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REVIEW ARTICLE

STUDY OF FATE AND TRANSPORT OF EMERGENT CONTAMINANTS AT WASTE WATER TREATMENT PLANT

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ABSTRACT

Contaminants of emerging concern such as pharmaceuticals, personal care products, bacteria, viruses, and pesticides are frequently found in waste water, groundwater, and surface waters. The search to find the sources of these compounds has routinely led to wastewater treatment plants (WWTPs) as an entry point of contaminants into the natural environment. The unknown effects of low concentrations of emerging contaminants in the aquatic ecosystem require scientists to study the occurrence, sources, fate, and transport of these compounds in wastewater treatment, to better understand and possibly identify mitigation opportunities. Reducing the contaminant levels in WWTP effluent helps minimize the contamination in lakes and rivers, which are both WWTP receiving waters as well as drinking water sources. Emerging contaminants end up in wastewater through several pathways including the disposal and use of consumer products, farm runoff, toxic spills, and excretion via the urine and feces of those consuming pharmaceuticals. The human body only metabolizes a percentage of each drug taken, expelling the rest into the municipal wastewater system. Another source is from consumer products such as soap, shampoo, disinfectant washes, and toothpaste which contain biologically active compounds that, when used, release these contaminants into the sewer system where they are then transported to a wastewater treatment plant (WWTP). Municipal wastewater treatment plants are not specifically designed to deal with the trace levels of emerging contaminates found in wastewater and many compounds pass through conventional treatment systems without removal. From the WWTP effluent, emerging contaminants are discharged into surface waters where they may have measurable effects on aquatic life at low concentrations. Once in surface waters, pharmaceuticals have been shown to interrupt the natural biochemistry of many aquatic organisms including fish and algae. Many of the problems associated with the removal of emerging contaminants from municipal wastewater stem from their low concentrations and chemical diversity, which make detection and analysis difficult. Low concentrations require extremely sensitive analytical equipment while the wide range of distinct chemical compounds necessitates techniques to identify many chemicals at once. Only recently have scientists become aware of the presence of some emerging contaminants in wastewater because analytical techniques able to detect them at the ng/l have only recently been developed. As laboratory procedures are developed and emerging contaminants can be accurately quantified, scientists are becoming increasingly able investigate the sources, removal pathways, and fate of pharmaceuticals in municipal wastewater. In addition to emerging contaminants, the potential entrance of prions into the wastewater system and their fate in wastewater treatment is an area of concern and a topic of interest in this study. A sampling program will implement to monitor the sources and fate of emerging contaminants in municipal wastewater treatment system. Laboratories will test for twelve different classes of emerging contaminants ranging from pharmaceuticals to flame-retardants. Hospitals, funeral homes, slaughterhouses and residential neighborhoods will monitor to determine possible point sources of contaminants into municipal sewer systems. Multiple locations within each wastewater treatment plant will monitor to trace the fate of each emerging contaminant class through the wastewater treatment process in an attempt to understand the fate removal pathways of each contaminant.

KEYWORDS

Contaminants, waste water, treatment plant, natural environment.

1. BACKGROUND

Emerging contaminants (ECs) such as pharmaceuticals, endocrine disruptors, pesticides, x-ray contrast media, and personal care products have been found in wastewater, groundwater, and surface waters [1-2]. These compounds can enter the environment through leaky sewers and septic systems, which can allow contaminants to infiltrate into the groundwater, and pass through wastewater treatment plants to discharge contaminants into receiving waters. The fate and concentrations of many EC's in the environment are mostly unknown, making the design of treatment strategies difficult. One class of EC attracting particular attention is known as endocrine disrupting compounds (EDCs). These natural or synthetic compounds mimic biological hormones disrupting an organism's natural processes. EDCs have been shown to be biologically active down to concentrations as low as the ng/L level and have been linked to significant change in wildlife including the feminization of fish [3-4]. Pharmaceuticals and personal care products (PPCPs) are another group of emerging contaminants gaining recent attention. These include x-ray media, analgesics, antiseptics, antibiotics etc. This class of contaminants often contains polar functional groups making the detection and removal process more difficult.

