Canadian Water Network

Canada's Challenges and Opportunities to Address Contaminants in Wastewater

Supporting Document 3

Contaminants in Municipal Wastewater Effluents

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Contents

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Acronyms and Abbreviations

When acronyms are used in the document, a definition is included with first use.

APE	Alkylphenol ethoxylate
ARG	Antibiotic resistance gene
BOD	Biochemical oxygen demand
cBOD	Carbonaceous biochemical oxygen demanding matter
CCME	Canadian Council of Ministers of the Environment
CEC	Contaminants of emerging concern
CSO	Combined sewer overflow
DWI	Drinking water intake
E1	estrone
E2	17β-estradiol
EE2	17α-ethinylestradiol
HAB	Harmful algal bloom
NH_3	Ammonia
NP	Nonylphenol
NPE	Nonylphenol polyethoxylate
PAH	Polycyclic aromatic hydrocarbon
PBDE	Polybrominated diphenyl ether
PBT	Persistent, bioaccumulative and toxic
PCB	Polychlorinated biphenyl
POP	Persistent organic pollutant
PPCP	Pharmaceuticals and personal care product
SS	Suspended solids
SSRI	Selective serotonin reuptake inhibitor
TRC	Total residual chlorine
VOC	Volatile organic compound
WSER	Wastewater Systems Effluent Regulations
WWTP	Wastewater Treatment Plant

Preface

As the list of chemicals we generate as a society grows, many find their way into wastewater and ultimately into our natural environments. Some of these substances are contaminants that can be harmful to human health, fish and wildlife, and to ecosystem services provided by Canada's waterways. To put into clearer context the opportunities to deal with wastewater contaminants in Canada, Canadian Water Network (CWN) led a national review of known contaminants and contaminants of emerging concern (CECs) in municipal wastewater and our options to deal with them.

Supported by a $400,000 investment from Environment and Climate Change Canada, and leveraging CWN's extensive network of research and practitioner communities, CWN convened a national expert panel from October 2017 to March 2018. The panel's mandate, as established by CWN, was to assess Canada's needs and opportunities in dealing with multiple contaminants in domestic wastewater through consideration of the following critical questions:

- Which wastewater contaminants do we need to worry about most, now and in the future?
- What are the options for our diverse Canadian communities to address these contaminants through wastewater treatment?
- What are the important opportunities and trade-offs involved in those treatment choices, including resource recovery, cost implications, socio-economic and cultural fit, and implications for related issues like greenhouse gas emissions?

The expert panel, chaired by Dr. Donald Mavinic, was composed of a group of eight leading experts from across Canada with diverse expertise in municipal wastewater treatment, conventional contaminants and contaminants of emerging concern, environmental and ecosystem impacts, wastewater resource recovery, and the broader legal and socioeconomic implications of wastewater effluent discharges.

The expert panel's primary task was to generate a synthesis report to convey providing a credible and useful framing of where we are, what we know and don't know, and a blueprint for how we can move forward to achieve benefits through more effective wastewater treatment in Canada. This blueprint was developed through panel discussions that were augmented by incorporating perspectives from an extended group of experts from across Canada. A broad range of geographic and topic-area insights were solicited from expert contributors with knowledge of municipal wastewater practice, environmental impacts and assessments associated with wastewater, as well as legal and community perspectives. Input from an expanded group of experts both invited participation at panel working meetings, as well as broader national input through an online questionnaire.

As a supporting document to the expert panel's report, the current document provides a high-level overview of contaminants in municipal wastewater effluents.

Purpose and Scope

The purpose of this document is to provide basic background information for the major groups of contaminants found in municipal wastewater effluent. This document is intended to provide a high-level overview of wastewater contaminants to a general audience in order to support the expert panel's main report. Where possible, we have made an effort to highlight Canadian environments and research.

A comprehensive review of contaminants in wastewater within a Canadian context has been prepared previously by Hydromantis for the Canadian Council of Ministers of the Environment (CCME, 2005), which is suggested as an additional resource if more detailed information is required.

Terminology

The term w*astewater treatment plant (WWTP)* is used in this report, although this term is being replaced in some jurisdictions with *water resource recovery facility (WRRF)*, in recognition of a more holistic approach to urban water management. Municipal WWTP refers to a local or regional government or utility, or provincial or Indigenous-owned facility that receives collected wastewater for treatment and release into the environment.

Within the report, the term *contaminant* has been used to capture pathogens, nutrients, metals, chemicals and physical constituents generated or concentrated by society, which can potentially pose adverse effects on receiving environments and public health.

The term *contaminants of emerging concern (CECs)* is used in this report to refer to non-conventional contaminants that have been detected in wastewater effluents, and for which the potential risks to public and environmental health are not yet fully understood. Various terms have been used in other literature, such as emerging contaminants, emerging substances of concern, trace contaminants, micropollutants and micro-contaminants.

