Environmental Chemistry Letters (2022) 20:2311–2338
https://doi.org/10.1007/s10311-022-01447-4

REVIEW

Worldwide cases of water pollution by emerging contaminants: a review

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Received: 9 March 2022 / Accepted: 31 March 2022 / Published online: 18 April 2022

Abstract
Water contamination by emerging contaminants is increasing in the context of rising urbanization, industrialization, and agriculture production. Emerging contaminants refers to contaminants for which there is currently no regulation requiring monitoring or public reporting of their presence in our water supply or wastewaters. There are many emerging contaminants such as pesticides, pharmaceuticals, drugs, cosmetics, personal care products, surfactants, cleaning products, industrial formulations and chemicals, food additives, food packaging, metalloids, rare earth elements, nanomaterials, microplastics, and pathogens. The main sources of emerging contaminants are domestic discharges, hospital effluents, industrial wastewaters, runoff from agriculture, livestock and aquaculture, and landfill leachates. In particular, effluents from municipal wastewater treatment plants are major contributors to the presence of emerging contaminants in waters. Although many chemicals have been recently regulated as priority hazardous substances, conventional plants for wastewater and drinking water treatment were not designed to remove most emerging contaminants. Here, we review key examples of contamination in China, Portugal, Mexico, Colombia, and Brazil. Examples include persistent organic pollutants such as polychlorinated biphenyls, dibenzofurans, and polybrominated diphenyl ethers, in lake and ocean ecosystems in China; emerging contaminants such as alkylphenols, natural and synthetic estrogens, antibiotics, and antidepressants in Portuguese rivers; and pharmaceuticals, hormones, cosmetics, personal care products, and pesticides in Mexican, Brazilian, and Colombian waters. All continents are affected by these contaminants. Wastewater treatment plants should therefore be upgraded, e.g., by addition of tertiary treatment systems, to limit environmental pollution.

Keywords Substances of global interest · Emerging contaminants · Wastewater treatment plants · Water pollution · Persistent organic pollutants · Pharmaceuticals · Personal care products · Pesticides

Introduction

Over the past two decades, the presence of emerging contaminants such as pharmaceuticals, cosmetics, personal care products, surfactants, and pesticides, in aqueous environmental compartments worldwide has become a major concern for our societies, as some of these chemicals are endocrine disruptors and others are proven carcinogens and mutagens. Indeed, the presence of chemicals in general, and particularly emerging substances, in aquatic compartments is a cause of concern and debate, as the risk they may pose to human health and the wildlife is not yet fully understood (Aristizabal-Ciro et al. 2017; Hernández-Padilla et al. 2017; Morin-Crini and Crini 2017; Botero-Coy et al. 2018; Crini and Lichtfouse 2018; Dong et al. 2018; Díaz-Casallas et al. 2019; Gallego-Schmid et al. 2019; Kumar et al. 2019; Reichert et al. 2019; Deviller et al. 2020; Balaram et al. 2022a, b).

The list of emerging contaminants is long, including:

- Pesticides, e.g., glyphosate and atrazine,

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- Pharmaceuticals, e.g., diclofenac, ibuprofen, antibiotics, and hormones,
- Licit and illicit drugs, e.g., caffeine, cocaine, and amphetamines,
- Preservatives such as parabens and triclosan,
- Personal care products, e.g., sunscreens and UV filters,
- Surfactants, cleaning products, industrial formulations, and chemicals, e.g., bisphenol A, chlorinated solvents,
- Food additives and food packaging, e.g., phthalates and plasticizers,
- Polycyclic aromatic hydrocarbons, polychlorinated biphenyls, halogenated polycyclic aromatic hydrocarbons, polychlorinated naphthalene, dioxins, hexachloro-1,3-butadiene, polyhalogenated carbazoles, and environmentally persistent free radicals
- Bromine-containing flame retardants, perfluorinated compounds and perfluorinated alkyl substances, brominated dioxins,
- Antibiotic-resistant pathogenic bacteria, e.g., *Escherichia coli* producing extended-spectrum *β*-lactamase,
- And other pollutants such as alkylphenols, metalloids, radionuclides, rare earth elements, nanomaterials, nanoparticles, microplastics, bioterrorism and sabotage agents, indoor pollutants, and pathogens.

The term "emerging contaminants" refers primarily to contaminants for which there is currently no regulation requiring monitoring or public reporting of their presence in our water supply or wastewaters. The US Environmental Protection Agency (EPA) defines an emerging pollutant as *a chemical or material which because of a recent source that it originates, or because of a new pathway that has developed, and for which a lack of published health standards exist poses a perceived, potential, or real threat to the human health or the environment* (source: US EPA 2012, Washington, DC). There is not yet an internationally recognized general classification for emerging substances. Nevertheless, many researchers agree with the definition given by the US EPA (Gogoi et al. 2018). Figure 1 proposes a general classification of emerging contaminants and issues of concern in water compartments into six broad categories, regardless of their risk to human health or the environment.

The bibliography on emerging chemical contaminants is particularly abundant and many analytical, biological, toxicological, ecotoxicological, and environmental topics are of interest to the scientific community. In particular, the presence, occurrence, transport of emerging contaminants in the water resources, and their impact on water quality are documented in thousands of publications reported worldwide during the last two decades, demonstrating an increasing concern about them (see for example the selected references: Patel et al. 2019, 2020; Reichert et al. 2019; Routti et al. 2019; Starling et al. 2019; de Oliveira et al. 2020; García et al. 2020; Gibson 2020; Iroegbu et al. 2020; Khan et al. 2020; Kovalakova et al. 2020; Olaniyan et al. 2020; Rigueto et al. 2020; Sousa et al. 2020; Wang and Zhuan 2020; Hartmann et al. 2021; Pastorino and Ginebreda 2021; Yusuf et al. 2021; Richardson and Ternes 2022).

Many chemicals are ubiquitous in every household because they are used in a wide variety of products and applications. As a result, they are found not only in domestic wastewater, but also in surface water and groundwater, and thus in drinking

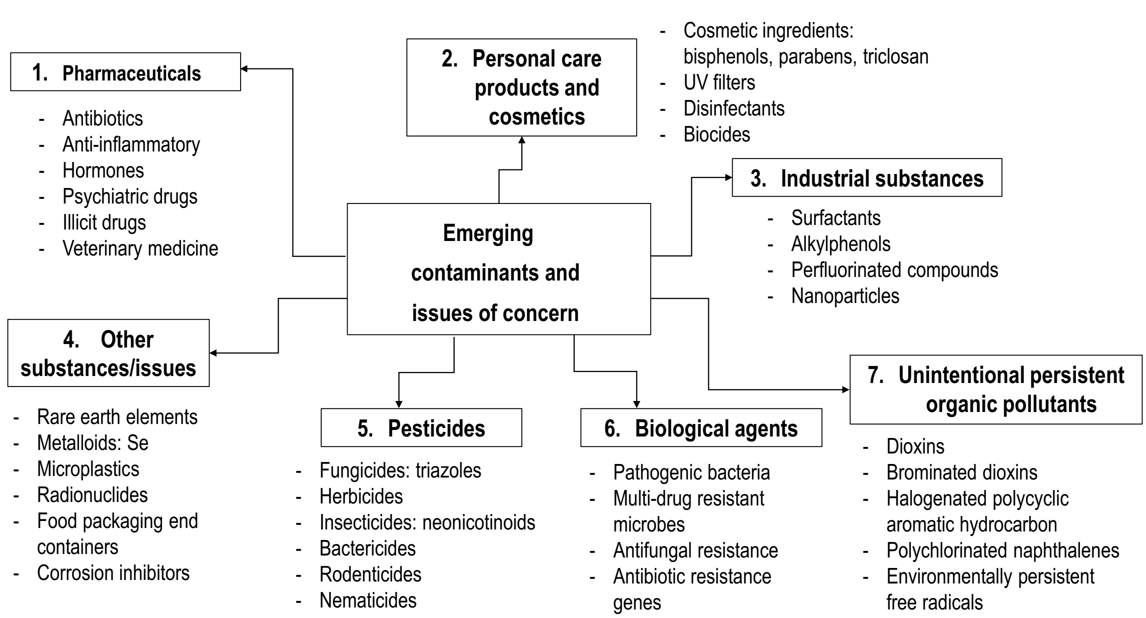

Fig. 1 Classification of emerging contaminants and issues of concern in water compartments

water and food sources, with the potential to cause known, unknown, or suspected adverse effects on human health. It is important to note that groundwater is the main source of drinking water in many countries (Crini and Badot 2007, 2010; Dévier et al. 2013; Kosma et al. 2014; Gee et al. 2015; Archer et al. 2017; Ebele et al. 2017). Emerging contaminants that enter the aquatic environment via domestic, industrial, and agricultural discharges may also have adverse effects on ecosystems. Due to their widespread use in our daily applications, agriculture, and industry, chemicals are ubiquitous not only in water and sediments, but also in soils, atmosphere, plants, living organisms, food products, and protected species (e.g., birds, polar bears), and thus even in areas where there is no anthropogenic activity (Datta et al. 2018; Routti et al. 2019), demonstrating their uncontrolled movement in the global environment. However, concentration levels are extremely variable from one continent to another, from one country to another, and even from one region to another. For example, Fekadu et al. (2019), comparing levels of contamination of pharmaceuticals such as carbamazepine and sulfamethoxazole in surface waters between Africa and Europe, observed that the maximum concentrations reported in Africa were 20,000 times higher than in Europe.

An important question is how rivers receive emerging contaminants. It is now recognized that the presence of some classes (e.g., pharmaceuticals) in aquatic environments is primarily correlated to the discharge of municipal wastewater treatment plant effluents. Numerous studies showed a correlation between the presence of emerging substances in the aquatic environment and discharges from wastewater treatment plants (Balaram 2016; Machado et al. 2016; Priac et al. 2017; Gilabert-Alarcón et al. 2018; Mezzelani et al. 2018; Fekadu et al. 2019; Gallego-Schmid et al. 2019; Inyinbor et al. 2019; Mohapatra and Kirpalani 2019; Montagner et al. 2019; Patel et al. 2019; Peña-Guzmán et al. 2019; Starling et al. 2019). However, other anthropogenic activities in the industrial and agricultural sectors, as well as other activities such as transport also contribute to water pollution by other types of contaminants of emerging concern (e.g., pesticides).

This review presents a selection of cases of water contamination by emerging pollutants described around the world, from China to Portugal, and Mexico, Brazil and Colombia. It is an abridged version of the chapter published by Morin-Crini et al. (2021) in the series Environmental Chemistry for a Sustainable World.

Persistent organic pollutants in lake and ocean ecosystems

Among the emerging pollutants, there is a list of persistent organic pollutants known as POPs. This term describes a wide range of organic compounds of anthropogenic origin present in the environment. This family of substances is subdivided into four main categories, namely polycyclic aromatic hydrocarbons known as PAH, chlorinated aromatic compounds, pesticides, including organochlorine pesticides, and brominated flame retardants. The best known substances include the herbicide 2,4-dichlorophenoxyacetic acid (known as 2,4-D), hexachlorobenzene (another chlorinated fungicide used to treat seeds, especially wheat), polychlorinated biphenyls commonly known as PCBs used as lubricants in transformers, or polycyclic aromatic hydrocarbons from combustion or pyrolysis (Puzyn and Mostrag-Szlichtyng 2012; Vestergren and Cousins 2013; DeWitt 2015; Eggen and Vogelsang 2015; Harmon 2015; Kallenborn et al. 2015; Zeng 2015; Xiao 2017; Dong et al. 2018; Lorenzo et al. 2018; Brusseau 2019; Ng et al. 2019; Klemes et al. 2019; Pan et al. 2020; Salthammer 2020).

Persistent organic pollutants represent a family of substances that is not strictly speaking "new" as it has been known for more than 50 years to date, but this group of pollutants is still of concern. Indeed, their presence in the aquatic environment is not a recent phenomenon, but this problem has become more widely evident over the last decade thanks to the constant improvement of analytical techniques and more regular monitoring of aqueous compartments, including sediments, and organisms. Persistent organic pollutants in the Arctic atmosphere have been investigated since the 1970s when the first atmospheric measurements revealed their presence in this pristine polar environment (Kallenborn et al. 2015). Nevertheless, this family of substances is regularly updated, for example, the emerging perfluoro alkyl and polyfluoro alkyl substances have been classified as persistent organic pollutants (Puzyn and Mostrag-Szlichtyng 2012; Vestergren and Cousins 2013; Zeng 2015). Only limited information is available for these "newly" identified substances.

