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RESEARCH TRENDS ON ENDOCRINE DISRUPTORS COMPOUNDS AND PERSONAL CARE PRODUCTS AS EMERGING CONTAMINANT IN SOUTH AFRICAN WATER SYSTEMS

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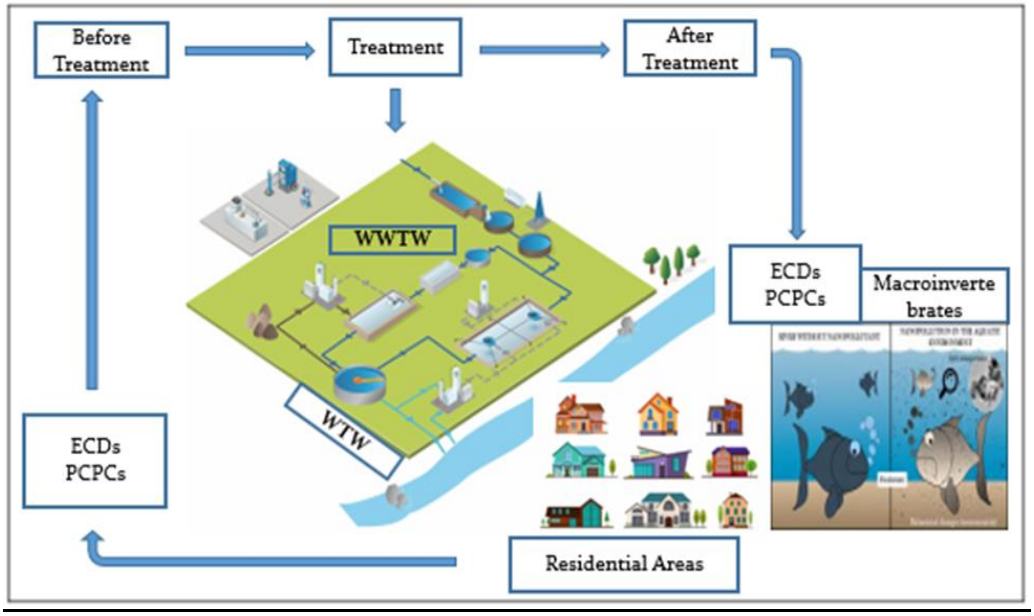
GRAPHICAL ABSTRACT



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ABSTRACT

The presence of pharmaceuticals, herbicides, endocrine disruptors and personal care products have recently attracted the interest of researchers due to their concern with regards to a wide variety of human health risk arising from their prevalence in surface waters resulting from inadequate waste water treatment processes. In this study, a recent trend in the fate and occurrence of personal care products such as (triclosan, nitro, polycyclic, macrocyclic musk, surfactants, Alkyl-phenol, Ethylates, 4-nonylphenol carboxylates, octylphenol) and endocrine disrupting compounds such as (17B-estradiol, estriol, estrone, alkylphenols, bisphenols A) will be reviewed for South Africa. These compounds are found at wastewater treatment plants, domestic and industrial pathways, agricultural sources and surface waters of South Africa. Sample analysis includes extraction with solid phase process of chemical compounds, with techniques such as liquid chromatography, Gas chromatography (GM), Mass spectrometry (MS) and high performance Liquid chromatography (HPLC). The identification and quantification of these emerging chemical compounds in surface waters and wastewaters has become a major scientific responsibility which is lacking regulation currently in South Africa especially when endocrine disruptors' compounds and personal care products have been detected to impact pregnant women and hormones of human adversely.

Keywords: emerging contaminants, endocrine disrupting compounds (EDCs), personal care product (PCPs), environmental waters

1. INTRODUCTION

Fresh water is one of the essential resources for the support of all life on earth. It is well recognized that anthropogenic activities are creating a huge amount of pressure on the water quality, and through such human activities, pollutants are entering fresh water systems [1]. There has recently been an exponentially growing concern regarding synthetic chemical pollutants that reach our aquatic environment [2]. Emerging contaminants (EC) are pervasive in environmental waters and are raising a concern due to their potential risks on human health and the environment [3]. Emerging contaminants are part of environmental pollutions which were linked to ancient times with the main focus then on the regulated pollutants that impacted human health with antecedent ecological effects [4].

[5] pointed out the history of emerging contaminants since it was discovered in water and aquatic environment due to the improvement in science and technology. Since the discovery of ECs, around 1970's in the USA, leading countries in studies of emerging contaminants are Canada, Japan, USA and China and Germany [5]. Africa is reported to lack behind in the study of EC even though there have been recent improvements and developments [6, 5, 7, 8, 9,]. However, countries like South Africa, Tanzania and Zambia has contributed into studying the extent of emerging contaminants in their water systems [1, 2, 6, 9, 10, 11].