1. Substances in Wastewater Regulated under Wastewater Systems Effluent Regulations

In Canada, the Fisheries Act prohibits discharge of substances that are deleterious to fish (Environment and Climate Change Canada, 2017). The Wastewater Systems Effluent Regulations (WSER; Government of Canada, 2012) allow the authorization or permit to deposit effluents containing selected deleterious substances (section 5), if effluent is not acutely toxic. These substances include carbonaceous biochemical oxygen-demanding matter (cBOD), suspended solids, total residual chlorine, and un-ionized ammonia (NH_3-N, at 15°C ± 1°C) at concentrations of 25, 25, 0.02, and 1.25 mg/L, respectively, in wastewater effluents (Government of Canada, 2012). In addition to the regulated parameters, wastewater effluent must be not acutely toxic at the point of discharge based on 96-hour test for rainbow trout (Government of Canada, 2012).

Oxygen-demanding substances consume dissolved oxygen during their degradation, which depletes oxygen required for the survival of fish and other aquatic organisms. Municipal wastewater contains high loads of organic material, including dissolved and particulate organic compounds. Release of oxygen-demanding materials results in characteristic oxygen declines in the receiving environment, a trend that has been recognized since the 1920s (Fan & Wang, 2008). Secondary (or biological) wastewater treatment uses microorganisms to oxidize and remove these oxygen-demanding substances, protecting receiving environments from oxygen depletion and the associated impacts on fish and other aquatic organisms.

Suspended solids (SS) are the small particles that remain in suspension in wastewater that are finer than coarse material but larger than colloidal matter. Once discharged into surface waters, these particles reduce light penetration into water, and contain nutrients that promote the growth of biofilms and phytoplankton, such as algae and cyanobacteria. Reduction in suspended solids in effluents has been associated with reduced growth of phytoplankton and periphyton, and increased water clarity in surface waters. Suspended solids are also reduced by secondary treatment, but can be present in high levels in combined sewer overflows (CSOs). In some cases, suspended solids in combined sewers have been found to primarily originate from wastewater, contributing up to 75% of the total suspended solids load (Anne-Sophie et al., 2015).

Humans excrete nitrogenous waste in urine in the form of urea, which rapidly breaks down to ammonia and carbon dioxide. As a result, domestic wastewater influents contain high concentrations of ammonia, which is acutely toxic to fish and other organisms, especially in the unionized form. In addition to toxicity, ammonia has an oxygen demand, as it is readily oxidized to nitrate in natural water bodies via the process of nitrification, which consumes oxygen. Nitrogen may also play a role as a limiting nutrient especially in estuaries, and excessive release into the environment may cause excess growth of algae (Forsberg et al., 1976).

Gastrointestinal pathogens are associated with fecal material, and these are regularly killed or inactivated before effluents are discharged to surface waters to protect public health. Chlorine has been widely used as a disinfectant to kill bacteria in wastewater prior to its discharge. While effective, this approach may result in the release of residual chlorine, which is toxic to aquatic life (Watson et al., 2012).

In addition to the substances regulated under WSER, wastewater effluents and residuals contain numerous additional chemicals, pathogens and nutrients that pose potential risks to human and ecosystem health. These include the diversity of chemicals used in households, industry, and agriculture. Although some of these chemicals are banned or undergoing risk management under various programs (and permits), they continue to persist in wastewater and many new pathogens and contaminants continue to emerge and raise concerns. Hundreds of different chemical compounds may be present in municipal wastewater effluents, which have the potential for additive, synergistic, or antagonistic effects on aquatic life (Hummel et al., 2006; Verlicchi et al., 2012). Mixtures of various chemical compounds complicate risk assessments on the effects of exposure to aquatic life in the environment because they can differ dramatically in their mechanisms of toxicity and exposure. However, approaches are being developed to better understand both the potential and actual risks of these mixtures in the environment (Diamond et al., 2017). The major groups of these contaminants are outlined the sections below.

2. Pathogens and Human Health Risks

Wastewater treatment originated and evolved as a response to public health and environmental concerns, with human health being an important driver. Pathogens are found in wastewater and stormwater effluents, and the exposure of human to these pathogens occurs through consumption of contaminated water and seafood (e.g., fish and shellfish) as well as activities such as swimming in contaminated waters (Health Canada, 1997, Loomer et al., 2008, Soller et al., 2010). These pathogens include a variety of bacteria, protozoa, helminths, and viruses. Although waterborne pathogens in municipal wastewater are generally from human sources, animals (e.g., livestock, pets, wildlife) can also be sources of many pathogens of concern for humans. Pathogens therefore arise from many sources across watershed and must be considered for public health protection. Altered hydrology, water quality and temperature resulting from climate change may allow for additional distribution and exposure to current and emerging waterborne pathogens in the future (Wu et al., 2016). Research is continuing to improve the techniques for analyzing the presence of pathogenic organisms and their potential for adverse health impacts.

2.1 Traditional threats to public health

Typhoid, cholera, dysentery, and diarrhea are major public health threats associated with fecal pathogens of human origin. *Salmonella*, *Shigella*, *Campylobacter*, and some strains of *Escherichia coli* (e.g., 0157 H7), are common bacterial pathogens found in municipal wastewater and thousands of deaths are caused by these pathogens every year globally (Metcalf and Eddy - AECOM, 2014).