1.1 Personal Care Products

Personal Care Products are a class of chemicals found in wide ranging consumer products from toothpaste to soap, to kitchen utensils. They are generally used for their antimicrobial and antifungal properties. In this study, wastewater samples were analyzed for triclosan (TCS) and triclocarban (TCC) concentrations. TCS and TCC are both anti-bacterial and anti-fungal agents commonly used in consumer products such as soaps, disinfectants, toothpastes, body washes, and medical disinfectant of which these products contain between 0.1% and 2% of TCS or TCC by 15 weight [5]. Approximately 96% of the triclosan is used in consumer goods, which are disposed of to the sewer system [6]. They have both been used since the 1970's and have been detected in wastewater treatment plant effluent and surface waters [7-8]. TSC and TCC interact with bacteria by binding to the enoyl-acyl carrier protein reductase enzyme (ENR) found in their cell membrane inhibiting fatty acid synthesis [9]. Bacteria need these fatty acids to build new cell membranes and without them they do not have enough material to build new cells and thus cannot reproduce. The following table outlines some triclosan and triclocarban concentrations found in other wastewater treatment plants.

Table 1: Selected Literature Concentration Values for Personal Care Products in WWT

Compound	Influent ng/L	Effluent ng/L	Removal efficiency	Location	Source	Treatment Type
Triclosan	1900	114	94%	Ontario	Lishmen et al.,2006	Activated Sludge Process
Triclosan	5200	260	95%	Columbus, Ohio	McAVoy et al., 2002	Activated Sludge Process
Triclosan	2500	625	75%	United Kingdom	Thompson et al., 2005	Rotating Biological Contactor as Secondary
Triclocarban	6100	170	97%	-	Heidler at el, 2007	-

The above papers report removal efficiency of 75 to 97% and suggest that an activated sludge process may remove more triclosan than a rotating biological contactor process though detailed comparisons are not possible without detailed operating conditions.

1.2 Antidepressants

Antidepressants are a class of pharmaceuticals that affect neurotransmitters, the chemicals that nerves within the brain use to communicate with each other. Examples of neurotransmitters include serotonin, dopamine and norepinephrine. Antidepressants are believed to work by inhibiting the release or affecting the action of neurotransmitters. The antidepressant compounds in this study are venlafaxine, O-desmethylvenlafaxine, citalopram, and desmethylcitalopram. Venlafaxine is a serotonin-norepinephrine reuptake inhibitor (SNRI) prescribed for the treatment of depression, depression with associated symptoms of anxiety, generalized anxiety disorder, social anxiety disorder and adult panic disorder. O-desmethylvenlafaxine, a major active metabolite of venlafaxine, also functions as an SNRI. It is also synthetically produced (desvenlafaxine) and was approved by Health Canada in 2009 for treatment of depression. Citalopram is a selective serotonin reuptake inhibitor (SSRI) prescribed for the management of depression as well as treating obsessive-compulsive disorder, panic disorder, premenstrual dysphoric disorder, anxiety disorder and posttraumatic stress disorder. Desmethylcitalopram, an active metabolite of citalopram, also functions as an SSRI. The following table outlines the concentrations of Citalopram and Venlafaxine found in other wastewater treatment plants.

Table 2: Selected Literature Concentration Values for Antidepressants

Compound	Influent ng/L	Effluent ng/L	Removal efficiency	Location	Source	Treatment Type
Citalopram	52	46	11%	Montreal, Canada	Lajeunesse et al.,2008	Primary Treatment Only
Venlafaxine	195	175	10%	Montreal, Canada	Lajeunesse et al.,2008	Primary Treatment Only
Venlafaxine	540	300	44%	Spain	Gracia-Lor et al., 2005	Not Specified

The removal rate for the wastewater treatment plant in Montreal, Canada was reported as 10 to 12% [10]. This low removal efficiency suggests that antidepressants are difficult to remove, however, the wastewater treatment plant in Montreal, Canada is only equipped with primary treatment and municipalities with secondary or tertiary treatment would likely expect a higher removal efficiency. The researcher does not specify the treatment type used in their study [11].

