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REVIEW

Pharmaceuticals, endocrine disruptors, personal care products, nanomaterials and perfluorinated pollutants: a review

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Abstract The presence of emerging micropollutants such as pharmaceuticals, endocrine disruptors, personal care products, nanomaterials and perfluorinated substances in the environment remains a great threat to the health and safety of humans and aquatic species. These micropollutants enter the environment via anthropogenic activities and have been detected in surface water, groundwater and even drinking water at nanogram per litre to microgram per litre concentration. To date, limited information exists on the fate, behaviours, and pathways of these micropollutants in the environment. The potential ecotoxicological effects on the receptors due to exposure to individual or mixture of these chemicals still remain unknown. This review provides an overview on pharmaceuticals, endocrine disrupting compounds, personal care products, nanomaterials and perfluorinated pollutants, with emphasis on their occurrences, effects, environmental fates, and potential risk of exposure in water, soil or sediment. Based on the literature survey, it was found that in spite of an extensive research and different developmental efforts on the challenges of emerging micropollutants, the solution to the problem of emerging micropollutants in the environment is far from being solved. The needs for behavioural change among citizens, strong political will and policy formulation on the part of government are identified as possible panacea for combating the growing influence of these potential damaging substances. Suggestions on proactive and precautionary measures that must be taken to protect the environment as well as guarantee the health and safety of humans and aquatic species are provided. Future research should concentrate on the development of a risk based screening models and framework that can predict the sources, fate and behaviours of emerging contaminants in the environment is recommended.

Keywords Emerging chemical contaminants · Endocrine disrupting compounds · Exposure risk · Environmental fate

Introduction

In recent times, the production and consumption of chemically manufactured products by consumers have been linked to growing environmental pollution and different health challenges. This growing environmental pollution which has attracted considerable global scientific attention cannot be attributed to the activities of the chemical and pharmaceutical industries or the climate change alone (Phillips et al. 2010). Other anthropogenic activities such as mining, agricultural, domestic activities and urbanisation also contribute immensely to the higher pollution index and hinder the availability of sustainable water supply. Currently, the pollution of the global water cycle with persistent organic contaminants remains one of the major challenges of the 21st century. The majority of these organic substances are only partially removed by conventional wastewater treatment plants. However, many of these contaminants escape into the environment and spread across different ecological compartments. These complex compounds are polar or semi-polar and are known to exhibit both acidic and basic functional groups in aqueous medium. Besides, these substances are mobile, ubiquitous, persistent and bioaccumulate in wildlife and

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human tissue, thus raising a number of questions regarding the safety of citizens (Fawell and Ong 2012; Focazio et al. 2008; Nam et al. 2014). Persistent organic contaminants are mostly unregulated or in the process of regulation, yet possess potent endocrine disrupting properties and interfere with the hormonal function in the body (Gavrilescu et al. 2015; Milić et al. 2013).

With advances in analytical techniques, such as high-performance liquid-chromatography-coupled mass spectrometry (HPLC–MS), liquid chromatography-mass spectrometry tandem mass spectrometry (LC–MS/MS), a wide array of previously undetected compounds in a complex environmental matrix have been identified and quantified at low microgram or nanogram-per-litre concentration (Fawell and Ong 2012; Rivera-Utrilla et al. 2013). These groups of substances have collectively been called contaminants of emerging concern or chemicals of emerging concern and have attracted significant research interest among the major international organisations such as World Health Organization (WHO), United States Geological Survey (USGS), United States Environmental Protection Agency (EPA) and the European Commission (EU). It is should be noted that while some previously used, harmful chemicals such as polychlorinated biphenyls, dichlorodiphenyltrichloroethane, chlordane, are being phased out of circulation. New chemicals are being manufactured and incorporated into products to meet human beneficial purposes namely toothpastes, soaps, perfumes, artificial sweeteners, insect repellents, deodorants, prescribed drugs, plastic bottles and pipes, among others. Most of these synthetic products containing chemicals of emerging concern are not really new in the market. Nevertheless, most chemical of emerging concern in the products are not yet properly regulated and may exert serious health effects on consumers. Contaminants of emerging concern include a wide array of different compounds and their metabolites such as pharmaceuticals, personal care products, endocrine disrupting compounds, pesticides, disinfection-by products, flame retardants, nanomaterials, veterinary medicines, among others (Al-Rifai et al. 2011; Duong et al. 2008, 2014; Gong et al. 2011; Heberer 2002; Houtman 2010; Fawell and Ong 2012; Kumar and Xagoraraki 2010; Lapworth et al. 2012; Postigo and Barcelo 2014; Sun et al. 2014).

The perceived toxicity of emerging contaminants in manufactured products has recently become a subject of intense debate and a global issue of public health concern due to their individual or synergistic actions on humans and the ecosystem. (Jiang et al. 2013; Kolle et al. 2013; Trapido et al. 2014; Manickum and John 2014). This interest is reflected in a growing number of scientific publications focusing on analytical method development, removal techniques, and environmental monitoring and risk assessment to name just a few aspects (Kümmerer 2011; Mompelat et al. 2009; Sanchez-Avila et al. 2012). However, chemicals of emerging concern have no clear definition; hence, no comprehensive list exists and their interactions are complex, and therefore different definitions have been proposed. Kümmerer (2011) defined emerging micropollutants as unregulated compounds or ones with limited regulation which are present in the environment at low µg/L range and below, irrespective of their chemical structure and which thus require monitoring. Marcoux et al. (2013) summarised emerging micropollutants as newly detected substances in the environment or those already identified as risky and whose use in items is prohibited, or substances already known but whose recent use in products can cause problems during their future treatment as waste. According to US Geological Society (2014), emerging contaminants are any synthetic or naturally occurring chemical or any microorganism or metabolites that is not commonly monitored in the environment but has the potential to enter the environment and cause known or suspected adverse ecological and/or human health effects.

Currently, there is a growing accumulation of emerging organic pollutants in the environmental matrix caused by careless disposal occasioned by the absence of stringent regulatory framework or lack of compliance with the existing environmental protection laws (Bell et al. 2011; Breivik et al. 2011; Esplugas et al. 2007; Manickum and John 2014). Reliable evidence from literature regarding associated adverse environmental and health effects of emerging contaminants on human beings has been limited, and the effects of exposure to multifaceted combinations of these modulators remain poorly understood (Bruce et al. 2010; Schaider et al. 2014; WHO 2011). However, there have been documented effects of exposure in aquatic species to chemicals of emerging concern. Currently, it has been established that continuous exposure to endocrine disruptors might result in serious transgenerational health effects on humans and wildlife, if care is not taken (Dmitruk et al. 2008; Fatoki and Opeolu 2009; Ferraz et al. 2007). It is therefore of paramount importance to understand the sources, pathways and the associated risk of exposure so as to prevent short- and long-term health implications.

The threat posed by emerging micropollutants with endocrine disrupting activity in the environment were first articulated in Carson's (1962) work "The Silent Spring" (Carson 1962, 2002). That was followed by extensive research conducted in the Western world (Fatoki and Opeolu 2009; Matthiesen 2000). Ever since then, extensive research vis-a-viz formulation of an adequate regulatory framework has been carried out in the developed world. Today, several reviews and published articles have confirmed the presence of endocrine disrupting pharmaceuticals in the environment (Bu et al. 2013; Dalvie et al. 2014; Olujimi et al. 2010, 2012; Manickum and John 2014; Petrie et al. 2015; Sauvé and

Desrosiers 2014). This was also corroborated by the WHO/UNEP (2013) World Health Organization and United Nations Environmental Programme report of 2013 which states that a number of nations including developing countries are at risk of imminent complicated health challenges due to the occurrence of pharmaceutically active compounds in their water system (Bergman et al. 2013). Currently, large numbers of these contaminants have been reported in water sampled in USA, China, Germany, Canada, Brazil, Holland, including South Africa due to lack of standardised discharge limits (Focazio et al. 2008; Jin and Peldszus 2012; Padhye et al. 2014a; Swati et al. 2008).

Despite the Stockholm Convention known as the Persistent Organic Pollutants Treaty of 2001 and the Berlaymont Declaration of 2013 to protect humans and aquatic species from risks associated with exposure to these contaminants and despite different monitoring programmes in most advanced countries, the number of exogenous chemicals in the environmental is on the increase (Fatoki et al. 2010; Phillips et al. 2010; Whitworth et al. 2012). Considering the growing environmental problems due to the ubiquitous increase in the number of emerging contaminants coupled with the shortcomings associated with the conventional wastewater treatment plants globally. It is imperative to have a better understanding of their occurrence, environmental fate and exposure risk on humans and other species in view of the wide research gaps. Therefore, this review presents an overview on the state-of-art as regards chemicals of emerging concern such as pharmaceuticals, endocrine disrupting compounds, personal care products, nanomaterials and perfluorinated pollutants. Furthermore, emphasis is placed on the different types of chemicals of emerging concern, their sources, effects, environmental fate and exposure risk. Finally, proactive and precautionary measures that must be taken to protect the environment as well as guarantee the health and safety of humans and aquatic species are suggested.

Categories of chemicals of emerging concern

Different categories of chemicals containing products manufactured to meet human needs ranging from cleansing agents, pharmaceuticals (both prescribed and over the counter drugs), cosmetics, fragrances, and personal care products to mention but a few are widely used globally. Others include artificial sweeteners, insect repellents, pesticides, industrial additives, nanoparticles, synthetic hormones, perfluorinated compounds, flame retardants (Kolpin et al. 2002). Most of these chemical compounds are either unregulated or in the process of being regulated but have endocrine disrupting properties which have generated public health concerns. In Europe significant reductions in the level of emerging micropollutants in water have been reported as contained in the European Water Framework Directive (EC Directive 2000/06/EC) and daughter directive 2008/105/EC (EC 2008). Developing countries are yet to come to terms with the need to standardise their environmental regulatory framework on emerging micropollutants (Pomiès et al. 2013). Recent monitoring studies have confirmed the presence of these xenobiotics in different environmental samples in developing countries (Olujimi et al. 2012; Sorensen et al. 2015). Different classes of chemicals of emerging concern which include pharmaceuticals, personal care products, endocrine disrupting compounds, nanomaterials and perfluorinated pollutants are represented in Fig. 1.