Persistent organic pollutants are transported from primary and secondary sources (industry, agriculture, urbanized areas, and transport) through the atmosphere, and they can be transported through natural atmospheric and oceanic processes over long distances to areas of the World through natural atmospheric and oceanic processes where there are no anthropogenic activities, e.g., in Arctic soils (Gioia et al. 2011; Cabrerizo et al. 2018) and in areas where these substances have never been used or produced (Puzyn and Mostrag-Szlichtyng 2012; Kallenborn et al. 2015). The majority of persistent organic pollutants identified so far are banned or restricted worldwide due to concerns about their harmfulness to human health and ecosystems. They are indeed substances of global concern due to their toxicity, persistence, bioaccumulation, and long-range transport (Puzyn and Mostrag-Szlichtyng 2012; Datta et al. 2018). Once released into the environment, persistent organic pollutants may remain intact for exceptionally long periods of time, as some long-banned substances persist in the environment.

Persistent organic pollutants are widely distributed in all environmental compartments such as water, sediment, soil, and the atmosphere. Their low solubility and high lipophilicity make them prone to accumulate in the fatty tissue of living organisms, and then biomagnify along food chains, being toxic to humans and wildlife (Mortimer 2013; Harmon 2015; Ding et al. 2022). Moreover, due to their special intrinsic physical and chemical properties, they are resistant to environmental degradation through physical, chemical, and biological processes (Harmon 2015; Zeng 2015). Exposure to persistent organic pollutants can lead to serious health effects, including certain cancers, birth defects, immune, and reproductive system dysfunction, increased susceptibility to disease and damage to the central and peripheral nervous systems. Some substances are also known as disrupting endocrine compounds.

In order to eliminate or reduce the release of persistent organic pollutants into the environment, several international treaties have been established, which call for efforts and actions by the international community, such as the Protocol to the UNECE (United Nations Economic Commission for Europe) Regional Convention on Long Range Transboundary Air Pollution (CLRTAP) and the Stockholm Convention on Persistent Organic Pollutants. The Stockholm Convention (signed in 2001; Puzyn and Mostrag-Szlichtyng 2012), which has 184 countries/regions member signatories, listed 12 original persistent organic pollutants in 2004 and has updated it over the last decades. These instruments have established strict international regimes for lists of persistent organic pollutants, including 16 chemicals in the UNECE Protocol and 12 original chemicals (the "dirty dozen" including dichlorodiphenyltrichloroethane (known as DDT), aldrin, endrin, heptachlor, polychlorinated biphenyls, polychlorinated dibenzofurans, and polychlorinated dibenzo dioxins) and 16 newly added chemicals in the Stockholm Convention.

Lake ecosystems, including the water body, sediments, organisms, surrounding soils, and the atmosphere, are one of the major sinks for persistent organic pollutants and an important part of surface water systems. In lakes, these substances can accumulate in the fatty tissue of living organisms through the food chain, posing potential risks to humans (Puzyn and Mostrag-Szlichtyng 2012; Duedahl-Olesen 2013; Schrenk and Chopra 2013; Mortimer 2013). Legacy persistent organic pollutants, such as polycyclic aromatic hydrocarbons, organochlorine pesticides, polychlorinated biphenyls, polychlorinated dibenzo-p-dioxins and dibenzofurans and polybrominated diphenyl ethers, have been commonly reported in various lake ecosystems, while emerging persistent organic pollutants such as perfluorinated compounds, polychlorinated naphthalenes, tetrabromobisphenol A and hexabromocyclododecane have been relatively less reported, despite their great concern. Figure 2 shows the average concentrations of various persistent organic pollutants in the Chinese Taihu lake ecosystem. Lake Taihu, located in the southern Jiangsu province and northern Zhejiang province, is the third largest freshwater lake in China and has provided local communities with valuable fisheries for centuries. There are 38 cities and more than 40 million people living around the lake, Taihu Basin being one of the most developed regions in China (Wang et al. 2009; Li et al. 2019).

Various carcinogenic persistent organic pollutants from the past are commonly detected in typical lake ecosystems in Europe and Asian. Organochlorine pesticides, polychlorinated biphenyls, and polycyclic aromatic hydrocarbons

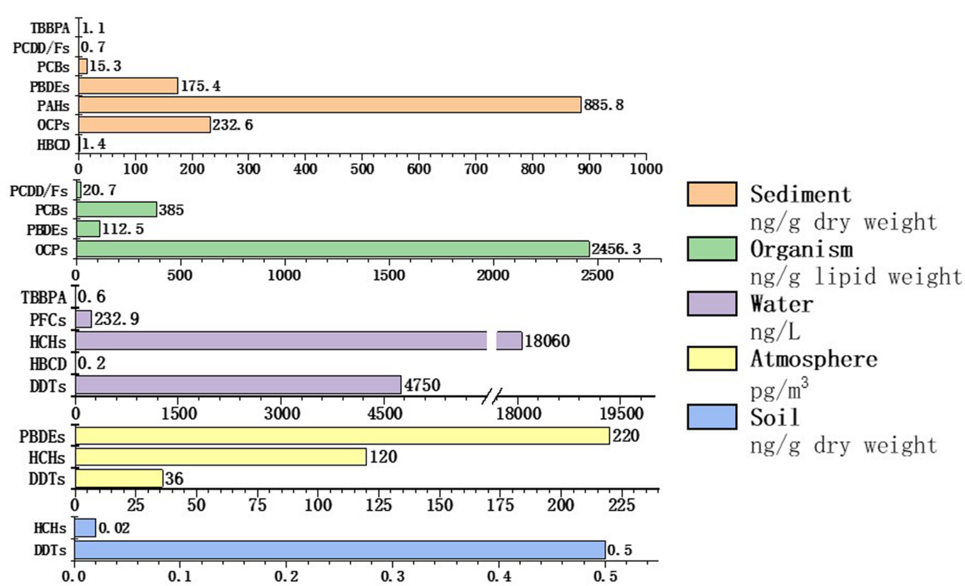

Fig. 2 Average concentrations of various persistent organic pollutants in the Chinese Taihu lake ecosystem. TBBPA: tetrabromobisphenol A; PCDD/Fs: polychlorinated dibenzo-p-dioxins and dibenzofurans; PCBs: polychlorinated biphenyls; PBDEs: polybrominated diphenyl ethers; PAHs: polycyclic aromatic hydrocarbons; OCPs: organochlorine pesticides; HBCD: hexabromocyclododecane; PFCs: perfluorinated compounds; HCHs: Hexachlorocyclohexanes; DDT: dichlorodiphenyltrichloroethane; DDTs: DDT-related compounds

are the main persistent organic pollutants in water bodies (Javedankherad et al. 2013), sediments (de Mora et al. 2004; Boehm et al. 2005; Nemirovskaya and Brekhovskikh 2008; Varnosfaderany et al. 2015; Baniemam et al. 2017), organisms (Rajaei et al. 2011; Mashroofeh et al. 2015), the surrounding soils, and atmosphere (Aliyeva et al. 2012, 2013; Shahbazi et al. 2012) of the Caspian Sea located between Europe and Asia, the World's largest lake.

The levels of these persistent organic pollutants in the Caspian Sea ecosystems are at a concentration order of approximately ng/g, with polycyclic aromatic hydrocarbons having a relatively higher concentration. However, persistent organic pollutants newly added to the Stockholm Convention, such as polychlorinated naphthalenes, perfluorinated compounds, and hexabromocyclododecane, are currently rarely studied in the Caspian Sea at present. Persistent organic pollutants, including organochlorine pesticides, polychlorinated biphenyls, polychlorinated dibenzo-p-dioxins and dibenzofurans, polycyclic aromatic hydrocarbons, polybrominated diphenyl ethers, polychlorinated naphthalenes, hexabromocyclododecane, perfluorinated compounds, and tetrabromobisphenol A, are also present in different environmental compartments and biota in Chinese lake ecosystems (Qiu et al. 2004; Zhang and Jiang 2005; Qiao et al. 2006; Wang et al. 2007, 2016a, 2016b; Wen et al. 2008; Yang et al. 2011; Yu et al. 2012; Zhang et al. 2012; Zhou et al. 2012a, 2012b; Xu et al. 2013, 2015; Cui 2018; Liu et al. 2018; Wei et al. 2019; Xiang et al. 2019).

Recently, the presence of perfluorinated compounds in the environment has been of great concern (Wang et al. 2022). Levels of perfluorinated compounds in Chinese lakes have averaged between 30 and 50 ng/L, being perfluorooctanoic acid and perfluorooctane sulfonate the most frequent perfluorinated compounds (Yang et al. 2011; Gao et al. 2016). Mean concentrations of organochlorine pesticides, polycyclic aromatic hydrocarbons, polychlorinated biphenyls, polybrominated diphenyl ethers, polychlorinated dibenzo-p-dioxins and dibenzofurans, perfluorinated compounds, and hexabromocyclododecane in samples of biota from various Chinese lakes are usually at ng/g levels (Liu et al. 2018). In summary, European and Asian lake ecosystems are contaminated to varying degrees by various persistent organic pollutants. There are large differences in the levels of these pollutants between different lake ecosystems and other environmental media. Further studies on newly added persistent organic pollutants are needed in the future to better understand their environmental fate and ultimate control.

Persistent organic pollutants, old and new, are also widely distributed in the water bodies, sediments, organisms, surrounding soils and atmosphere of lake ecosystems of North America and Africa. Lake Superior is the largest freshwater lake in the World and also the largest of the Great Lakes in North America. Polychlorinated biphenyls and organochlorine pesticides occurring in the Lake Superior water body in the 1990s were at ng/g range and by 2018 they were at pg/g levels, which is lower than in lakes in Europe and Asia (Jeremiason et al. 1994; Ruge et al. 2018). Polychlorinated dibenzo-p-dioxins, dibenzofurans, polybrominated diphenyl ethers, and polychlorinated biphenyls were found in Lake Superior sediments at ng/g magnitude, which was comparable to that of European and Asian lakes.

In addition to organochlorine pesticides and polychlorinated biphenyls, some emerging persistent organic pollutants such as polybrominated diphenyl ethers, hexabromocyclododecane, polychlorinated naphthalenes, and perfluorinated compounds were also present in Lake Superior organisms (Kannan et al. 2000; Luross et al. 2002; Dykstra et al. 2005; Fernie and Letcher 2010; Furdui et al. 2015). Polycyclic aromatic hydrocarbons, polybrominated diphenyl ethers, organochlorine pesticides, polychlorinated naphthalenes, and polychlorinated biphenyls were present in the surrounding atmosphere (Hillery et al. 1997; Strandberg et al. 2001; Fernandez et al. 2002; Helm et al. 2003).

Lake Tanganyika is a freshwater lake located in Central Africa. Reports of persistent organic pollutants in Lake Tanganyika are mainly related to lake organisms. Organochlorine pesticides, polychlorinated biphenyls, polybrominated diphenyl ethers, and hexabromocyclododecane were detected in fish samples from Lake Tanganyika (Mahugija et al. 2018; Polder et al. 2014). Among these reported substances, the concentration of polychlorinated biphenyls in fish samples ranged from 36 to 167 ng/g, which is generally lower than in Asian lakes. In addition, the contamination of persistent organic pollutants in Lake Tanganyika was lower or comparable to that in other parts of Africa (Manirakiza et al. 2002).

