Coping with Emerging Contaminants in Potable Water Sources

Heather E. Gall and Odette Mina

Abstract Humans use a large variety of chemicals in their everyday lives including over-the-counter medications, prescription drugs, and personal care products. The chemicals that comprise these items enter wastewater treatment systems when they are manufactured by companies and used by consumers. Wastewater treatment plants have various removal efficiencies, causing these chemicals, generally referred to as "emerging contaminants," to enter surface water bodies. In addition to human sources of emerging contaminants, veterinary pharmaceuticals and hormones are given to livestock raised in concentrated animal feeding operations. The land application of biosolids and animal waste to agricultural fields as a fertilizer source also introduces emerging contaminants into the environment. Recent advances in technology have allowed researchers to detect these compounds in water samples at significantly lower concentrations, thereby allowing researchers to assess the exposure of humans and aquatic species to concentrations at the partsper-trillion level. This chapter provides an overview of the types of emerging contaminants found in potable water sources, their major sources, issues associated with their removal in treatment plants, and a social perspective of the public's concerns regarding emerging contaminants in their potable water.

Keywords Pharmaceuticals • Endocrine disrupting compounds • Drinking water • Wastewater treatment

H.E. Gall $(\boxtimes) \cdot$ O. Mina Department of Agricultural and Biological Engineering, The Pennsylvania State University, University Park, PA, USA e-mail: heg12@engr.psu.edu

1 Introduction

On a global scale, access to clean, safe drinking water is an ongoing concern. Nearly one billion people do not have access to this very basic necessity. Globalization and the increase in human population have led to a wide range of problems related to water quantity and quality. Thousands of tons of personal care products are produced annually [\[1](#page-28-0)] and the number of prescriptions has increased from 2 to 3.9 billion in the United States from 1999 to 2009 [\[2](#page-28-0)]. In the United States, the number of chemicals produced tripled from 1941 to 1995 [\[3](#page-28-0)], with 80,000 chemicals in currently in use [[4\]](#page-28-0). These products bring benefits along with great concerns for human health and the environment. The risks associated with exposure to consumption of such products are not yet well understood, and therefore regulations for surface water and finished drinking water have not been established. This class of compounds, generally referred to as "emerging contaminants" (ECs), includes hormones, pharmaceuticals, and personal care products (PPCPs). These chemicals continue to raise significant concerns among public health professionals, engineers, and scientists, as many are known or suspected to have endocrine disrupting properties. The occurrence of these contaminants has been documented in surface water and groundwater, but the extent of their distribution and the consequences of their presence are largely unknown.

In 2002, the United States Geological Survey (USGS) brought attention of the widespread detection of emerging contaminants (ECs) to scientists with results of the first nationwide study of organic wastewater contaminants (OWCs) [\[5](#page-28-0)]. This seminal study, which has been cited more than 2,800 times, included the collection of surface water samples from nearly 140 locations across the United States. The collected samples were tested for pharmaceuticals, hormones, and other OWCs, and 80 % of the sampled locations tested positive for at least one EC. It was this study that formed the basis for new areas of water quality research, including the fate and transport of ECs, the removal efficiency of water and wastewater plants of these unregulated compounds, and the understanding of their effects on human health and aquatic ecosystems.

In 2008, an Associated Press investigation surveyed 62 major water providers and found that at least 46 million US residents receive drinking water from water bodies contaminated with trace levels of one or more prescription drugs [\[6](#page-28-0)]. Sources of these contaminants include treated and untreated municipal sewage, industrial chemical wastes, surface application of manure and biosolids, and agricultural livestock wastes. The detected concentrations of ECs can be orders of magnitude lower than "traditional" contaminants, which are regulated by the Environmental Protection Agency (EPA) and have drinking water standards. However, concentrations as low as 1 ng/L are known to cause adverse effects on sensitive aquatic species [\[7](#page-28-0)] and the long-term effects on humans are not well understood. In addition, these pollutants undergo various physical, chemical, and biological transformations, with the products potentially causing additional risk to human health and aquatic ecosystems. Perhaps more importantly, these chemicals are known to behave synergistically, causing poorly understood magnified effects when multiple compounds are present in the water simultaneously.

This chapter will explore major sources of ECs, their presence in drinking water sources, and the types of contaminants that have been detected and have the potential to adversely affect human health and aquatic ecosystems. In this chapter, we will also review the capabilities of currently available treatment technologies to remove these contaminants, and will close with an overview of public concerns regarding the presence of these unregulated contaminants in drinking water. The underlying motivation for this chapter is to provide readers with a general appreciation for the widespread nature of this problem and the need for ongoing research to adequately address this growing problem.

2 Emerging Contaminants: What Is in Drinking Water Sources?

The USGS defines ECs as "any synthetic or naturally occurring chemicals or any microorganisms that are not commonly monitored in the environment but have the potential to enter the environment and cause known or suspected adverse ecological and/or human health effects" [[8\]](#page-28-0). As is mentioned in the definition, ECs fall into two major categories, depending on their sources. Humans and animals naturally excrete hormones (androgens and estrogens), and these hormones are therefore classified as natural or endogeneous chemicals. The second category includes a wide range of chemicals manufactured by humans that comprise pharmaceuticals, household products, personal care products, industrial products, and livestock implants and are classified as synthetic or exogenous chemicals. This section provides an overview of the various types of ECs and their sources.

2.1 Natural Compounds

All humans and animals naturally excrete steroid hormones from their bodies. These excretions are introduced into the environment through wastewater treatment plant (WWTP) effluent, combined sewer overflow events, and the land application of biosolids and animal manure. Lange et al. [\[9](#page-28-0)] suggested that humans and livestock excrete hormones at a rate that is on the same order of magnitude. However, human wastes are generally treated, albeit at varying efficiencies, whereas livestock wastes do not typically undergo treatment prior to their introduction to the environment. As a result, it has been estimated that the land application of animal wastes has the potential to introduce 200 times more estrogens to the environment than the land application of biosolids [[7\]](#page-28-0). However, without estimates of hormones discharged to surface water bodies during the release of raw sewage via combined sewer overflow events, the relative contributions of humans versus livestock remain unclear. The estrogens of natural origin, which are of concern in drinking water supplies, are 17β - and 17α -estradiol (E2) and their metabolites estrone (E1) and estriol (E3). Natural androgens include testosterone (TST) and its environmental metabolite, androstenedione (AND).

Several other ECs have natural origins. However, their presence in the environment at elevated levels is directly related to human activities. For example, caffeine and nicotine originate from plants, but consumption of coffee and cigarette smoking have increased the presence of these compounds in surface water bodies. The effects of these increased concentrations in water bodies are not well understood.

2.2 Synthetic Compounds

A vast array of chemicals are contained in products that people use every day as medical treatments and household conveniences. These products benefit industry, agriculture, people, and animals; however, they often contain bioactive chemicals that affect living tissue and the environment. Synthetic compounds include chemicals used in PPCPs. Pharmaceuticals include both prescription and overthe-counter (OTC) drugs, and personal care products span a wide range of products used for personal hygiene.

The use of prescription medications and OTC drugs is a major source of ECs in the environment. People take medications for many health-related issues, including physical and mental health. Not all of the medicine is absorbed by the patient's body, and therefore active ingredients and their metabolites are excreted and enter the wastewater stream. The types of pharmaceuticals that have been found in drinking water sources are summarized in Table [1](#page-4-0). Additionally, the synthetic hormone 17α-ethinylestradiol (EE2), which is used in birth control pills, is also commonly detected in wastewater and surface water bodies. This is of particular concern, as it is more potent than natural estrogens and therefore has the potential to cause greater adverse impacts on humans and aquatic organisms.

