for the removal of PEC, activated carbon is one of the most economical, and does not present high removal efficiencies. Reverse osmosis and nanofiltration are treatments with better removal of these contaminants. Not enough information has been found regarding pilot-scale or real-scale applications of treatments for the removal of PEC in the country, which is required to determine the real efficiency of these treatments.

It is necessary for health-promoting institutions to implement education strategies for citizens in general regarding self-medication so that in addition to preserving the patient's health, the generation of discharges of pharmaceutical compounds is reduced.

This study contributes to strengthening knowledge in the environmental and public health fields and provides information that can be considered for the formulation of public policies. In addition, it contributes to the analysis of the applicability of treatment solutions in different contexts, such as in the case of Colombia, as well as their costs, advantages, and disadvantages.

Experimentation with different advanced treatment systems at the pilot or full scale is necessary to determine the different removal capacities of treatment systems, since many of the studies reviewed were carried out under laboratory conditions. It is proposed to combine conventional and advanced methods to determine their efficiencies when removing emerging pharmaceutical contaminants, thus providing information for future research.

Declarations

Author Contributions

Conceptualization, M.E.B.-G. and J.D.S.-P.; methodology, M.E.B.-G., M.M.-M. and A.M.R.-C.; formal analysis, M.E.B.-G. and M.J.V.-O.; writing—original draft preparation, all authors contributed equally; writing—review and editing, M.E.B.-G.; visualization, S.S.-L.; project administration, M.E.B.-G. All authors have read and agreed to the published version of the manuscript.

Data Availability Statement

The data presented in this study are available in this article.

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Conflicts of Interest

The authors declare that there is no conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancies have been completely observed by the authors.

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