



Review Article

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Pharmaceutical and Personal Hygiene Products (PPcPs): A Threat Little Studied in Colombian Waters



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Abstract

Pharmaceutical and personal care products (PPcPs) are common expended in modern society. One of the main input sources of these pollutants into the environment is the wastewater, those in Latin America and currently in Colombia is not strictly treated. The objective of the present review was to make a synthesis about straightaway information in Colombia about PPCPs through bibliographic search in scientific sources as Scopus and web of science. For this purpose, is being argued about PPCPs persistence and risk in aquatic ecosystems. Finally, it was found that diclofenac, carbamazepine, naproxen and ibuprofen are the pharmaceuticals (PPcPs) most frequently found in water. In addition, there are cities which do not have systems to treat wastewater hence the situation is even worse being that any pollutant is treated before reaching surface waters such as rivers.

Keywords: PPCPs; Emergent pollutants; Pharmaceutical products; Wastewater (Figure1)

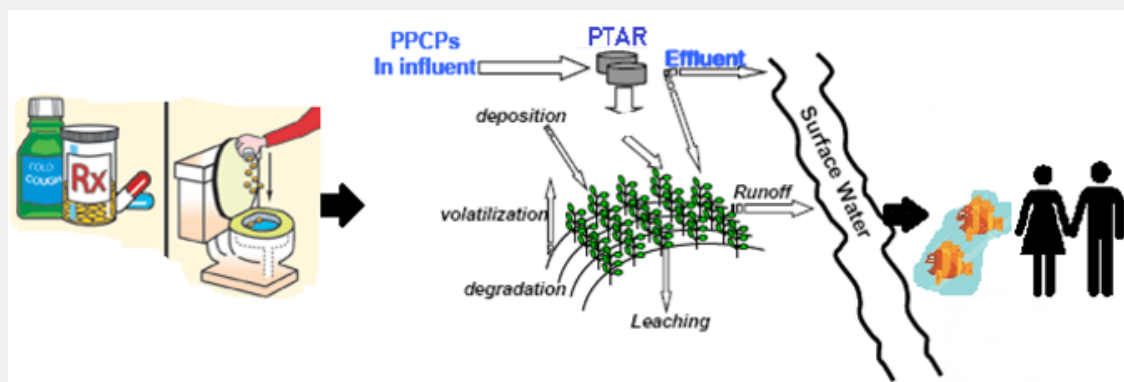


Figure 1

Introduction

Pharmaceutical and personal hygiene products (PPcPs) belonging to the group of so-called emerging organic pollutants are a threat little studied in Colombia and in general in Latin American countries, while in developed countries they are part of a wide range of investigations due to the alleged consequences generated by its interaction with organic matter and with living beings through the food chain. That is why, during the last 15 years its presence in water and soil has been identified and quantified. In this way, they are today known as potentially dangerous pollutants that can generate damage still unknown to ecosystems

and therefore to living beings [1]. Today, PPcPs are widely used by modern society. In the case of drugs, they are therapeutic drugs used to prevent or treat human and animal diseases; some of them, such as anti-inflammatories, antibiotics or diuretics, are designed to generate a therapeutic effect on the body, crossing biological membranes and activating at very low concentrations [2], which vary from 1/ng to 1/μg, representing seemingly little risk. On the other hand, there are the synthetic and chemical products used to make fertilizers, fragrances, soaps, toothpastes, deodorants, household cleaning products, detergents, softeners, shampoos, shaving lotions or in general personal care or hygiene products,

which have a high consumption rate in Colombia and around the world, so there is an increase in the presence of PPcPs in different environments such as water, sediments and biota [3,4].