Pharmaceuticals

Pharmaceuticals are any synthesised or natural chemical compounds designed to cure and prevent the spread of diseases as well as adding value to human and animal life (Maletz et al. 2013). According to Daghrir and Drogui (2013) pharmaceuticals are active substances given to animals to accelerate their feeding efficiency and growth rate. Pharmaceuticals have different chemical structure, behaviour, applications and metabolism in the human and animal body and hence the environment (Fawell and Ong 2012; Jiang et al. 2013). Pharmaceuticals are classified based on their therapeutic uses into the followings: antibiotics (ciprofloxacin), anti-diabetics (sulfonylurea), anti-epileptic (carbamazepine), antimicrobials (penicillins), anti-inflammatories and analgesics (ketoprofen, diclofenac), antiulcer and antihistamine drugs (ranitidine and famotidine), anti-anxiety/hypnotic agents (diazepam), lipid regulators (Clofibrate) to mention but a few (Esplugas et al. 2007; Jiang et al. 2013; Kanakaraju et al. 2014; Rivera-Utrilla et al. 2013). Mompelat et al. (2009) classified humans and animals pharmaceuticals and metabolites into 24 different classes, out of which 4 classes were most predominantly found in water, which includes non-steroidal anti-inflammatory drugs (NSAIDs), anticonvulsants, antibiotics and lipid regulators. Very recently, Bruce et al. (2010) and Rivera-Utrilla et al. (2013) independently categorised pharmaceutical into: anti-inflammatories and analgesics (paracetamol, ibuprofen), antidepressants (benzodiazine-pines), antiepileptics (carbamazepine), lipid-lowering drugs (fibrates), β-blockers (atenolol, metoprolol), antiulcer and antihistamines drugs (famotidine), anticancer drugs (cyclophosphamide, ifosfamide), antibiotics (tetracyclines), tranquilizers, antipyretics and stimulants. Van Doorslaer et al. (2014) reported that more than 5000 pharmaceuticals were synthesised and made available in the market for human and animal consumption. Currently, the global annual drug consumption figures is in the range

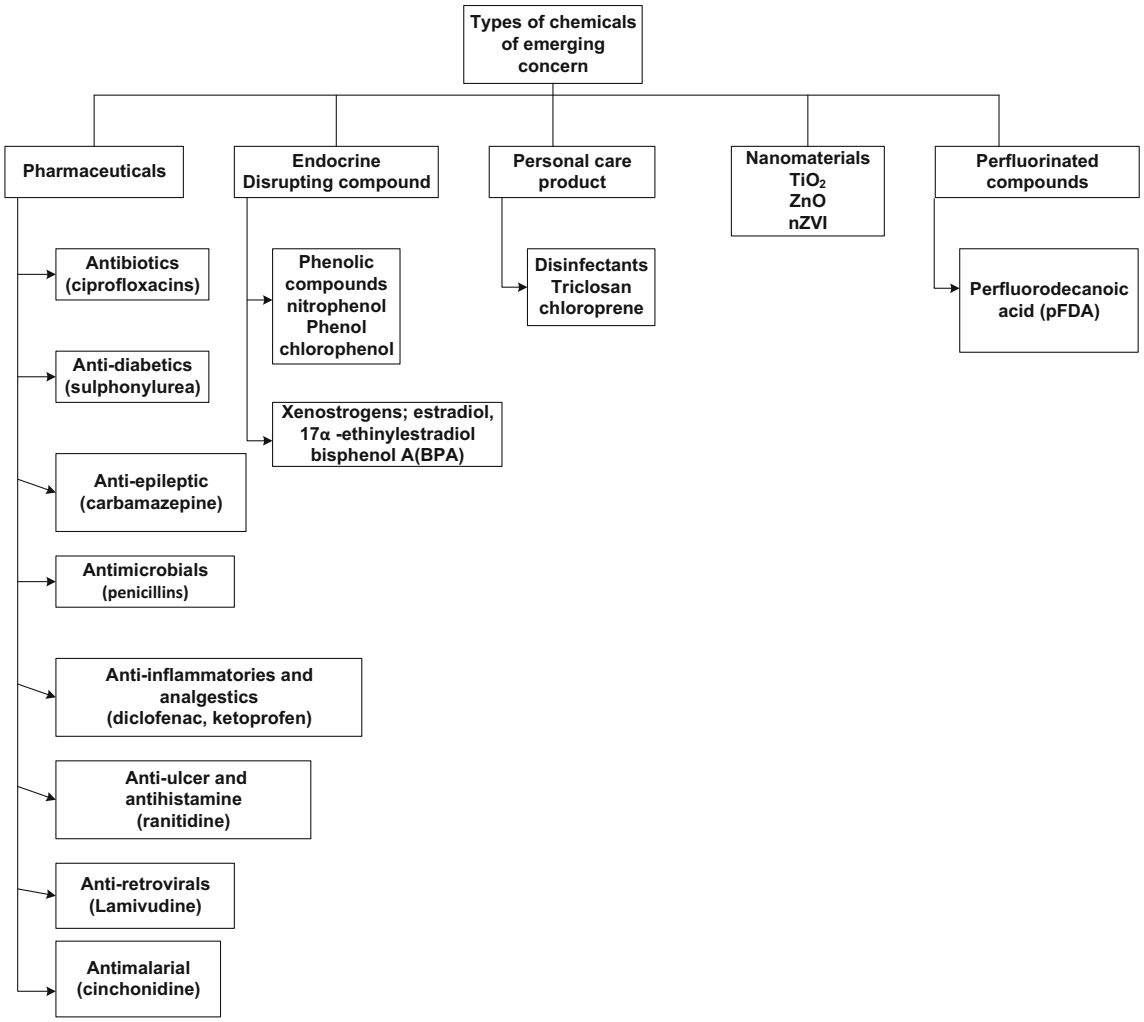

Fig. 1 Various class of emerging micropollutants in the environment (Fawell and Ong 2012; Jiang et al. 2013). Legend nZVI (nano zero valent iron particles)

of 100,000–200,000 tons with countries such as Brazil, Russia, India, China, and South Africa having greater proportion. Most of these drugs can be administered orally or by injection but due to their incomplete metabolism in humans and animals, part of the drugs may be excreted in urine or faeces and eventually end up in wastewater treatment plants. Due to growing utilisation of pharmaceuticals by human and animal, couple with non-complete assimilation in the body, original or partially metabolised drugs have been identified in the environment. These contaminants are stable and difficult to degrade by conventional wastewater treatment plants, thus escape the wastewater treatment plants into the environment (Baker and Kasprzyk-Horden 2013). Ever since early 2000s, there have been extensive research focusing on the detection of pharmaceuticals and pharmaceutical residue in water sources (Kanakaraju et al. 2014). Kleywegt et al. (2011) reported the detection of over 30 different pharmaceuticals in finished drinking water across the world. Exposure to pharmaceuticals and its metabolites via food or water may have short- and long-term health impacts on human and aquatic species depending on the dose and the durations (Daghrir and Drogui 2013). Some of the adverse effects on humans and other ecological species including disruptions of endocrine system, chronic toxicity and increase drug-resistant bacterial strains. For instance, Daghrir and Drogui (2013) reported that the exposure to tetracycline residue resulted in the slow growth of terrestrial and aquatic species, signifying that tetracycline antibiotics possessed endocrine disrupting properties. Thus, excessive consumption of tetracycline should be avoided. The main concern of the environmentalist is not about the acute toxic nature of the pharmaceuticals but rather their chronic toxicity on exposed organisms (Jiang et al. 2013). These pharmaceutically active compounds have been identified in different environmental water samples such as surface,

ground or wastewaters in China, USA, Holland, Spain, Germany, Canada, Brazil and even in South Africa (Chen et al. 2011; Rivera-Utrilla et al. 2013; Yan et al. 2014). In a related study, Matongo et al. (2015) investigated the presence of pharmaceuticals such as sulfamethazine, sulfamethoxazole, erythromycin, metronidazole, trimethoprim, acetaminophen, caffeine, carbamazepine, clozapine and ibuprofen in the Umgeni River, in the city of Durban in KwaZulu-Natal, South Africa. The authors found that clozapine had the highest concentration of 78.33 μg/L in the surface waters of Umgeni River while ibuprofen was found most in the sediment with a concentration of 62.0 μg/L. The authors ascribed the high level of these compounds to inefficiency on the part of the wastewater treatment processes as well as human activities. According to Hughes et al. (2013), over 200 pharmaceutically active substances have been identified in river waters with ciprofloxacin having maximum concentration of 6.5 mg/L. The concentration of detected pharmaceuticals in water is conspicuous; nevertheless, the quantified amounts differ from country to country depending on a number of critical factors such as the consumption pattern, or population size to mention but two. The fate and behaviour of most pharmaceuticals in the environment are also diverse and complex, and at the moment there is limited knowledge in the literature in this regard. Most pharmaceutical products are hydrophilic in nature, soluble in water, easily break down and have a short life span (Fent et al. 2006; Fent 2008), while some pharmaceuticals such as naproxen, or sulfamethoxazole can remain in the environment without degradation for more than a year. Clofibric acid, on the other hand, takes several years to decompose depending on the environmental media. Depending on the local industrial output, the age of wastewater treatment plant, rainfall pattern and other factors, a significant amount of the contaminants are conjugated and are found in the natural aquatic environment (Baker and Kasprzyk-Horden 2013). Pharmaceuticals enter wastewater treatment plants via multiple routes such as domestic sewage, septic tanks, landfill sites, industrial effluents, urban wastewater, agricultural practices, showering and bathing and eventually enter the water cycle as shown in Fig. 2 (Rodil et al. 2012). Others routes through which emerging contaminants get introduced into the environment including recreational activities, human excretion, direct discharge from pharmaceutical production plants (Daghrir and Drogui 2013).