The livestock industry is also a major source of ECs in the environment. Approximately 80 % of antibiotics produced in the United States are given to animals raised to produce food $[10]$ $[10]$. In 2009, more than 13 million kg of antibiotics were sold for farm animal use [\[11](#page-28-0)]. Antibiotics given to animals are listed in Table [2.](#page-4-0) Because the majority of livestock are raised in concentrated animal feeding operations (CAFOs), there is a high potential for diseases to spread quickly in these facilities. Therefore, animals are given regular doses of antibiotics to suppress the spread of illness. Additionally, dairy cattle generally receive rBGH (recombinant bovine growth hormone), which is a synthetic hormone that increases milk production, and beef cattle are commonly given implants that contain synthetic androgens to increase their growth, allowing slaughter at a younger age. Although these compounds enable CAFOs to be more efficient and are not generally thought to have negative consequences for human consumption of milk and beef, these

General type	Specific examples
Prescription	
Antacid	Cimetidine, ranitidine
Antiasthmatic	Salbutamol
Anticoagulant	Warfarin
Anticonvulsant	Carbamazepine
Antidepressant	Fluoxetine, paroxetine
Antihypertensive	Diltiazem, enalaprilat
Beta-blocker	Atenolol
Contraceptive	17α -Ethinylestradiol, mestranol
Over the counter	
Analgesic	Acetaminophen, ibuprofen, codeine
Antihistamine	Diphenhydramine
<i>Antibiotics</i>	
Macrolides	Azithromycin, erythromycin, roxithromycin
Quinolines	Ciprofloxacin, lomefloxacin, norfloxacin, ofloxacin
Sulfonamides	Sulfadiazine, sulfamethoxazole, sulfathiazole
Tetracyclines	Doxycycline, oxytetracycline, tetracycline
Other human antibiotics	Chloramphenicol, lincomycin, trimethoprim

Table 1 Emerging contaminants originating from human medications

Table 2 Emerging contaminants originating from veterinary antibiotics

General type	Specific examples
Macrolides	Tylosin, virginiamycin
Ouinolines	Enrofloxacin, sarafloxacin
Sulfonamides	Sulfachloropyridazine, sulfadimethoxine, sulfamethazine
Tetracyclines	Chlorotetracycline
Other veterinary antibiotics	Ormetoprim

compounds and their metabolites are excreted by animals and enter the environment during land application of their wastes.

Many synthetic compounds in industrial and personal care products have been found in the environment. A list of general types of ECs from industrial and household products is given in Table [3.](#page-5-0) Many of these compounds are known or suspected endocrine disrupters. Musk is a common base in fragrances and is known to bind to hormone receptor cites. Although significant research has been conducted on the impacts of musk, more than a dozen other fragrance compounds have the potential to cause endocrine disruption [\[12](#page-28-0)]. Triclosan, a common antimicrobial used in many household products, is known to affect hind limb development in tadpoles at concentrations as low as $0.15 \mu g/L$ [[13\]](#page-28-0).

While research on the impacts of some individual ECs on aquatic life is ongoing, these compounds are rarely, if ever, found in surface water bodies in the absence of other contaminants. Many ECs are known to behave synergistically, triggering endocrine disruption at concentrations lower than would be expected based on the

General	
purpose	Specific examples
Antioxidant	3-tert-Butyl-4-hydroxyanisole, 5-methyl-1H-benzotriazole
Cosmetics	Triethyl citrate
Detergent	Nonylphenol
Disinfectant	Triclosan
Flame retardant	Tri(dichloroisopropyl) phosphate, tributyl phosphate, tri(2-butoxyethyl) phosphate
Flavor	Camphor, menthol
Fragrance	Musk, isoborneol
Herbicide	Atrazine, bromacil, prometon
Insecticide	Carbaryl, carbazole, chloropyrifos, diazinon, N,N-diethyl-meta-toluamide (DEET)
Pesticide	Metalaxyl, metolachlor
Plasticizer	Diethyl phthalate, bis(2-ethylhexyl) phthalate, bisphenol A (BPA), triphenyl phosphate
Preservative	Anthracene, para-cresol
Sunscreen	Avobenzone, dioxybenzone, oxybenzone, sulisobenzone

Table 3 Emerging contaminants originating from industrial and household products

concentrations of each individual compound. Therefore, the synergistic behavior exhibited by "contaminant cocktails" needs to be better understood to assess risks that these contaminants pose to aquatic life and humans.

2.3 Emerging Pathogens

In addition to emerging chemicals of concern, new microorganisms are being discovered that are considered "emerging pathogens." Little is known about these emerging pathogens, and in some cases, microorganisms that had not previously been considered pathogenic are now being recognized as pathogens [\[14](#page-29-0)]. In 2005, the EPA listed several bacteria (Aeromonas hydrophilia, Helicobacter pylori, Mycobacterium avium intracellulare), viruses (Caliciviruses, Adenoviruses, Coxsackieviruses, Echoviruses), protozoa (Microsporidia), and cyanobacteria (blue-green algae) on its Contaminant Candidate List 2 [[15\]](#page-29-0). This list was created as a way to identify chemicals and pathogens that pose a threat to human health and may require regulation in the future. Egli and Rust [[16\]](#page-29-0) list several other bacteria and viruses as pathogens of emerging concern in drinking water.

The major sources of these pathogens include livestock, stormwater, and human recreational activities; however, some sources may also include wildlife and aquatic species [\[17](#page-29-0)]. In general, little is known about the transmission routes of these pathogens, their minimum infective dose, or their virulence [[14\]](#page-29-0). Additionally, little is known about the effectiveness of disinfection on inactivation of these emerging pathogens. There is potential for regrowth during distribution from the drinking water treatment plant to consumers. Therefore, even if the levels of these pathogens leaving the treatment plant could be considered safe, levels in the tap water coming out of the faucet may be too high. Many drinking water treatment plants add a secondary disinfectant to the finished drinking water to help prevent additional growth in water distribution systems. However, without data regarding the minimum infective dose and the effectiveness of these disinfectants on emerging pathogens, safe levels and adequate primary and secondary disinfectant doses cannot be established.

2.4 Regulations

One of the main issues surrounding ECs compared to other "traditional" contaminants is the lack of regulations. Various agencies, such as the Chemical Material Risk Management Directorate, the State of Massachusetts, and the State of South Carolina, have even included the lack of regulations as part of the definition of ECs [\[18](#page-29-0)]. Although their widespread presence in the environment has only recently been identified, it is likely that these contaminants have been in the environment for as long as they have been manufactured.

Currently, the data on the fate and transport of ECs in the environment and their risk to human and aquatic ecosystem health are insufficient for regulations to be developed. The Toxic Substances Control Act (TSCA) in the United States provides the EPA with the power to regulate new chemicals before they enter the market and existing chemicals, when they are found to pose a significant risk to human or environmental health. While the TSCA applies to any stage within a chemical's life cycle, the lack of life cycle data for ECs would make regulation under the TSCA difficult. Additionally, when it was passed in 1976, it exempted approximately 62,000 chemicals that were currently in use [\[4](#page-28-0)]. While testing is currently being conducted on approximately 500 chemicals [[19\]](#page-29-0), this is a small fraction of the 80,000 chemicals currently produced, of which 8,000 are known to be carcinogenic [\[3](#page-28-0)]. The chemicals that are currently regulated include lead, asbestos, chlorofluorocarbons (CFCs), and polychlorinated biphenyls (PCBs).