Protozoa is another category of pathogens that is of major concern for public health. *Cryptosporidium parvum* and *Giardia lamblia* are the two major protozans that can be found in wastewaters, and have been the causes of most waterborne disease outbreaks in Canada (CCME, 2006). The potential for giardiasis outbreaks is greater in northern regions, since cold water and ice cover provide ideal conditions for the proliferation of parasites (Environment Canada, 2001). *C. parvum* and *G. lamblia* have resistant cysts and therefore cannot be effectively inactivated or destroyed during conventional disinfection with chlorine (Environment Canada, 2001).

The discharge of wastewater effluent that is contaminated with infectious viruses represents another potential risk to human health (Qiu et al., 2015). Several groups of enteric fecal viruses are found in raw municipal wastewater, the most important of which are enterovirus, norovirus, rotarovirus, reovirus, adenovirus, and hepatitis A virus. Norovirus and rotarovirus are major causes of viral diarrhea, while reovirus and adenovirus are known to cause gastroenteritis, respiratory problems, and eye infections (Metcalf and Eddy - AECOM, 2014). In a study in the Gold Bar WWTP in Edmonton (Alberta), seven viruses including norovirus, notarovirus, sapovirus, astrovirus, adenovirus, enterovirus and JC virus were detected in primary wastewater effluents in which infectious viruses were present (Qiu et al., 2015).

2.2 Emerging threats to human health

2.2.1 Antimicrobial resistance genes

Antibiotic resistance genes (ARG) are emerging environmental concerns that occur naturally but may be selected for in environments containing high concentrations of antibiotics (Sanderson et al., 2016). ARGs are often found on mobile genetic elements of bacteria such as transposons, integrons and plasmids. These mobile elements can facilitate the evolution of ARGs by transferring them to bacteria of the same or different species within wastewater systems or receiving environments (Allen et al., 2010, Baquero et al., 2008). Multidrug-resistant bacteria pose an imminent threat to global health and economies because of the reduced susceptibility of these pathogens to most antibiotics used in medical treatments (World Bank, 2016). When ARGs are discharged to the environment from municipal wastewater effluents, they may affect the natural resistance of bacteria in biofilms, sediments, and aquatic life and ultimately increase the occurrence of antibiotic resistant strains of bacteria over time (Marti et al., 2013, Singer et al., 2016). Impacts of WWTP effluents on the distribution of ARGs can be measured at significant distances from the point of discharge. For example, ARGs detected in highest abundances downstream of a Saskatchewan municipal WWTP were consistent with the genes found at the highest abundances in the treated effluent. None of the genes surveyed returned to their upstream levels, 5 km downstream of the effluent release point (Marti et al., 2013).

3. Nutrients

Large amounts of nutrients, especially nitrogen and phosphorus, enter aquatic ecosystems through municipal wastewater, stormwater, and overland runoff carrying manure and synthetic fertilizers. Nutrients present in wastewater effluents act as fertilizers and promote the growth of algae and cyanobacteria that form the base of aquatic food chains. This increase in nutrient status is known as eutrophication, and can result in harmful algal blooms, oxygen depletion, and fish kills in fresh- and coastal waters (Schindler & Vallentyne, 2008). Excessive nutrient loading has been suggested as a cause for observed decline in invertebrate densities and loss of sensitive taxa downstream of wastewater outfalls (Grantham et al., 2012), and eutrophication has been called the most widespread water quality problem facing the world today (Schindler & Vallentyne, 2008; Schindler, 2012). Although it varies depending on the site, municipal and industrial WWTPs can contribute significant nutrient loads to receiving waters in urban areas (Schindler et al., 2012). In the context of wastewater effluents, nitrogen and phosphorus are the nutrients primarily associated with eutrophication and remains a widespread concern for healthy ecosystems.

3.1 Phosphorus

Municipal wastewater can have a considerable contribution to the phosphorus load into freshwater systems. However, the relative contributions of phosphorus from various sources is highly dependent on the local characteristics of a particular watershed. For example, in the Lake Simcoe watershed (Ontario, Canada), only 7% of the total phosphorus load originates from municipal wastewater, while 6% comes from septic systems located near the lake, 31% from urban storm water, 29% from agriculture, and 27% from atmospheric deposition (Ministry of the Environment and Climate Change, 2010). In this watershed, stringent phosphorus caps have been in place for WWTPs since the 1980s, resulting in additional treatment strategies resulting in very effective phosphorus removal.

Phosphorus has been known to be a limiting nutrient in freshwater systems for decades, when whole-lake experiments confirmed that addition of phosphorus was essential to creating freshwater algal blooms Schindler, 1974, 1977). As a result, phosphorus inputs to many lakes were reduced, and several case histories exist that demonstrate successful reversals of lake eutrophication (Schindler, 2012). More recent reports based on whole-ecosystem, long-term experiments continue to support the assertion that phosphorus limits primary productivity (and therefore eutrophication) in freshwater environments (Higgins et al., 2017; Schindler, Hecky, et al., 2008). However, high levels of phosphorus have accumulated in the sediment from historic loading and can become re-suspended in the water column (Carey & Migliaccio, 2009; Nürnberg & LaZerte, 2016; Orihel et al., 2017; Tammeorg et al., 2016) and delay water body recovery even when external phosphorus loads have been reduced (Nürnberg et al., 2016; Schindler, 2012). Therefore, many watersheds remain impacted due to legacy phosphorus loadings from both agriculture and wastewater despite nutrient management practices.