Endocrine disrupting compounds

Endocrine systems comprise the endocrine glands, hormones and receptors that regulate the body's physiological activities such as reproductive processes including embryonic development, sex differentiation and metabolic development (Flint et al. 2012). Endocrine glands secrete hormones which circulate within the body through the blood stream. However, it has been found that a certain group of compounds mimic or disrupt endocrine glands from functioning properly. These compounds have been called endocrine disrupting compounds and are also known as endocrine disrupting chemicals, or endocrine disruptors, or endocrine modulators. Endocrine disrupting compounds can be natural or artificial chemicals; however, they interact with the oestrogenic receptors and enhance or inhibit the hormones from functioning properly (Jackson and Sutton 2008). Diamanti-Kandarakis et al. (2009) defined endocrine disruptors as compounds that interfere with the synthesis, secretion, transport, binding, action, or elimination of natural hormones in the body that are responsible for development, behaviour, fertility, and maintenance of homoeostasis (normal cell metabolism). These xenobiotics are present in most manufactured consumer products ranging from plastic bottles, children toys, cosmetics, toothpaste, detergent, polyvinylchloride pipes among others. Endocrine disrupting compounds cause adverse effects on aquatic organisms or their progeny via alteration of chemical messengers of the body or binding to receptors of the endocrine system at exposure levels up to a million times lower than carcinogen exposure levels of concern (Burger and Nel 2008; Olujimi et al. 2010). Cases of intersex alteration among alligators, frogs and fish upon exposure to endocrine disrupting chemicals are no longer disputable facts (Aneck-Hahn et al. 2009). Among the endocrine disrupting compounds that have attracted recent scientific attention due to their production output and consumption pattern include phthalates, phenolic compounds (bisphenol-A, nitrophenol, nonylphenol, alkylphenol, and chlorophenols), triclosan, ethinylestradiol, diethylstilbestrol, 17β-estradiol. More chemicals may be recognised as endocrine disrupters as the list of compounds increases due to unanticipated effects (Ferraz et al. 2007; Fatoki and Opeolu 2009). There are concerted efforts in the western world to provide an up to date list and regulation of persistent emerging contaminants (Pomiès et al. 2013). Despite the fact that there is little epidemiological data regarding the impact of endocrine disruptors exposure on human health, there are increasing incidences of adverse effects such as abnormal sperm count among male and females, high rates of infertility, accelerated ovarian, prostate, testicular and breast cancer among humans. Exposure to endocrine disrupting compounds has been reported to cause immune deficiency, neurological effects, impairment of intellectual and childhood development and psychological effects. The absence of a precautionary principle and regulated monitoring is responsible for the increasing concentration of micro-pollutants in the

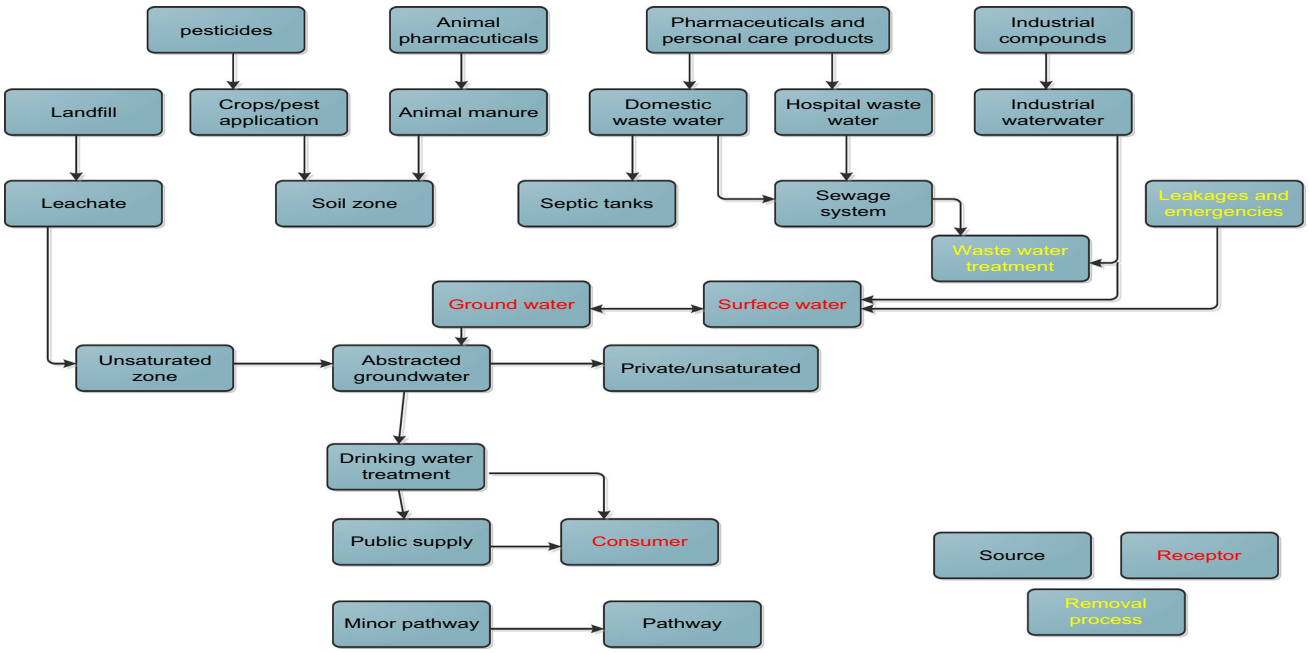

Fig. 2 Potential sources and pathways of some emerging micropollutants to receptors and aquatic environment (*Sources*: Stuart et al. 2012)

environment and has given rise to increasing public concern over the presence of endocrine disrupting compounds in drinking water. Proper identification, quantification as well as effective treatment strategies will be needed to remove these compounds from the water system.

Mechanism of endocrine disruption within the body

The mechanism of endocrine disrupting pharmaceuticals on the hormonal system of the body is a complex phenomenon and difficult to predict since compounds exist as mixtures. However, a review of various studies reveal that disruption can take place via multiple routes listed below:

- The contaminants can bind to the nuclear and hormonal receptor cells or sometimes block or mimic the chemical messengers in the body and cause considerable adverse ecological health effects. Endocrine disruptors sometimes alter the response activity of genes. For instance, bisphenol-A can bind to oestrogen receptor β based on its lower affinity compared to estradiol (Rogers et al. 2013).
- The concentrations or level of the chemical messengers/hormones in the body may be affected via alteration of their metabolic or synthetic pathways (Olujimi et al. 2010).
- Interference with the hormonal-controlled physiological signals responsible for body homoeostasis and development is often evident.
- Lastly, modification or modulation of certain numbers of chemical messenger receptor within the body cell responsible for the immune system.

Personal care products

Personal care products are made up of a large group of active and inert substances including prescribed and non-prescribed pharmaceuticals utilised by people and animals (Jiang et al. 2013). They include analgesics, synthetic hormones, sun screens, insect repellent, cosmetics, fragrances, preservatives, shampoos and toiletries. Unlike pharmaceuticals that are ingested, personal care products are applied directly on the human body to change appearance, taste and odour. Personal care products are categorised into polycyclic musks and parabens used to inhibit bacterial decay (Fawell and Ong 2012; Houtman 2010). Furthermore, disinfectants such as triclosan and chloroprene have been used industrially in the manufacture of consumer products such as air-fresheners, hand soap, toothpaste, sportswear, plastics, toys, lotions, medical disinfectants and mouthwash (Fawell and Ong 2012). In the same vein, benzophenone and alkylated siloxanes are incorporated in sun-screen lotions and hair-care products. The micropollutants enter the aquatic environments including surface water through recreational activities such as swimming and also via showering and bathing as well as other technological process (Larsson et al. 2007; Kasprzyk-

Hordern et al. 2009; Rahman et al. 2009). Over the years, a substantial number of personal care products and their transformed products have been identified in wastewater treatment plants. Some of these metabolites get converted into harmless inorganic compounds such as carbon dioxide and water within a wastewater treatment plants or are partially adsorbed onto sedimentation sludge due to their lipophilic nature and non-biodegradability or adhere to other hydrophilic components (Jiang et al. 2013). Others escape the wastewater treatment plants and are more persistent in the environment either in their original or metabolised form. Houtman (2010) reported that triclosan and chloroprene accumulate in the bile from bream in the Dutch River Dommel, Netherlands. Lastly, most endocrine disrupting compounds, pharmaceuticals and personal care products have been shown to disrupt endocrine systems, yet they are still unregulated and are carelessly discharged into the immediate environment, especially in developing countries where there is no stringent regulatory frame work.