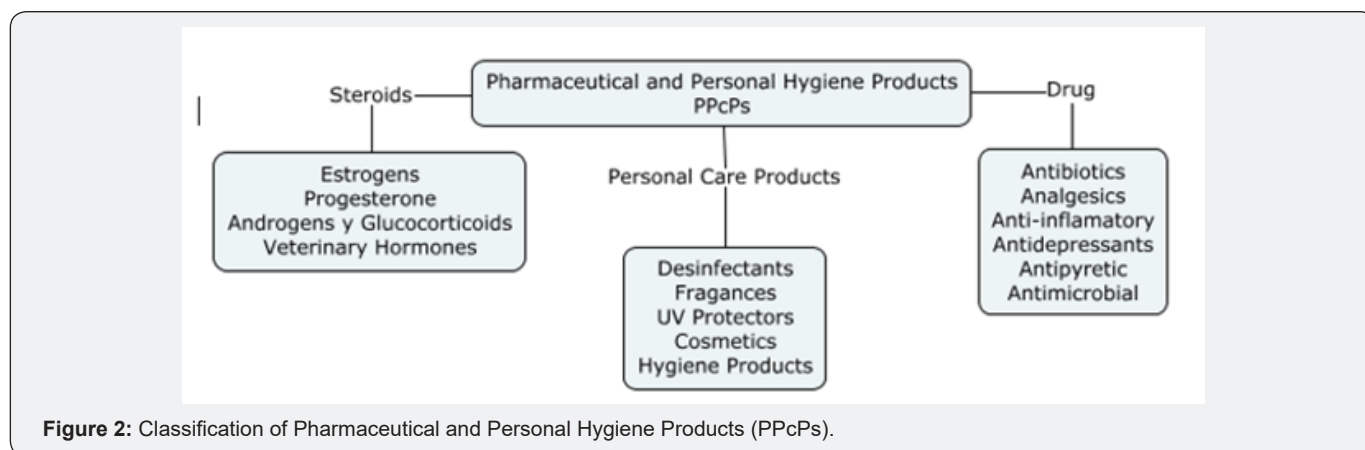
One of the main sources of entry of these substances into the environment is generally hospital wastewater, agricultural or industrial waste and domestic activities through the urine and feces of humans or animals that consume or use them [4-6], from there they are transported to the wastewater treatment plants (WWTP) which in Latin America and specifically in Colombia are not currently designed to treat this type of substances [1,7,8], therefore, they are mobilized through its effluents to various bodies of water such as streams, streams, groundwater, rivers or seas. In Europe alone, it is estimated that more than 300 different pharmaceutical components are used and many of them are likely to reach the water cycle [9], in that case, reference is only made to drugs, without taking into account the products of grooming and personal hygiene, which are increasingly common, reaching the point of being essential for all human beings regardless of where in the world they are (developed or developing countries). In this way, the amount of pollutants that are thrown minute by minute through the sewer systems to the various water sources around the world, can affect living beings and possibly generate effects still unknown to humans.

Since these pollutants are not completely mitigated by the WWTPs, they are discharged into the aquatic environment, tending to accumulate for long periods of time in the ecosystems; thus allowing an increase in their concentrations, intensifying through the trophic chain through biomagnification processes [10], generating various types of mixtures between them; increasing the levels of contamination of water and soils and generating, in addition, transformation products or metabolites, which in the specific case of drugs become even more dangerous than the original compounds. In this way, these bioaccumulative

pollutants can generate mutagenic effects, endocrine alteration and antibiotic resistance in the organisms to which they are exposed [11]. As PPcPs have been reported separately or in admixture as found in the environment, they generate numerous adverse effects on the behavior and physiology of various types of organisms. In this way, authors report to: *Daphnia magna*, *Dreissena polymorpha*, *Danio rerio* and *Gammarus pulex* as some of the most used species as biomarkers to carry out these studies [12-15]. These species are used since they have a variety of the basic physiological characteristics found in vertebrates, so that, the assessment of certain physiological parameters in these simple organisms, allows an estimation of the effects of these pollutants in more complex organisms [16].