Polybrominated diphenyl ethers have raised new concerns in recent decades. Some studies have focused on polybrominated diphenyl ether contamination in the North Pacific Ocean, where the World's largest fishery, that of Hokkaido, is being lacerated. It is the main source of seafood for people living in countries along the North Pacific coast, especially those living in Japan, South Korea and China. However, this coast is also the World's largest producer and consumer of polybrominated diphenyl ethers. The use and emissions of polybrominated diphenyl ethers in this region are significant, and pollution by polybrominated diphenyl ethers is becoming an increasingly serious problem. Xiang et al. (2019) showed that the current concentration of polybrominated diphenyl ethers in the atmosphere, water, sediments and organisms in the North Pacific Ocean is 0.14 to 896 pg/m^3, 3 to 4360 pg/L, 0.24 to 7397.7 ng/g dry weight and 0.56 to 3930 ng/g lipid weight, respectively. The predicted concentration of polybrominated diphenyl ethers in the Hokkaido fishery could be 4.6 to 9 pg/m^3 (air), 106 to 341 pg/L (water), 3.4 to 248 ng/g dry weight (sediment), and 5 to

64.7 ng/g lipid weight (biota). The concentration of polybrominated diphenyl ethers in the atmosphere, water bodies, sediments, and organisms in the North Pacific has a characteristic near-shore > ocean or low latitude > high latitude distribution (Xiang et al. 2019).

From a global perspective, lake ecosystems are contaminated to varying degrees by several persistent organic pollutants, both from past and recent contamination. Although many studies have been conducted on these substances in lake ecosystems, these reports have mainly focused on traditional persistent organic pollutants such as organochlorine pesticides, polychlorinated biphenyls, polychlorinated dibenzo-p-dioxins, and dibenzofurans, while emerging persistent organic pollutants, such as polychlorinated naphthalenes, perfluorinated compounds, and hexabromocyclododecane, are relatively less reported. The extent and significance of all these substances are still largely unknown. Environmental risk assessment of persistent organic pollutants remains particularly difficult, not only in lake ecosystems, but also in freshwater and marine ecosystems.

These ecosystems are fragile, living at the rate of anthropogenic disturbances (pollution, artificialization, withdrawals), increasingly vulnerable for example to climate change, but essential to life in general. Indeed, lake ecosystems are important freshwater resources closely linked to human life, but our understanding of pollution by persistent organic pollutants in lake ecosystems is not yet sufficient. A better understanding of these substances, in terms of their degradation, behavior, accumulation, transfer, toxicity, and impact, still requires research in the different environmental compartments and living organisms of these systems. More comprehensive international health policies and standards are also needed in the future.

Monitoring emerging contaminants in Portuguese rivers

Portugal is the top third country with more studies in the field of surface water even by normalizing the amount of reports by the number of inhabitants per country, according to a review of data published between 2012 and 2018 on the environmental monitoring of water organic pollutants (Sousa et al. 2018). That review paper provides an overview of the worldwide occurrence of contaminants included in the European Union guidelines (priority substances of European Directive 39/2013/EU and contaminants of emerging concern of Watch Lists of European Decisions 495/2015/EU and 840/2018/EU), presenting also comprehensive data about other specific organic pollutants not considered in those European Union documents and with reported high concentrations and frequencies.

Another review paper covering the data published between 2001 and 2015 systematically overviewed the monitoring of industrial compounds, natural and synthetic estrogens, phytoestrogens and phytosterols, pharmaceuticals, and pesticides in the Portuguese waterbodies (Ribeiro et al. 2016b). The data collected in that review demonstrated that some substances were detected at quite high levels in the studies performed between 2001 and 2015, namely industrial and household compounds (e.g., bisphenol A, alkylphenols, and alkylphenol polyethoxylates) found from few ng/L to dozens of μg/L, in agreement with other European countries such as Spain, Switzerland, Netherlands, and France.

In the reviewed literature, natural and synthetic estrogens were detected always at ng/L levels in Portuguese surface waters, similar or above those concentrations reported in European countries. While phytoestrogens were reported as frequently detected at μg/L range, the authors pointed out that most publications about pharmaceuticals in Portuguese surface waters published before 2016 referred only few therapeutic classes in the north of Portugal and most consisted in the application of new analytical methods rather than consistent monitoring data (Ribeiro et al. 2016b; Balaram et al. 2022a). The reports on a broad range of classes of pesticides including priority substances of European Directive 39/2013/EU were collected in that review from 2001 to 2015, some of them up to μg/L, including atrazine that was already banned in Europe. The present section aims to update the information about the occurrence and distribution of emerging contaminants in Portuguese rivers (Fig. 3) and results from a literature survey comprising reports published since 2015 in Scopus database, using as keywords: "river water," "surface water" or "estuarine water" and "Portugal" or "Portuguese," and "monitor" or"occurrence."

Natural organic compounds have been studied lately in Portuguese rivers, namely phytoestrogens and phytosterols that are found in many plants, as well as mycotoxins produced by fungi. In a monitoring study in the Douro River estuary, the maximum concentration of daidzein (up to 277.4 ng/L) was detected in the summer (Ribeiro et al. 2016a), whereas the highest level in another seasonal study performed by the same research group in the Ave River was found in the spring (up to 404.0 ng/L) (Ribeiro et al. 2016c). In the Ave, a seasonal trend for phytoestrogens and phytosterols was demonstrated and the largest level of coumestrol (up to 165.0 ng/L) was registered in the summer. Remarkably, this compound was not found in any sample collected in the Douro monitoring campaign (Ribeiro et al. 2016a). Both studies reported that the mycotoxin deoxynivalenol was ubiquitously found up to 373.5 ng/L in the Douro (Ribeiro et al. 2016a) and between 59.5 and 642.4 ng/L in the Ave (Ribeiro et al. 2016c), whereas the mycotoxin zearalenone and its metabolite α-zearalenol were only detected in spring and summer in

Fig. 3 River pollution by emerging contaminants and related challenges for their management. Ave River, left, and Leça River, right. Source: Ana Rita Lado Ribeiro, Porto, Portugal

the Douro (Ribeiro et al. 2016a). This last finding agrees with another monitoring study in seven Portuguese rivers and one creek, where the highest concentration of zearalenone was reported during spring. In that study, a contamination frequency of 23.7% was reported for zearalenone, with concentrations ranging from 5.6 to 82.6 ng/L (Laranjeiro et al. 2018).

Generally, the highest concentrations found since 2015 are quite lower than those reviewed in the period 2001–2015 by Ribeiro et al. (2016b). Industrial and household compounds were also monitored in Portuguese rivers. In a monitoring program developed in Mira River over a 1-year period, including natural and pharmaceutical estrogens, phytoestrogens, and the phytosterol sitosterol, both estrogens and industrial/household pollutants were determined at high levels in all sampling sites, including some located in environmental protected areas (Rocha et al. 2016). Particularly, the annual average of industrial/household pollutants was of approximately 1.3 µg/L, whereas ng/L levels were described for phytoestrogens and sitosterol (Rocha et al. 2016). Another recent study targeting endocrine disruptor compounds in Minho, Ave, and Mondego River estuaries, reported very high concentrations of alkylphenols and alkylphenol ethoxylates (up to 4855 ng/L) despite those of the natural estrogens were noticeably lower than previous monitoring programs.

Regarding personal care products, one study performed in northern Portuguese rivers in the summer (2018) targeted 43 substances and detected 28 (Celeiro et al. 2019): fragrances up to 200 ng/L (citronellol), limonene, α-isomethylionone, tonalide and galaxolide in all samples, the latter up to 379 ng/L; phthalates up to 92 ng/L (benzylbutylphthalate), including the priority substance diethylhexylphthalate determined at 88 ng/L in one sampling location; the priority substance antioxidant butylhydroxytoluene between 8 and 25 ng/L in half of the samples; and UV filters up to 254 ng/L (benzophenone 3).

Since 2015, pharmaceuticals have been extensively monitored by some research groups. A wide set of 66 human and veterinary pharmaceuticals from seven therapeutic groups was monitored in Tejo estuarine waters and the therapeutic classes with an overall frequency of detection higher than 90% were antibiotics, β-blockers, antihypertensives, lipid regulator, and anti-inflammatories (Reis-Santos et al. 2018). The highest concentrations of each therapeutic family were 304 ng/L for antidepressants (sertraline), 51.8 ng/L for non-steroidal anti-inflammatories (diclofenac), 77.0 ng/L for lipid regulators (gemfibrozil), 161.9 ng/L for antihypertensives (irbesartan) and 128.0 ng/L for antibiotics (doxycycline). Another 1-year monitoring study targeting 23 pharmaceuticals in Tejo and Mondego Rivers (Pereira et al. 2017a) reported one site in Tejo River with comparable concentrations of diclofenac, gemfibrozil, and bezafibrate. In that occurrence study, pharmaceuticals were determined in 27.8% of the samples at ng/L levels, with the following therapeutic classes having the highest frequencies and mean concentrations: selective serotonin reuptake inhibitors, anti-inflammatories, and antibiotics (Pereira et al. 2017a). The authors confirmed the impact of wastewater treatment plants by the rising mean concentrations downstream and the impact of lower river flow rates on increasing frequencies and concentrations. Although the risk quotients were above one only for two pharmaceuticals, the authors stressed the ecotoxicological pressure especially due to water scarcity in drought periods, which may raise the risk in rivers located in the Iberia region.

A study carried out in the Lisbon's drinking water supply system targeted 31 pharmaceuticals in surface waters from Tejo and Zêzere Rivers, where 15 and 10 pharmaceuticals were detected at ng/L levels, respectively (de

Jesus Gaffney et al. 2015). Caffeine, erythromycin, acetaminophen, sulfadiazine, sulfapyridine, sulfamethoxazole, carbamazepine, and atenolol were quantified in both rivers; whereas propranolol, sulfamethazine, gemfibrozil, indomethacin, ibuprofen, diclofenac, and naproxen were detected in the Tejo River; and nimesulide was determined only in the Zêzere River. The risk assessment was performed for human health in that study and it was suggested that the exposure to residual pharmaceuticals in drinking water would be improbable. Considering that cancer incidence has grown worldwide and the consumption of cytostatics has clearly increased over the last years, this is a class of great environmental relevance. From seven cytostatics (cyclophosphamide, capecitabine, mycophenolic acid, imatinib, bicalutamide, prednisone, and 5-fluorouracil) determined in river water samples in a recent study encompassing also wastewaters, only mycophenolic acid was detected in Uíma, Douro and Leça Rivers, at average concentrations of 210, 211 and 541 ng/L, respectively (Santos et al. 2018b, a). Twenty-seven pharmaceuticals including antibiotics and psychiatric drugs were also analyzed in Douro and Leça Rivers (Fernandes et al. 2020). That study reported higher frequency and concentrations of pharmaceuticals in the Leça River, where six antibiotics (azithromycin, ciprofloxacin, clarithromycin, moxifloxacin, ofloxacin, and trimethoprim) were found, the highest concentration was registered for azithromycin (2,819 ng/L), and fluoxetine was the most detected pharmaceutical. Interestingly, in the same report, none of the studied antibiotics was detected in river samples of Douro River.

Some monitoring studies targeting pharmaceuticals and other classes of compounds simultaneously (namely pesticides) have been carried out in the last years. One recent monitoring program developed recently in four rivers (Ave, Antuã, Cértima, and Leça) reported the spatial and temporal variations of 39 priority substances and contaminants of emerging concern, some included in the European Directive 39/2013/EU, European Decisions 840/2018/EU and 495/2015/EU (Barbosa et al. 2018). From the 13 pollutants detected in all the rivers, several pharmaceuticals were detected (up to 396 ng/L) and the priority pesticide isoproturon was found up to 92 ng/L. The industrial priority substance perfluorooctanesulfonic acid was detected below its quantification limit in the Antuã, Cértima, and Leça. Atrazine, a priority substance of European Union Directive 39/2013 already banned, was also detected in Ave, Antuã, and Leça (up to 41 ng/L), whereas simazine was found in the Cértima and Leça (Barbosa et al. 2018), both pesticides being found at higher concentrations than those reported in the Arade River by Gonzalez-Rey et al. (2015). The neonicotinoids acetamiprid and imidacloprid of the European Decision 840/2018/EU were only detected during the dry season in the Ave (Barbosa et al. 2018).