Types of ECs that has adverse effects on human health and the environment are antibiotics, pharmaceuticals, personal care products (PCPs), hormones, artificial sweeteners and illicit drugs. These are recognised class of water contaminants in surface waters and underground waters due to their unregulated state. Globally, these contaminants are known as emerging because they lacked standards and guidelines for their environmental monitoring [5, 12]. The attention these emerging contaminants are receiving is due to recent development of

analytical technologies and in the use of new compounds prevalent in fighting emerging diseases, insect repellent (DEET) and societal demands in cosmetic product.

The presences of a group of emerging contaminants have been documented as significant water pollutants such as organic contaminants that have been having adverse effects on human and wild life endocrine system [13]. It is believed that natural attenuation and conventional water treatment processes are not capable of removing these micro-pollutants which are reported to bioaccumulate in humans, micro invertebrates and other organisms in aquatic food web [13]. Only a small fraction of the hundreds of pharmaceuticals products that found their way into our water sources have been detected. The few ECs found to be present in drinking water has generated significant concerns regarding the risk of estrogenic and other adverse effects on human beings [13, 14]. The detection of these new compounds in South Africa are found in sources such as surface water, ground water, drinking water, sewage treatment works and waste water treatment works raises considerable public concern, especially when human health based guidelines values are not available both globally and locally [1, 12]. It is still to be questioned if and how much of the ECs detected concentration affects human health?

Generally, pharmaceuticals such as endocrine disruptors compounds (EDC) and personal care products makes up one of the most familiar groups of contaminants in surface waters due to their over usage that has been documented [15, 16] and little is known concerning their impact on aquatic life [17]. Fluoxetine and antidepressant is prevalent in aquatic environment and a range of concentration can result in alterations in the life of aquatic macro invertebrates [17].

The advancement of suitable quantitative methods for the analysis of EDC and PCP contaminants in South Africa water is continuous with increasing attention aimed at analysis of water contaminants [11, 12, 18]. Liquid chromatographic procedures were used to investigate and determine emerging chemical compounds in surface waters [19], and "both gas and high liquid performance chromatography (HPLC) has been reported in the literature to analyse the EC in South Africa water with high performance liquid chromatography preferred method for non-volatile chemical compounds [18, 20]. Other methods used for analysis of EDC and PCPs are the LC-MS/MS [21]. However, there is a need for the development of standardised analytical technologies that allows for EDC, PCP and other ECs identification and quantification of chemical compounds at very low concentrations in drinking water [10, 22].

Emerging contaminant sources differ both in their numbers and nature. They can be categorised into two sources mainly point source and non-point sources. Point sources can include excretion in human and animal urine and faeces, flushing of un-used antibiotics, house detergents etc. one of the major point sources of emerging contaminants is treated municipal and industrial wastewater at outflow of wastewater treatment plants in urban, industrial and in agricultural areas [6] Non-point sources include storm-water runoff, urban areas and agricultural land. The use of pesticides is one example of non-point source that is considered a major agricultural contaminant [15]. Recently research marks that the human infertility rate is gradually increasing, and reasons for the growing of infertility rate is hypothesised to extend that it might be due to environmental contaminants that are spread throughout our environmental water and human exposure is highly unavoidable [23]. Due to anthropogenic activities, freshwater systems worldwide including in South Africa are confronted with thousands of EC compounds [24].

2. TYPES OF EMERGING CONTAMINANTS

2.1. Endocrine Disrupting Compounds

The human endocrine system is that human system that is responsible for production of hormones in the body, in the 1990's, evidence has been collected that certain synthetic and natural chemicals in the environment can disturb the hormonal (endocrines e.g., testosterone) system of exposed organisms by mimicking or blocking the action of hormones. The human endocrine system is comparable to that of a fish, therefore exposure to endocrine disrupting contaminants can cause certain health risks to humans also [25]. Types of EDC are 17B-estradiol, estriol, estrone, alkylphenols, bisphenols A and alkylphenolethoxylates. Figure 1 below shows chemical structures of some endocrine disruptor's compounds.