Regarding its elimination, there are biological, chemical and / or physical techniques that allow its removal in different proportions; However, in the WWTP of developing countries such as Colombia, these techniques are not used in their entirety, generating a previously unknown accumulation of such pollutants in ecosystems. Yang et al. [17] report a low efficiency of elimination of PPcPs from conventional wastewater treatment plants in European and Asian countries, because the most used treatment system is secondary, which is designed primarily to remove organic matter and solids Suspended, not such pollutants. As mentioned earlier, in Colombia there are few wastewater treatment systems whose operation is carried out within international standards [7] in addition, prior treatments in hospital waters are necessary before reaching the sewer systems of all Colombian cities. The objective of this review is to make a synthesis about the information that is available in Colombia on pharmaceutical and personal hygiene products PPcPs, through a bibliographic search in scientific sources such as Scopus and Web of Science. For this, we discuss the persistence and risk of PPcPs in aquatic ecosystems.

What are pharmaceutical and personal hygiene products (PPcPs)?



In the last decade pharmaceutical and personal hygiene products (PPcPs) have been recognized as emerging pollutants due to their persistent presence in aquatic environments; This has been demonstrated and proven according to scientific publications.

The term PPcPs refers to any personal care, grooming or medicinal product for humans and animals. [7,8,17-21] report that these pollutants they have been found at different concentrations and mixtures in surface, underground, residual and even drinking

water in concentrations that vary from (ng /L) to (µg /L) and their elimination by conventional systems is low. They can be classified into several groups depending on their properties and functions (Figure 2): steroids (estrogens, progesterones, veterinary growth hormones, androgens and glucocorticoids); drugs (antibiotics, analgesics, anti-inflammatories, antidepressants, antipyretics, antimicrobials); and personal care and hygiene products (disinfectants, fragrances, UV protectors, cosmetics, personal hygiene). With regard to drugs, most of these are weak acids and bases, so that their distribution in the aquatic environment once they are discharged depends on the pH and the acidity constant of the medium [22], which indicates their potential for bioaccumulation in aquatic organisms, persistence or toxicity.

Persistence of PPcPs

Persistence is defined as the duration, existence or permanence of something, in this particular case about the fate of PPcPs once they are released to the environment. The physicochemical properties of many PPcPs mean that they cannot be easily eliminated through conventional wastewater treatments, which are managed in a high number of WWTPs in Latin America and Colombia, generating uncertainty about the final destination of these substances, as they remain in the ecosystem, in different links of the food chain

(Figure 3) and therefore have a high probability of returning to the human being through food or water; generating consequences still unknown to health, because this type of research is relatively new. The persistence of these substances varies depending on their chemical structure, as some can easily be transformed by natural processes such as photodegradation, biodegradation or absorption by plants or microorganisms; However, the consumption of PPcPs in Colombia and in the world is so high that their release into the environment is equivalent to tons of these substances per day and of these, the drugs are the most commonly found PPcPs in aquatic environments [23-25]. Due to the complexity in their chemical structures, the active ingredients used in the manufacture of medicines and eventually their bio transformed metabolites, they can become persistent entities in ecosystems [24]. According to the dissipation time in water/sediment samples Loffler et al. [25] categorized the persistence of some pharmaceutical products in low, medium and high, resulting in paracetamol and ibuprofen being in the low persistence category; considered as moderate persistence are: oxazepam, iopromide, and ivermectin; and finally clofibric acid, diazepam and carbamazepine as substances of high persistence. Which means that these last three pharmaceutical products tend to remain in the environment for long periods of time, accumulating and intensifying through the food chain.