In the same report, higher concentrations were observed downstream of industrial areas and urban wastewater treatment plants (Barbosa et al. 2018). In another study published by the same group, all the emerging contaminants preconized in the Watch List in force at the time of the monitoring program (European Decision 495/2015/EU) were monitored during four seasons in two Portuguese rivers positioned in the northern region, Ave and Sousa (Sousa et al. 2019). In that study, 13 contaminants of emerging concern were found, being diclofenac, 2-ethylhexyl 4-methoxycinnamate, and azithromycin the most frequent compounds and diclofenac, 2-ethylhexyl 4-methoxycinnamate and imidacloprid those determined at the highest levels (Sousa et al. 2019). The same research group reported the first spatial distribution of 37 pollutants in the entire Portuguese coast, including the ocean shore and the nearest river discharging on it. This occurrence program included priority substances and contaminants of emerging concern of the European Watch List (European Decisions 495/2015/EU and 840/2018/EU; EU 2015, 2018). In general, the authors reported high concentrations of diclofenac, tramadol, and carbamazepine, the latter being attributed to medium to high risk for algae (Sousa et al. 2020). While some pharmaceuticals and perfluorooctanesulfonic acid were widely distributed, atrazine and alachlor were also determined in most samples, with the concentrations of alachlor in some sites being considered as medium to high risky. Remarkably, higher levels were found in some seawater samples in comparison to that of the nearest estuarine location, which was ascribed to the possible direct discharge into the sea. The first study on the presence of pharmaceutical compounds and pesticides in the Arade River estuary included 19 pharmaceuticals and 47 pesticides, some of which defined as priority substances of European Directive 39/2013/EU or included in the European Decision 495/2015/EU (EU 2013; Gonzalez-Rey et al. 2015). In that study, the compounds detected varied temporally and seasonally, except the stimulant caffeine (804 ng/L) occurring without any remarkable differences. Other study targeting caffeine as anthropogenic marker in the Lis River over a period of 11 months, described its presence in all river samples ranging from 25.3 to 321 ng/L, with its maximum levels found downstream of effluent discharge points, confirming that it could be an effective indicator of human-born pollution (Paíga et al. 2019). Besides caffeine, those compounds found at highest levels in the Arade River included the antiasthmatic theophylline (184 ng/L), the analgesic paracetamol (88 ng/L), and the fungicide carbendazim (45 ± 18 ng/L) (Gonzalez-Rey et al. 2015). Atrazine, diuron, isoproturon, and simazine were found at few ng/L and below the environmental quality standards.

Some monitoring surveys focusing on pesticides have been also published. Surface waters of the Lezíria do Tejo agricultural area were monitored and the measured environmental concentrations for the 19 pesticides were compared with their environmental quality standards for risk assessment, being the risk demonstrated in 100% of the samples containing insecticides, which represented 60% of the total risk identified (Pereira et al. 2017b). One monitoring study performed between 2002 and 2004 in the river basins Mondego, Sado, and Tejo, aimed to assess the risk of 29 pesticides and metabolites, including priority substances and other compounds used in dominant crops of a number of agricultural areas within the catchment of these Mediterranean river basins (Silva et al. 2015a, 2015b). In that study, 20 substances were detected, from which seven were priority substances (alachlor, atrazine, chlorfenvinphos, chlorpyrifos, endosulfan, simazine, and terbutryn) and curiously all exceeded their individual environmental quality standards.

Other specific pollutants (molinate, oxadiazon, pendimethalin, propanil, terbuthylazine, and the metabolite deethylatrazine) had non-acceptable aquatic risks (Silva et al. 2015a). The authors highlighted the importance of monitoring such substances not only at European Union, but also at local/river-basin/national level, allowing to define the exceedances of the environmental quality standards. The most recurrently found substances in a previous study reporting 29 pesticide compounds (21 herbicides, 5 insecticides, and 3 metabolites) in surface waters of Mondego, Sado, and Tejo from 2002 and 2008 were: chlorfenvinphos (mean 0.16 µg/L), propanil (mean 0.007 µg/L) and the metabolite 3,4-dichloroaniline (mean 0.33 µg/L) in all rivers; molinate (mean 1.03 µg/L) and oxadiazon (mean 0.006 µg/L) in Mondego and Sado Rivers; and atrazine (mean 0.16 µg/L), simazine (mean 0.08 µg/L) and metolachlor (mean 0.06 µg/L) in Mondego and Tejo Rivers (Silva et al. 2015a).

Endosulfan was also detected in all rivers, but less commonly (3–7%) and with a relatively lower mean value of 0.0008 µg/L. In that study, the aquatic risk of the measured pesticide mixtures for primary producers, arthropods, and fish, was predicted in the three Portuguese river basins and it was proposed a ranking per taxonomic group and river basin for the relative contribution of individual compounds or groups sharing the same mode of action. In this approach, the highest toxicity role was observed for oxadiazon on primary producers and for the organophosphorus insecticides chlorfenvinphos and chlorpyrifos on arthropods and fish, respectively. It is remarkable that a study conducted between 2010 and 2011 in the Mondego estuary, encompassing 56 priority pesticides (insecticides, herbicides, and fungicides), described more than half of the quantified pesticides above the maximum values set by the European Directives (98/83/EC and 2013/39/EU), with a potential risk for the pesticide mixture at the maximum concentrations reported, and a significant toxic effect for *Artemia salina* (Cruzeiro et al. 2016).

In another spatial and temporal monitoring program comprising the pesticides atrazine, azoxystrobin, bentazon, λ-cyhalothrin, penoxsulam, and terbuthylazine in the Mondego estuary, azoxystrobin was the most frequently detected (Rodrigues et al. 2018), a fact that was also reported in all aqueous samples collected during 2012–2013 in a wetland of worldwide interest (Ria Formosa Lagoon) (Cruzeiro et al. 2015a), where pesticide concentrations surpassed those recommended by European Directive 2013/39/EU in two monitoring studies (Cruzeiro et al. 2015a, 2015b). In the Mondego (Rodrigues et al. 2018), atrazine was the second most detected, reinforcing that this priority substance should be monitored despite its ban. Although a low risk to estuarine organisms was estimated for the concentrations of pesticides found, all pesticides were bioaccumulated by bivalves and triazine pesticides were found also in macroalgae (Rodrigues et al. 2018).

In the same study, the pesticides were quantified typically during summer concurring with the pesticide usage period and no severe contamination occurred during a flood event that was expected to promote the runoff of pesticides from the adjacent agricultural areas. The quantified residues of pesticides were also more frequent during the summer and thus in the pesticide application period in another spatial and temporal monitoring program performed in 2017 in the Sado estuary (Rodrigues et al. 2019). That report suggested a long-term aquatic exposure for five herbicides (alachlor, bentazon, metobromuron, metribuzin, and triclopyr), which were found in the water samples before and after the production season. Regarding the potential adverse effects of the application of agricultural pesticides on the aquatic organisms in this estuary, it was not found any severe effect, even considering the potential mixture effect of pesticides.

The problematic of microplastics in river waters has received little attention in comparison to marine system. Two recent studies performed in Antuã River (Rodrigues et al. 2018) and Douro river estuary (Rodrigues et al. 2019) targeted this environmental concerning issue. The first study reporting the occurrence of microplastics in freshwater systems in Portugal was developed in the Antuã River during March and October 2016, showing a correlation of their occurrence with the urbanization and industrial activities, with an abundance in water up to 193 items/m^3 in March and up to 1265 items/m^3 in October. A higher abundance in October was found for almost all sampling sites (one exception) and a decrease from upstream to downstream areas was reported. In a study performed in the Douro between 2016 and 2017, the authors found a total of 2152 particles, with an average density of 17.06 items/100 m^3 (Rodrigues et al. 2019), with an average ratio of fish larvae to microplastics greater than one being found in some months, highlighting the possible ingestion by fish and the resulting impact in these communities.

All these results show that protecting water resources is essential to guarantee the sustainability of the ecosystems from which Portuguese economy depends on. The distribution of pollutants along the entire Portuguese mainland coast shows the need for mitigation actions to boost the elimination of pharmaceuticals in wastewater treatment plants and measures to control the use and discharge of pesticides and industrial compounds.

Emerging contaminants in the aquatic environment in Latin America

With about one-third of the World's water resources and 24,400 m^3 of water per capita per year, Latin America is a region with abundant freshwater resource availability. This resource is nevertheless fragile and particularly impacted by increasing economic development (Peña-Guzmán et al. 2019). Indeed, in many Latin America countries, there is a high population density in areas suffering from poor sanitary conditions, which contributes to the contamination of rivers and reservoirs supplying water (Caldas et al. 2019; Peña-Guzmán et al. 2019; Starling et al. 2019; Oyedotun and Ally 2021). The chemical and physicochemical properties, e.g., water solubility, high adsorption capacity, and low biodegradability, of many substances in daily use facilitate their distribution, triggering problems not only for the environment but also for the health of living organisms (Janssens et al. 1997; Knepper et al. 1999; Kuster et al. 2008; Hernández et al. 2011a, 2011b; Peña-Guzmán et al. 2019).

In a recent comprehensive review by Peña-Guzmán et al. (2019), water quality was addressed involving various aqueous compartments of Latin American countries (Argentina, Bolivia, Brazil, Chile, Colombia, Costa Rica, Ecuador, Guatemala, Mexico, Uruguay, and Venezuela): water reservoirs for human consumption, drinking water treatment plants, groundwater, surface waters, rivers, and wastewater treatment plants. The authors concluded that the most commonly detected emerging substances included pharmaceuticals, mainly antibiotics, followed by personal care products, being the most common molecules 17-β-estradiol, bisphenol A, estrone, and caffeine. The majority of these chemicals are not yet regulated by any environmental law. Peña-Guzmán et al. (2019) reported that a total of 51 emerging pollutants were determined in studies related to wastewater, with Ecuador being the country with the largest number of samples, followed by Mexico, Brazil, and Colombia.

In wastewater, the highest concentrations reviewed were found for caffeine (5597 μg/L), followed by the cocaine metabolite benzoylecgonine (1065 μg/L) and carbamazepine (830 μg/L). In the drinking water supply, the compound with the highest concentration reported (625 μg/L) was ibuprofen (Brazil, Colombia, and Venezuela). For surface water sources, the contaminants with the highest levels were cholesterol (301 μg/L), caffeine (106 μg/L), stigmasterol (85.5 μg/L), and bisphenol A (64.2 μg/L) (Brazil, Costa Rica, Colombia, Bolivia, Chile, Mexico, and Argentina). Mexico is the only country that has recorded cases of emerging contaminants in groundwater in the urban water cycle, with the highest concentrations determined for naproxen (2 μg/L), sulfasalazine (0.78 μg/L), ibuprofen (0.51 μg/L), and salicylic acid (0.464 μg/L). In drinking water treatment plant effluents, the substances identified in Brazil were caffeine (4.083 μg/L), nonylphenol (0.228 μg/L), 17-α-ethinylestradiol (0.798 μg/L), bisphenol A (0.0005 μg/L), estriol (0.0003 μg/L), and estrone (0.00003 μg/L).

Other recent studies have also shown that pharmaceuticals, personal care products, industrial substances, and pesticides are widely detected at concentrations of the same order of magnitude as those reviewed (Machado et al. 2016; Hernández-Padilla et al. 2017; Botero-Coy et al. 2018; Gilabert-Alarcón et al. 2018; Sodré et al. 2018; Montagner et al. 2019; Starling et al. 2019). All these studies concluded that these types of pollutants are persistent in the environmental compartments of Latin America, their continuous introduction into rivers yielding a complex mixture with potential synergistic effects that can result in a serious ecological risk.

As in many other countries around the world, emerging substances enter the aquatic environment of Latin America by many pathways, with the main one being the conventional wastewater treatment plants not designed to eliminate these type of contaminants, which discharged effluents contain organic substances and metabolites (Dougherty et al. 2010; Hernández-Padilla et al. 2017; Starling et al. 2019). Moreover, it is estimated that only 20% of municipal wastewater is treated, and in some Latin America countries, many wastewater treatment plants are abandoned due to the high operating costs or low performance (Hernández-Padilla et al. 2017). In Mexico, less than 50% of wastewater is treated and wastewater treatment plants are not financially self-sustaining, resulting in uneven water quality (Gilabert-Alarcón et al. 2018). In Brazil, a country with more than 200 million inhabitants, the percentage of households with access to the wastewater treatment system does not exceed 50%, and a large part of the collected wastewater does not receive appropriate treatment before being discharged into rivers (Machado et al. 2016). In Colombia, raw wastewater is sometimes discharged directly into surface waters in certain Colombian regions such as Florencia and Tumaco (Botero-Coy et al. 2018). When untreated water is released, this contamination further reduces the availability of freshwater for domestic purpose and other uses, causing also damage to ecosystems.