Sources and effects of chemicals of emerging concern

Chemicals of emerging concern enter the environment via multiple point and non-point sources such as pharmaceutical industries, mining activities, hospitals, and health service centres, or agricultural practices (Agunbiade and Moodley 2014). In metropolitan areas, chemicals of emerging concern could also be introduced into the environment through sewer overflows, run-off from farmland, disposal of animal waste and septic tank effluents. Other routes through which chemicals of emerging concern enter the environment include household use and disposal of personal care products, cleaning agents, prescribed, illicit and unused drugs into septic tanks or sewerage systems as well as defecation of partially metabolised drugs by humans and animals (Swartz et al. 2006; Labadie et al. 2007; Dougherty et al. 2010). Depending on their level of persistency, most compounds pass unchanged through waste water treatment plants and enter the terrestrial or aquatic environment (Barnes et al. 2002; Pryor et al. 2002; Harrison et al. 2006). Thousands of these compounds and their metabolites have been detected in the aquatic environment and the metabolites in most cases are even more toxic than the original compounds (Daghrir and Drogui 2013). Sometimes they exerted synergistic effects on the target organisms. Thus, the aquatic environment is acting as a sink and dumping ground for most contaminants. Published research has supported the claim that current wastewater treatment plants and untreated urban wastewater discharges are not specifically designed for removal of these compounds. This is because the molecular structure of these compounds are complex. Another challenge is due to their low concentration in water. Specifically, wastewater treatment plant effluents are considered to be an important route through which endocrine disrupting pharmaceuticals enter the environment. As a result of only partial elimination in the treatment plants, most endocrine modulators and residual pharmaceuticals are frequently encountered in the solids and water environmental compartments at low concentrations (ng/L to μg/L). Table 1 and Fig. 2 summarises the potential sources of chemicals of emerging concern in the environment. Other sources of emerging contaminants not included in Fig. 2 according to Heim and Schwarzbauer (2013) include diagenetic activities in the sediments, atmospheric deposition, and groundwater recharge. The multitude of persistent micropollutants present in the aquatic environment have become a major source of negative impacts upon living organisms ranging from increased feminisation of male, or masculinizing effects upon female species, bacterial resistance to antibiotics, birth defects, long-term toxicological effects, prostate cancer, thyroid and other cancers (Marcoux et al. 2013). While the causes of these health challenges are not clearly understood, there is growing evidence that most detected micropollutants might play a crucial role and perhaps be responsible for the occurrence of intersex fish, cancers and low sperm count in species found in the aquatic environment (Shaw 2011). Thus, there is need for global sensitisation regarding the pathways and the health effects associated with exposure to these xenobiotics and most environmental regulatory bodies are trying to establish minimum discharge limits (Agunbiade and Moodley 2014). The health effects associated with exposure to the individual endocrine disrupting compounds and pharmaceuticals, personal care products are described in Table 2. The effects of exposure to endocrine modulators, pharmaceuticals or personal cares may be temporary, permanent or even transgenerational depending on the exposure dose and time. While the health effects on aquatic species have been widely reported, there are diverse opinions regarding the direct impact of environmental endocrine disrupting chemicals on human health. However, several investigations have suggested that the negative health effects of exposure of aquatic species to endocrine disruptors may include low sperm count, reduced fertility and reproductive malfunctions (Andersson et al. 2007; Bolong et al. 2009). While the health effects on human beings is still a subject of debate and needs further investigation, low sperm count, reduced fertility and reproductive malfunctions is on the increase in humans these days. There is a growing understanding that most detected micropollutants might play a crucial role and perhaps be responsible for these manifestations. However, the observed decrease in the sperm count varies from region to region. As a result of

Table 1 Sources of different emerging micropollutants and important classes in the aquatic environment

Category	Important classes	Major sources (distinct)	Major sources nonexclusive
Pharmaceuticals	Nonsteroidal anti-inflammatory drugs (NSAIDs), lipid regulator, anticonvulsants, antibiotics, β-blockers and stimulants	Domestic wastewater (from excretion), hospital effluents	Sources that are not exclusive to individual categories includes: industrial wastewater (from product manufacturing discharges) landfill leachate (from improper disposal of used, defective or expired items)
Personal care products	Fragrances, disinfectants, UV filters, and insect repellents (triclosan)	Domestic wastewater (from bathing, shaving, spraying, swimming)	
Steroid hormones	Oestrogens	Domestic wastewater (from excretion), run-off from CAFOs and aquaculture	
Surfactants	Non-ionic surfactants	Domestic wastewater (from bathing, laundry, dishwashing and etc.), industrial wastewater (from industrial cleaning discharges)	
Industrial chemicals	Plasticisers, fire retardants (bisphenol-A; phthalates	Domestic wastewater (from leaching out of the material)	
Pesticides	Insecticides, pesticides and fungicides	Domestic wastewater (from improper cleaning, run-off from gardens, lawns and roadways and etc., agricultural runoff	

CAFOS Concentrated animal feeding operations *Sources* (Luo et al. 2014)

Table 2 Effects of different endocrine disrupting compounds in the humans and aquatic species

Endocrine disrupting compounds	Health effects due to exposure	References
Bisphenol-A used in epoxy resin and polycarbonate plastics (in food and drink packaging)	Proven to have oestrogenic effects in rats and hormonal effects which increase breast cancer risk in human reported to act as anti-androgen, which causes feminising side-effects in men	Rogers et al. (2013)
Phthalates—used as plasticisers in plastic, polyvinylchloride baby toys, flooring, pesticides	Exposure to high levels reported to cause miscarriage and pregnancy complication	Staples et al. (1997), Liang et al. (2008)
Disinfectants/antiseptics, i.e. triclosan—used in toothpaste, hand soaps, acne cream)	Found in receiving waters, linked to toxic, biocidal effects (killing helpful microorganisms) and bacterial resistance development towards triclosan	Kookana et al. (2011)
Polychlorinated biphenyls—used in electrical equipment (capacitors and transformers)	The metabolites mimic estradiol (female hormone), and cause carcinogenic changes. Exposure was reported to cause delayed brain development and IQ decrease in children	Jacobson and Jacobson (1997), Routledge et al. (1998)
Estrone and 17-β estradiol (steroidal oestrogens) and 17-α ethynylestradiol (synthetic contraceptive)—contained in contraceptive pills	Cause feminisation which observed for fish in sewage treatment. The discharge causes mimicking oestrogen/hormone effect to non-target	Witte (1998)
Antibiotics (such as penicillin, sulphonamides, tetracyclines)	Shown to cause resistance among bacterial pathogens, that lead to altered microbial community structure in nature and affect higher food chain	Daughton and Ternes (1999)
Fragrances (musk)	Musk xylol—proved carcinogenic in a rodent bioassay and significantly absorbed through human skin, Musk ambrette may damage the nervous system	Bronaugh et al. (1998)
Preservatives, i.e., parabens (alkyl-hydroxybenzoate)—used for anti-microbiological preservatives in cosmetics, toiletries and even foods	Shows weak oestrogenic activity	Routledge et al. (1998)
Disinfectants/antiseptics, i.e., triclosan—used in toothpaste, hand soaps, acne cream)	Shown to cause toxic, biocidal effects and also cause bacterial resistance development towards triclosan	Okumura and Nishikawa (1996), McMurry et al. (1998)

Sources (Adopted from Bolong et al. 2009)

different health challenges associated with exposure to endocrine disruptors and other chemicals, WHO/UNEP (2013) World Health Organization and United Nations Environmental Programme in one of their studies jointly called for proper understanding of the links between health risks and the exposure dose or route for individual as well as multiple endocrine disrupting compounds. For instance, triclosan, bisphenol-A and phthalates are essential industrial chemical components incorporated into multiple commercial household products and as a result are widely detected in different environmental matrices. Indeed, exposure to triclosan has been linked to an increase in antibiotic resistance and thyroid malfunction with increasing medical costs globally. Thus, the occurrence and behaviour of endocrine disrupting pharmaceutical compounds in the aquatic environment remains a complicated issue (Zhou et al. 2012), as pollutants infiltrate the aquatic environment through different sources and various pathways including: agricultural run-off, household discharge, industrial, sewage and municipal wastewater as shown in Fig. 2. Since wastewater treatment plants are not designed for effective removal of these xenobiotics, original compounds or metabolites have been identified in water sources at nanogram per litre and microgram per litre. However, the pathways by which most pollutants penetrate from different sources to the human or animal receptor remain unclear as this depends on the physical and chemical properties of the micro-pollutants. Daghrir and Drogui (2013) submitted that in view of the increasing concentration and accumulation of emerging persistent organic pollutants in the environment, potential adverse effects on animals or perhaps humans would continue to rise unless adequate measures are taken. The authors called for cautions in the usage of manufactured products containing emerging chemicals and suggested effective coordination strategies among the different regulatory bodies to curb abuse or misuse of the manufactured products by consumers.