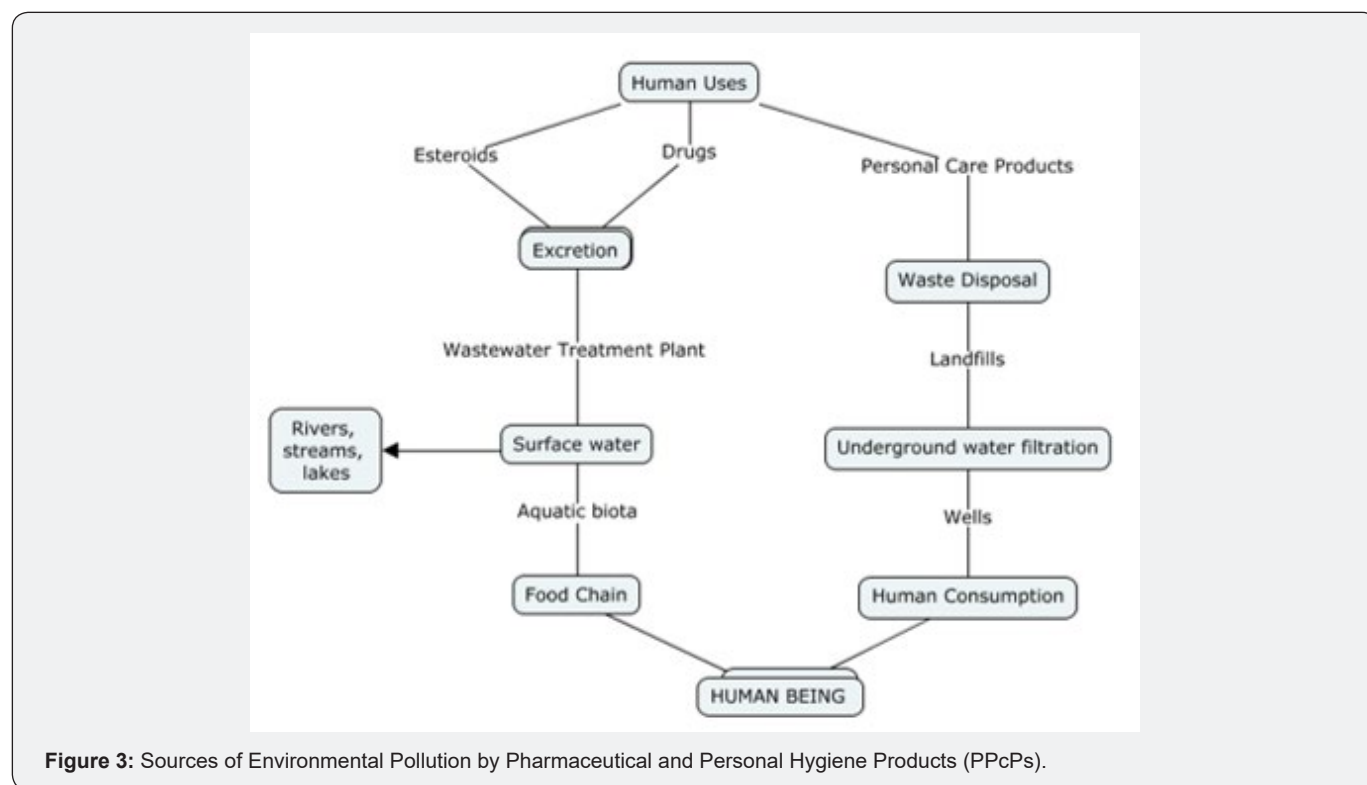


Figure 3: Sources of Environmental Pollution by Pharmaceutical and Personal Hygiene Products (PPcPs).

Toxicity of (PPcPs)

Toxicity refers to the ability of any substance to damage or produce harmful effects on a living being. Steroids and drugs are specifically designed to maximize their biological activity at low concentrations and point to certain metabolic, enzymatic

or cellular signaling mechanisms; In spite of this, these can be pharmacologically activated in non-target organisms such as aquatic biota, which are unintentionally exposed to their natural environment [26]. Once pharmaceutical products such as antibiotics, analgesics, anti-inflammatories, antidepressants, antipyretics or antimicrobials are discharged into aquatic

environments, unknown mixtures of these are formed, affected individually or jointly to the organisms. Such is the case of the transformation products that are generated after a series of natural physical, chemical and biological processes and can become highly toxic [27]. Authors such as [5,18,23,26] report bacterial resistance due to the excessive use of antibiotics in human and animal medicine and their non-elimination in WWTPs.