However, this is not just a domestic water problem. Indeed, treated industrial wastewater from the pharmaceutical, chemical, textile, or metal industries, as well as water from agricultural activities including irrigation, livestock, and aquaculture, can also contain a cocktail of substances, which end up in rivers. Before being discharged, like municipal wastewaters, industrial effluents must comply with regulations concerning a number of "conventional" substances such as metals and water parameters, namely suspended solids, chemical, and biochemical oxygen demand, but organic substances are rarely registered in industrial wastewater (Hernández-Padilla et al. 2017; Gilabert-Alarcón et al. 2018).

The assessment of the occurrence of new contaminants in rivers is one of the major challenges for the scientific community and there is a lack of information in most Latin American countries (Hernández-Padilla et al. 2017; Peña-Guzmán et al. 2019; Starling et al. 2019). Unlike Europe and other regions, many Latin American countries do not have an established list of priority pollutants or laws controlling their production or release into the environment. However, regulations are beginning to evolve, particularly in Mexico, Colombia, and Brazil, for example with regard to the treatment of municipal and industrial wastewater before discharge. Treatment plants and industries in Latin America do not have a formal structure to detect, assess, control, and manage new compounds. Researchers and industrialists in Mexico, Colombia, and Brazil draw on the experience of other developed countries in order to protect the aquatic environment of their respective regions, which have different conditions, environmental culture, and pollutions levels (Hernández-Padilla et al. 2017).

Pharmaceuticals, personal care products, and pesticides in the aquatic environment in Mexico

Based on data of the Federal Agency of Consumer (PROFECO) of Mexico, this country is one of the main economies in Latin America, which has registered a strong growth in the personal care products market during the last decade, being ca. 40% of the demand related to hair and skin care products. On the other hand, according to the Ministry of Health (Mexico), it is estimated that more than 80% of the Mexicans self-medicate, meaning that they use drugs regardless medical prescription. Unfortunately, similar to other countries, the uncontrolled use of drugs and personal care products, coupled to the discharge of untreated domestic wastewater or the inefficient removal of this type of substances in conventional wastewater treatment plants trigger the contamination of the aquatic environment in various Mexican regions (Gilabert-Alarcón et al. 2018; Lesser et al. 2018; Pérez-Alvarez et al. 2018).

In some States, wastewater reuse has been implemented without integrative planning and assessment of the impact on ecosystems and public health (Gilabert-Alarcón et al. 2018). Despite the fact that emerging contaminants are rarely monitored in Mexico, the occurrence of pharmaceuticals and personal care products in effluents and surface waters has been investigated. Trimethoprim (0.11–0.15 µg/L, 0.28–0.32 µg/L), clarithromycin (0.37–0.45 µg/L, 1.10–1.40 µg/L), naproxen (6.2–6.74 µg/L, 2.84–3.16 µg/L), and diclofenac (0.25–0.34 µg/L, 0.42–0.50 µg/L) were determined in wastewater from Mexico City-Mezquital Valley (Pumping station "Gran Canal" and Emisor Profundo "El Salto"). The concentrations of these substances were found to be similar to levels reported for sewage in Europe, Japan, and USA. The determined concentrations of metoprolol (0.21–0.25 µg/L) in water from the Gran Canal (Siemens et al. 2008) were similar to those reported in wastewater in Switzerland (0.14–0.29 µg/L) and higher than those found in sewage in Spain (< 0.005–0.09 µg/L).

Lesser et al. (2018) carried out analyses of untreated wastewater used for agricultural irrigation and collected from different points in Mezquital Valley (located in Central Mexico), reporting that from the group of 7 reproductive hormones and 118 pharmaceuticals measured, 3 hormones and 65 drugs were detected. Some examples of pharmaceuticals determined at quite high concentrations include acetaminophen (39,900 ng/L), ciprofloxacin (1190 ng/L), sulfamethoxazole (5360 ng/L), oxytetracycline (134 ng/L), naproxen (11,800 ng/L) and metformin (82,900 ng/L).

In hospital wastewaters from Toluca City (the capital city of the State of Mexico), different pharmaceuticals were quantified, namely antidiabetics (e.g., 1.92 µg/L and 1.31 µg/L for glibenclamide and metformin, respectively), β-blockers (0.20 µg/L for atenolol and 2.02 µg/L for metoprolol), β-lactams (3.77 µg/L for penicillin G and 0.42 µg/L for penicillin V), hormones (0.08 µg/L for 17-β-estradiol), and anti-inflammatory drugs (0.59 µg/L for diclofenac, 0.62 µg/L for ibuprofen, 1.79 µg/L for naproxen and 2.66 µg/L for acetaminophen) (Pérez-Alvarez et al. 2018).

Similarly, the presence of some emerging contaminants was also investigated in five effluents of domestic wastewater produced in the coastal zone of Cihuatlan, Jalisco (a State of Mexico on the edge of the Pacific Ocean) (Arguello-Pérez et al. 2019). In that study, diclofenac, ibuprofen, and ketorolac were determined in all the analyzed effluents at concentrations considered as "toxic," estradiol being in two of the effluents at levels considered as very toxic range.

Díaz and Peña-Alvarez (2017) reported an analytical method to detect ibuprofen, 2-benzyl-4-chlorophenol, naproxen, triclosan, ketoprofen, diclofenac, bisphenol A and estrone in river sediments, showing that sediments from the Tula River (Hidalgo, in central Mexico) had ibuprofen, naproxen and triclosan concentrations in the order of ng/g. The report mentioned that the highest naproxen concentration (240 ng/L) was found in a sample taken from the riverside where wastewater from Ixmiquilpan City was discharged.

Others monitoring program analyzed the occurrence of pharmaceuticals in influent/effluent of the wastewater treatment plant "Acapantzingo" and surface water (Apatlaco River in Cuernavaca, the capital of the State of Morelos in south-central Mexico) (Rivera-Jaimes et al. 2018). The total concentrations of pharmaceuticals were higher in surface water and influent wastewaters than those detected in effluent wastewaters, showing the critical role of wastewater treatment plants. Rivera-Jaimes et al. (2018) indicated that the discharge of untreated wastewater increased the concentration of pharmaceuticals in the river, where naproxen (732–4880 ng/L), acetaminophen (354–4460 ng/L), diclofenac (258–1398 ng/L) and bezafibrate (286–2100 ng/L), were the most abundant.

Recently, Pérez-Coyotl et al. (2019) analyzed water samples from an urban reservoir (Madín dam, located in the municipalities of Naucalpan and Atizapán in the metropolitan area adjacent to Mexico City) that is a source of drinking water and represents an area for recreational activities, including carp fishing. Their results showed different concentrations of personal care products (especially sunscreens) and pharmaceuticals. As example, 2,2′,4,4′-tetrahydroxybenzophenone (13.7 ng/L), 2-ethylhexyl 4-(dimethylamino) benzoate (29 ng/L), 4-methylbenzylidine camphor (134 ng/L) and methyl benzotriazol (250 ng/L), were detected. Very high levels were determined for the antidiabetic agents glibenclamide and metformin (2148 ng/L and 9557 ng/L, respectively) and for the analgesic acetaminophen (9156 ng/L). Hence, it is essential to implement a monitoring program of these and other emerging contaminants in drinking water sources of this region.

The presence of pesticides in water resources is another important problem in Mexico, as in other Latin American countries (García-de la Parra et al. 2012; Arellano-Aguilar et al. 2017). The high availability and convenient cost of agrochemicals, as well as flexible environmental laws have led to an overuse of pesticides in Mexico, in an attempt to satisfy the global demand of agricultural products. Nevertheless, residual agrochemicals in agricultural runoff are rarely monitored by farmers, and some of their leachates drain these pollutants to lakes or rivers (sources of drinking water), which eventually contaminate bays and coasts (Fig. 4) (Osuna-Flores and Riva 2002; Arellano-Aguilar et al. 2017). In particular cases, people of various villages are in contact with this type of hazardous leachates; furthermore, private wells for drinking water, which is untreated (used directly), are located close to agricultural fields (Picos-Corrales et al. 2020). Based on an environmental regulation (NOM-127-SSA1-1994) by the Federal Commission for the Protection against Sanitary Risk (organ of the Health Secretary of Mexico), the maximum limits for the main organochloride pesticides in drinking water are: 30 μg/L for 2,4-D (2,4-dichlorophenoxyacetic acid), 20 μg/L for methoxychlor, 2 μg/L for γ-lindane, 1 μg/L for $\sum$ dichlorodiphenyltrichloroethane and hexachlorobenzene, 0.2 μg/L for chlordane, 0.03 μg/L for heptachlor, aldrin and dieldrin (separated or combined).

Sinaloa is an example of an industrialized agricultural region in Northwestern Mexico that is characterized by a variety of crops and high production. In this State, pesticides have been found in sediments of agricultural drainages (García-de la Parra et al. 2012), with concentrations of organochlorine compounds (15 substances) and organophosphorus compounds (8 substances) within the ranges 0.1–20.19 ng/g and 0.03–1294 ng/g, respectively, being diazinon the compound with the highest concentration. Arellano-Aguilar et al. (2017) also reported that this industrialized agricultural region has a documented pesticides usage of 700 t/year, at least 17 molecules under use being classified as moderately to highly toxic.

With the same focus, Leyva-Morales et al. (2017) assessed the quality of river water from central Sinaloa (Humaya, Tamazula, and Culiacan rivers) by monitoring residual pesticides and suggested that their contamination derived mainly from runoffs of polluted soils. Their results showed the presence of the following chemicals: lindane (0.0041–0.0104 μg/L), endosulfan (0.0198–0.03601 μg/L), heptachlor, dichlorodiphenyltrichloroethane (0.0525 μg/L), diazinon (0.0211–0.0403 μg/L), chlorpyrifos, methyl parathion, cyromazine (0.1961 μg/L), permethrin (0.2351 μg/L), ethion, carbofenothion, lambda cyhalothrin, pirimicarb, aldrin (0.0099–0.1023 μg/L), and malathion.

The relationship between agrochemical loading in drains/rivers and pollution in coastal lagoons of the Mexican Pacific was studied (Arellano-Aguilar et al. 2017), describing that the organochlorine chemicals were clearly accumulated in the coastal lagoons from the drains and rivers, with hexachlorocyclohexanes showing the highest concentration. In another work focused on accumulation of pesticides, dichlorodiphenyltrichloroethane ($\sum$DDTs ~ 0.6–137 μg/kg) was quantified in sediments samples taken from Texcalac River in Tlaxcala (Mexico), exceeding the guidelines for protection of aquatic life according to the authors (García-Nieto et al. 2019).

Yucatan (southeastern Mexico) is reported as a state with high levels of groundwater pollution, where different pesticides have been detected (3.2 mg/L for endrin, 10.86 mg/L for δ-lindane, 5.23 mg/L for γ-lindane, 6.53 mg/L for α-lindane, and 12.54 mg/L for heptachlor) (Polanco-Rodríguez et al. 2018). This type of hazardous chemicals has been also identified in the abovementioned study targeting the drinking water source "Madín dam," namely diazinon up to 12.592 ng/L and fenthion sulfoxide

Fig. 4 Contamination sequence involving river pollution in Sinaloa from agricultural wastewater. Sinaloa is an important agricultural region of Mexico. Source: Lorenzo A. Picos-Corrales, Sinaloa, Mexico

up to 2.465 ng/L (Pérez-Coyotl et al. 2019). Based on the current literature, it is recommended to increase the monitoring work regarding pesticides content in surface water in Mexico.