3.2 Nitrogen

Nitrogen is a major component of municipal wastewater that presents risks to aquatic ecosystems. Ammonia is acutely toxic to fish and can result in oxygen depletion as a result of in-river nitrification. However, ammonia and other nitrogenous compounds (especially nitrate) are nutrients that can also be used by heterotrophic bacteria, cyanobacteria, and algae. Eutrophication causes predictable increases in the biomass of algae in freshwater and coastal marine ecosystems. Increases in cyanobacteria and suspended algae, as a result of eutrophication, have been reported worldwide for natural lakes and large rivers (Smith, 2003). Some researchers have suggested that nitrogen is the most limiting nutrient in estuaries and therefore, controlling nitrogen inputs is essential for control of coastal eutrophication (Howarth & Marino, 2006; Schindler, 2012). However, this suggestion has been criticized as being based on bottle bioassays, mesocosms, nutrient ratios, and other short-term indicators of nitrogen limitation, which have questionable real-world significance (Schindler, 2012). Several studies have also asserted that, in some cases, it may be necessary to reduce nitrogen loading to reverse eutrophication of lakes (Conley et al., 2009; Lewis et al., 2011; Lewis & Wurtsbaugh, 2008; Scott & McCarthy, 2010; Xu et al., 2010).

However, others have argued that reduction of nitrogen is very expensive and that inducing nitrogen-limitation may have the unintended negative consequence of shifting algal communities to favour of nitrogen-fixing cyanobacteria, while prevents a response from the lake in terms of nutrient status (Higgins et al., 2017; Schindler, 2012; Schindler, Hecky, et al., 2008).

3.3 Harmful algal blooms

One of the most serious problems caused by nutrient enrichment is its contribution to increased prevalence of harmful algal blooms (HABs), which are associated with high densities of cyanobacteria or algae. Some cyanobacterial taxa produce toxins that are poisonous to aquatic organisms and humans, but non-toxic cyanobacteria and algae also pose problems (Metcalf & Codd, 2014). For example, decomposition of these phytoplankton by heterotrophic bacteria depletes dissolved oxygen, and algal blooms reduce light penetration, resulting in the loss of submerged vegetation (Carey et al., 2013). In addition, toxins produced be algal blooms may result in health impacts in humans if ingested through drinking water or aquatic food sources such as shellfish (Glibert et al., 2005; Watson et al., 2016) Occurrence of HABs is strongly correlated with elevated phosphorus concentrations and increased temperatures, but the role of nitrogen is less clear (Carey et al., 2013). It has been demonstrated that low nitrogen:phosphorus ratios shift phytoplankton community compositions to favour nitrogen-fixing species of cyanobacteria (e.g., *Nostocales*), which are more likely to produce toxins (Schindler et al., 2012). However, in the presence of very high phosphorus concentrations, HAB occurrence can be positively correlated with nitrogen concentrations (Dolman et al., 2012). In addition, a study of 102 German lakes found that algal and cyanobacterial taxa had diverse responses to differential nitrogen versus phosphorus concentrations, and that differences between taxa were not consistent with the hypothesis that nitrogen-fixing taxa would be favoured in low nitrogen conditions (Canadian Water Network, 2017; Dolman et al., 2012). There is strong evidence that excessive nutrient input into water bodies promotes eutrophication and harmful algal blooms. Although phosphorus acts as the primary limiting nutrient and is the primary target for management action in freshwater environments, the role of nitrogen also needs to be considered, at least under some circumstances.

4. Metals

Metals are naturally occurring, but inputs to aquatic environments have increased due to widespread use in commercial products. Worldwide, municipal wastewater effluents contain numerous metals, which originate from sources such as dental practice wastes, paints, consumer electronics, and flame retardants (Hargreaves et al., 2016), and occur primarily bound to particulate matter (Baldwin et al., 2016; Carletti et al., 2008). Metals such as copper (Cu), lead (Pb), nickel (Ni), zinc (Zn) mercury (Hg), antimony (Sb), chromium (Cr), arsenic (As) and cadmium (Cd) are potentially toxic and pose risks such as acute or chronic health effects in animals, phytotoxicity, and bioaccumulation (Alhadrami et al., 2016; Carletti et al., 2008; Darko et al., 2016; Gagnon & Saulnier, 2003; Hargreaves et al., 2018; Lisa Jones et al., 2017; Marcogliese et al., 2015; Marsalek et al., 2006; Mudhoo & Kumar, 2013; Zheng et al., 2007). Metal toxicity may be lethal or produce adverse biological effects on an organism's activity, growth, metabolism, or reproduction. For example, mercury is a known neurotoxin that causes structural damage to the brain and inhibits the activity of enzymes that are needed for neurotransmission (Wright & Welbourn, 2002). In aquatic ecosystems, even low mercury concentrations may be problematic because this metal bioconcentrates in tissues and bioaccumulates in the upper trophic levels of a food chain. In contrast, lead bioconcentrates in the skin, bones, kidneys, and liver of fish but does not biomagnify up the food chain. Chromium and cadmium have also been shown to inhibit growth of aquatic plants, crustaceans, and fish (Solomon, 2008).