Treatments used to remove PPCPs from wastewater

The impact of human activities on the aquatic environment increases due to population growth; which has intensified the discharge of hospital, domestic, industrial and agricultural waters, these being the main source of pollution. Globally there are more and more efforts made to conserve the environment and therefore, regulations are increasingly demanding. It should be noted that the treatment of wastewater is mandatory before being discharged to the aquatic environment, for this, primary, secondary or tertiary treatment techniques are executed depending on its final destination, where various types of biological, physical techniques are carried out and / or chemicals to remove all types of contaminants.

Biological treatments (bioremediation-phytoremediation): Biological degradation treatments are considered the most important within the mechanisms of elimination of organic contaminants. Resistant microorganisms have the facility to use PPCPs as a source of carbon and energy, thus allowing efficient and environmentally friendly degradation; and in some cases, different microorganisms are associated to eliminate contaminants [28]. Lin et al. [29] showed that strains CE21 and CE22 of *Pseudomonas* sp isolated from activated sewage sludge from treatment plants are capable of removing 75% cephalixin in 12 hours. Domaradzka et al. [30] report that *Planococcus* sp. It has the ability to degrade 75.14±1.71 of naproxen to an initial concentration of 6mg / l in the presence of glucose as an additional source of carbon. [31-33] show that Beta-proteobacteria and Planctomycetes; *Stenotrophomonas maltophilia* KB 2 and *Trametes versicolor* respectively, have the ability to degrade naproxen. While Rodarte-Morales et al. [34] suggest the *Phanerochaete chrysosporium* fungus to completely degrade diclofenac and ibuprofen under different aeration conditions. The enzymatic induction of microorganisms is the key to the biodegradation of PPCPs, since their degradation depends on the ability of microorganisms to produce the specific enzyme that breaks them down; for example, triclosan can induce European *Nitrosomonas* to produce ammonium monooxygenase, which breaks down triclosan [35]. On the other hand, there is phytoremediation as a biological technique that allows soil decontamination or sewage treatment, due to the restorative capacity of some plants [17]. This technique includes a set of methods responsible for degrading, assimilating, metabolizing or detoxifying contaminants; through on-site treatments [36]. Phytoremediation is based on the naturally occurring processes by which rhizospheric plants and microorganisms degrade and sequester organic and inorganic contaminants [37]. Rezanian et al.

[38] report to *Eichhornia crassipes*, because of their high capacity to absorb water contaminants, Akinbile & Monh [39] suggest *Pistia stratiotes* and *Eichhornia crassipes* to reduce water turbidity with an effectiveness of 92.70% 87.05% respectively. *Brachiaria mutica*, has the potential to remove organic and inorganic contaminants from water effluents, in particular from wetlands [40]. Hijosa-Valsero et al. [41] report the adsorption of PPCPs by the roots of the *T. angustifolia* and *P. australis* macrophytes in a WWTP in Spain, with caffeine and galaxolide being the most adsorbed. Chehrenar et al. [42] report the elimination of ibuprofen by the *Epipremnum aureum* plant enhanced with ferrous iron. Finally, Lin & Li [37] report elimination of carbamazepine, ibuprofen, sulfamethoxazole sulfadiazine and triclosan by *Eichhornia crassipes* and *Pistia stratiotes*, although the high concentrations of these PPCPs cause toxic effects in these species, reducing their elimination efficiency. In this way, different types of microorganisms and plant species are able to recover environments contaminated with PPCPs, which is interesting for future research on bioremediation and phytoremediation techniques.