In the United States, the government (i.e., the EPA) is responsible for the assessment of chemical risks. However, in the European Union, this responsibility falls on industry through the REACH (Registration, Evaluation, Authorisation, and Restriction of Chemicals) regulation. REACH is currently being phased in, but will take full effect in 2018. Under REACH, a chemical cannot be manufactured or imported into the EU unless it has been registered and passed REACH's regulations. As a result of tighter regulations, some compounds that are used in the United States are banned in Europe. One such example is the use of growth promoting antibiotics for livestock, which were banned by Sweden in 1986, Denmark in 1995,

and the European Union (EU) in the 1999, but are widely used in the United States in CAFOs [[20\]](#page-29-0).

While some regulations do exist on the production side of the issues surrounding ECs, no drinking water regulations currently exist. In the United States, the EPA is predominantly focused on addressing PPCPs and has set a four-pronged strategy for addressing them: (1) improve science; (2) improve public understanding; (3) identify partnership and stewardship opportunities; and (4) take regulatory action when appropriate [\[21](#page-29-0)]. The EPA has launched educational campaigns to help consumers learn how to properly dispose of unwanted medications in order to reduce their threat to the environment. Additionally, the EPA has conducted various studies to assess the impacts of ECs on fish and the water quality implications of ECs used in livestock and poultry production. However, much research is still needed before regulations can be made. In preparation for future regulations, the EPA has created Contaminated Candidate List 3, which includes 104 chemicals and 12 microbiological contaminants that have been identified as occurring in public water systems and that pose a potential threat to public health [[22\]](#page-29-0). The EU's Priority Substances Directive limits the concentrations of priority substances in ground and surface water bodies under the Water Framework Directive (WFD) in order to protect ecological health and drinking water sources. In 2011, the development of a watch list was proposed that would target the monitoring of ECs as part of the WFD's ongoing monitoring activities across the EU [[23\]](#page-29-0). In 2013, 15 ECs were added to the watch list, including two estrogenic compounds and a painkiller [[24\]](#page-29-0). After more research has been conducted, contaminants from the watch list may be placed on the priority substances list.

3 Pathways to Drinking Water Sources

ECs enter the environment from various pathways. The major sources include runoff and leaching from agricultural fields and effluent from wastewater treatment plants (domestic, hospital, and industrial). These sources have been studied at a variety of scales and locations nationally and internationally. ECs have also been detected in water that does not appear to be impacted by wastewater effluent or agricultural runoff. Other sources of ECs in the environment include leaching from landfills and septic tanks and the discharge of raw sewage into rivers and streams during combined sewer overflow (CSO) events. This section provides an overview of these pathways that ECs take to reach surface and groundwater.

3.1 Pathways to Surface Water

Contaminants in surface water originate from municipal wastewater, industrial wastewater, combined sewer overflow, hospital wastewater, and land application of human and animal wastes.

3.1.1 Municipal Wastewater

Products that we use in our everyday lives become part of the wastewater stream after use. We excrete medications that we take, as our bodies do not absorb all of the active ingredients. Active ingredients in products that we use for personal hygiene, such as shampoo, soap, deodorant, and cosmetics, are washed down the drain and become part of the wastewater stream.

Municipal WWTPs were not designed to remove ECs. Many plants in the United States were built in the mid-1900s, long before many of the contaminants classified as ECs even existed. Additionally, the EPA does not currently regulate the concentrations of these chemicals in wastewater effluent. Therefore, any removal during the wastewater treatment process is coincidental rather than intentional. The size, age, type of treatment processes, and operation of the WWTP all influence the removal rate of ECs.

Effluent from WWTPs is generally discharged to rivers and streams. Water from these surface water bodies may be the source water for drinking water treatment plants downstream. Figure [1](#page-9-0) illustrates how wastewater generated from one household may become drinking water for someone living downstream. Because wastewater and drinking water treatment plants have varying removal efficiencies, some studies have found ECs in finished drinking water. The removal efficiencies of treatment technologies are discussed in Sect. [4](#page-16-0).

3.1.2 Industrial Wastewater

Wastewater generated during the manufacturing of industrial and household products is a source of ECs to the environment. Often, wastewater generated by these manufacturing plants is treated prior to being sent to municipal wastewater treatment plants. However, ECs are not regulated, and therefore the discharge permits for these manufacturing plants do not include ECs. The USGS conducted a national study to assess the contribution of manufacturing plants to the release of pharmaceuticals to the environment. This study found that municipal WWTPs that received a significant amount of wastewater from pharmaceutical manufacturing facilities ($>$ 20 %) had effluent EC concentrations 10–1,000 times higher than municipal WWTPs that did not receive this type of wastewater [\[25](#page-29-0)].

Fig. 1 Schematic of pathway emerging contaminants taken from use at one person's home to consumption at another

3.1.3 Combined Sewer Overflow Events

In the early 1900s, many cities in the United States installed combined sewer systems (CSSs) to collect both storm water and wastewater. During rain events, the combined volume of storm water runoff and wastewater can exceed the capacity of the WWTP. To prevent backup of sewage in residential areas, the mixture of storm water and wastewater is released to receiving waters (typically rivers or streams) untreated. Such an event is called a combined sewer overflow (CSO) event. More than 700 cities in the United States, primarily those located on the east coast, near the Great Lakes, and in the Pacific Northwest, have CSSs. CSSs are also present in Europe and other parts of the world.

Various studies have been conducted to assess the contribution of CSOs to ECs in receiving water bodies. Buerge et al. [[26\]](#page-29-0) used caffeine as an anthropogenic marker and conducted a mass balance to estimate the contribution of CSOs to streams in Switzerland. Caffeine loads exported by the streams were normalized on a per capita basis and found to be up to 10 times higher during CSO events than during normal flow conditions, suggesting that CSO events were responsible for a large contribution of ECs to these receiving streams. In the United States, Boyd et al. [[27\]](#page-29-0) found ibuprofen and triclosan concentrations in New Orleans stormwater canals receiving water from both sanitary and storm sewers. The study attributed the increased EC concentrations to the discharge of untreated sewage following rainfall events of 7 cm or greater.

In the Lake Champlain Basin, Phillips and Chalmers [[28\]](#page-29-0) initiated a study of the occurrence of organic wastewater compounds (OWCs), which are classified as ECs. They found that CSO events and urban storm runoff contributed to the presence of OWCs in Lake Champlain. Specifically, the OWCs that were effectively removed during wastewater treatment were found in CSO effluent at concentrations that were similar to or greater than in WWTP effluent. Caffeine, a flame retardant, and cholesterol had higher mass loadings in the CSO effluent than in the WWTP effluent. The results of this study emphasized the importance of identifying and treating waters that bypass normal wastewater treatment processes as part of the efforts invested in decreasing the amounts of OWCs entering receiving waters.

3.1.4 Hospital Wastewater

The wastewater generated by hospitals contains a wide variety of pharmaceuticals, disinfectants, and other compounds used for medical purposes that are classified as ECs. The diagnostic, research, and laboratory activities contribute to the presence of these compounds in wastewater, along with the excretion of pharmaceuticals and their metabolites by patients. Despite the differences in the levels of contaminants in domestic and hospital wastewater, hospital wastewater is generally sent to municipal WWTPs, often without any pretreatment. Sometimes hospital wastewater is disinfected using chlorine, but when pretreatment is used, it is generally for wastewater generated by the infectious disease ward of the hospital and not necessarily for all of the wastewater generated by the entire hospital [[29\]](#page-29-0). Therefore, hospital wastewater has the potential to be an important source of ECs to municipal WWTPs.