Chemicals of emerging concern in the environment

It is well established that emerging micropollutants with endocrine disrupting and bio-magnification properties have been identified in global water cycle including drinking water (Aneck-Hahn et al. 2009; Petrie et al. 2015). This has raised concerns regarding water quality considering the current pollution status. The level of these xenobiotics in the environment vary from country to country and mostly depends on the levels of industrial activity, consumption patterns, compliance with existing regulatory frameworks, population growth rate, efficacy of wastewater treatment plants' performance, among others. This section focuses on reviewing different case studies of emerging contaminants that have been identified and quantified in different countries. Bu et al. (2013) detected over 100 pharmaceuticals and personal care products at μg/L and ng/g level spread across different strata of the natural and aquatic environment in Eastern China. According to the screening level risk assessment, six out of the several pollutants namely erythromycin, roxithromycin, diclofenac, salicylic acid, ibuprofen, and sulfamethoxazole were classified as priority pollutants. Duong et al. (2014) reported the presence of approximately 940 micro-organic pollutants in the sediments collected in rivers located in Vietnam, Japan using gas chromatography—mass spectrometry technique. The authors found that sediments collected from metropolitan areas precisely Hanoi and Ho Chi Minh City were mostly contaminated with high concentration of phthalates, sterols, polyaromatic hydrocarbons, pyrethroids, and deltamethrin. The presence of these pollutants was attributed to run-off of petroleum products and vehicular emissions. Rodil et al. (2012) demonstrated the occurrence of 53 different compounds in wastewater, surface and drinking water collected in the North West region of Spain. The solid-phase extraction procedure followed by liquid chromatography-electrospray-tandem mass spectrometry (LC–ESI–MS/MS) was adopted for the extraction and quantification of the compounds. Among the studied pollutants, salicylic acid, ibuprofen, benzophenone-4 were identified as compounds with concentration >1 μg/L. López-Serna et al. (2013) investigated the occurrence over 72 pharmaceuticals and 23 pharmaceuticals residue in selected groundwater in Barcelona, Spain. Solid-phase extraction followed by liquid chromatography-electrospray ionisation-tandem mass spectrometry was used to identify and quantify the contaminants. Antibiotics namely erythromycin, sulfadiazine, sulfamethazole, tetracycline, tylosin, ciprofloxacin remained predominant in 10 out of the 13 sampling sites with a total concentration greater than 1000 ng/L. Kleywegt et al. (2011) investigated the occurrence of 48 emerging contaminants in untreated water source and finished drinking water in Ontario, Canada, using solid-phase extraction and liquid chromatography-mass spectroscopy technique. It was found that 27 of these contaminants were identified in both the source water and the finished drinking water. Of all the identified compounds, carbamazepine (749, 601 ng/L), gemfibrozil (9, 4 ng/L), ibuprofen (79, 25 ng/L), bisphenol-A (87, 99 ng/L), remained predominant in source and finished drinking water, while roxithromycin (155 ng/L) and enrofloxacin (13 ng/L) were detected in the environmental samples for the first time. Valcárcel et al. (2011) investigated the presence of 33 pharmaceutically active compounds in the Rivers and tap water sample collected at different points in Madrid, Spain. Out of the 33 compounds, 25 pharmaceuticals and metabolites were quantified in ten different sampling points. Carbamazepine, caffeine, cotinine,

ifosfamide, venlafaxine were mostly detected at higher concentrations in the surface water. According to the authors, continuous exposure to these compounds portends short- and long-term health risk. Jiang et al. (2014) utilised solid-phase extraction followed by liquid chromatography coupled with tandem mass spectrometry to detect and quantify 31 different emerging contaminants in the coastal waters of Taiwan. Acetaminophen, ibuprofen, ketoprofen, codeine, ampicillin, erythromycin, cephalexin, ketamine, pseudoephedrine, caffeine, carbamazepine, and gemfibrozil were detected in the median concentration range of 1.47–156 ng/L. Sorensen et al. (2015) detected *N*, *N*-Diethyl-m-toluamide, triclosan and trihalomethane at a concentration of 1.8, 0.3 and 50 μg/L, respectively, in the groundwater sources in Kabwe, Zambia. The authors attributed the presence of these contaminants to the absence of well protection, poor sanitation and household disposal of solid or liquid waste. Padhye et al. (2014a) investigated the occurrence and removal of 30 representative emerging micropollutants in an urban drinking water treatment plant located in the South East United States. *N*, *N*-Diethyl-m-toluamide and nonylphenol were mostly found in the water while triclosan, bisphenol-A, ibuprofen, atrazine and caffeine were detected at low concentration. The average concentration of the pharmaceuticals and endocrine disruptors in the studied water was 360 ng/L. Padhye and coworkers established a correlation between the pharmaceuticals and the endocrine disrupting chemicals in surface and drinking water within the studied period. Similarly, Peng et al. (2008) reported the occurrence of endocrine disrupting chemicals in three urban streams and a major river at Guangzhou, South China. The authors detected high concentration of nonylphenol, bisphenol-A, triclosan and 2-phenylphenol. Salicylic acid, clofibric and ibuprofen were detected in most water samples with concentration of 2098, 248 and 1419 ng/L, respectively, and naproxen was not highly prevalent. In the same vein, Jonkers et al. (2009) studied the occurrence and behaviour of bisphenol-A, phenylphenol and parabens in municipal wastewaters in the river Glatt waters near Zurich. The authors recommended continuous chemical monitoring of the water sources as a means of determining the fates and behaviour of the studied compounds. Kim et al. (2007) reported the presence of different classes of emerging contaminants in South Korea surface and drinking waters. Specifically, hormones, pharmaceuticals, flame retardants, antibiotics, and personal care products were identified in wastewater and drinking water, respectively. Hass et al. (2012) examined the presence of primidone, phenobarbital, oxazepam, diazepam, meprobamate, pyrithyldione and phenylethylmalonamide in effluents from WWTPs, surface water, groundwater including final drinking water, respectively. Primidone and phenylethylmalonamide were largely presented in all the water samples with concentrations of 0.87 and 0.42 μg/L, respectively, while phenobarbital (0.96 μg/L), oxazepam (0.18 μg/L), pyrithyldione (0.04 μg/L), meprobamate (0.50 μg/L) were identified virtually in all the water samples apart from raw and finished drinking water. On the other hand, diazepam was not found in any of the water samples possibly due to its complete withdrawer from German market. Besides, China, Germany, USA, Canada, South Korea, Japan, emerging contaminants have been found in developing country such as South Africa. A study has established that aquatic species such as fish and other amphibians were affected upon exposure to 17β-estradiol (Burger and Moolman, 2006). Slabbert et al. (2005) discovered that different surface waters and effluents tested in the Gauteng Province presented high oestrogenic activity. Olujimi et al. (2010) submitted that South Africa waters contained high concentration of oestrogens such as 17β-estradiol. Similarly, Barnhoorn et al. (2004) revealed high concentration of some endocrine disruptors in fat tissue of catfish in South Africa. These reports verify that endocrine disrupting compounds are present in South African environments in different proportions depending on the use and consumption patterns. Burger and Moolman (2006) stated that the occurrence of endocrine disrupting compounds in South African water systems may have a long-term cumulative health impact on its citizenry. In an attempt to better understand the pollution status of emerging contaminants in South Africa water sources, Burger and Moolman (2006) conducted a surveillance study of these sites; Makhathini flats, Vaal River Barage, Hartbeespoort and Rietvlei Dam in South Africa where significant endocrine disruptors activity was established. Fatoki et al. (2010) showed that the concentration of phthalates such as dibutylphthalates in water samples ranged between 0.16 and 10.17 mg/L in rivers and dams in the Venda region. The latter value surpassed the 3 μg/l maximum recommended by the United States Environmental Protection Agency (USEPA) for the survival of fish and other aquatic species. Olujimi et al. (2012) investigated the presence of eleven priority phenols and six phthalate esters in five selected wastewater treatment plants and freshwater systems in Cape Town. The study established that certain numbers of the wastewater treatment plant accounted for the highest concentrations of dibutylphthalates compared to other treatment plants. Phenol, 2-chlorophenol and PCP were equally detected, though at low concentrations. Manickum and John (2014) quantified the level of steroid hormones such as 17-β-estradiol (E2), estrone (E1), estriol (E3), synthetic oestrogen (17-β-ethinylestradiol (EE2), testosterone and progesterone in Pietermaritzburg wastewater treatment plants, South Africa using the non-analytical enzyme-linked immunosorbent Assay (ELISA) technique. The authors reported that close to 92 % of EDCs were eliminated by activated wastewater treatment plants, while

the residual 8 % found their way into the environment. The authors ascribed different levels of pollutants to seasonal variation, different rainfall patterns, raw wastewater effluent flow rates, and activated sludge performance capacity among others. The authors recommended further treatment of the water and promulgation of effective legislation with respect to maximum allowable levels of these compounds in water matrices. Similarly, the scoping studies conducted by Patterton (2013) on the status of South African drinking water sampled mostly from Johannesburg, Pretoria, Bloemfontein, Durban and Pietermaritzburg, Cape Town and Port Elizabeth over a period of four season revealed the presence of high concentration of pesticides such as atrazine, terbuthylazine as well as pharmaceuticals (carbamazepine). Other compounds such as hexazinone, phenytoin, and tebuthiuron (Durban), telmisartan, simazine, oxadixyl, metolachlor, amphetamine imidacloprid tebuthiuron (Johannesburg), and fluconazole, phenytoin and tebuthiuron (Bloemfontein) were also quantified during the last three season. Differences in concentrations were ascribed to seasonal variation and dilution factors. The authors identified agricultural run-off, medical waste and pesticides, leaching of pharmaceuticals and pesticides into groundwater reservoirs as possible pathways through which the contaminants enter the drinking water. The results also indicated that the levels of the contaminants in drinking water were generally below the concentrations that could trigger or raise serious health issues (Fawell and Ong 2012). However, the annual or daily exposure rate was not calculated. Osunmakinde et al. (2013) categorised most pharmaceuticals identified in the Daspoort WWTP, Pretoria, water environment into six categories such as hypertension, analgesics, antiretroviral, antibiotics, vitamins and antidiabetic drugs. The results of their investigations revealed the presence of ribavirin, pindolol, famciclovir, carbamazepine, ketoprofen, fenoprofen and ibuprofen in various concentrations in effluents from WWTPs. Besides, the pharmaceuticals, bisphenol-A, Estrone (E1), 17β-Estradiol (E2), Estriol (E3), 17α-ethinylestradiol (17α-EE2) and ethinylestradiol were also detected in the wastewater. The authors revealed that carbamazepine and bisphenol-A remained the most prescribed drug and used industrial chemical, respectively identified in the investigated wastewater effluent. Currently the list of persistent emerging organic pollutants in South Africa is still growing owing to population growth, industrial activities and absence of monitoring of standard regulatory and discharge limits. Very recently, Agunbiade and Moodley (2014) reported the presence of caffeine, nalidixic acid, atenolol and acetaminophen predominantly at very high concentrations in the estuary mouth and lagoon of the Umgeni River water in KwaZulu-Natal. These studies show that the South African water system is exposed and acts as a sink for persistent environmental contaminants as well as their transformation products. And since these rivers serve for drinking purposes and for indirect re-use as potable water, there are anticipated concerns that continuous consumption of such water could lead to undesirable toxicological health effects among citizens in the near future, unless proactive measures are employed. Thus, these compounds must be quantitatively eliminated within treatment plants before ultimate discharge into rivers and lakes used for drinking water purposes. Because the wastewater is discharged into rivers or dams that are sources of drinking water, it constitutes indirect potable reuse. Other case studies involving chemicals of emerging concern detected in different countries (South Africa inclusive) are indicated in Table 3. A gap analysis of literature reviewed showed that little information exists on the identification of intermediate compounds which are considered more toxic than the parent compounds and even less on the availability of treatment technologies for the decomposition and eventual elimination of these micropollutants from water. Equally, the eco-toxicity and chronic effect of the transformation products evolved during treatment including their mode of action on the target organisms have not been fully established. According to Table 3, it is obvious that the concentrations of chemicals of emerging concern in different environmental samples contrasts from country to country, region to region, which can be attributed to several factors such as population figures, demands level, consumption habit, wastewater treatment plants capacity and performance, agricultural activities and water utilisation rate. A deeper understanding of the toxicity and ecotoxicity with respect to the exposure rate to endocrine disrupting compounds is indispensable to predict possible risk patterns of micropollutant infiltration. A follow-up publication to address these issues, ranging from detection, treatment techniques involving advance treatment process to toxicity will be presented subsequently by the authors