Physical and / or chemical treatments: Once the basic processes (filtration, sedimentation, anaerobic and aerobic lagoons) are carried out, advanced PTARs use physical-chemical treatments to provide better conditions for decontamination of wastewater. It has also been shown that these pollutants are capable of altering the endocrine system, blocking or disrupting the hormonal functions of organisms, causing feminization and hermaphroditism of the same, decreased fertility and mating efficiency and may even increase the incidence of different types of cancer [39]. On the other hand, the presence of PPCPs in the environment is a strong potential for the creation of antimicrobial resistance, due to the excessive use of antibiotics in human and animal medicine, which threatens the effective prevention and treatment of various types of infections caused by microorganisms.

A look at the Colombia case

Colombia is a country favored by its geographical position, this allows the ecosystem processes to recover despite the constant discharge of pollutants into the environment; nevertheless, there is so much pollution that is generated and continues to be generated that the time comes when the ecosystem alone cannot recover; This is the case of the Bogotá River, one of the rivers with the highest pollutant load in the country, due to the constant effluent it receives along its route, so it is difficult for self-purification processes to be observed [40]. In Colombia there are only 562 wastewater treatment systems distributed in 480 municipalities and about 89 systems located in 78 municipalities are abandoned [43]. To this is added the aforementioned, in relation to the fact that conventional systems for the treatment of wastewater worldwide are not designed to eliminate PPCPs, which means that the Colombia case is much more serious than is believed, Since not only are these types of pollutants being removed from the water, but also all types of pollutants, since the treatment systems do not work 100%. Although the WWTPs have the function of decontaminating wastewater and thus being discharged to

surface waters without greatly affecting the aquatic environment, the reality is different. In the cities of Florencia Caquetá and Tumaco Nariño, hospital, domestic and industrial wastewater is discharged directly to the rivers (without any restrictions), these cities are part of two of the country's richest biodiversity regions: the Pacific region and Amazon [44], because these two cities do not have WWTP, with this, the contamination by sewage is increasing due to processes such as runoff or evaporation, threatening important water sources and complete ecosystems. The Colombia case regarding pollution by PPCPs in water needs to be thoroughly studied; since not enough bibliography is recorded today; In addition, it is necessary to insist on the need to establish demanding policies when it comes to protecting water sources, since these are the basis for the sustenance of life.

PPcPs in Colombian waters: In Colombia there is a large gap of information on the presence of this type of pollutants in effluent discharges. Therefore, it is not very common to find scientific sources with specific figures of PPcPs in water sources, on the contrary there are very few investigations that deepen it, this demonstrates the need to expand the research field. Only in Bogotá 9'310.123 pharmaceutical units (tablets, dragees, capsules, ampoules), of special control products are destroyed annually, without considering those discarded at the domestic level [27], which reach directly to sewage or sanitary landfills given its inadequate deposit. This causes pollution levels in the country to increase.

The medicines with the highest input and therefore the most likely to reach the water sources are generally the analgesics, the most used analgesics are acetaminophen and aspirin followed by diclofenac, fenopropfen, ketoprofen, mefenamic acid, indiomethacin, naproxen and ibuprofen [3]. Ibuprofen and diclofenac are common anti-inflammatory analgesics consumed around the world and

especially in Colombia are easily acquired by consumers due to their sale without a prescription. Similarly, the acceptant is widely consumed due to pressure from the pharmaceutical industry, the ease of acquisition when pain occurs and the lack of knowledge about the implications of its high consumption. On the other hand, synthetic profiles and chemical products used to make fragrances, soaps, household cleaning products, detergents, softeners, shampoos, shaving lotions and even herbicides also have a high consumption in Colombia, which end up in wastewater and later in water sources where due to their chemical structure and physicochemical properties they have a potential for bioaccumulation and bioconcentration in the soil and in the adipose tissue of aquatic organisms, generating consequences still unknown to humans, through food consumption or water.