Verlicchi et al. [[30\]](#page-29-0) compared the quality of urban and hospital wastewater using data collected and published in dozens of studies. Standard parameters used to evaluate wastewater quality include biochemical oxygen demand $(BOD₅)$, chemical oxygen demand (COD), and suspended sediment concentrations. Verlicchi et al. [[30\]](#page-29-0) found that each of these values was 2–3 times higher for hospital than urban wastewater. The average concentrations of ECs in hospital wastewater ranged from 1 to 150 times higher than EC concentrations in urban wastewater. Hormones and beta-blockers were on the same order of magnitude in both wastewater types. Analgesics, antibiotics, and cytostatics were present in hospital wastewater at concentrations up to 10–15 times greater than in urban wastewater. Some heavy metals (gadolinium and platinum) were 55–90 times higher in hospital wastewater. Iodine-based contrast media (ICM), used in radiology, was up to 150 times higher in hospital wastewater compared to urban wastewater. These elevated levels of many ECs in municipal WWTP effluent suggest that hospital wastewater can be a significant source of ECs to the environment, as the municipal WWTPs are not able to significantly reduce the elevated levels of ECs coming from hospital wastewater sources.

3.1.5 Land Application of Human and Animal Wastes

Land application of treated sewage sludge, commonly referred to as biosolids, offers the benefits of improving soil structure and fertility, given their richness in nutrients and organic matter [\[31](#page-29-0)]. In 2004, more than 6 million metric tons of biosolids were generated in the United States, and more than 50 % were land applied to agricultural fields $[32]$ $[32]$. Similarly, animal waste generated by animal feeding operations (AFOs) is often applied to agricultural fields as a nutrient source. Each year, over 50 million metric tons of manure is produced in the United States, with the majority being land applied. These wastes are known to be a source of ECs to the environment. Biosolids from municipal WWTPs contain natural hormones and a wide array of PPCPs. Manure contains natural hormones and veterinary pharmaceuticals.

While the land application of biosolids and manure is an excellent mechanism for managing waste, the contaminants that they contain can reach surface water via surface runoff. Various studies have reported ECs in surface runoff from fields following application, with some evidence of elevated concentrations in surface runoff for extended periods of time (i.e., several months) after application [[33–](#page-29-0)[37\]](#page-30-0).

The presence of tile drains in agricultural fields also contributes to the presence of ECs in surface water bodies. Tile drains are a network of perforated pipes installed \sim 1 m below the soil surface to keep the water table below corn roots. They are generally installed in poorly drained soils in which corn is grown in order to provide the roots with the aerobic conditions they need to achieve desired yields. Although tile drains improve the efficiency of corn production, the reduction in the water holding capacity of the fields results in an increased amount of water discharged into nearby ditches and streams. Studies have shown that tile drains contribute to the presence of ECs in surface water bodies [[36–38\]](#page-30-0).

3.2 Pathways to Groundwater

Contaminants in groundwater can originate from rapid flow pathways, landfill leaching, and septic tanks.

3.2.1 Rapid Flow Pathways

The extent to which soil can act as an effective biogeochemical filter is directly related to the amount of time that a solute spends within the soil profile before reaching groundwater. This residence time is a function of physical soil properties (e.g., depth to groundwater, porosity), hydroclimatic properties (e.g., rainfall frequency, intensity, and depth), and solute properties (e.g., sorption coefficient, degradation rate constant). The ratio of the residence time to the rate of solute degradation is a useful indicator of the extent to which the soil can effectively filter the soil prior to release to groundwater. The more effectively the soil acts as a filter, the lower the solute delivery ratio, which is calculated as the mass of solute that reaches the groundwater as a fraction of the mass applied to the soil surface. If the amount of time the solute resides in the soil is long compared to the rate at which it degrades, then the solute delivery ratio will be small. Conversely, the amount of solute mass that reaches groundwater is larger when the residence time is short compared to the degradation rate.

Soil structure plays an important role in controlling solute residence time and delivery ratio. Soils without preferential flow pathways are more effective biogeochemical filters because the residence time is longer compared to soils with extensive preferential and macropore flow pathways. These pathways enable solute transport to be short-circuited through the soil profile to groundwater and provide little opportunity for the solute to interact with the soil matrix, limiting sorption. Therefore, the activation of these flow pathways can cause significant solute transport. In cases when groundwater is shallow, solute residence time in these pathways can be on the order of hours. Without significant loss to the soil matrix via sorption and without significant loss via degradation due to the short travel times, the delivery ratio through these pathways can approach 1. The overall delivery ratio of the solute through the soil profile can be calculated as the sum of the delivery ratio through each pathway. Because the matrix delivery ratio is likely to be small for most ECs, the overall delivery ratio is limited by the fraction of water that is transported through these preferential flow pathways.

The results of some field-scale studies observed rapid transport of hormones to tile drains following manure applications to agricultural fields [\[36](#page-30-0), [38](#page-30-0)], suggesting the importance of rapid flow pathways to EC transport. Although some studies have found ECs in groundwater, the presence of ECs in surface water bodies is much more widespread. Reif et al. [[39\]](#page-30-0) analyzed well water samples collected from six sites in Pennsylvania for 44 ECs, and only one compound (cotinine, a metabolite of nicotine) was detected. However, as many as 51 different compounds were detected in samples collected from streams across the state.

3.2.2 Landfill Leaching

The vast majority of the products we use eventually make their way to a landfill. Additionally, much of the biosolids that are not land applied are disposed of in landfills. Although landfills are lined to minimize leaching of contaminants to groundwater, leaching still occurs. Landfill leachate has the potential to be another source of ECs to the environment [[40,](#page-30-0) [41\]](#page-30-0).

Several studies have identified ECs in landfill leachate. Andrews et al. [\[42](#page-30-0)] sampled leachate from three landfill cells containing wastes of various ages: >25 years old, 3–16 years old, and an operating cell with wastes less than 5 years old. ECs were detected in leachate from each cell, with concentrations ranging from 0.11 to 114 μg/L. The ECs detected included sterols of human and plant origin, PPCPs, industrial compounds, hydrocarbons, and pesticides. The type of ECs found in leachate from each cell varied, with four ECs detected exclusively in leachate from the oldest cell, two exclusively in leachate from the intermediate cell, and none exclusively in the leachate from the operational cell. These results suggest that the age of the waste contained in a landfill may influence the types of ECs leaching. Eggen et al. [[43\]](#page-30-0) collected leachate samples from three landfills in Europe. Leachate samples contained flame retardants, plasticizers, insecticides, and PPCPs (ibuprofen, naproxen, and musk compounds) at concentrations on the ng and μg/L level. ECs were detected in both the aqueous and particulate phases of the leachate and were found to pose a threat to groundwater quality. Treatment technologies, which are commonly applied for landfill leachates, and based on short-term biological degradation, aeration, and sedimentation processes, may not be effective for removal of ECs. It may be necessary to apply other treatment processes such as membrane filtration or reverse osmosis in order to effectively reduce leachate concentrations [\[43](#page-30-0)].