In addition to monitoring, examples related to the impact of emerging pollutants on human health and ecosystems in Mexico have also been reported. In a study determining some residual personal care product ingredients in hydroponic French lettuce from a local supermarket in Mexico (Cabrera-Peralta and Peña-Alvarez 2018), the results indicated that using wastewater to irrigate crops can result in contaminated food since the absorption by plants is favored in the presence of compounds with intermediate polarity.

Martinez-Gomez et al. (2015) previously assessed the effect of pharmaceuticals and personal care products on aquatic life using the rotifer *Plationus patulus*, which is a member of riverine food networks in Mexico. The results indicated that the continuous exposure to emerging contaminants could reduce rotifer population growth. Hence, the constant discharge of treated wastewater to aquatic ecosystems could represent a risk for the zooplankton community.

In another work, the toxicological risk of a hospital effluent from Toluca (the capital of the State of Mexico) was evaluated using two bioindicator species, i.e., *Xenopus laevis* and *Lithobates catesbeianus* (Pérez-Alvarez et al. 2018). This research group suggested that the hospital effluents represent a latent risk for the ecological balance and diverse organisms, being necessary a prior treatment for the removal of the pharmaceuticals (Pérez-Alvarez et al. 2018).

Based on hazard quotients for pharmaceuticals detected in the Apatlaco River in Cuernavaca (south-central Mexico), Rivera-Jaimes et al. (2018) found that contamination levels of this river could represent a risk for the aquatic ecosystem, due to the high concentrations of naproxen, ibuprofen, diclofenac, and sulfamethoxazole. Similarly, experiments performed using samples from the drinking water source "Madín dam" (Naucalpan-Atizapán, State of Mexico) revealed that residual pharmaceuticals, personal care products and pesticides originated embryotoxicity, embryolethality, congenital abnormalities, and oxidative stress on the common carp embryos. This was a rational explanation regarding the reduction in the population of *Cyprinus carpio* species of this ecosystem (Pérez-Coyotl et al. 2019). Also, in past years in the Bay of Ohuira (Topolobampo, a port on the Gulf of California in northwestern Sinaloa), the decrease in shrimp production was associated, among other causes, with the high coastal pollution by pesticides (Osuna-Flores and Riva 2002). In Chiapas (Southern Mexico), concentrations of organochlorine pesticides were found in a greater amount in cow's milk and forage than in water samples. Despite the content of contaminants was below the limits established by Mexican government, a monitoring program was recommended by the authors (Murga et al. 2016).

It is also estimated that in Yucatan (southeastern Mexico) around 30% of the residents drink water from contaminated sources, and investigations revealed high levels of pesticides (7.352 mg/L for endosulfan I, 2.336 mg/L of 4,4'-dichlorodiphenyldichloroethane, 3.695 mg/L for aldrin and 1.434 mg/L for heptachlor) in the blood of women having cervical uterine cancer. Additionally, other studies showed high levels of these compounds in breast milk (18.436 mg/L for heptachlor epoxide and 2.10 mg/L for dieldrin) (Polanco-Rodríguez et al. 2018).

Concluding, Mexico is facing the environmental problem of emerging substances occurring in its aqueous compartments, which is accentuated by the diversity of substances found and the concentrations levels. Table 1 summarizes examples of contamination by emerging substances reported in water compartments in Mexico, comparing them with those reported by Colombian and Brazilian research groups. It is necessary to mobilize all the stakeholders and the population to protect aquatic ecosystems in order to ensure the production of quality water. To achieve this goal, a thorough monitoring of surface waters is necessary and indispensable as part of a vast program at several spatial and temporal scales, not only at the national level but also as part of a more comprehensive water policy in collaboration with Mexico neighboring countries. In addition, attention should also be paid to treatment plants and their decontamination performance, as part of a more organizational water management at national and regional levels.

Emerging contaminants in Colombian rivers

Colombia is a country with an area of 1,141,748 km^2, a large marine area covering 928,660 km^2 and a population of 46,581,823 (*Departamento Administrativo Nacional*, DANE 2011). Colombia has a privileged geographical position in South America with direct access to both ocean Atlantic and Pacific. Colombia has many water sources throughout its territory and the most important biodiversity not only in the region, but also in the World. The country has many rivers, the most important of which are the following: Amazon (6992.6 km, shared with Peru and Brazil), Caquetá (Japura, 2,816.3 km, shared with Brazil), Negro (2230.5 km, shared with Brazil and Venezuela), Orinoco (2140.4 km, shared with Venezuela), Putumayo (1609.3 km, shared with Peru and Brazil), Guaviare (1496.6 km, shared with Venezuela), Arauca (1049.2 km, shared with Venezuela), Cauca (965.6 km), Goal (804.6 km, shared with Venezuela), Magdalena (528.8 km), and its affluent Rio Bogotá. Bogotá River starts 3,300 m above sea level and flows for 380 km to Girardot, where it merges into the Magdalena River. Its basin is located in the center of the country, in the department of Cundinamarca, with an area of approximately 6.000 km^2 (Hernández et al. 2015; Bedoya-Ríos et al. 2018; Bedoya-Ríos and Lara-Borrero 2018).

Colombia income is mainly originated on the production of raw materials to export and to manufacture consumer products for the domestic market. Its main activities include the production of oil for exportation (fourth in Latin America and sixth in the continent) and mining, particularly carbon, gold, emeralds, sapphires and diamonds (Biazil, 2016). The strongest industrial sectors in Colombia are petrochemicals, textiles, automobiles, and chemicals.

Like most countries in the World, Colombia is vulnerable to the problem of emerging compounds and Colombian developing regions are concerned (Martínez Vidal et al. 2006; Barceló and Petrovic 2008; Ibáñez et al. 2008, 2009; Klamerth et al. 2009; Teijón et al. 2010; Tóbon-Marulanda et al. 2010; Gil et al. 2012; Díaz-Casallas et al. 2019; Reichert et al. 2019). As in many other developing countries, Colombia's indiscriminate industrial growth has led to severe pollution of its waterbodies. Several studies have shown that many water sources are heavily contaminated, including by emerging contaminants. The sources of contamination are industrial, agricultural, and domestic, the latter often uncontrolled. Once entered into the environment, chemicals, and their metabolites may produce subtle effects on aquatic and terrestrial organisms, especially on the former since they are exposed to long-term continuous influx of wastewater effluents.

Recently, Díaz-Casallas et al. (2019) proposed a comprehensive analysis of the water quality at the upper basin of the Bogotá River between 2008 and 2017, pointing out an insufficient quality of water. This study highlighted the necessity for further efforts on the continuous monitoring of Colombian rivers basins. Indeed, Colombia is currently focusing its efforts on the detection and the quantification of emerging substances, in order to compile a list of priority substances adjusted to the needs and characteristics of Latin America. Research programs are also underway to provide recommendations for future monitoring, prevention, and elimination programs for these chemicals. It is important not only to monitor discharge waters from municipal treatment plants and industrial wastewaters but also to encourage

Table 1 Contamination by emerging substances reported in water compartments in Colombia, Mexico and Brazil, from 2013 onward

Country	Water type	Emerging substance	Concentration (ng/L)	References
Colombia (La Fe)	Drinking water supply reservoir	Ibuprofen	7–39	Aristizabal-Ciro et al. (2017)
Colombia (Rio Grande)	Drinking water supply reservoir	Ibuprofen	5–62	Aristizabal-Ciro et al. (2017)
Mexico (Mexico City)	Surface water	Ibuprofen	15–45	Félix-Cañedo et al. (2013)
Mexico (Toluca City)	Hospital wastewater	Ibuprofen	620	Pérez-Alvarez et al. (2018)
Mexico (Morelos)	Surface water	Diclofenac	258–1398	Rivera-Jaimes et al. (2018)
Mexico (Mexico City)	Surface water	Diclofenac	28–32	Félix-Cañedo et al. (2013)
Mexico (Morelos)	wastewater	Diclofenac	600–2500	Rivera-Jaimes et al. (2018)
Mexico (Cuernavaca)	Wastewater	Diclofenac	258–1398	Rivera-Jaimes et al. (2018)
Mexico (Toluca City)	Hospital wastewater	Diclofenac	590	Pérez-Alvarez et al. (2018)
Mexico (Morelos)	Surface water	Naproxen	732–480	Rivera-Jaimes et al. (2018)
Mexico (Tula River)	Surface water	Naproxen	240	Díaz and Peña-Alvarez (2017)
Mexico (Mexico City)	Surface water	Naproxen	52–186	Félix-Cañedo et al. (2013)
Mexico (Mezquital Valley)	Wastewater	Naproxen	11,800	Lesser et al. (2018)
Mexico (Cuernavaca)	Wastewater	Naproxen	732–4880	Rivera-Jaimes et al. (2018)
Mexico (Morelos)	Wastewater	Naproxen	800–4200	Rivera-Jaimes et al. (2018)
Mexico (Toluca City)	Hospital wastewater	Naproxen	2660	Pérez-Alvarez et al. (2018)
Brazil (São Paulo)	Surface water	Paracetamol	30,421	Campanha et al. (2015)
Brazil (São Paulo)	Surface water	17-α-ethinylestradiol	777	Montagner et al. (2019)
Brazil (São Paulo)	Surface water	Caffeine	19–127,000	Montagner et al. (2019)
Brazil	Surface water	Caffeine	40–19,000	Machado et al. (2016)
Brazil (São Paulo)	Surface water	Caffeine	129.585	Campanha et al. (2015)
Brazil	Drinking water	Caffeine	1.8–2000	Machado et al. (2016)
Brazil (São Roque)	Drinking water	Caffeine	4083	Peña-Guzmán et al. (2019)
Brazil (São Paulo)	Drinking water	Caffeine	121	Montagner et al. (2019)
Brazil (Rio Grande)	Drinking water supply reservoir	Caffeine	18,828	Machado et al. (2016)
Brazil (São Paulo)	Surface water	Bisphenol A	2–13,016	Montagner et al. (2019)
Brazil (Rio Grande)	Drinking water supply reservoir	Bisphenol A	11	Machado et al. (2016)
Brazil (São Paulo)	Drinking water	Bisphenol A	23	Montagner et al. (2019)
Brazil (São Roque)	Drinking water	Bisphenol A	0.5	Peña-Guzmán et al. (2019)
Brazil (São Paulo)	Surface water	Cocaine	10	Montagner et al. (2019)
Brazil (São Paulo)	Surface water	Benzoylecgonine	133	Montagner et al. (2019)
Colombia (Bogotá)	Wastewater	Benzoylecgonine	4000	Bijlsma et al. (2016)
Colombia (Bogotá)	Wastewater	Amphetamines	12–68	Bijlsma et al. (2016)
Brazil (São Paulo)	Drinking water	Triclosan	2–37	Montagner et al. (2019)
Brazil (Rio Grande)	Surface water	Atrazine	5–49	Caldas et al. (2019)
Brazil (São Paulo)	Drinking water	Atrazine	687	Montagner et al. (2019)
Brazil (Rio Grande)	Drinking water	Atrazine	5–37	Caldas et al. (2019)
Brazil	Drinking water	Atrazine	2–6000	Machado et al. (2016)
Mexico (Yucatan)	Groundwater	Endrin	3.2 mg/L	Polanco-Rodríguez et al. (2018)
Mexico (Yucatan)	Groundwater	γ-lindane	10.86 mg/L	Polanco-Rodríguez et al. (2018)
Mexico (Yucatan)	Groundwater	Heptachlor	12.54 mg/L	Polanco-Rodríguez et al. (2018)

the improvement of their systems and treatment techniques in order to achieve better removal efficiencies.

The list of substances that have been found in effluents from wastewater treatment plants, and consequently in Colombian waters (surface water, groundwater) is extr

emely diverse, ranging from pharmaceuticals, hormones and steroids to surfactants, cosmetics, and pesticides (Hernández et al. 2005, 2012, 2015; Martínez Vidal et al. 2006; Ibáñez et al. 2008, 2009; Tóbon-Marulanda et al. 2010; Bijlsma et al. 2016; Sarria-Villa et al. 2016; Portilla et al. 2017; Botero-Coy et al. 2018; Serna-Galvis et al. 2019; Reichert et al. 2019). These studies also helped to improve the understanding about the dynamics and behavior of the target compounds.