Botero-Coy et al. [24] report drugs found in sewage and surface waters in Bogotá and Antioquia (acepminophen, carbamazepine, diclofenac, clarithromycin, lincomycin, losartan, valsartan), where according to their study the highest concentration found was acepminophen (39.2 and 9,2µg/L) and the antibiotic azithromycin (6.3 and 5.8µg /L) respectively. Similarly, Aristizabal-Ciro et al. [25] report a poor elimination of PPcPs in PTAR because all the compounds detected in the tributaries studied in Medellín were also detected in the effluents of the PTAR, which demonstrates the low efficiency of removal of this type of contaminants in Colombian WWTPs similar to those reported by Wang & Wang [28], Ebele et al. [3], Tamura et al. [8] and Yang et al. [17], in other countries. An analysis of treated water showed that these compounds are not completely eliminated once the purification, coagulation, flocculation, sedimentation, filtration and chlorination disinfection processes are finished, this demonstrates that conventional treatments, which are designated to eliminate as much Organic matter is not efficient enough to remove polar pollutants at low concentrations [4].

Table 1: Pharmaceutical and personal hygiene products PPcPs identified in Colombian waters.

PPcPs	Type of PPcPs	Source
Ibuprofen	Analgesic	Arrubla et al. [5], Botero-Coy et al. [24], Aristizabal-Ciro et al. [25]
Naproxen	Antiinflammatory	Arrubla et al. [5], Botero-Coy et al. [24]
Diclofenac	Antiinflammatory	Arrubla et al. [5], Botero-Coy et al. [24], Aristizabal-Ciro et al. [25]
Aspirin	Analgesic	Arrubla et al. [5]
Ketoprofeno	Antiinflammatory	Arrubla et al. [5]
Caffeine	Stimulating	Arrubla et al. [5]
Galaxolide	Toilet product and hygiene	Arrubla et al. [5]
Tonalide	Toilet product and hygiene	Arrubla et al. [5]
Metildihidrojasmonato	Toilet product and hygiene	Arrubla et al. [5]
Chloribic acid	Antilipémico	Aristizabal-Ciro et al. [25]
Benzofenona	Toilet product and hygiene	Aristizabal-Ciro et al. [25]
Lorazepam	Antidepressant	López. (2016)
Butilprobano	Parabeno	Aristizabal-Ciro et al. [25]
Etilparabeno	Parabeno	Aristizabal-Ciro et al. [25]

Metilparabeno	Parabeno	Aristizabal-Ciro et al. [25]
Aceptaminofen	Analgesic	Aristizabal-Ciro et al. [25]
Clarithromycin	Antibiotic	Botero-Coy et al. [24]
Clindamycin	Antibiotic	Botero-Coy et al. [24]
Doxycycline	Antibiotic	Botero-Coy et al. [24]
Erythromycin	Antibiotic	Botero-Coy et al. [24]
Carbamazepine	Anticonvulsivo	Arrubla et al. [5], Botero-Coy et al. [24] Aristizabal-Ciro et al. [25]
Irbesartan	Antipyretic	Botero-Coy et al. [24]
Losartan	Antipyretic	Botero-Coy et al. [24]
Metronidazole	Antiparasitic	Botero-Coy et al. [24]
Norfloxacin	Antibiotic	Botero-Coy et al. [24]
Sulfamethoxazole	Antibiotic	Botero-Coy et al. [24]
Trimetropina	Antibiotic	Botero-Coy et al. [24]
Valsartan	Antihypertensive	Botero-Coy et al. [24]
Venlafaxine	Antidepressant	Botero-Coy et al. [24]

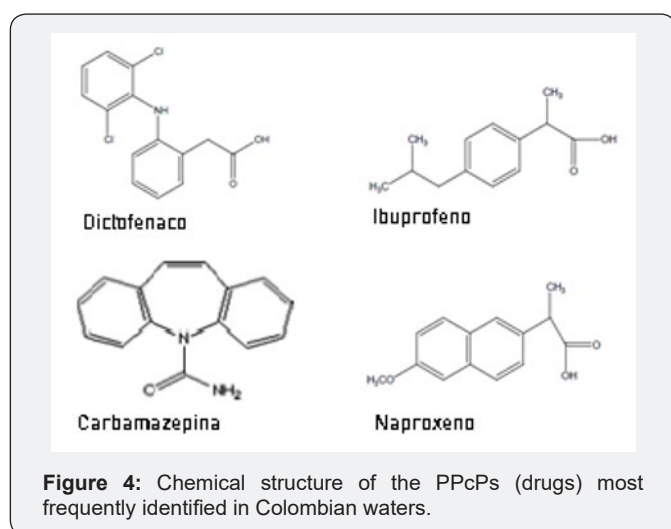


Figure 4: Chemical structure of the PPcPs (drugs) most frequently identified in Colombian waters.