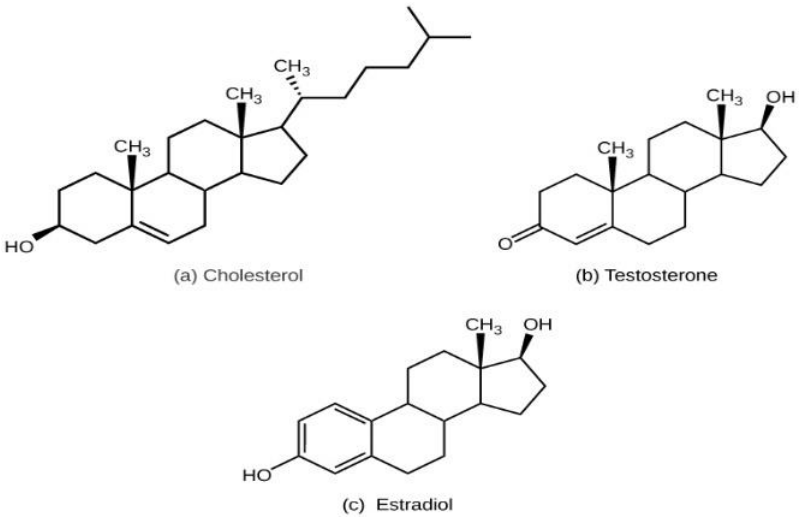

Figure 1. Shows chemical structures of endocrine disruptors [26].

2.2. Pharmaceutical Compound

Pharmaceuticals are chemical compounds that are used for the diagnosis, treatment, or prevention of disease in humans and animals. Hundreds and thousands of different pharmaceuticals are currently used and distributed both in public and private sector, antibiotics, anti-diabetics, B-Blockers, lipid regulators, anti-depressants, anti-epileptics [25]. The study of pharmaceuticals and their metabolites in the environment has rapidly become a field of scientific research under environmental studies with an increasing number interest [27]. Varieties of drugs are used for medicinal purposes and human health care globally, and diverse pharmaceuticals are used for veterinary science for animal treatment. Over about 3,000 thousand pharmaceutical compounds are approved as medicinal products. The occurrence of pharmaceuticals in the environment also poses a threat both to the scientific community and the public [27]. Figure 2 shows chemical structures of pharmaceutical compounds.

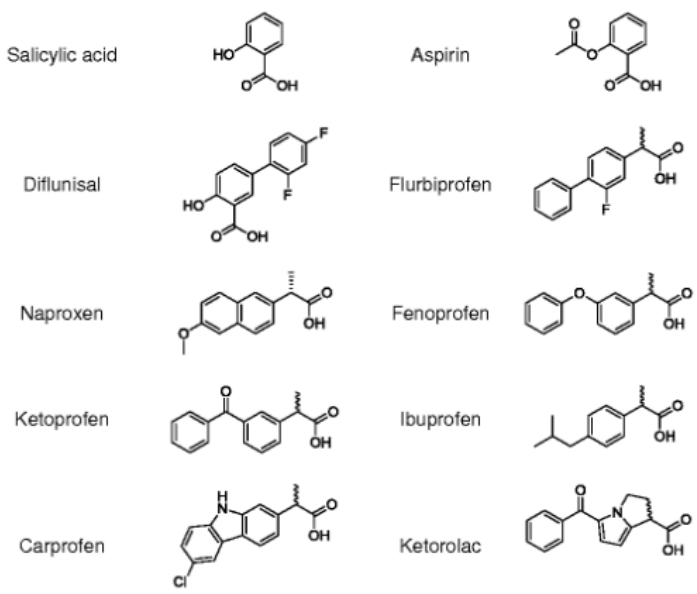

Figure 2. Shows chemical structures of pharmaceutical compounds [28].

2.3. Personal Care Products

Personal care products are those products that are applied directly to the human body and are not ingested, such products comprise of cosmetics, toiletries and fragrances. One of the group of personal care products used in as fragrance is polycyclic musk's, and the second group comprises of preservatives like parabens applied in shampoos, creams and body lotions. An emerging chemical contaminant known as Triclosan has been used for years in a wide variety of consumer products e.g., toothpaste, bath soaps, hand soaps etc. [25].