3.2.3 Septic Tanks

Approximately 25 % of the US population has on-site septic tanks to treat their wastewater [\[84](#page-32-0)]. Discharge from septic tanks is released to groundwater, which is potentially problematic, as people with septic tanks generally get their drinking water from groundwater. Despite the potential importance of septic systems as a source of EC contamination in groundwater, Schaider et al. [\[44](#page-30-0)] conducted a literature review and found that less than 20 studies have been conducted to investigate the ability of septic systems to remove ECs from wastewater influent. Of the studies that were conducted, removal is low within the septic tank itself due to anaerobic conditions that slow degradation processes [\[46](#page-30-0), [47\]](#page-30-0). However, EC removal in leach fields is high due to sorption and aerobic conditions that promote faster degradation [\[45](#page-30-0), [48](#page-30-0)]. The ECs found in septic tank leachate included antibiotics, prescription medications, over-the-counter medications, hormones, plasticizers, compounds from personal care products, flame retardants, and detergent metabolites [[44\]](#page-30-0). The maximum concentrations of ECs detected in septic tank effluent were five orders of magnitude higher than those in leach field effluent; however, the median concentrations detected in leach field effluent were similar to those in municipal wastewater treatment effluent [[44\]](#page-30-0). In addition to potentially contaminating groundwater, surface water may also be impacted by septic systems. Standley et al. [[49\]](#page-30-0) reported hormone concentrations in surface water whose headwaters were aquifers that were negatively impacted by septic systems.

3.3 Spatial and Temporal Inequality

This section describes the concept of inequality as it applies to transport of contaminants in natural systems.

3.3.1 Quantifying Inequality

In order to effectively reduce the export of ECs to water bodies, "hot spots" within watersheds and the periods of time during which they are most active (i.e., "hot events") must first be identified. The heterogeneity and mixed land use typical of many watersheds generally cause small portions of the watershed to export disproportionately large solute loads to receiving water bodies. For example, in a watershed consisting of 20 % urban area and 80 % forest, hormone loads generated by the urban area are likely to contribute nearly 100% of the loads exported by the entire watershed (neglecting endogenous hormones excreted by wildlife), despite comprising a much smaller fraction of the watershed's area. Additionally, if the urban area contains combined sewer systems, the highest hormone fluxes are likely to occur during large rainfall events that cause the greatest amount of raw sewage to be discharged to nearby surface water bodies during combined sewer overflow events. Despite occurring perhaps 20 % of the time over the course of a year, these events might generate 80 % of the watershed's annual hormone load. This example illustrates the Pareto Principle, which is also known as the 80-20 rule. In general, it means that a small portion of a population generates a disproportionately large percentage of an outcome. It is commonly used in economics to describe the inequality of distribution of money among people at various scales, but also has common applications in the natural world.

3.3.2 Importance of Temporal Inequality

While the quantification of temporal inequality in water quality and quantity data is new [\[50](#page-30-0)], prior studies demonstrated a general understanding of the disproportionate contribution of high-flow events to annual discharge and loads. One such study conducted by Royer et al. [[51\]](#page-30-0) collected long-term (8–12 years) discharge and nutrient data at three tile-drained watersheds in Illinois ranging from 101 to 481 km^2 . The results indicated that the export dynamics are highly seasonal, with the majority of export occurring in the late winter and spring months. These months generally experience large rainfall events resulting in periods of high flow. Discharges classified as extreme (i.e., \geq 90th percentile) were responsible for more than 80 % of the dissolved reactive phosphorus loads and more than 50 % of the nitrate loads. Additionally, Richards et al. [\[52](#page-31-0)] found that over a 20-year period (1975– 1995), more than 70 % of total phosphorus, soluble reactive phosphorus, nitrate, and suspended solids were exported from four predominantly agricultural

catchments in the Lake Erie Basin $(88-16,400 \text{ km}^2)$ during storm runoff periods that accounted for \sim 1/3 of the time.

To conduct this kind of analysis, water quality data must be collected during periods of high flow. Grab samples collected at periods of time such as monthly are not likely to enable such an analysis, unless data were collected for a sufficiently long period of time to capture the full range of discharge. While such data sets are not uncommon for nutrients and other traditional contaminants, few sufficient data sets exist for ECs, and therefore our understanding of the temporal dynamics of EC export is weak. The data that do exist, however, suggest that EC loads exhibit high temporal inequality. Most field studies have found positive relationships between concentration and discharge. These chemograph dynamics cause the majority of export to occur during large storm events, as high concentrations are generally associated with periods of high flow. Gall et al. [[37\]](#page-30-0) calculated hormone loads exported during a 17-month study period at a tile-drained agricultural site in Indiana. The results of the study indicated high temporal inequality for hormone export, with 80 % of hormone loads exported during high flow occurring during 9– 26 % of the study period. Similar to conclusions made by Royer et al. [\[51](#page-30-0)] and Richards et al. [[52\]](#page-31-0), the majority of the hormone export occurred during late winter and early spring during large rainfall events.

3.3.3 Importance of Spatial Inequality

Similar to temporal inequality, spatial inequality implies that portions of a watershed contribute disproportionately large contaminant loads to a receiving water body. This is generally done by calculating annual loads exported from sub-watersheds on a per area basis (i.e., $kg/km²$) and identifying locations that have the highest values. Such studies have been conducted to identify nutrient "hot spots" or "critical zones" in the Chesapeake Bay Watershed and Mississippi River Basin in order to better understand the sources that are contributing to hypoxia in these water bodies. This type of analysis has led to the identification of four critical nutrient hot spots in the Chesapeake Bay Watershed that coincide with animal production operations [[53\]](#page-31-0).

Due to the widespread presence of ECs in streams and rivers, such a spatial analysis for these compounds would prove useful in locating EC hot spots. The land use at these locations could then be investigated to identify the sources causing the highest loads within a particular watershed. Such an analysis would help us to better understand the relative contributions of the various pathways discussed in Sect. [3.1](#page-8-0) to ECs in surface water bodies. Appropriate management plans to reduce loads could then be focused on these hot spots.

3.3.4 Implications for Best Management Practices

Due to local, regional, and national concerns regarding erosion, eutrophication, and coastal hypoxia, widespread efforts have been made to encourage the adoption of best management practices (BMPs). In general, BMPs is a term that refers to methods of controlling and reducing the transport of contaminants to receiving ground and surface water bodies. Urban BMPs aim to reduce the amount of stormwater runoff generated during rainfall events in an effort to mitigate the increase in runoff volume associated with an increase in impervious area and to improve water quality. Common examples of urban BMPs include green roofs, rain gardens, rain barrels, and porous pavement. Agricultural BMPs generally focus on reducing the transport of sediment, nutrients, and pesticides. BMPs include source management, such as the timing and amount of nutrient and pesticide applications; on-field practices, such as conservation tillage and cover crops; and practices that help to mitigate contaminants in stormwater runoff, such as constructed wetlands and conservation buffers.

Additional efforts have been made to understand the barriers to adoption of BMPs and to evaluate the effectiveness of various BMPs. However, many paired watershed studies have shown that the implementation of BMPs has little to no reductions in nutrient concentrations and loads [[54,](#page-31-0) [55\]](#page-31-0). The large uncertainty regarding BMP effectiveness and long lag times between implementation and water quality improvement are problematic for promoting BMP adoption [\[56](#page-31-0)]. Additionally, the types of BMPs generally implemented by land owners are most effective at reducing loads during periods of low flow, rendering their overall benefits minimal to water quality improvements at larger temporal and spatial scales.

The consistent spatial and temporal patterns suggest that BMPs must effectively reduce loads during high-flow events from high load-generating areas in order to significantly improve downstream water quality. Properly identifying these "windows of opportunities" and "hot spots" is essential for targeting BMPs to mitigate loads in the most effective manner. These trends suggest that reducing loads during only a few high-flow events from a few locations (i.e., "hot spots") can have a disproportionately large benefit to water quality. Therefore, properly assessing and quantifying the temporal and spatial inequality of ECs will facilitate the optimal design and implementation of BMPs. This approach has the potential to ensure that investments made in BMPs will translate to water quality benefits.