In addition to municipal wastes, urban runoff contributes metals to aquatic environments, which may originate from sources such as automobiles, tire wear, vehicle exhaust, commercial and industrial activities, building facades, structures, soil erosion, and road pavement (Ma et al., 2016). Contributions from stormwater are seasonally dependent, and are typically high in early spring, coinciding with snowmelt (Bartlett et al., 2012). Although urban runoff often enters waterways independently from municipal wastewater, stormwater inputs contribute to municipal effluents in combined sewer systems.

High removal efficiency of several metals has been reported in municipal WWTP effluents using activated sludge treatment (Carletti et al., 2008; Nielsen & Hrudey, 1983). Removal of mercury in wastewater treatment plants has positive correlations with suspended solids removal and chemical oxygen demand, which may be explained by its high-sorption characteristics (Hargreaves et al., 2016). However, metals cannot be degraded or destroyed, and removed metals are correspondingly concentrated in waste sludge, shifting the metal-associated risk to the solid waste (e.g., sludge, biosolids).

Environments downstream of wastewater effluent outfalls are often enriched with metals compared to control sites. For example, elevated metal concentrations have been found in the sediments and amphipods downstream of sewer outfalls and street drainages (Schertzinger et al., 2018), and elevated cadmium and metallothionein levels have been found in fish (pearl dace, *Semotilus margarita*) exposed to municipal wastewater effluents (Klaverkamp et al., 2006). In the Grand River (Ontario), wild freshwater mussels exposed to effluents from municipal WWTPs and stormwater from four urban centres had significant increases in Cu, Pb, Zn, Al, Cr, and Ni in their gills (Gillis, 2012). In Montreal, where chemically-enhanced primary treatment is used, municipal wastewater contributes relatively high loads of Cd, Cu, Zn, Ag to the St Lawrence River, although mercury concentrations in the St. Lawrence have declined over the past 40 years (Marcogliese et al., 2015).

Although toxic impacts of metals at high concentrations are well documented, metal toxicity is complex and chronic impacts on the aquatic biota are poorly understood. Metal bioavailability (and therefore toxicity) depends a variety of physicochemical characteristics of the environment, including temperature, pH, hardness, salinity, and dissolved organic carbon (Hargreaves et al., 2018; Wright et al., 2002). Metal toxicity is further complicated by the variable concentrations between sediments and the water column, bioaccumulation and mobility of metals, the intermittent nature of stormwater inputs, and the uncertainty of exposure in migratory animals.

5. Legacy Pollutants

Legacy pollutants are chemicals that remain in the environment long after they were first introduced, and often exist as a result of commercial and industrial activities. Many of these persistent organic pollutants (POPs) have been classified as persistent, bioaccumulative and toxic (PBT) and have been banned under various programs (e.g., the Canadian Environmental Protection Act). Many chemicals have been identified for virtual elimination and/or risk management after undergoing risk assessments. Interestingly many of these legacy contaminants can also act as endocrine disruptors (Rahman et al., 2001; Tyler et al., 1998) and because they tend to be hydrophobic many legacy chemicals detectable in wastewater effluents are associated organic particles such as sediments and sludges (Gomes et al., 2003; Petrović et al., 2001). There a many different legacy contaminants and examples of some common groups of legacy pollutants are outlined below.

5.1 Volatile organic compounds (VOCs)

Volatile organic compounds are among the detected legacy contaminants in wastewater that are of public health concern. Although VOCs can be released from the air, effluent and sludge of wastewater treatment facilities (CWWA, 2002) the boiling point and vapour pressure of these compounds makes it so they are likely to be released into the environment in their vapor state (Metcalf and Eddy - AECOM, 2014). Long-term exposure to high levels of some VOCs (i.e., benzene and formaldehyde) may be associated with increased rates of cancer (Health Canada, 2017).

5.2 Polychlorinated biphenyls (PCBs)

Polychlorinated biphenyls (PCBs) were synthesized in North America for electrical equipment, heat exchangers, hydraulic systems, and other specialized applications from 1929 until the late 1970s (Environment and climate change canada, 2017b). Although, these compounds were never produced in Canada, they were widely used until their import sale and use were banned in the late 1970s and in 1985, respectively. However, PCBs are very persistent in the environment (with reported half-lives in soil and sediment ranging from months to years) (Environment and climate change canada, 2017b) and can still be released into the wastewater treatment plants and receiving waters from industrial discharges and contaminated disposal sites. PCBs have very low solubility in water, and most of these compounds are contained in sediments that may continue to be mobilized over time, especially during flooding events. Many PCBs are highly lipophilic (fat soluble) and resistant to biodegradation, and therefore bioaccumulate in animal tissues, and can biomagnify (concentrate) in food webs. Exposure to PCBs in humans is predominantly through fatty foods such as meat, poultry, eggs, and dairy products, and fish (USEPA, 1999) but wastewater may continue to act as source to the environment. PCBs have also been shown to affect endocrine function in aquatic animals (Fossi & Marsili, 2003; Gilroy et al., 2017) and have been linked to cancer in humans (IARC, 2015).