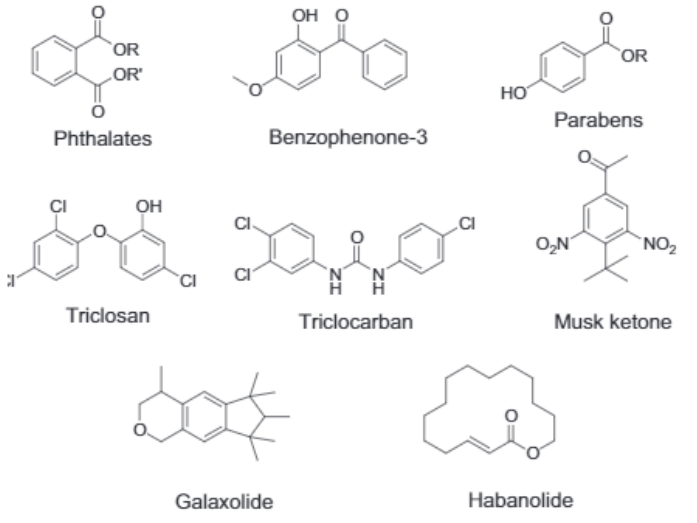

Figure 3. Shows chemical structures of personal care products [31].

Some personal care products are a very unique category of micro-pollutants due to their special degradation characteristics. Some examples of this class of emerging contaminants are fragrances (nitro, polycyclic, macrocyclic musk's), surfactants, Alkyl-phenol and Ethylates, 4-nonylphenol carboxylates, octylphenol ethoxycarboxylate, and some of them were included in the priority list of hazardous substances in the water policy by the European Union [29]. Some PCP do not fall within cosmetic regulations and are constantly finding its way into aquatic environment [30]. Figure 3 shows chemical structures.

2.4. Herbicides and Pesticides

Pesticides are products that are used merely in agriculture and crop production, they vary from herbicides, fungicides, insecticides and bactericides etc., they have also become a concern in surface water quality for years. Extensive use in agricultural practices and industrial emission during their production are important sources of pesticides and their residues in aquatic environment. Currently a number of registered pesticide compounds which include glyphosate, triazines, organophosphorus herbicides, thiocarbomates, and chlorophenoxy acetic acid. These compounds after being applied in agricultural fields and they contaminate the surface water by run-off, drainage and leaching [25]. Figure 4 shows different classification of pesticides by use that emanate as agricultural emerging contaminants.

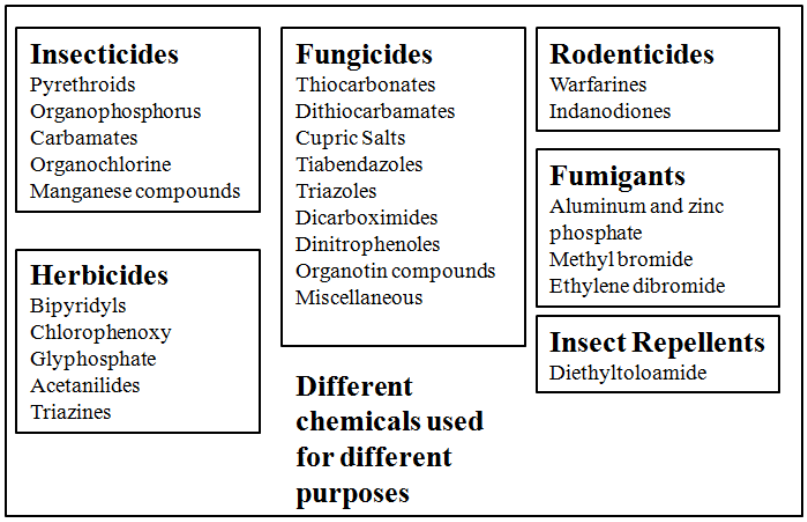

Figure 4. Shows different types of pesticides that emanate as agricultural emerging contaminants (Adapted from [32]).

3. Sources and Pathways of EDC and PCPs

Surface water is one of the largest water sources after oceans and it emanates from rivers, lakes, canals, dams and streams, and these sources of water serve as sources of drinking water for consumption by the public. Ground water is generally of certain amounts of

bacteriological concentrations with slightly constant chemical quality, whereas surface water quality differs considerably due to anthropogenic upstream activities with varying discharge volume and flow rate [25]. Emerging contaminants are brought in by human activities, which include agriculture, household use detergent, wastewater treatment, industry and spillages. Wastewater (sewage water) is considered one of the most significant sources of personal care products in environmental water [30]. Figure 5 shows sources and pathways of emerging contaminants that reach various receptors.