The substances most frequently found in Colombian water sources were pharmaceuticals such as antihypertensives, antibiotics, analgesics and psychiatric products, and illicit substances, e.g., cocaine and its main metabolite, heroine, and amphetamines (methamphetamine). They are usually detected at trace levels, usually between ng/L and μg/L, depending on the target molecule. Raw wastewater from the cities of Bogotá, Medellín, and Florencia contain levels of several pharmaceuticals above 1 μg/L, while the concentrations of antibiotics are commonly above 5 μg/L in raw hospital wastewater.

Indeed, Colombia is facing the problematic of the high use of pharmaceutical products and cosmetics. Among these substances, antihypertensives (valsartan, irbesartan), antibiotics (ciprofloxacin, norfloxacin), and endocrine disruptors are of great concern and their hazardous effects are already reported (Darbre 2015; Gee et al. 2015). Triclosan is also a substance of concern, with hydrophobic nature and thus it can accumulate in fatty tissues, such as fish and human samples (urine, breast milk, and serum). Moreover, triclosan has been shown to originate toxic effects in aquatics organisms at environmental concentrations and its degradation products can be more toxic and persistent than the parent compound (Kosma et al. 2014; Juliano and Magrini 2017; Machado et al. 2016; Montagner et al. 2019).

Ciprofloxacin, one of the top five most common antibiotics prescribed in Colombian hospitals, was reported at high frequency in hospital effluents, with a variable load from hospital wastewater between studies (Botero-Coy et al. 2018; Serna-Galvis et al. 2019). Another problem is the environmental presence of endocrine disruptors (e.g., natural (17-β-estradiol) and synthetic (17-α-ethinylestradiol) hormones), which are capable of mimicking the hormones produced by living organisms and interfere with various functions (Darbre 2015). Natural and synthetic hormones may end up in water via domestic, industrial, and/or hospital discharges. As referred above, conventional municipal wastewater treatment plants are not designed to deal with these substances and their metabolites, which can both reach the aquatic environment. Endocrine disrupting effects such as intersexuality and reproductive disorders have been reported in fish and other aquatic organisms living downstream from wastewater treatment plant outfalls (Vajda et al. 2008; Gagné et al. 2011; Tetreault et al. 2011).

Cocaine is a highly consumed illicit drug in populated Colombian cities such as Bogotá, and consequently, its main metabolite (benzoylecgonine) has a high prevalence in wastewaters (Bijlsma et al. 2016; Hernández et al. 2015; Serna-Galvis et al. 2019). Bijlsma et al. (2016) conducted for the first time a wastewater-based epidemiology in Colombia, reporting that cocaine was the most consumed illicit drug, particularly in Medellin (department of Antioquia), whereas the excreted metabolite benzoylecgonine exceeded 4000 ng/L in all wastewater treatment plant samples. Amphetamines were also found in all samples at concentrations between 12 and 68 ng/L (Bijlsma et al. 2016). Illicit substances, together with pharmaceuticals, are commonly measured in Colombian effluents as both groups of substances are not completely removed with conventional treatments (Serna-Galvis et al. 2019).

Another important issue is the aquatic presence of pesticides, such as glyphosate, dichlorodiphenyltrichloroethane, parathion, and clofibric acid, since Colombia is a major agricultural country that relies on the use of phytosanitary products. A typical example is the presence of glyphosate used to protect illicit crops. The frequent detection of this substance suggests that it is a widespread environmental contaminant. Another typical example is the insecticide dichlorodiphenyltrichloroethane that has well-known adverse health consequences, namely an established link with malformations in newborns and significant impacts on fish and reptiles (deformities, reduced fertility, and abnormal behavior).

There are also industrial products such as phthalates, bisphenol A and its analogues (bisphenol F and bisphenol S) used in the production of resins and plastics, paints/lacquers and binding materials, or cleaning agents (nonylphenol). Another example is triclosan used as a broad spectrum antibacterial agent into many consumer products and as preservative in products of domestic use. It is also found in cosmetics, textiles, and antibacterial fibers. These substances have been identified and quantified in industrial wastewaters and in rivers.

In Colombia and in other Latin America countries, the situation is worrying not only from a water pollution point of view but also from a health point of view, with the appearance of resistance phenomena for certain bacteria. Antibiotics are antimicrobial drugs that kill or reduce the growth of bacteria. They have been used in large quantities for several decades around the world, favoring the selection and the spread of antibiotic-resistant pathogens causing difficult-to-treat infections (García et al. 2020). Resistance to antibiotics has been for long a focus of research in clinical settings and, more recently, in environmental research. One widely reported case is that of *Staphylococcus aureus*, which has developed resistance to methicillin (MRSA, methicillin-resistant *Staphylococcus aureus*). There are three main routes of entry of antibiotics into freshwater: (1) effluents from wastewater treatment plants, (2) chemical manufacturing plants, and (3) livestock, agricultural, and aquaculture sites. Indeed, antibiotics can bypass water treatment processes and end up directly in the environment. They are detected in Colombian rivers at very low concentrations and are diluted more than a million times compared to concentrations in the human body. However, these residual concentrations are highly likely to generate or to favor antimicrobial resistance, having direct and indirect effects on the microbial component of aquatic communities. Indeed, even at low concentrations, these antibiotics could significantly impact ecosystems and human health.

In conclusion, the presence of emerging substances in Colombian waters is a concern that is becoming increasingly important in society. The demand for water, whether for domestic, agricultural, or industrial use, is constantly increasing, with the consequences of water pollution and ecosystem degradation, as a significant amount of wastewater is either untreated or poorly treated. This scenario is complicated by a climate context that is affecting the predictability of our most precious resource. All water stakeholders are concerned by this issue and mobilized. There is a growing consensus that the challenges can be tackled by adopting an integrated approach for water resources management. In particular, special efforts need to be made in the operation of municipal wastewater treatment plants and in the protection of the ecosystems and water resources.

Analysis of emerging contaminants in Brazilian water resources

Like Mexico and Colombia, the presence of the same wide group of emerging contaminants (pharmaceuticals, personal care products and cosmetics, industrial substances, and pesticides) in Brazil's water resources, including rivers, lakes, drinking water reservoirs, and tap water, has been reported at the same orders of magnitude of concentrations, showing that the conventional wastewater treatments are not effective for their removal, even in regions where exist adequate sanitation. Indeed, numerous studies indicated that the conventional applied wastewater treatments are not efficient enough to eliminate these substances from wastewater and sludge, resulting in their presence in the environment (Barbosa et al. 2015; Campanha et al. 2015; Albuquerque et al. 2016; Machado et al. 2016; Caldas et al. 2019; Starling et al. 2019).

The collection, treatment, and reuse of wastewater from households and industry, the reduction of diffuse pollution, and the improvement of water quality are major challenges for Brazilian water stakeholders. Freshwater quality is indeed under threat. Its pollution is widespread and increasing in many regions. In Brazil, regulations concerning emerging contaminants in both natural waters and drinking water require a commitment among researchers and regulatory authorities, since this issue has not been considered as a priority by the government (Machado et al. 2016).

The substances that have been analyzed in Brazilian waters include mainly, hormones, antibiotics, endocrine disruptors, caffeine (as licit drug), illicit drugs (methamphetamine, cocaine, heroin), industrial compounds (bisphenols, triclosan), and pesticides. Among them, pesticides in particular have been monitored, as Brazil is the World's largest consumer of herbicides, fungicides, and insecticides (Barbosa et al. 2015; Albuquerque et al. 2016; Caldas et al. 2019; Starling et al. 2019), which means that these substances may end up in the environment, especially in aquatic systems and reservoirs used for drinking water production. There are 381 active ingredients authorized by the Brazilian Ministry of Agriculture for pesticides used on crops and 1,670 formulated plant protection products in the market (Albuquerque et al. 2016). Brazil has regulations on the use of pesticides and their presence in water. For example, the Brazilian drinking water norm (Ordinance 2914/2011 of the Ministry of Health) includes 64 chemical substances, of which 27 are pesticides that must be monitored every 6 months. However, this number represents < 10% of the current pesticide active ingredients approved for use in the country (Barbosa et al. 2015). Of the 27 pesticides, 21 are banned in Europe due to the risks they offer to health and the environment. A comprehensive review on the presence of pesticides in Brazilian freshwaters was published by Albuquerque et al. (2016). They reported that data in peer-reviewed literature are scarce and often incomplete, concluding that, without the implementation of a nationwide pesticide freshwater monitoring program and a clear definition of proper water quality standards, it is not possible to evaluate the risks posed by pesticides to the aquatic life in Brazil.

Among the most widely used pesticides in Latin America, glyphosate and atrazine occupy prominent positions as the best-selling active ingredients in Brazil (Brazil 2011; Barbosa et al. 2015; Marin et al. 2019). Since its introduction as an herbicide active ingredient in 1971, glyphosate became and remains the market leading herbicide worldwide (glyphosate-based formulations are mainly represented by Roundup). The limit of glyphosate in drinking water in Brazil is 0.5 µg/L (Brazil 2011; Barbosa et al. 2015), while in Europe only 0.1 µg/L is permitted (0.9 µg/L for World Health Organization, 0.8 µg/L for Canada). The 0.1 µg/L value is also the maximal residue level of glyphosate in European surface and groundwater. The maximum contaminant level in the U.S. in drinking water is 700 µg/L (sum of glyphosate and its residues) and the health-based guideline value in Australia is 1000 µg/L (Székács and Darvas 2018). The contamination of the environment by glyphosate is worrying since it impairs enzymatic activity, causes cytotoxicity and DNA data in human cells (Marin et al. 2019).

In drinking and surface waters, acceptable limit for atrazine (herbicide of the triazine class) and methyl parathion (organophosphate insecticide) are 9 and 2 µg/L, respectively (Ordinance 2914/2011 of the Ministry of Health). These limits are based on toxicological and neurotoxicological trials, but they do not take into account possible estrogenic effects that atrazine can cause when human and wildlife are chronically exposed to low concentrations (Machado et al. 2016). In Canada, the maximum acceptable concentration for atrazine in drinking water is 5 µg/L (this value is applicable to the sum of atrazine and its metabolites) while for the United States Environmental Protection Agency, the maximum concentration in water is 3 µg/L. In the European Union, a limit of 0.1 µg/L has been set for all pesticide residues in drinking water and groundwater. This value of 0.1 µg/L for each substance represents a standard for water quality, not for the health risk. If more than one pesticide is found in drinking water, the sum of all pesticides has a legal limit of 0.5 µg/L (source: French Agency for Food, Environmental and Occupational Health and Safety).

Sodré et al. (2010) investigated the occurrence of endocrine disruptors in drinking water of the city of Campinas. Six emerging contaminants (stigmasterol, cholesterol, bisphenol A, caffeine, estrone, and 17-β-estradiol) were found in 12 samples. Estrone and 17-β-estradiol were detected only during the dry season, with concentrations below quantification limits. The highest average concentration was reported for stigmasterol (0.34 ± 0.13 µg/L), followed by cholesterol (0.27 ± 0.07 µg/L), caffeine (0.22 ± 0.06 µg/L), and bisphenol A (0.16 ± 0.03 µg/L). Worryingly, the level of bisphenol A detected in drinking water (0.0005 µg/L) was higher than the average effluent from effluent treatment plants (0.00002 µg/L) (Sodré et al. 2010). Ten years later, similar levels of concentrations were reported by Peña-Guzmán et al. (2019). Sodré et al. (2010) reported that the emerging contaminants levels in drinking waters of Campinas were higher than median values compiled for drinking and finished water samples around the World, which is related to surface drinking water supplies receiving large amounts of raw sewage inputs. Montagner and Jardim (2011) also reported that among endocrine disruptors, bisphenol A (a compound used as a manufacturing intermediate) is distinguished by its widespread presence in the environment. In the State of São Paulo, this substance was detected in all surface water sampling campaigns with concentrations between 204 and 13,016 ng/L (Montagner and Jardim 2011).