4 Removal in Treatment Plants

The growing awareness of ECs in drinking water sources has led to the need to understand the removal of these compounds during wastewater treatment processes. Since these compounds generally do not have surface or drinking water standards, there is no regulatory requirement for their removal during wastewater and drinking water treatment. Therefore, any removal is coincidental rather than intentional. Treatment plants have a wide range of removal efficiencies, depending in part on the types of treatment technologies employed at the specific plant. The EPA synthesized the results of more than 80 studies into a report, which provides an assessment of the ranges of removal efficiencies for 16 selected ECs (bisphenol-A, caffeine, carbamazepine, DEET, diclofenac, estradiol, estrone, galaxolide, gemfibrozil, ibuprofen, iopromide, naproxen, nonylphenol, sulfamethoxazole, tri (chlorethyl) phosphate, and triclosan [[57\]](#page-31-0). This section provides an overview of the current capabilities and efficiencies of commonly used treatment technologies to reduce the concentrations of ECs and brings attention to areas of innovative technologies that have the potential to improve removal efficiencies.

4.1 Treatment Technology Removal Efficiencies

Recent studies have been conducted to evaluate the removal efficiencies of various municipal wastewater and drinking water treatment processes: activated sludge, granular activated carbon, reverse osmosis, and disinfection. These treatment technologies and their removal efficiencies are discussed below.

4.1.1 Activated Sludge Treatment Process

The activated sludge wastewater treatment process is the most commonly used secondary treatment in the United States. It consists of two stages, as shown in Fig. [2.](#page-18-0) The first stage consists of an aerated tank in which microbial growth is promoted to remove organic matter. The second stage is a clarifier, which is a settling tank designed to remove solids via gravitational settling. The solids are referred to as activated sludge. A portion of the activated sludge is returned to the aeration tank, and the rest becomes part of the treatment plant's waste stream.

This treatment process has a wide range of removal efficiencies for ECs, with an average removal of 22 % for carbamazepine and 94 % for caffeine. The overall average removal efficiency for all 16 ECs included in the EPA's report in the activated sludge treatment process is approximately 70 $%$ [\[57](#page-31-0)]. These removal efficiencies refer to the removal of ECs from the treated water. The primary removal mechanisms are biodegradation in the aeration tank and sorption to the solid materials that enter the waste stream. Therefore, the half-life and partition coefficient play important roles in the amount of ECs removed from the water during this treatment process. Because a portion of the activated sludge stream is returned to the aeration tank, the ECs contained in this activated sludge reenter the tank. Additionally, the ECs in the activated sludge that becomes part of the waste stream may be introduced into the environment if these biosolids are land applied.

Fig. 2 Removal of emerging contaminants, represented as pills, in the activated sludge treatment process by biodegradation (left) and settling of particulate matter to which they are sorbed (right)

4.1.2 Granular Activated Carbon

Granular activated carbon (GAC) is a commonly used media in granular filtration. It is widely used in household filtration systems to remove constituents in finished drinking water that cause an undesirable taste. At the larger scale, GAC is used in fixed bed columns (see Fig. [3](#page-19-0)) in the drinking water treatment processes, for further treating treated wastewater for water reuse, and can be used for tertiary wastewater treatment. GAC is an excellent sorbent, as it has a very high surface area. It performs best when the inflowing water is relatively clean, and is generally used to remove soluble organic compounds and inorganic compounds.

Many ECs are amenable to removal via adsorption onto GAC. The average removal efficiency for drinking water treatment ranges from 42 % for sulfamethaxazole to 79 % for gemfibrozil [[57\]](#page-31-0), with an overall average of $~60~\%$. For water reuse applications in which GAC is used to further treat the treated wastewater, the average removal efficiency of ECs ranges from 3.6 % for naproxen to 63 % for DEET [[57\]](#page-31-0), with an overall average EC removal efficiency of \sim 30 %. This lower average removal efficiency compared to drinking water treatment is likely due to the levels of EC concentrations present in the influent. As the wastewater effluent had already been treated, the 30 % average removal would be in addition to any removal that had already occurred during previous treatments. Therefore, the overall removal efficiency from untreated wastewater influent through treated effluent to water reuse standards would likely be significantly higher than 30 %, depending on the types of secondary and/or tertiary treatment used earlier in the treatment process.

Fig. 3 Removal of emerging contaminants, represented as pills, via adsorption to granular activated carbon (GAC) in a fixed bed column

4.1.3 Disinfection

Disinfection is primarily used to inactivate pathogens present in water and wastewater. In drinking water treatment, this process is needed to protect consumers from exposure to pathogens at doses that could cause adverse health effects. In wastewater treatment, disinfection protects receiving water bodies and keeps pathogen levels low enough to enable the water to be used for recreational purposes. In some states, disinfection is not required during winter months when human contact with surface water bodies for recreational activities (e.g., swimming and fishing) is not expected.

Fig. 4 Emerging contaminants, represented as pills, removed by three types of disinfection: chlorination, ozonation, and ultraviolet disinfection

There are three main types of disinfection: chlorine, ultraviolet (UV), and ozone (see Fig. 4). Overall average EC removal efficiencies for wastewater treatment were calculated based on the EPA's literature review [\[57](#page-31-0)] and were greatest for ozone $(88 \pm 12 \%)$ and lowest for chlorine $(65 \pm 27 \%)$. The average removal efficiency for treating drinking water was slightly higher for chlorine (39 \pm 16 %) compared to UV disinfection $(36 \pm 28 \%)$.

Disinfection is able to reduce ECs both directly and indirectly. Chlorine and ozone are strong oxidizers that can oxidize ECs, transforming them into other chemicals. UV disinfection cleaves bonds in ECs, transforming them into other compounds. UV disinfection can also indirectly reduce EC concentrations. The energy of UV light reacts with water, generating hydroxyl radicals. These radicals then react with ECs, causing them to transform into other compounds. The by-products of these reactions can sometimes be carcinogenic, as is often the case with chlorine disinfection. More research is needed on the products that form to assess whether the product compounds retain any endocrine disrupting properties. Although the disinfection by-products generated by ozone and UV are not as well understood as the by-products generated by chlorination, some disinfection by-products themselves are classified as ECs. For example, bromoform is a carcinogenic disinfection by-product classified by the USGS as an EC [\[39](#page-30-0)].

4.1.4 Reverse Osmosis

Reverse osmosis is a type of membrane filtration that uses pressure to drive contaminant-laden water across a membrane (see Fig. [5\)](#page-22-0). There are four types of pressure-driven membrane filtration, with the classification based on the size of the membrane pores: microfiltration (0.1–5 μm), ultrafiltration (0.01–0.1 μm), nanofiltration $(0.001-0.01 \text{ µm})$, and reverse osmosis $(0.0001-0.001 \text{ µm})$. Membranes can be selected to remove contaminants ranging from suspended particles (microfiltration) to dissolved compounds (nanofiltration and reverse osmosis). Therefore, reverse osmosis is necessary to remove ECs that are dissolved in the influent. Due to the very small pore size, reverse osmosis requires a significant amount of pressure to drive the influent across the membrane and is therefore an expensive treatment technology. However, the removal efficiencies for ECs are very high, with an overall average of 95 ± 6 % for water reuse applications for which treated wastewater is the influent [\[57](#page-31-0)].

4.1.5 Transformation Products

The removal efficiencies discussed in this section should not necessarily be interpreted as being equivalent to the removal of endocrine disrupting properties. The removal efficiencies were calculated based on the removal of a specific EC, but degradation metabolites and products of chemical reactions may possess endocrine disrupting or carcinogenic properties. Therefore, more research is needed to understand the products that are generated during treatment. For removal mechanisms that are physical rather than microbiological or chemical, the removal efficiency is likely equivalent to the removal of associated endocrine disrupting properties, as the parent compound has been removed via sorption, settling, or size exclusion (i.e., membrane filtration) and no products or metabolites were produced during these removal mechanisms.