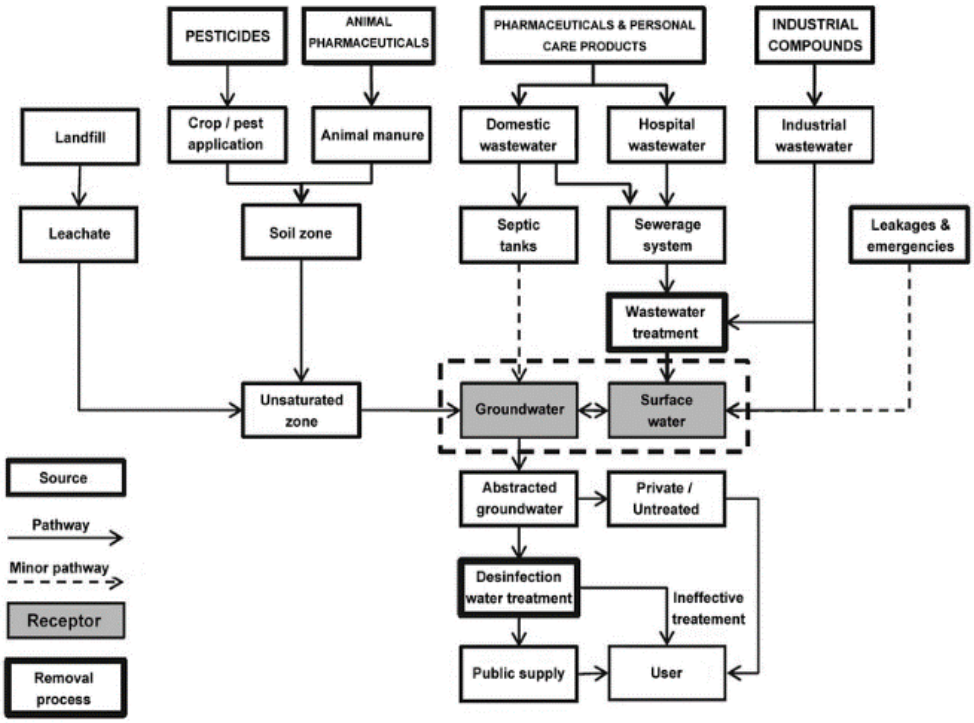

Figure 5. Sources and pathways for emerging contaminants that reach various receptors [33].

3.1. Waste Water Treatment Plants (WWTP)

Huge amounts of emerging contaminants are persistent in surface waters and the latter is that they create pressure on the wastewater treatment plants (WWTP) for their detection and removal. A study conducted by [7] on wastewater treatment works and environmental waters showed that in about eight emerging contaminants that were classified as illicit drugs were identified in wastewater influents [1]. Emerging contaminants have become a concern in wastewater treatment works, and primary sources (hospital, funeral homes, residential houses and abattoirs) [34], that effectively contribute to the wastewater treatment plants, makes ECs from WWTP as a secondary source, as effluent that contaminant our surface waters,

According to [35], emerging contaminants such as endocrine disrupting compounds and personal care products have ever been present in surface water, ground water as well as in drinking water and in wastewater treatment plants (WWTPs) discharge, (Municipal

wastewater works are viewed as one of the discharge sources for the emanation of emerging contaminants like non-point and point sources, industries and storm water, wastewater from households and water treatment facilities into the environment [35]. Many sources of pharmaceutical compounds are effluents from wastewater treatment works, efavirenz (a pharmaceutical compound) concentrations found in wastewater treatment plants were ranging between 5500 to 14000 ng/L according to an article by [2]. Wastewater treatment plants have been studied in depth as one of the major sources of emerging organic contaminants (EOCs) in particular municipal solid waste [36]. Numerous studies have demonstrated the presence of PCPs and EDCs in urban wastewater, excreted original or metabolized in sewage from hospitals and surface waters. They have also been detected in ground water and even in some drinking water [4].

3.2. Agricultural Use

Agriculture is responsible for the spread of CECs from domestic and industrial sources where bio solids and manure are used as soil conditioners and fertilizers and wastewater for irrigation [37]. Human exposure to pesticides can occur in the workplace, in the households and through the ambient environment. It was found that the women with a history of working in agricultural industry had an elevated risk of infertility. To determine the effect of pesticide exposure reproductive health, agricultural workers were evaluated in several different countries. In a Canadian study, exposure to the phenoxy herbicides 2, 4 dichlorphenoxy acetic acid (2, 4-D) was associated with spontaneous abortion. High exposure to organophosphate pesticide may be considered as an important cause of idiopathic preterm delivery in women [38].

3.3. Industrial and Domestic Sources and Pathways

Over recent years, the manufacturing and widespread applications of synthetic chemical compounds have become crucial in industrial sectors. Significant interest, therefore, continues to grow in the treatment and remediation of these compounds including endocrine disruptors, hormones, pharmaceuticals and synthetic textiles dye pollutants, because these substances may cause disorders of the nervous, hormonal and reproductive system, thus posing adverse health outcome [39]. About 300 million tons of synthetic compounds annually used in industrial and consumer products, partially find their way to natural waters [24].