Studies in Colombia have identified PPcPs as contaminants in both wastewater and surface water (Table 1). As reported by Arrubla et al. [5], Botero-Coy et al. [24], Aristizabal-Ciro et al. [25], it was found that diclofenac, carbamazepine, naproxen and ibuprofen are the drugs (PPcPs) most frequently found in water bodies of the study cities: Medellín, Bogotá, Pereira, Florencia and Tumaco (Figure 4). Due to its chemical characteristics, carbamazepine has been reported by several authors as a persistent drug in water and soil. With regard to ibuprofen, it is one of the most important drugs included in the “Essential drugs list” of the world health organization that gives rise to its mass production worldwide has high polarity and low volatility [8]; Therefore, its repeated presence in Colombian waters. Sun et al. [18] report to the carbamazepine antiepileptic as a marker due to its conservative behavior and because it is not easily eliminated by the WWTPs, therefore its persistence in surface waters is presented. Pompei et al. [45], they report the use of synthetic organic compounds in the domestic, agricultural and industrial context in Africa which is on the increase, additionally 70% of

the total urban population in several of the main cities of Africa do not have a water system wastewater and 80% of this water is discharged untreated to surface water or soil. Depending on the physicochemical properties of the drugs, their metabolites and degradation products and soil characteristics, these substances can reach groundwater and contaminate aquifers or be retained in the soil and accumulate and can affect the ecosystem and humans through the chain trophic.

Ebele et al. [3], they report in Rio de Janeiro in Brazil that carbamazepine and clofibric acid produce many more effects in *Daphia magna* than other drugs at the same concentration, in addition clofibric acid, diclofenac and naproxen were detected at low concentrations (0.01-0.06 μ g/L) in surface waters intended for human consumption. Gil et al. [46] find ibuprofen, diclofenac, carbamazepine and clofibric acid in drinking water. Wang & Chu [47] conclude that carbamazepine is one of the most frequently detected PPcPs in the effluents of WWTPs, it is resistant to biodegradation and biological treatments, therefore, it has little elimination effect even if its concentration is higher in effluents than in the tributaries possibly by the hydrolysis of conjugates of their original compounds, which not only occur in carbamazepine but also in all persistent, toxic and/or bio accumulative compounds.

Conclusion

The presence of pharmaceutical and personal hygiene products in aquatic ecosystems in Colombia is a reality due to several factors such as: lack of knowledge when making a correct final disposition and the poor or null processes carried out in the treatment plants of wastewater, which generates that charged waters of all types of pollutants reach rivers, streams, lagoons or seas. Authors around the world report numerous toxicological tests in different animal species, where they manifest various types of alterations in their physiology, morphology or behavior due to constant exposures to PPcPs in aquatic environments. The WWTPs in cities with Bogotá and Medellín, do not completely eliminate this type of

emerging pollutants such as PPCPs. Diclofenac, carbamazepine, naproxen and ibuprofen are the drugs (PPcPs) most frequently found in water bodies of: Medellín, Bogotá, Pereira, Florencia and Tumaco. In addition, cities such as Tumaco and Florence do not have systems to treat wastewater, so the situation is even worse since no pollutants are being treated, they all reach surface waters directly. The bibliographic information that refers to this type of contaminants in Colombian environments is still very little; However, the studies found express concern about this issue, which is motivating to continue with future research.

Conflicts of Interest

This review does not present conflicts of interest.

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