Fate of chemicals of emerging concern in the environment

The study of emerging micropollutants has dominated the public and scientific discussion in the last 15 years due to their growing accumulation in the environment. The discharge of pharmaceutically active compounds such as pharmaceuticals, endocrine disrupting compounds, personal care products into the environment may perhaps continue in the view of the growing human population figures and demands for more manufactured products. The continuous identification and detection of these compounds in water sources particularly drinking water have raised serious concerns regarding the fate and transport of these contaminants among the water experts and consumers.

Table 3 Occurrence of chemicals of emerging concern in the environment showing the types of water, countries, compounds detected, analytical detection techniques and concentration range

Type of water	Countries	Compounds detected	Analytical method of detection	Concentration range (ng/L)	References
Industrial water in Pretoria	South Africa	p-nonylphenol, bisphenol-A, phthalate esters, and others	Recombinant yeast cell bioassay and GC–MS method were used	Oestrogenic activity was detected in all the samples collected from these sites while phthalate esters were detected at some of the other sites	Mahomed et al. (2008)
Mbokodweni River water	South Africa	Triclosan and ketoprofen	Solid-phase extraction followed by high performance liquid chromatography	Triclosan and ketoprofen were detected in all wastewater (influent and effluent) samples at a range of 1.2–9.0 μg/L and in some river water samples	Madikizela et al. (2014)
River and dam water in Venda	South Africa	dimethyl phthalate (DMP), diethyl phthalate (DEP), dibutyl phthalate (DBP), di-2-ethylhexyl phthalate (DEHP)	Liquid–liquid extraction, followed by column chromatographic clean-up and capillary gas chromatography coupled with FDI detector were used	The levels of phthalate esters reported in this study for the water in rivers and dams ranged from 0.16 to 10.17 mg/L and varied between 0.02 and 0.89 mg/kg in sediments for DBP and DEHP respectively. The obtained values are higher than the US Environmental Protection Agency (USEPA) criterion of 3 μg/L for the protection of fish and aquatic life in rivers	Fatoki et al. (2010)
Wastewater effluent	South Korea	The following compounds were atenolol, iopromide, (tris(chloroisopropyl) phosphate (TCPP), (tris(2-chloroethyl)phosphate (TCEP), musk ketone, naproxen, (*N*,*N*-diethyl-meta-toluamide), (DEET), carbamazepine, trimethoprim, sulfamethoxazole, and benzophenone	Liquid chromatography coupled with tandem mass spectrometry (LC–MS/MS) and electrospray ionisation (ESI) and atmospheric pressure chemical ionisation (APCI) were used	The average concentrations of all the identified compounds were in the range of 98–663 ng/L. On the other hand, steroid hormones, atrazine and octylphenol were not detected in any of the samples	Ryu et al. (2011)
Raw waters	Spain	Phenytoin, atenolol and hydrochlorothiazide, sotalol and carbamazepine Epoxide were frequently and most found in the finished drinking water	Solid-phase extraction procedure followed by ultra-performance liquid chromatography (UPLC) system containing turbo Ion spray source	The concentration of Phenytoin, atenolol and hydrochlorothiazide was close to 10 ng/L while sotalol and carbamazepine epoxide was >2 ng/L. Above all, the removal rate of the five compounds were above 95 %	Huerta-Fontela et al. (2011)
Surface waters	South Korea	Iopromide, atenolol, tris(chloroisopropyl (TCPP), (tris(2-chloroethyl)phosphate (TECP), musk ketone, naproxen, (*N*,*N*-diethyl-meta-toluamide), (DEET), carbamazepine, caffeine, and benzophenone	Solid-phase extraction followed by liquid chromatography with tandem mass spectroscopy (MS/MS) containing electrospray ionisation (ESI)	The average concentrations of the identified pharmaceuticals and metabolites both in the river and creek samples ranged between 56 and 1013 ng/L and 102 and 3745 ng/L respectively	Yoon et al. (2010)
Groundwater	USA	Acetaminophen, caffeine, carbamazepine, codeine, p-xanthine (a caffeine metabolites), sulfamethoxazole and trimethoprim	Solid-phase extraction followed by high performance liquid chromatography coupled with mass spectrometer	The concentrations of the pharmaceuticals in the groundwater: acetaminophen (1.89 μg/L), caffeine (0.29 μg/L), carbamazepine (0.42 μg/L), codeine (0.214 μg/L), p-xanthine (0.12 μg/L), sulfamethoxazole, (0.17 μg/L) and trimethoprim (0.018 μg/L)	Fram and Belitz (2011)
Groundwater	USA	Sulfamethoxazole, perfluorooctane sulphonate, phenytoin	Solid-phase extraction followed by high performance liquid chromatography coupled with mass spectrometer	Maximum concentration of sulfamethoxazole was (113 ng/L) and phenytoin (66 ng/L) while perfluorooctane was ((97 ng/L)	Schaider et al. (2014)

Table 3 continued

Type of water	Countries	Compounds detected	Analytical method of detection	Concentration range (ng/L)	References
Natural and drinking water	USA	Sixteen chemicals of emerging concern which belongs to antibiotics, hormones, analgesics, stimulants, antiepileptics, and X-ray contrast media	Solid-phase extraction (SPE) of water samples, followed by liquid chromatography coupled with tandem mass spectrometry (LC–MS/MS)	Caffeine, ibuprofen, and acetaminophen were predominately found in the water samples with concentrations of 224, 77.2, and 70 ng/L, respectively. Higher concentrations were obtained during winter as compared to summer due to low precipitation during winter months	Wang et al. (2011)
Rivers and dams in Eastern Cape	South Africa	Dimethyl phthalates (DMP), diethyl (DEP), dibutyl phthalates (DBP) and diethylhexyl phthalates (DEHP)	Solid-phase extraction followed by capillary GLC	The results showed that the level of dimethyl (DMP), diethyl (DEP), dibutyl (DBP) and diethylhexyl (DEHP) were in the range of 0.03–2306 ± 9.4 μg L^{-1}. Among the investigated phthalates ester, DEHP remained the most predominant in both the harbour and freshwater samples, which raises concern for aquatic species because the reported value for rivers is greater than the USEPA water criteria of 3 μg L^{-1}	Fatoki and Noma (2002)
Wastewater influent and effluent from Diepsloot, Johannesburg	South Africa	Naproxen, ibuprofen, and triclosan	Polar organic chemical integrative sampler followed by high performance liquid chromatography (HPLC) system coupled with ultraviolet (UV) and fluorescence (FLD) detectors	The concentrations of the three pollutants in wastewater influent ranged from 55.0 to 78.4 μg/L in Goudkoppies and 52.3 to 127.7 μg/L in Northern WWTPs. While in the treated effluent, the concentration ranged from 10.7 to 13.5 μg/L and 20.4 to 24.6 μg/L in Goudkoppies and Northern WWTPs respectively. The concentrations of the studied compounds in both treatment plants did not exceed 127.7 μg/L	Amdany et al. (2014)
Jukskei River catchment	South Africa	Dimethyl phthalate (DMP), and diethylhexyl phthalates (DEHP), diethyl phthalate (DEP), dibutyl phthalate (DBP)	Liquid–liquid extraction and soxhlet extraction	The level of phthalates in unfiltered and filtered water samples ranged from 0.04 (±0.00) ng mL^{-1}, (DMP), 9.76 (±0.1) ng mL^{-1} (DEHP), 0.09 (±0.01) ng mL^{-1} DMP, 4.38 (±0.06) ng mL^{-1} (DEHP) respectively. The concentration of phthalates in sediment were 0.05 (0.00) (DMP) and 4910 (0.36) ng/g (DEHP). The obtained values were below water quality guideline of United States Environmental Protection Agency (USEPA)	Sibali et al. (2013)
Surface water	China	15 different Phthalates	Solid-phase extraction followed GC–MS analysis	The concentration of 15 studied phthalates was between 355.8 and 9226.5 ng/L, with the mean value of 2943.1 ng/L. DBP and DEHP were more prominent. The PAE concentrations ranged from 52.5 to 4498.2 ng/L and 128.9 to 6570.9 ng/L	Liu et al. (2013)