5.3 Polybrominated diphenyl ethers (PBDEs)

Polybrominated diphenyl ethers (PBDEs) are a class of flame retardants used in building materials, electronics, polyurethane foams, furnishings, thermoplastics, textiles and vehicles. They are commonly found in municipal effluent and sludge (Song et al., 2006) and sediments near discharge points of municipal effluents (Samara et al., 2006). Their persistence in the environment ranges from days in air, to months in water and soil, and years in sediment (Environment Canada, 2006). While PBDEs have low acute toxicity, they have long-term effects on the endocrine system, such as interference with steroid and thyroid hormone metabolism (Gilroy et al., 2017; Morgan & Lohmann, 2010). In addition, bioaccumulation and the trophic transfer of PBDEs has been observed in aquatic food webs (e.g., Lake Winnipeg (Law et al., 2006)), resulting in heightened risk for predators at higher trophic levels, including humans.

5.4 Nonylphenols, nonylphenol polyethoxylates and alkylphenol ethoxylates

A major group of contaminants in effluents are nonylphenols (NPs) and their polyethoxylates (NPEs). Concentrations of NPs and NPEs are attributed to human activities, as these compounds along with alkylphenol ethoxylates (APEs) were widely used in industrial and household products, including laundry detergents, shampoos, cosmetics, latex paints, and spermicides (Servos et al., 2003). APEs with longer ethoxylate chains undergo rapid biodegradation, with some of their degradation products being more persistent and toxic. Nonylphenol is a potential endocrine disrupting chemical with estrogenic properties in mammals and aquatic life (Bjerregaard et al., 1998; Metcalfe et al., 1996, 2001).

5.5 Polycyclic aromatic hydrocarbons (PAHs)

Polycyclic aromatic hydrocarbons (PAHs) comprise several hundred compounds that arise from both natural and anthropogenic activities. PAHs occur primarily in soil, sediment, oily substances, and in particulate matter in air (Watson et al., 2012). Their half-lives vary widely from minutes in air (degrading quickly with UV exposure) to years in soil (CCME, 1999; Wild et al., 1991). Oil spills and refinery effluents are major sources of PAH contamination of freshwater and marine environments. Domestic sewage, stormwater runoff, landfills, leachate from the wood preservative industry (e.g., creosote), and waste disposal sites are further contributors of anthropogenic PAHs to the environment (CCME, 1999, Yunker et al., 2002). PAHs in municipal and stormwater effluents originate primarily from petroleum, and their concentrations in municipal effluents varied based on factors such as energy sources (e.g., electricity vs. fossil fuel), motor vehicle density, and the types of industries discharging to the sewer systems (Pham et al., 1999). Several PAHs are acutely toxic to aquatic organisms in the presence of solar UV radiation (CCME, 1999), and sub-lethal effects have also been observed (e.g., Wilson et al., 2000).

6. Pharmaceuticals and Personal Care Products

Wastewater is a complex mixture of substances, including wide variety of natural and synthetic compounds used for health or cosmetic reasons, collectively known as pharmaceuticals and personal care products (PPCPs). Some of these compounds are not effectively removed in the wastewater treatment process and therefore are found in final effluents and surface waters receiving these discharges (Boxall et al., 2012). A wide variety of PPCPs, such as antibiotics, antimicrobials, pain relievers, anti-depressants, fragrances and caffeine, as well as their metabolites, have been identified in North American water bodies, sediments, and the tissues of aquatic organisms (Daughton & Ternes, 1999; Ebele et al., 2017; Focazio et al., 2008; Kolpin et al., 2002). PPCPs have also been detected in drinking water sources (Heberer, 2002), although typically at concentrations that are orders of magnitude below therapeutic doses (Cizmas et al., 2015; Jones et al., 2005). Both the US and EU have identified PCPPs as substances of concern that may require regulation (European Commission, 2012, 2013; US EPA, 2014), and the European Commission has proposed amending its list of priority substances (which are monitored) to include diclofenac, estradiol, and ethynylestradiol (European Commission, 2012, 2013).