In addition, to their introduction through human use, pharmaceuticals are also used in livestock, poultry, and fish farming. A variety of drugs are commonly given to farm animals to reduce illnesses and diseases in these animals and also to increase the size of the animals. Despite the fact that approximately 3000 substances are used in pharmaceutical ingredients, only a small fraction has been examined in the environment [40]. The presence of pharmaceutical and personal care products in the aquatic environment may pose potential threat to the ecosystem and human health; hence PCP’s have aroused much concern over the world [42]. An increased amount of personal care products have been detected in South Africa aquatic environment in recent years [42]. These synthetics organic products are found in Africa through domestic (pit latrines), agricultural and industrial sources [6].

4. SOUTH AFRICAN STUDIES ON EDCS AND PCPS

South Africa is one of the developing countries in Africa with a population that is estimated at about 56.5 million people from its 2017 census results [43], and it also has an increased urbanization. A study conducted by [9] in North West South Africa showed a presence of triclosan a personal care product compound with a precursor of 286 m/z and a product of 35:141.8 with a cone voltage of 22 (v) and energy collision of 11 (eV), Endocrine disruptors where also studies in Northwest namely Estrone (E1) with a precursor of 269.2m/z, products of 145.10;143.05 cone voltage of 149(v) and collision energy of 45 (eV). 17β-estradiol (β-E2) with a precursor of 271.2 m/z, product of 145; 10; 183.10, cone voltage of 14(v) and energy collision of 47. Estriol (E3) has a precursor of 287m/z and product of 143.05; 171.15, cone voltage of 15(v) and energy collision of 43 (eV). 17α-ethinylestradiol (EE2) has a precursor of 295.3 m/z, product of 145.05; 187, cone voltage of 11(v) and collision energy of 47(eV).

Study conducted by [18] in Kwazulu Natal South Africa showed the presence of triclosan in three different sources i.e., surface water at 0.4-0.9 ug/L, Wastewater treatment Works influent (WWTW) at 2.1-9.0 ug/and Wastewater treatment Works effluent at 1.3-6.4 ug/L. In Gauteng South Africa, also the presence of triclosan was traced, in Wastewater treatment Works influent at 78.4 ug/L and Wastewater treatment Works effluent at 10.7 ug/Studies on steroidal Hormones a group of endocrine disruptor compounds were studied across South African environmental waters see Table 1. Initially detection of endocrine disruptor studies in South Africa is mainly steroidal hormones particularly estrogens in water systems.

This is as a result because of the usage of synthetic estrogen contraceptives and hormone replacement therapy (HRT) by a large number of South African populations [1]. However, it is well known in South Africa that personal care products and endocrine disruptors are accumulating in the water systems in same rate as contraceptive medications. Table 1 below shows a summary of endocrine disruptors and personal care product in South African water systems at different sources and locations.

5. EXPOSURE AND HUMAN HEALTH RISK

Human exposure to environmental contaminants occurs through various routes, and detected at low concentration in drinking water, which raises considerable human health concerns. It acknowledged that environmental exposures to emerging chemical compounds are potential risk factors for infertility and pregnancy in women. Endocrine disrupting compounds interferes with the synthesis, secretions, and transport, metabolism and elimination processes of hormones in the human body that are responsible for reproduction [23]. In a study that was conducted by [44], it was found that xenoestrogen bisphenol-A (BPA) an endocrine disruptor compound “increases the propensity to develop mammary cancer during adulthood, long after cessation of exposure.” Exposures to BPA are associate with female infertility and affect the functions of the uterus and the ovaries as well as the embryo implantation [23]. Figure 6 show BPA exposure of PCPs and EDCs to human organs.

Table 1. A review summary of Endocrine Disruptors (EDCs) and Personal Care Products (PCPs) in South African surface waters (Adapted from [1, 9])