Table 3 continued

Type of water	Countries	Compounds detected	Analytical method of detection	Concentration range (ng/L)	References
Wastewater and wastewater sludge	USA	Bisphenol-A	Solid-phase extraction followed by LC–MS/MS	BPA concentration in wastewater range from 0.07 to 1.68 μg/L while the wastewater sludge contain 0.104–0.312 μg/g	Mohapatra et al. (2011)
Surface and underground water	China	Dimethyl phthalates (DMP), Diethylphthalates (DEP), di-*n*-butyl phthalates (DBP), butyl benzyl phthalates(BBP), diethylhexyl phthalates (DEHP), DnOP	Solid-phase extraction followed by gas chromatography-mass spectroscopy	The concentrations of six PAE compounds in groundwater and surface water range from 6.7 to 33.8 ng/L, with an average value of 0.9 and 11.1 ng/L, respectively. Di (2 ethylhexyl) phthalate (DEHP) and di-butyl phthalate (DBP) were mostly detected	Huang et al. (2012)
Surface water, drinking water and plasma	Malaysia	Bisphenol-A	Solid-phase extraction followed by GC–MS	The concentration of bisphenol-A in tap water ranged from 3.5 to 59.8 ng/L with the highest concentration found in tap water connected to PVC pipes and water filter devices. The bottled mineral water had lower levels of BPA (3.3 ± 2.6 ng/L). Only 17 % of the plasma samples contained BPA with concentration ranges from 0.81 to 3.65 ng/mL	Santhi et al. (2012)
Waste, river and sea water	China	Triclosan	Solid-phase extraction followed with gas chromatography–ion trap mass spectrometry	The concentration of Triclosan in water sources were as follows: Sea water: 16.2 ± 1.3., 99.3 ± 10.6., 31.9 ± 2.1 ng/L, River water: 37.6 ± 3.8., 26.0 ± 1.9., 31.6 ± 4.1 ng/L, Wastewater: 142.0 ± 16.5., 170.2 ± 18.3., 22.5 ± 1.4 ng/L	Wu et al. (2007)
Wastewater	China	Dimethyl phthalate (DMP), diethyl phthalate (DEP), di-*n*-butyl phthalate (DBP), butyl benzyl phthalate (BBP), bis (2-ethylhexyl) phthalate (DEHP) and di-n-octyl phthalate (DOP)	Liquid-phase extraction followed by GC–MS	DEHP was found to be highest with a concentration of 16.86 ± 11.67 ng/mL followed by DBP, DOP, and DEP with concentrations of 14.34 ± 7.03, 8.08 ± 3.48, and 8.07 ± 6.32 ng/mL respectively	Gao et al. (2014)
Wastewater and sewage sludge	Greece	Nonylphenol, nonylphenol ethoxylates, triclosan and bisphenol-A	SPE followed by GC–MS	More than 60 % of the studied compounds both in liquid and solid samples except for 4-n-NP were recovered. While more than 35 and 65 % 4-n-NP were also recovered in more than 35 % of wastewater and sludge samples respectively	Gatidou et al. (2007)
Surface waters	Portugal	Bisphenol-A	SPE extraction followed by GC–MS	The analysed samples revealed a widespread contamination of BPA especially in Ave, Cavado, Douro, Ferro, Sousa and Vizela Rivers. Achieving 98.4 ng/L for the highest concentration	Rocha et al. (2013)
Surface runoff, untreated and treated wastewater	Austria	Dimethyl phthalate (DMP), diethyl phthalate (DEP), dibutyl phthalate (DBP), butylbenzyl phthalate (BBP), bis(2-ethylbenzyl) phthalate (DEHP) and dioctyl phthalate (DOP)	Ultrasonic extraction followed by GC–MS	Of all the investigated phthalates, DEHP was found to be most abundant with concentration range 3.4–34 μg/L and 0.083–6.6 μg/L in influent and effluent respectively	Clara et al. (2010)

DMP dimethyl phthalate, *DEP* diethyl phthalate, *DBP* dibutyl phthalate, *DEHP* di-2-ethylhexyl phthalate, *TCPP* tris(chloroisopropyl)phosphate, *TCEP* tris (2-chloroethyl)phosphate, *DEET* N,N-diethyl-meta-toluamide, *DOP* di-n-octyl phthalate, *BBP* butylbenzyl phthalate

Therefore, there is need for a better understanding of their fate and ecotoxicological impacts on the environment after the discharge and escapes from the urban wastewater into the environment. The fate of these chemicals either in water or soil depends on their physicochemical properties such as partition coefficient (Kow), solubility index and other environmental factors (temperature, pH, salinity level, humus content) among others (Lapworth et al. 2012). The possible fate of chemicals of emerging concern once entering wastewater treatment plants include (1) conversion into harmless inorganic compounds such as carbon dioxide and water via biological, chemical or physicochemical transformation in the environment (2) partial adsorption or retention upon the sediments or sludge due to their lipophilic nature and non-biodegradability (3) adherence to other hydrophilic components and eventual discharge either as bulk or conjugate product into receiving waters (Richardson and Ternes 2011; Richardson 2012). Nevertheless, the reviewed literature noted that some emerging chemical contaminants and their metabolites that escape the treatment plants remained more persistent in the terrestrial and aquatic environment. A number of challenges are encountered in trying to understand the fate of emerging contaminants in the aquatic environment. According to Heim and Schwarzbauer (2013) aquatic environment are of three different ecological order namely water, sediment and suspended particulate matters. Most contaminants that passed through the wastewater treatment plants preferably hydrophobic pollutants are often accumulated or trapped in the particulate matter. Thus the fates of such compounds are tied to the sediment phases since they can no longer undergo either photo or biodegradation. The authors suggested the need to establish prior background concentration values of each lipophilic pollutants in the sediment phase as this will guide in the prediction of their environmental fate. On the other hand, diclofenac, synthetic estradiol and nonylphenols are not persistent in the aquatic environments and are easily isolated from aqueous solutions due to high octanol–water partition coefficients (Kow) (Mohapatra et al. 2010). These compounds easily undergo photodecomposition and biodegradation at ambient conditions and sometimes get absorbed into solid sludge in the treatment plants. The photo and biodegradations of emerging chemical contaminants within the treatment plants or in the environment is limited by their adsorption onto sludge or sediments. Their retention and sorption onto sludge or suspended particular matter is a functions of pollutants physico-chemical properties and the soil structural properties. The degradation of these xenobiotics via photo-irradiation can be direct or indirect (free radical concept) depending on seasonality, water levels, intensity of ray of light among others (Mompelat et al. 2009). However, the mobility of emerging contaminants in the environment as well as their retention onto sediments or sludge is often based on n-octanol water distribution coefficient (log Kow) (Mompelat et al. 2009). It is worthy of note that while some pharmaceuticals such as fluoxetine, diclofenac, triclosan, albuterol, diazepam, fenofibrate, iodoarene are susceptible to photodegradation, phthalates, tetracycline and others remain fixed in the environment. These plasticisers undergo a series of transformation, bioaccumulate and exposure to them produces endocrine disruptions such as carcinogenic and teratogenic effects (Padhye et al. 2014; Loos et al. 2010). For instance, acridine, a photodecomposition products of carbamazepine has been reported to be toxic, carcinogenic and highly mutagenic (Mompelat et al. 2009). In addition, bisphenol-A does not hydrolyse in water because of its chemical structure and as such more than 50 % BPA get adsorbs or binds onto sludge/sediments. However, it undergoes rapid photo-degradation in wastewater treatment plants and receiving waters. BPA is readily decomposed by bacteria under both aerobic and anaerobic conditions. It is regarded as a pseudo-persistent pollutant with a short half-life between 2.5 and 4 days, though other conjugates or residual by-products are associated with longer half-lives of up to a month (Ike et al. 2000, 2006; Oehlmann et al. 2009). Even though, bisphenol-A or other know industrial chemicals have shown to decompose in laboratory experiments—their levels in the environment are constantly being augmented by new discharges. This means in practice that no overall reduction can be achieved until the source discharge is prevented. Apparently, triclosan is one of the most ubiquitous pollutants partly removed in the wastewater treatment plants and have been detected in virtually all environmental wastewater samples including drinking water (Kantiani et al. 2008; Zhao et al. 2010). Due to its hydrophobic behaviour, triclosan including triclocarban after leaving the water phase get adsorbs onto the sediment and suspended particulate matters (Heim and Schwarzbauer (2013). In the water phase precisely, triclosan can undergo photo or biodegradation and transform into toxic and persistent metabolites. Some of the triclosan transformation products include chlorinated phenols, methyl triclosan, dioxins (toxin), polychlorinated benzodioxins, chloroform, and other chlorinated compounds at high pH value (Wu et al. 2007). In the same vein, triclosan reacts with the chlorine in water forming a human carcinogenic product called chloroform. On the other hand, the trapped or accumulated triclosan in the sediment or suspended particulate matters can no longer undergo photo or biodegradation and as a result the concentration begin to build up. Since 1980, the concentration of triclosan and its metabolites in the sediments namely limnic, fluvial, and estuarine have increased owing to high-level usage of products containing triclosan by consumers. Apart from

triclosan, the screening analysis conducted on the sediments indicated geometrical increases in the level of other emerging contaminants in the sediments, thus raising concern about the future safety of aquatic species (Heim and Schwarzbauer 2013). Heim and Schwarzbauer (2013) suggested continuous geochronological investigations of aquatic environment as a viable approach to monitor and possibly predicts the pollution status of the sediments. Phthalates are another group of emerging contaminants that readily decomposed within the treatment plants due to high lipophilicity and low solubility, thus get adsorbed upon particulate matter and subsequently discharged into the environment in the form of sludge (Liang et al. 2008). Thus, due to its hydrophobic behaviour, substantial amount of phthalates originating from the urban runoff, drainage, and domestic and industrial discharges also settles as sludge in the environment. The decomposition depends on the various environmental conditions. The fate and behaviour of the individual xenobiotic does not only depend on its hydrophobic-hydrophilic properties but also on environmental conditions such as water solubility, pH, adsorption coefficient, redox condition, temperature and bioaccumulation potential (Rahman et al. 2009). This observation has been widely reported and confirmed in several studies and review articles (Barnabé et al. 2008; Liang et al. 2008; Staples et al. 1997). The quantity of residual pharmaceuticals and endocrine disruptors in aquatic environments are frequently influenced by factors such as the amount of wastewater produced, consumption pattern, geographical locations, lifestyle, appropriate treatment techniques, regulations among others (Pal et al. 2010; Rogers et al. 2013). Petrie et al. (2015) recommended adoption of integrated standardised analytical approaches to accurately predict and understand the environmental fate of most emerging contaminants. Petrie et al. (2015) argued that several emerging contaminants accumulated in the sludge and the traditional analytical techniques such as low-resolution mass spectrometry failed to detect either the original or the metabolites. According to the authors, the integrated approach should be complimented with appropriate biological assays.