Seasonality may affect the occurrence and concentration of some PPCPs in the environment. For example, higher concentrations of CECs have been detected in wastewater effluent and receiving waters in the winter, presumably due to less effective PPCP removal by wastewater treatment processes in lower temperatures (Hedgespeth et al., 2012; Vidal-Dorsch et al., 2012; Vieno et al., 2005). Likewise, in northern Canada, low temperatures have been found to inhibit removal of PCPPs in wastewater treatment (Chaves-Barquero et al., 2016). Conversely, some pharmaceuticals have marked usage patterns, such as the use of DEET (an insect repellent) in the summer (Luo et al., 2014)

6.1 Observed impacts of pharmaceuticals and personal care products on aquatic organisms

Low concentrations of PPCPs are typically detected in natural waters (Lishman et al., 2006; Metcalfe et al., 2003) but, given that pharmaceutically active compounds are often designed to produce biological responses at low concentrations, some of these compounds may pose risks to aquatic organism health. In addition, some PPCPs may accumulate in living tissues, particularly if they are lipid soluble. For example, some aquatic species (e.g., mussels and plants) have been shown to bioaccumulate a variety of PCPPs including antibiotics (de Solla et al., 2016). Fish and other aquatic life can be affected by various pharmaceuticals because the mechanisms may be similar (retained through evolution) across organisms (Brown et al., 2014). For example, a common class of antidepressant, selective serotonin reuptake inhibitors (SSRIs), has been found to alter behaviours in fish and aquatic invertebrates, which may reduce their ability to survive (Brodin et al., 2013; Fong et al., 2017; Peters et al., 2017). A benzodiazepine anxiolytic drug (oxazepam) altered behaviour of wild European perch (*Perca fluviatilis*) at concentrations encountered in surface waters receiving treated municipal effluents (Brodin et al., 2013). Pharmaceuticals may also act through a variety of unexpected mechanisms and affect non-target organisms, such as aquatic plants and algae. Assessing the environmental risk of pharmaceuticals in the environment therefore is associated with considerable uncertainty (Brain et al., 2008; Oakes et al., 2010).

6.2 Uncertainty regarding the impacts of pharmaceuticals and personal care products

Although most individual PPCPs are at low concentrations in wastewater effluents and surface waters and likely do not represent an unacceptable risk (Corcoran et al., 2010), some may be approaching concentrations that could have chronic effects on aquatic life because discharges and exposures are continuous and the compounds have high potency. However, the risk they pose to the receiving environment remains uncertain and needs further research (Boxall et al., 2012).

7. Endocrine-Disrupting Chemicals

A variety of compounds found in municipal effluents, including PPCPs, pesticides, PAHs, metals, and household and industrial chemicals, have endocrine-disrupting effects on aquatic organisms (Bergman et al., 2012; Hewitt & Servos, 2001). Natural hormones, which enter the environment predominantly through human and animal excretion, such as 17β-estradiol, estrone, androstenedione and testosterone, have been detected in municipal wastewater effluents and receiving waters (Hamid & Eskicioglu, 2012; Meador et al., 2016; Servos et al., 2005; Ternes et al., 1999).

7.1 Endocrine disruption impacts on aquatic organisms

Endocrine disruption associated with municipal wastewater is widespread and has the potential to alter populations of aquatic organisms globally (Mills & Chichester, 2005; Tyler et al., 1998). Early interest in the endocrine disrupting effects of wastewater was sparked by observations in the United Kingdom that fish downstream of wastewater outfalls exhibited reproductive changes including a high incidence of intersex (developing eggs and/or female reproductive ducts in testicular tissue of male fish) (Jobling et al., 1998; Sumpter & Jobling, 2013). These responses were associated with the presence of estrogens and estrogen mimics in the effluents (Desbrow et al., 1998). The estrogenicity of effluent has typically been associated with natural estrogens (17β-estradiol, E2; estrone, E1), the active ingredient in birth control (17α-ethinylestradiol; EE2), and, to a lesser extent, industrial chemicals such as alkyphenols and bisphenol-A. Since these early observations, numerous reports worldwide have associated wastewater exposure with the increased occurrence of intersex in fish (Bahamonde et al., 2013). The significance of low levels of intersex in the environment remains ambiguous, but moderate to high levels of intersex have been weakly associated with impaired population fitness (Harris et al., 2011; Lange et al., 2011). Antiandrogens such as triclosan (Jobling et al., 2009) and chemicals such as metformin (Niemuth & Klaper, 2015) found in wastewaters have also been shown to cause endocrine disruption in fish.

Numerous laboratory experiments have demonstrated that estrogenic chemicals found in municipal effluents, such as E2, EE2 and nonylphenol (and many others), can cause endocrine disruption and reproductive changes in fish (Länge et al., 2001; Nash et al., 2004; Parrott & Blunt, 2005). The concentrations of EE2 reported in some municipal wastewater effluents are sufficient to cause reproductive changes in fathead minnows (Parrott et al., 2005). Although estrogenicity of treated effluents are typically lower, even effluents from tertiary treatment facilities appear to have some endocrine disrupting properties (Baynes et al., 2012; Filby et al., 2010; Ings et al., 2011).

Increased prevalence of an intersex condition in fish downstream of WWTPs has been recently documented in the Grand River (Ontario, Canada). For example, the incidence of intersex condition in fish downstream of Waterloo and Kitchener WWTPs in Ontario was shown to be significantly higher than at reference sites (Tanna et al., 2013; Fuzzen et al. 2016). Other studies have demonstrated that sewage-exposed male darters had reduced ability to produce male sex hormones (Tetreault et al., 2011; Bahamonde et al. 2015) and impaired reproductive performance (Fuzzen et al., 2015). During a recent upgrade of the treatment plant in Kitchener it was shown that there was a rapid decline in estrogenicity of the effluent that was associated with changes in gene expression, sex steroid production and reductions in intersex in wild fish populations (Hicks et al., 2017; Marjan et al., 2017, 2018).