Name and group of EC contaminant.	Concentration in ug/L	Location	Source	References
Endocrine disrupting Compounds (EDCs)				
Estrogen (E1)				
	0.001-0.03	Kwazulu Natal	WWTW Downstream	Manickum and John,2014
	0.01-0.02	Western Cape	STW Downstream	Swart et al., 2011
	0.009-0.011	Western Cape	STW Effluent	Swart and Pool,2007
	0.01	Western Cape	STW Effluent	Swart and Pool,2007
	0.003-0.02	Kwazulu Natal	STW Effluent	Manickum et al., 2011
	0.01-0.35	Kwazulu Natal	WWTW influent	Manickum and John,2014
	0.003-0.08	Kwazulu Natal	WWTW effluent	Manickum and John,2014
	0.02-0.02	Western Cape	STW influent	Swart et al., 2011
	0.01-0.02	Western Cape	STW Downstream	Swart et al., 2011
	0.002-0.004	Gauteng	Drinking Water	Van Zijl et al., 2017
	0.0004-0.001	Western Cape	Drinking Water	Van Zijl et al., 2017
Estradiol (E2)				
	0.001-0.03	Kwazulu Natal	WWTW Upstream	Manickum and John,2014
	0.002-0.07	Kwazulu Natal	WWTW Downstream	Manickum and John,2014
	0.001	Western Cape	STW Effluent	Swart and Pool,2007
	0.005	Western Cape	STW Effluent	Swart and Pool,2007
	0.01-0.02	Kwazulu Natal	STW Effluent	Manickum et al., 2011
	0.02-0.20	Kwazulu Natal	WWTW influent	Manickum and John,2014
	0.004-0.11	Kwazulu Natal	WWTW effluent	Manickum and John,2014
	0.001-0.03	Mpumalanga	Surface Water	Van Wyk et al., 2014
	0.04-0.37	Gauteng	Drinking Water	De Jager et al., 2013
	0.05-0.37	Western Cape	Drinking Water	De Jager et al., 2013
	0.00003	Gauteng	Drinking Water	Van Zijl et al., 2017
	0.00002	Western Cape	Drinking Water	Van Zijl et al., 2017

Name and group of EC contaminant.	Concentration in ug/L	Location	Source	References
Ethynyl-oestradiol (EE2)				
	0.003	Kwazulu Natal	WWTW Upstream	Manickum and John,2014
	0.001-0.004	Kwazulu Natal	WWTW Downstream	Manickum and John,2014
	0.01-0.095	Kwazulu Natal	WWTW influent	Manickum and John,2014
	0.001-0.008	Kwazulu Natal	WWTW effluent	Manickum and John,2014
	0.001-0.01	Mpumalanga	Surface Water	Van Wyk et al., 2014
	0.00002	Gauteng	Drinking Water	Van Zijl et al., 2017
Progesterone(P)				
	0.01	Kwazulu Natal	WWTW Upstream	Manickum and John,2014
	0.06	Kwazulu Natal	WWTW Downstream	Manickum and John,2014
	0.16-0.90	Kwazulu Natal	WWTW influent	Manickum and John,2014
	0.03	Kwazulu Natal	WWTW effluent	Manickum and John,2014
Testosterone (T)				
	0.005-0.02	Kwazulu Natal	WWTW Upstream	Manickum and John,2014
	0.003-0.02	Kwazulu Natal	WWTW Downstream	Manickum and John,2014
	0.12-0.64	Kwazulu Natal	WWTW influent	Manickum and John,2014
	0.03	Kwazulu Natal	WWTW effluent	Manickum and John,2014
Personal Care Products (PCPs)				
Triclosan				
	78.4	Gauteng	WWTW influent	Amdany et al., 2014
	10.7	Gauteng	WWTW effluent	Amdany et al., 2014
	127.7	Gauteng	WWTW influent	Amdany et al., 2014
	22.9	Gauteng	WWTW effluent	Amdany et al., 2014
	0.4-0.9	Kwazulu Natal	Surface Water	Madikizela et al., 2014
	2.1-9.0	Kwazulu Natal	WWTW influent	Madikizela et al., 2014
	1.3-6.4	Kwazulu Natal	WWTW effluent	Madikizela et al., 2014
	35m/z	North West	WWTW influent	Kanama et al., 2018