4.2 Technology Innovations

In March of 2013, the EPA issued its first version of a "Blueprint for Integrating Technology Innovation into the National Water Program." The document emphasized the serious water-related challenges that our country faces, including ECs. The purpose of the blueprint is to promote technological innovations that can help the United States to meet today's demands for clean and safe water and to cope with the many challenges that will continue to face the US water resources. This section provides an overview of published work regarding lab- and pilot-scale studies of traditional treatment technologies and also presents an overview of state-of-the-art

emerging technologies that have received high-profile funding for their potential to treat emerging contaminants.

4.2.1 Lab- and Pilot-Scale Studies of Traditional Treatment Technologies

In addition to the full-scale treatment technologies discussed in the previous section, some pilot- and lab-scale studies have been conducted to test the removal efficiencies of various treatment technologies for ECs. Far fewer studies have been conducted for removal during drinking water treatment compared to wastewater treatment. One pilot-scale study on drinking water treatment using ozonation found a 99 % removal efficiency of clofibric acid and naproxen [[57,](#page-31-0) [58](#page-31-0)]. Two lab-scale studies assessed the removal efficiency of chlorine disinfection on caffeine, salicylic acid, trovalfoxacin, and estradiol and found average removal efficiencies ranging from 42 % to 60 % [[57,](#page-31-0) [59](#page-31-0)]. No data have been reported for the removal efficiencies of these compounds in full-scale drinking water systems.

Typically, removal efficiencies are higher in lab-scale studies than full-scale studies. However, data across scales are not currently comparable for many compounds of interest, and therefore studies that assess removal efficiencies of the same compounds for the same types of technologies across scales are needed. Then, lessons learned from bench-, lab-, and pilot-scale studies can be applied to improve removal efficiencies in full-scale treatment plants.

4.2.2 Pilot-Scale Subsurface Flow Constructed Wetlands

Constructed wetlands are sometimes used in wastewater treatment to remove nitrogen and phosphorus prior to discharge to receiving water bodies to prevent eutrophication. Some pilot-scale studies have been conducted to assess the ability

of subsurface flow constructed wetlands to remove ECs. Matamoros et al. [\[60](#page-31-0)] found that shallow beds performed better than deeper beds, likely due to more oxidized conditions. Additionally, they found the removal efficiencies in shallow beds were higher than removal in WWTPs in Germany and Brazil. Matamoros and Bayona [\[61](#page-31-0)] tested a pilot-scale subsurface flow constructed wetland's ability to remove PPCPs from residential wastewater in a 200-person urban housing development. They classified the compounds based on their removal efficiencies. Those classified as efficiently removed had a removal efficiency >80 % and included caffeine and salicylic acid. Moderately removed compounds had removal efficiencies of ~50 % and included ibuprofen and naproxen. Recalcitrant compounds included ketoprofen and diclofenac.

Although subsurface flow constructed wetlands may be appropriate for the removal of some ECs, others can actually exhibit greater endocrine disrupting properties after treatment. Some data suggest that under anaerobic conditions, the estrogen metabolite, E1, may convert back to its parent compound, 17β-E2 [\[62](#page-31-0)]. 17β-E2 is a more potent estrogenic compound than E1, and therefore the use of anaerobic treatment technologies has the potential to cause more harm than good with respect to the treatment of estrogenic compounds. Therefore, more research is needed to determine when different types of treatment technologies are appropriate. Results of this research will be critical to the development of recommendations for best management practices for treating wastewater.

4.2.3 Emerging Research for Emerging Contaminants

Current research on innovative technologies for treating emerging contaminants shows promise for improving the removal efficiency of ECs in wastewater. A brief overview of three areas of innovation is given in this section: photocatalysis, membrane bioreactors, and the Eco-MachineTM.

Some research has shown that ECs can undergo phototransformation in aquatic environments. This phototransformation is generally thought to play a minor role in the fate and transport of ECs [[63\]](#page-31-0). However, there is potential for this process to be exploited for water treatment purposes. Photocatalysis uses a catalyst to accelerate the rate at which photoreactions occur. Liang et al. [[64\]](#page-31-0) built titanium dioxide $(TiO₂)$ anatase phase nanobelts to photocatalyze the oxidation of pharmaceuticals in wastewater. Over a period of 90 min, naproxen, theophylline, and carbamezapine concentrations were reduced by more than 90 %, suggesting that even some of the most persistent ECs can be removed effectively with photocatalysis [[64\]](#page-31-0). Encinas et al. [\[65](#page-31-0)] found that the presence of other organic and inorganic compounds interferes with the effectiveness of photocatalysis. Therefore, it is likely that this technology would be best used as a tertiary treatment [\[65](#page-31-0), [66\]](#page-31-0).

Membrane bioreactors are an emerging technology in wastewater treatment. They are particularly attractive for water reuse applications, as they are able to produce high-quality effluent [\[67](#page-31-0)]. Trinh et al. [[67\]](#page-31-0) found that although the ability of membrane bioreactors to remove regulated organic contaminants was very high

($>90\%$), the removal efficiencies for pharmaceuticals were lower (24–68 %). Forrez et al. [[68\]](#page-31-0) applied biogenic metals (manganese oxides and palladium) in membrane bioreactors at the laboratory scale as oxidative and reductive agents. This increased the removal efficiency of membrane bioreactors for PPCPs, with efficiencies ranging from 52 % to 95 %. The removal efficiencies for naproxen, codeine, and ibuprofen were greater than 90 $\%$, while triclosan was \sim 70 $\%$, and sulfamethoxazole was $\sim 50\%$ [\[68](#page-31-0)].

The National Science Foundation (NSF) has currently invested in a project at The Pennsylvania State University to investigate the potential of fungi and bacteria to remove emerging contaminants in wastewater treatment plants [\[69](#page-31-0)]. Dr. Rachel Brennan's research group is currently conducting research at Penn State's Eco-MachineTM, a living, sustainable wastewater treatment plant. The goal of the research is to first identify the mechanisms used by fungi and bacteria to remove these contaminants and then develop ways to enhance these removal mechanisms. This research is expected to provide a significant cost savings over traditional wastewater treatment technologies and has the potential to establish a new paradigm for treating wastewater in a holistic manner.

5 Public Awareness

Over the past decade, the public has become increasingly aware of the presence of pharmaceuticals in drinking water sources. Numerous news articles have been published on this topic, and as the public's awareness increases, people are confronted with new decisions to make regarding their everyday habits. This section explores the issues surrounding ECs from a public perspective.