Potential risk from exposure to chemicals of emerging concern

There are concerns regarding the potential risk from exposure to pharmaceutically active agents in the environment (Fawell and Ong 2012). Depending on their fate and behaviour in wastewater treatment plants and even in drinking water treatment plants, the probability of human exposure to these compounds is high. In order to conduct a thorough risk assessment of emerging micropollutants for humans, there is a need to assess the exposure rate and the actual dose; this will assist in determining the associated adverse health effects. Since the concentration of these compounds in water is low, the acute toxicity may be difficult to evaluate, but the precautionary principle should be kept in mind because of chronic and long-term exposure. Given also that long-term exposure data are not available, the risk assessment might be technically hard to calculate. Aquatic species have a greater risk of exposure to individual agents or combinations of these compounds. It has been established that feminisation of fish in freshwater systems is a result of exposure to certain endocrine disruptors. Further research is needed to find whether this exposure had a major impact on entire populations. Strauch (2011) affirmed that the effects of exposure to pharmaceuticals and endocrine disruptors irrespective of their concentration in the water supply upon human toxicity are yet to be ascertained. However, research carried out by Ternes et al. (2004); Topp et al. (2008) revealed that oestrogenic compounds have a very high bioaccumulation potential with considerable negative effects on aquatic organisms. This environmental bioaccumulation aggravates abnormal hormonal control, as well as reproductive impairments and causes persistent antibiotic resistance. The acute and chronic toxicity experienced by aquatic species such as fish upon exposure to these compounds in the freshwater system is similar to that of health effects caused by exposure to low concentration of metallic elements (Sharpe and Irvine 2004; Xia et al. 2005). Studies conducted by Michael (2001) also revealed that exposure of aquatic species to endocrine disruptors causes low sperm count and reproductive malfunctions. Safe (2000) also observed that exposure of aquatic organisms to organochlorines cause feminisation of fishes and gulls, and sexual abnormalities in alligators. However, among the aquatic species, fish remain most susceptible to the high dose of these chemical substances. Studies have shown that exposure to diclofenac and 17α-ethinylestradiol in the aquatic environment induced structural deformities of kidneys and intestines as well as gene alteration which affected the body metabolic activities (Kümmerer 2011). In humans, recent reports of increasing occurrences of reproductive and developmental abnormalities in infants and children, of temporal downward trends in semen quality and testosterone levels as well as increased rates of testicular and thyroid cancers (Stuart et al. 2012). Among adult male populations has generated concern regarding the potential risk of environmental endocrine disrupting chemicals to men's health. Safe (2000) attributed the declining sex ratios in Canada and the USA to over exposure to endocrine modulators. The potential risk associated with water consumption varies between compounds and might depend on the concentration, exposure

time, volume, and metabolism rate. Currently, it is difficult to link human health effects to exposure to chemicals of emerging concern due to the existence of background natural diseases in the human body. It has been reported that direct exposure to emerging contaminants in drinking water portends no danger because the concentrations of these compounds are too low to cause serious health effects (Stanford et al. 2010). According to Houtman (2010) the effect of exposure to low dose of chemicals of emerging concern in drinking water will begin to manifest after 80 years. On the other hand, a series of abnormalities have been observed in the case of aquatic species, for instance, disruption of endocrine system of fish via exposure to low dose of oestrogenic hormones leading to severe adverse effects (Kümmerer 2011). The exposure rate is determined by comparing the exposure dose with the toxicity-based benchmarks. These toxicity-based benchmark standards vary and can be related to World Health Organization tolerable daily intake as well other standards (Fawell and Ong 2012; WHO 2011). Caldwell et al. (2010) conducted a comprehensive risk assessment of drinking water in the USA and found out that exposure to oestrogenic hormones caused no adverse health effects to the overall receptor (US population). Very recently, Stanford et al. (2010) conducted a comparative survey on the rate of exposure to oestrogenic activity and other compounds present in US drinking water, food, beverages, and air. The authors concluded that humans being are only exposed to a small fragment of emerging contaminants via consumption of municipal drinking water and there is no evidence of adverse effects on human health due to exposure to US drinking water. Human being is also exposed to these chemicals via consumption of fruits or vegetables irrigated with polluted water. Fromme et al. (2009) assessed the rate of exposure to perfluorinated octanoic acid and oestrogenic hormone via consumption of drinking water in Germany and the USA and found that the daily exposure rate ranged between 0.7 and 2 %. Thus, the level of individual or mixtures of pharmaceutically active substances in drinking water was considered too low to cause a considerable chronic or acute health effects on humans (Bull et al. 2011). Claessens et al. (2013) demonstrated the occurrence of blocker propranolol in Belgian marine waters and its acute toxic effects on the *Phaeodactylum tricornutum*. The authors observed no instant risk of exposure to this compound by the *P. tricornutum*. On the other hand, a potential chronic risk was identified when exposed to these compounds in the two Belgian coastal harbours. This shows that environmental conditions also played a role during the exposure period. However, the extent and long-term effects of exposure still require further studies.

Conclusion

It is has been established that endocrine disrupting chemicals are present in the global water cycle. There is incontrovertible evidence that these compounds have a detrimental effect upon aquatic species, and their ubiquitous presence globally is causing serious harm to the environment. This has generated considerable concern among governmental and non-governmental organisations with respect to human health. It is indisputable that these compounds are toxic in the environment and that it is only a matter of time before the levels build up to a point where many species die out and human toxicity will become evident. The vast majority of people are not aware of the health risks associated with continuous exposure or consumption of some of these chemicals. Therefore, the precautionary principle is advisable because the long-term potential for considerable harm to the environment is high and health impacts cannot be ruled out. Chemicals of emerging concern with higher health risks due to long-term exposure should be banned. Likewise, citizens should be cautioned or prevented from using products containing these compounds since there is insufficient capacity for continuous monitoring. Further, because most studies on emerging contaminants remain scattered and even uncoordinated, an integrated, collaborative, multi-disciplinary research approach involving different stakeholders in various fields is required in order to gather sufficient evidence and garner the proofs needed to support efforts to prevent continuous release of these chemicals into the environment. Various ministries, departments, agencies with responsibility for protecting the environment should be involved. Seeing that many existing wastewater treatment plants have failed to effectively treat wastewater or to remove pollutants, development of improved low-cost physicochemical and advanced oxidation technologies should be given priority. This will protect the health and safety of citizens as well as the environment and offer some protection to aquatic species. In view of the increasing scientific evidence regarding the number of xenoestrogens in the environment, it is imperative to establish a screening protocol and build endocrine disrupting compounds test laboratories equipped with highly sophisticated and sensitive analytical instruments to perform continuous monitoring. Humans and aquatic species are exposed to numerous compounds on a daily basis depending on the routes of exposure and dose. In order to conduct a thorough health risk assessment especially for humans, the exposure dose as well as the safe limit needs to be determined in both acute and chronic exposure scenarios. Several factors need to be taken into cognisance when applying the benchmarks for these pollutants in drinking water as the

exposure rate and sensitivity among humans differ. In a situation where there are more than one contaminant in the drinking water, it might be practically impossible to predict the health risk caused by each contaminant, as they may act independently or in synergy even over shadow each other. As a matter of fact, detailed studies regarding human health risk do not exist. This is due to the high cost of conducting the risk assessment for emerging contaminants on humans and lack of technical experts to practically identify and quantify the detection limits. Further investigations on the risk assessment of individual or combinations of contaminants present in drinking water at low concentrations will help to understand and then minimise the risk. The call for epidemiological risk assessment should not be ignored as this would give an improved understanding of the impacts of the pollutants in human and wild life. A systemic management approach to screening, detection and removal of endocrine disruptors should be considered; this should also include quantification of the fate of other persistent pollutants emitted from water treatment plants. In order to understand and predict the fate of micro-pollutants in the environment, development of concise and precise models for tracking micropollutants' fate and decomposition rate should be developed. Predictive tools could be created to quantify the mass influx of contaminants into the aqueous environment as this will assist in future formulation of policies and overall risk assessment and containment. There is a need for concerted efforts to protect our limited available water resources and remaining aquatic species via pollution control and implementation of a comprehensive programme on water safety. However, pollution control requires strong political will for the formulation, legislation, implementation and policing of the various environmental laws. There is a need to create public awareness among citizenry on the health risks associated with exposure to chemicals of emerging concern

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