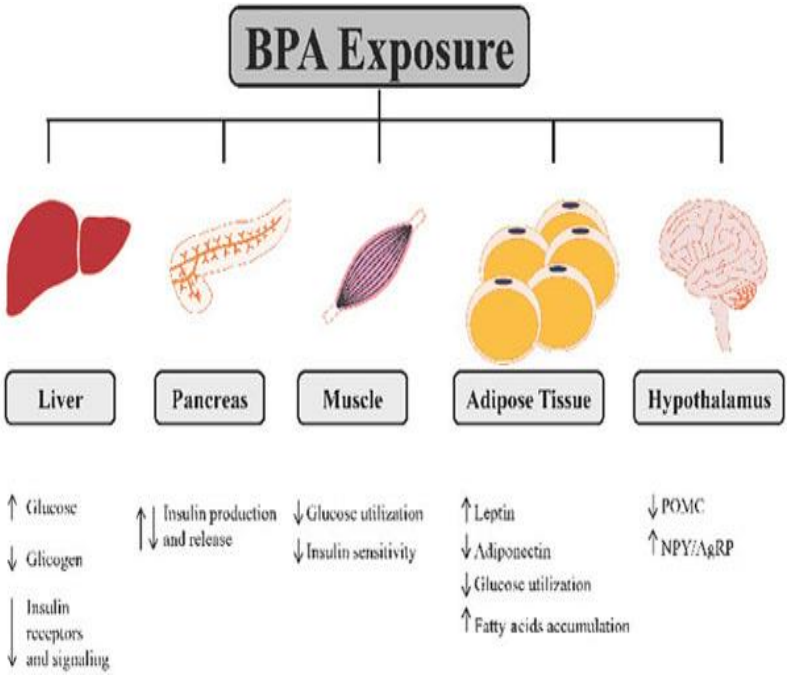

Figure 6. Shows Human organs that are exposed to BPA an endocrine disrupting compound [45].

6. REGULATIONS ON EMERGING CONTAMINANTS

According to the World Health Organisation (WHO, 2017) guideline for drinking water-quality and the US Environmental Protection Agency, the levels of Concern of Emerging Contaminants were below maximum levels, but the range of Emerging Contaminants detected in drinking water requires a seasonal and more frequent screening programme [22]. Many of the Emerging Contaminants elevates significant toxicological and public health concern particularly when human health guideline values are not available [24]. In addition, there has been a lack of publication to provide health standards and guidelines in treating Emerging pollutants [36].

Developing and enforcing regulations pertaining to pharmaceuticals and personal care products may be needed. A review has highlighted that critical information on interventions to improve the use of antibiotics and other pharmaceutical products is needed [46]. It is well recognized that the existing South African water quality management policy (WQM) was last dated in 1991 and resource directed management (RDM) of water quality in 2006, while innovations are taking place, revision of the policy needs to be dealt with in order to align with current overarching policy and legislative frameworks [47]. Currently there are no guidelines and values in South Africa regarding emerging chemical compounds for pharmaceuticals, endocrine disruptors, and personal care products, exception is only for pesticides which are agricultural contaminants. However preventive measures such as rational drug use and education of those who prescribe the medication and the public to reduce disposal and discharges to the environment will likely reduce human exposure [48].

7. METHODOLOGY, QUANTIFICATION, AND ANALYSIS

7.1. Solid Phase Extraction

[22] described methodology of quantifying and analysing EC through solid phase extraction. Samples of compounds were prepared with extraction and reconstitution in 1 Ml of H_2O/0.1% formic acid. Different solid phase extraction cartridges with varying sorbent characteristic are analyzed to identify the cartridges with the best optimum recovery. Before extraction, cartridges are equilibrated with 6 Ml PURE MeOH. After equilibration, samples are loaded at flow of 6 ml/min. After loading of samples, cartridges will be washed with 6Ml tubes using a 2 Ml of MeOH and 2 Ml and 2Ml of acetonitrile. Eluates are evaporated using a savant SC 210A speed-vac concentrator with a thermo RVT 4104 refrigerated vapour trap. Extract are be reconstituted in 1Ml of H_2O/0.1% formic acid and suspended using a vortex (Velp Scientifica. Italy) as well as by sonication (Branson, USA) [22].

7.2. LC-MS/MS/GC Analysis/Quantification

The analysis using LC-MS is performed on a High Performance Liquid Chromatography (HPLC, Agilent 1200) linked to a 3200 QTRAP hybrid triple quadrupole mass spectrometer (AB Sciex, Framingham, MA USA. The HPLC is fitted with 3-micro-meter Gemini-NX-C18 110-Å (150 x 2 mm) column (Phenomexe, CA, Torrance, USA). Formic acid (0.1% v/v) in water (solvent A) and formic acid (0.1% v/v) in MeOH (solvent B) are used as elution to charge analytes positively [22].

CONCLUSION

Literature has shown that emerging contaminants are available in South African surface water system with increasing human health risk associated with these contaminants. Urbanization and over population has led to the increasing use of pharmaceuticals product, personal care products and endocrine disrupting compounds in South Africa. These products require a more in-depth research in terms of their occurrence, fate and behavior. Since there are no regulations in South Africa there will be a need to educate the public about emerging contaminants and their associated health effects to human and animals.

ETHICAL COMPLIANCE

The authors have stated all possible conflicts of interest for this work and all sources of funding for this work.

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