5.1 Is Ignorance Bliss?

As the general public becomes increasingly aware of the presence of ECs in their drinking water sources, are concerns that people have with respect to their personal health justified? Or is ignorance bliss? Although ECs are known to be harmful to aquatic organisms, these species are much more sensitive to the endocrine disrupting properties of many of these compounds than humans. Currently, there is no proof that consumption of ECs at such low levels is harmful to humans. The extremely low concentrations of ECs in drinking water suggest that people would need to consume a very large amount of water in order to receive the equivalent dose of one medication. Benotti et al. [[70\]](#page-31-0) collected data regarding the presence of ECs in drinking water for more than 28 million people in the United States. The most commonly detected pharmaceuticals were atenolol, carbamazepine, gemfibrozil, naproxen, and sulfamethoxazole. Based on the maximum concentrations detected in drinking water sources in this study, Table [4](#page-25-0) shows the volume of water

Compound (type of medication)	Maximum concentration in drinking water sources ^a (ng/L)	$Dose^b$ (mg)	Amount of water to consume one dose (L)
Atenolol (beta-blocker)	36	$50 - 100$	1,400,000-2,800,000
Carbamazepine (anticonvulsant)	51	$800 -$ 1,200	15,700,000-23,500,000
Gemfibrozil (lipid regulator)	24	600	25,000,000
Naproxen (anti- inflammatory)	32	$500-$ 1,500	15,600,000-46,900,000
Sulfamethoxazole (antibiotic)	110	800	7,300,000

Table 4 Volume of water needed to consume to receive one dose of various medications

^a Source: Benotti et al. [\[68\]](#page-31-0)
^b Source: http://www.rylist

Source: http://www.rxlist.com

that would need to be consumed to receive the equivalent of one dose. For reference, the EPA recommends consuming 2 L of water per day. Therefore, the risk associated with drinking water containing even the maximum concentration of pharmaceuticals detected appears very low. However, ECs are known to behave in poorly understood synergistic ways in fish. The presence of multiple compounds in water appears to have a multiplicative effect, with the adverse impacts higher than would be predicted based on known effects of the concentrations of each individual compound. Therefore, there are potential concerns regarding the risks associated with consuming water tainted by low levels of many different contaminants (i.e., "contaminant cocktails"). The synergistic effects of compounds present in contaminant cocktails must be better understood so that the risks associated with consumption of drinking water containing multiple ECs can be better calculated.

An increase in public awareness has led to an increase in proper disposal of medications. Many educational campaigns have been launched in the United States at local and national levels to discourage the flushing of unwanted medications down toilets and discarding them in the trash. Municipalities across the United States have set up unwanted medication drop boxes, in which people can bring medications that expired or are no longer needed. These drop boxes have been very successful, with some bringing in hundreds of pounds of medications over the course of a year. In the United Kingdom, called "Only Order What You Need" [\(http://www.medicinewaste.com\)](http://www.medicinewaste.com/) was launched to educate the public about issues surrounding the improper disposal of unwanted medications and to provide information on where unwanted medications can be brought. Any unused and expired medications can be returned to pharmacies for proper disposal.

5.2 Is Bottled Water Better?

As people become more concerned about the quality of the drinking water supplied to them from their municipality or private well, bottled water consumption over the past few decades has experienced significant growth [\[71](#page-32-0)]. Regulations set by the EU's Drinking Water Directive apply to all drinking water, including bottled water. However, the EPA drinking water standards for tap water do not apply to bottled water, and therefore the exposure to potentially harmful contaminants in bottled water is largely unknown and difficult to assess [[72\]](#page-32-0). Potential sources of organic pollutants in the bottled water may include the presence of pollutants in the water source, contaminant from the bottling plant, or the plastic containers themselves [\[73](#page-32-0), [74](#page-32-0)].

Devier et al. [[75\]](#page-32-0) conducted an analysis to test for the presence of ECs, including hormones and other endocrine disrupting compounds, in Evian[®] and Volvic[®] brands of bottled water. The study found no detectable levels of 120 organic compounds that were tested, but did detect pharmaceuticals, alkylphenols, and phthalates. The detected pharmaceuticals were ketoprofen, salicylic acid, and caffeine. However, the study also reported the presence of the same contaminants in laboratory procedural blanks, which are used as part of the quality assurance and quality control protocol. This suggested that the contamination was likely introduced by the laboratory during analysis rather than in the bottled water itself. The source of water for these two brands is groundwater. Therefore, the absence of ECs in these two bottled water brands confirmed the effectiveness of the natural geologic protection and the long-term protection policies implemented on their watersheds. The study also tested for contaminants that may potentially originate from the plastic bottle itself, such as polypropylene terephthalate (PET), and the results confirmed that no leaching of the targeted compounds occurred under the test conditions.

The presence of bisphenol A (BPA) in water bottles is another issue that has gained widespread public recognition and led to an increased development of BPA-free products. BPA is known to be estrogenic and to possess endocrine disrupting properties. Cooper et al. [[76\]](#page-32-0) conducted a study to assess BPA release from reusable water bottles known to contain BPA (i.e., made from polycarbonate plastics) and those that claimed to be BPA free. Water stored in polycarbonate bottles had BPA concentrations of 0.2–0.3 mg/L. Water stored in aluminum bottles with epoxy-based resin lining had BPA concentrations ranging from 0.08 to 1.9 mg/ L. Under extreme circumstances (e.g., boiling the water and then storing it in the reusable bottles), BPA leached at even higher levels. However, their results were encouraging in that they found that as long as the BPA-free bottles were used according to the manufacturer's instructions, products marketed as BPA free did not release BPA and effectively protected water from BPA contamination.

6 Conclusions

In general, the public sees any detectable levels of contaminants in their drinking water as undesirable, even if the risk associated with consumption of low levels of contaminants is extremely low. People accept much greater risks in their everyday lives, such as driving in a car, than the risks associated with drinking water that has been treated to EPA standards. However, since the risks associated with chronic consumption of low levels of unregulated ECs are not well understood, it is understandable that people would prefer ECs to be undetectable in their drinking water sources. As technology continues to improve, the detection limits for compounds decrease, making it possible to detect contaminants at increasingly lower concentrations. The cost associated with treating water to a level at which ECs would be present at concentrations below instrument limits of detection would be many times higher than the rates we currently pay for tap water. Therefore, the public is likely going to have to accept the presence of detectable levels of ECs in their drinking water, especially if the drinking water source is a surface water body. The EPA currently has a list of ECs that are candidates to be regulated. However, significant research is still needed before the risks associated with the long-term consumption of "contaminant cocktails" are better understood, and regulations are likely still years away.

Without regulations to reduce the discharge of ECs to the environment, one of the best ways to reduce the presence of ECs in the environment is to manage their sources more effectively. This approach is a preventative one, which aims to reduce ECs at their sources rather than treating them once they enter the water cycle. Education and outreach programs are likely the best chance we currently have to change human behavior and, in turn, reduce the presence of ECs in drinking water sources.

Because a significant source of ECs in the environment is from the land application of animal and human wastes, management strategies that reduce the presence of ECs in these wastes prior to their land application would reduce the amount of ECs introduced into the environment. Often, land application of wastes occurs based on the nitrogen (N) demand of crops. Due to different demands of crops for N and phosphorus (P) compared to the amounts of these nutrients in animal manure [[77\]](#page-32-0) changing to applications based on the P demand of crops would reduce application rates, thereby reducing the amount of ECs inadvertently applied to agricultural fields [\[78](#page-32-0)]. Additionally, various studies suggest that composting animal manure reduces EC concentrations [\[79](#page-32-0)[–85](#page-30-0)].

Because the manufacturing and everyday usage of PPCPs contribute to ECs in the environment, we should make informed decisions about the products we buy and the ways in which we dispose of unwanted medications. It is becoming more common for municipalities to hold unwanted medication collection drives to encourage the proper disposal of these products. Because of successful educational and outreach programs, people are responding positively to these collection drives and are happy to be provided with the opportunity to be good environmental

stewards. Additionally, we drive industry with the purchasing decisions we make. Choosing to purchase personal care products made with natural ingredients can reduce our EC footprints.

It will take a collective effort to make source management successful. Although each individual action of land managers and consumers may seem small, an individual's decisions can influence the behavior of others. Collectively, these individual decisions can help to establish grassroots support for policy changes. Overall, the health of our aquatic ecosystems and the long-term sustainability of our water resources depend on the collective outcome of our individual decisions.

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