A special substance in Latin America is caffeine. It is consumed by people of all ages, cultures and socioeconomic status (Campanha et al. 2015; Sodré et al. 2018; Peña-Guzmán et al. 2019; Starling et al. 2019). As caffeine is present in everyday beverages such as coffee, tea, chocolate, and cola drinks, a large amount of caffeine waste is generated, which enters the aquatic environment. Many drugs also contain caffeine, namely analgesics, antihistamines, diet pills, cold remedies, and stimulants of psychophysical activity. Campanha et al. (2015) reported a 3-year study on the occurrence of pharmaceuticals and hormones in surface waters of a central urban region of São Paulo State of Southeast Brazil (the Monjolinho River in São Carlos). The most frequently detected compounds at highest concentrations were caffeine, paracetamol, and atenolol (maximum concentrations 129.585 µg/L, 30.421 µg/L, and 8.199 µg/L, respectively), while the hormones estrone and 17-β-estradiol were the least detected, at levels up to 0.0148 ng/L. The results also showed that there was an increasing trend in concentrations of most of the compounds along the river course, especially downstream of the river where there are discharges of both wastewater treatment plant effluent and raw sewage from a particular region of São Carlos city. The concentrations of contaminants were higher during dry periods as a result of the decline in the water levels. A decrease in concentrations occurred near the river mouth at different extents for each compound, being high for caffeine and atenolol and very low for carbamazepine and diclofenac.

The first nationwide survey of emerging pollutants in Brazilian waters has been published by Machado et al. (2016), including one hundred drinking water samples investigated in 22 Brazilian State capitals during two campaigns between July and September of 2011 and 2012, as well as seven source-water samples from two of the most populous regions of the country were evaluated. The monitoring program showed that caffeine, triclosan, atrazine, phenolphthalein, and bisphenol A were detected in at least one of the samples collected in the two sampling campaigns. Caffeine and atrazine were the most recurrently detected compounds in both drinking and source water, in agreement with the data published by Campanha et al. (2015). Caffeine concentrations in drinking water ranged from 1.8 ng/L to values above 2.0 µg/L while source-water concentrations varied from 40 ng/L to approximately 19 µg/L. The frequency of detection of caffeine corresponded to 93%, corresponding to a higher frequency than those found in similar Spanish and Chinese studies (< 88% in the two cases). For atrazine, concentrations were found ranging from 0.002 to 0.006 µg/L in drinking water and at concentrations of up to 0.15 µg/L in source water. The levels of atrazine were between two and three orders of magnitude below the maximum limit established by the Brazilian regulations (2.0 µg/L is the authorized value in drinking and surface water). Atrazine was found in 75% of the drinking water samples collected in both campaigns. Only two other substances, triclosan, and phenolphthalein, were detected in drinking water samples. Emerging contaminants such as pharmaceuticals, hormones, and industrial products were not detected in the samples. Caffeine, atrazine, and bisphenol were detected in all seven surface samples. The highest concentrations of caffeine and bisphenol A detected were 18.828 µg/L and 0.011 µg/L in a reservoir connected to the Rio Grande River. Machado et al. (2016) concluded that in both drinking and source waters, caffeine reached concentration values from a few ng/L to µg/L, depending on the region of collection. The detection frequency of caffeine in drinking water was higher than other values reported in the literature. Its widespread presence in samples of treated water (detected in 93% of samples) suggested the presence of domestic sewage in the source water, considering that caffeine is a compound of anthropogenic origin, exhibiting great residence time in the environment and low susceptibility to degradation. Caffeine can be proposed as an indicator of contamination by sewage (Campanha et al. 2015; Machado et al. 2016).

Montagner et al. (2019) summarized the results of 10 years of analyses carried out in the State of São Paulo that has one of the highest population densities in Brazil and intense agricultural and industrial activities. In this report, 58 compounds (9 hormones, 14 pharmaceuticals and personal care products, 8 industrial compounds, 17 pesticides, and 10 illicit drugs) were analyzed (2006 – 2015). Samples were collected in 13 cities in the State of São Paulo (drinking water), in 10 rivers (surface waters) and 4 reservoirs, and in 5 municipal wastewater treatment plants. The detection frequency of each molecule intensely varied among samples. The average concentration for the synthetic hormone 17-α-ethinylestradiol was 777 ng/L and the highest frequencies of detection in surface water were in the following order: caffeine (97%), atrazine (69%), triclosan (43%), estriol (31%), estrone (28%) and testosterone (13%). Caffeine varied from 19 to 127,000 ng/L, showing the differences between sampling points according to their anthropogenic impact, similar to the results reported by Campanha et al. (2015) in the Monjolinho River, the main water body of São Paulo. Bisphenol A was quantified in 145 samples in a wide range of concentrations, between 2 and 13,016 ng/L.

Cocaine ant its metabolite (benzoylecgonine) were detected in 53% (average concentration: 10 ng/L) and 84% (average concentration: 133 ng/L) of the samples, respectively. For drinking water collected between 2007 and 2015, bisphenol A was the most frequently analyzed substance (average concentration of 23 ng/L and a maximum concentration of 178 ng/L) followed by caffeine (average concentration of 548 ng/L), testosterone (average concentration of 3 ng/L), 4-n-nonylphenol (average concentration of 114 ng/L), triclosan (concentrations ranging from 2 to 37 ng/L), and atrazine (average concentration of 36 ng/L). All hormones were analyzed in more than 100 samples, but the frequencies of detection were between 0 and 4%, the maximum concentration being 125 ng/L for estriol.

Other substances such as dioctylphthalate and octylphenols were found in drinking water. From the 33 groundwater samples collected, the most ubiquitous substance was atrazine (86% of samples 6/7) followed by caffeine (55%, 17/31) and bisphenol A (50%, 16/32). Finally, a preliminary risk assessment for aquatic life protection identified potential risks for many compounds: caffeine, paracetamol, diclofenac, 17-α-ethinylestradiol, 17-β-estradiol, estriol, estrone, testosterone, triclosan, 4-n-nonylphenol, bisphenol A, atrazine, azoxystrobin, carbendazim, fipronil, imidacloprid, malathion, and tebuconazole. As expected, raw wastewaters had high levels of concentrations, with the highest values reported for caffeine, benzoylecgonine, and cocaine. Peña-Guzmán et al. (2019) reported similar findings, but some emerging contaminants were still detected even after treatment at relatively high concentrations in the treated wastewaters. Montagner et al. (2019) suggested that the wastewater treatment technology used in São Paulo State (Brazil) was not effective for the elimination of most of the target compounds. Campanha et al. (2015) also indicated that the main source of emerging contaminants in the river was related to the discharges from wastewater treatment plants.

Drinking water criteria were available only for 22 compounds of those studied by Montagner et al. (2019) and for those substances no adverse effects were expected at the concentrations found, except for 17-β-estradiol and atrazine. Montagner et al. (2019) concluded that, among the 58 studied contaminants, caffeine, estrone, 17-α-ethinylestradiol, 17-β-estradiol, bisphenol A, atrazine, carbendazim, fipronil, malathion, and imidacloprid were underlined as substances of priority concern, due to the high frequency of detection above water quality criteria. Another important conclusion was the fact that caffeine and benzoylecgonine were pertinent anthropogenic indicators, especially for regions highly inhabited and with poor sanitation structure. Since the cocaine metabolite can persist in the aquatic environment even after 2 weeks from the discharge, this substance may be a reliable marker of contamination of surface water and also for drinking water.

Caldas et al. (2019) monitored 33 pesticides in surface and drinking water in Southern Brazil (data of 4-year monitoring; 48 samples) and reported that 30 of them were identified and quantified, with four compounds (tebuconazole, atrazine, azoxystrobin, clomazone) being identified in more than half of the samples. Five class of pesticides, i.e., triazines, triazoles, carbamates, strobilurins, and imidazolinones, were identified. Among them, the herbicide atrazine was the most commonly detected pesticide, with concentrations ranging from 0.005 to 0.049 µg/L in drinking water and from 0.005 to 0.037 µg/L in surface water. In this study, atrazine was identified as a priority substance among those that pose a significant risk to the aquatic environment. The authors hypothesized a relationship with agricultural activities since atrazine is widely used in Brazil as a weed-killer in the cultivation of crops (rice, soybeans, sugarcane, and corn).

Other pesticides detected in the whole 4-year monitoring period included the fungicide azoxystrobin (concentrations 0.041–0.233 µg/L in surface water and 0.1–0.192 µg/L in drinking water), the herbicide clomazone (identified in more than half of the samples at concentrations up to 0.164 µg/L) and the triazole compounds such as cyproconazole, difenoconazole, epoxiconazole, propiconazole, and tebuconazole (identified in more than 80% of the samples at concentrations up to 0.46 µg/L). Triazole substances are used not only as fungicides in agriculture and as biocides but also as antifungal agents in human and veterinary pharmaceuticals. Caldas et al. (2019) concluded that the concentrations quantified (ranging from 0.004 to 1 µg/L) were of the same order of magnitude as those detected in other Brazilian regions. The concentrations of the target pesticides detected in drinking water in the city of Rio Grande were below the limits established by the Brazilian legislation and by the World Health Organization. However, considering the values established by the European Union for individual pesticides in drinking water of 0.1 µg/L, the maximum values detected for some pesticides (e.g., azoxystrobin, cyproconazole, clomazone, quinclorac, and tebuconazole) exceeded this limit.

Numerous Brazilian studies come to the conclusion that, in many Brazilian regions that suffer from poor sanitary conditions, there is an urgent need to study the occurrence of emerging contaminants in natural waters. Those reports show the importance of continuous monitoring of this type of substances and other water parameters for proper water management. The use of pesticides should be minimized, and if necessary, agricultural practices should be modified. Another important action should focus on improving the treatment processes applied in the municipal wastewater treatment plants (Sodré et al. 2018; Montagner et al. 2019; Peña-Guzmán et al. 2019; Caldas et al. 2019; Starling et al. 2019).

In conclusion, the extent of industrial pollution is not yet well known in Latin America, as discharges are poorly monitored and rarely aggregated at the national level. Although some domestic and industrial wastewater is on-site treated, few information is available and combined in national and regional assessments, unlike in Europe. Many countries lack the capacity to collect and analyze the data needed for a comprehensive assessment. Reliable monitoring of water quality is nevertheless essential to guide investment priorities. It is also important for assessing the status of aquatic ecosystems and the need to protect and/or restore them.

Conclusion

This review shows that emerging contaminants are ubiquitous in water resources worldwide, exemplified by data from China, Portugal, Mexico, Colombia, and Brazil. All continents are affected by the presence of pharmaceuticals, hormones, cosmetics, antibiotic-resistant bacteria, and pesticides in waterbodies. This occurrence can be explained by municipal, hospital, industrial, and agricultural discharges and by the insufficient removal achieved by the traditional treatments used in domestic wastewater treatment plants. Upgrading wastewater treatment plants with more efficient treatment technologies, such as the implementation of tertiary treatment systems, would help reduce the pollution of waterbodies.

Humanity's water needs are constantly increasing (population growth, rapid urbanization, overexploitation of resources, economic issues). However, the multiform pollution resulting from our lifestyles and growing consumption patterns are major threats to this vital element that is water, with consequences also for the environment and human health. Indeed, water pollution worries us all. We must (continue) to mobilize to protect this resource.

Acknowledgements Nadia Morin-Crini and Grégorio Crini (Besançon, France) thank the FEDER (*Fonds Européen de Développment Régional*) for its financial support (NIRHOFEX Program) and the *Région Bourgogne Franche-Comté* (France) for the research grant awarded to Guest Research Director G. Torri. Ana Rita Lado Ribeiro (Porto, Portugal) would like to thank FCT funding under the DL57/2016 Transitory Norm Programme and the scientific collaboration under projects: PTDC/QUI-QAN/30521/2017—POCI-01-0145-FEDER-030521—funded by FEDER funds through COMPETE2020—*Programa Operacional Competitividade e Internacionalização* (POCI) and by national funds (PIDDAC) through FCT/MCTES and Base-UIDB/50020/2020 and Programmatic-UIDP/50020/2020 funding of LSRE-LCM of the Associate Laboratory LSRE-LCM, funded by national funds through FCT/MCTES (PIDDAC).

Funding The following funders are thanked for their contribution: FEDER, Région Bourgogne Franche-Comté, Portuguese National Programmes.

Declarations

Conflict of interest The authors declare no conflict of interest.

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