Kidd et al. (2007, 2014) examined short- and long-term changes within an experimental lake dosed with EE2. Three years of EE2 additions to the lake led to intersex in the fathead minnow and a near extinction of this species from the lake (Kidd et al., 2007, 2014). Likely, because of its short lifespan, the fathead minnow was the first species to decline, implying short-lived species may be at greatest risk from estrogens and estrogen mimics. This loss of fathead minnow also triggered indirect effects in the food web such as decreases in lake trout, a top predator fish, from loss of prey fish and increases in invertebrates due to decreased predation. A few years after the EE2 additions stopped, the fathead minnow population recovered (Blanchfield et al., 2015). As mentioned previously for the Grand River, this suggests that reducing the estrogens in municipal wastewater effluents has benefits for fish living in the surface waters.

8. New Risks to Aquatic Environments

As new industrial processes and consumer products are developed, new groups of contaminants will continue to be identified in municipal wastewaters. Two recently identified substances that may pose risks to aquatic ecosystems are microplastics and nanoparticles. Although some evidence exists that these compounds may have negative environmental impacts, there is a high degree of uncertainty regarding their environmental exposure, bioavailability and effects on aquatic life.

8.1 Microplastics

Microplastics are plastic particles less than 5 mm in diameter, are diverse in size and form, and can be present as fibres, pellets/beads, foams, and films. Sources of microplastics to water include the breakdown of larger plastic items, product spillage from industry or shipping, and synthetic textiles. Historically, microbeads have been used in personal care products and cleaning supplies, but Canada has recently prohibited the use of microbeads in toiletries and natural health products (Government of Canada, 2018). Other forms of microplastics continue to enter water bodies through municipal wastewater, such as polyester and acrylic microfibers from clothing. A single fleece garment has been shown to produce up to 110,000 microfibres each time it is washed (Carney Almroth et al., 2018), and microplastics originating from polyester fibres from clothing have been found in sediments near wastewater discharges (Woodall et al., 2015).

Microplastics are an emerging concern in marine and freshwater environments, with some negative impacts observed on aquatic ecosystems (Eerkes-Medrano et al., 2015). For example, microplastics stimulate biofilm formation throughout the water column and have the potential to enhance the growth of biohazardous bacteria (Eckert et al., 2018). In mussels, microplastics are drawn into the gills and ingested, resulting in effects on the immune system and membranes (Von Moos et al., 2012). In addition, toxic constituents of microplastics such as monomers and plastic additives might be accumulated within organisms (e.g., planktons), (Wright et al., 2013). Moreover, microplastics may also move up a food web through trophic transfer (Farrell & Nelson, 2013). However, the ecological implications of microplastic uptake into aquatic species and their trophic transfer in aquatic food webs are currently unclear.

Larger plastic particles are typically removed through mechanical wastewater treatment processes. Secondary and tertiary treatment plants are shown to be more effective in the removal of microplastics.

These plants removed 99.7% of microplastics ≥ 300 µm in size from wastewater influent and left 10–40 particles per cubic meter in the wastewater effluent, while primary treatment plant effluent had a residual of approximately 1500 particles per cubic meter (Magnusson et al., 2016). However, given the large volume of effluents discharged, there may still be risks associated with these particle discharges. Moreover, microplastics that are physically removed during the wastewater treatment process will end up in biosolids, with a potential alternate route of exposure to the environment.

8.2 Nanoparticles

Nanoparticles are complex and represent a very diverse group of substances, which share the common property of being <100 nm in at least one dimension. Engineered nanoparticles are used in a wide variety of areas including biomedicine, environmental remediation pharmacology, agriculture, cosmetics and sunscreens, electronics, and renewable energies (Berkner et al., 2016; Nowack & Bucheli, 2007). As many of these groups are household products, release of them into the environment through municipal wastewater appears to be inevitable.

There is some evidence of harmful impacts of nanoparticles on aquatic life. For example, toxicity of copper nanoparticles has been reported in fish (Griffitt et al., 2007), invertebrates (Gomes et al., 2011), bacteria (Yoon et al., 2007), and algae (Aruoja et al., 2009). Biological accumulation of copper nanoparticles may also elicit adverse chronic effects in animals, but this is not well understood (Croteau et al., 2014). Iron oxide nanoparticles have been demonstrated to cause malformations, hatching delays, or mortality in zebrafish embryos (Zhu et al., 2012). In addition, chronic impacts in *Daphnia* and fathead minnow have resulted from exposure to the engineered nanoparticles (Zhu et al., 2012).

Nanoparticles tend to aggregate in water and therefore settle into sediment/sludge, although they can be released back into the water from sediments (Dwivedi et al., 2015). Assessing the risk of nanoparticles is complicated as their form and bioavailability affect their toxicity (Vale et al., 2016). Ultimately, there is still some degree of uncertainty regarding these particles, and continued research will help to ascertain the relative risks to aquatic ecosystems and human health that may be posed by various types of nanoparticles.

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