1.3 Pharmaceutical (Acid/Basic/Natural)

Pharmaceuticals can be divided up into acidic, basic, or neutral compounds and can be used to treat pain, inflammation and a variety of other conditions. They range in prevalence from Naproxen, which is a prescription drug, to caffeine, which is found in coffee, tea, and chocolate among other goods. The following table outlines examples of the WWTP concentrations found in literature for some of the compounds in this study.

Table 3: Selected Literature Concentration Values for Pharmaceuticals in WWTPs

Compound	Influent ng/L	Effluent ng/L	Removal efficiency	Location	Source	Treatment Type
Acetaminophen	23202	100	99%	Spain	Rosal et al.,2009	Biological Treatment with Nitrification and Denitrification
Ibuprofen	2687	135	94%	Spain	Rosal et al.,2009	Biological Treatment with Nitrification and Denitrification
Ibuprofen	9500	18	99%	Virginia	Thomas and Foster 2004	Activity Sludge and Gravity Filtration
Naproxen	2363	923	60%	Spain	Rosal et al.,2009	Biological Treatment with Nitrification and Denitrification
Caffeine	22849	1176	94%	Spain	Rosal et al.,2009	Biological Treatment with Nitrification and Denitrification
Caffeine	43800	36	99%	Virginia	Thomas and Foster 2004	Activity Sludge and Gravity Filtration
Caffeine	2700	560	79%	Spain	Santos et al.,2005	Activated Sludge
Ibupofen	83500	6500	92%	Spain	Santos et al.,2005	Activated Sludge
Napoxen	6000	2560	57%	Spain	Santos et al.,2005	Activated Sludge

The reported removal rates for these pharmaceuticals range from 79 to 99% with the exception of Naproxen in Rosal et al. and Santos et al. which reported 60% and 57% respectively. This highlights the differences between specific pharmaceuticals and the need to study each individually. Overall, however, literature suggests that most pharmaceuticals except for naproxen are greater than 90% removed from wastewater.

1.4 Musks

Musks are a class of compound known for their fragrance. They have broad uses from cosmetics to detergents. They can be separated into three classes: aromatic nitro musks (musk ketone, musk xylene), polycyclic musks (tonalide, galaxolide, celestolide), macrocyclic musks. The following table outlines some musk concentrations found in municipal wastewater treatment plants.

Table 4: Selected Literature Concentration Values for Musks in WWTPs

Compound	Influent (ng/L)	Effluent (ng/L)	Removal Efficiency	Location	Source	Treatment Type
Galaxolide	4000	80	98%	USA	Horii et al., 2007	Activated Sludge
Galaxolide	1701	876	48%	Ontario	Lishman et al., 2006	Activated Sludge
Tonalide	3000	150	95%	USA	Horii et al., 2007	Activated Sludge
Traseolide	131	47	64%	Ontario	Lishman et al., 2006	Activated Sludge
Tonaline	687	298	56%	Ontario	Lishman et al., 2006	Activated Sludge
Phantolide	22	-	-	Ontario	Lishman et al., 2006	Activated Sludge
Celestolide	34	20	41%	Ontario	Lishman et al., 2006	Activated Sludge
Galaxolide	1941	695	64%	Germany	Bester 2004	Activated Sludge
Tonalide	583	212	63%	Germany	Bester 2004	Activated Sludge

Musk ketone	569	99	82%	Ohio	Simonich et al., 2000	Activated Sludge
Musk xylene	376	5	98%	Ohio	Simonich et al., 2000	Activated Sludge
Tonalide	10700	1180	88%	Ohio	Simonich et al., 2000	Activated Sludge
Galaxolide	13700	1170	91%	Ohio	Simonich et al., 2000	Activated Sludge

The removal rates for musks in the wastewater treatment plants described above range from 41 to 99%. This high variability suggests that the removal of musks is inconsistent between wastewater treatment plants.

1.5 PBDEs

Polybrominated diphenyl ethers or PBDEs are commonly used as flame-retardants in a variety of products including electronics, cars, textiles, and plastics. The PDBE family includes 209 individual compounds, which are generally used as a mixture rather than a pure compound. The following table outlines the concentrations of common PDBEs found at different municipal wastewater treatment plants.

Table 5: Selected Literature Concentration Values for PBDEs in WWTPs

Compound	Influent (ng/L)	Effluent (ng/L)	Removal Efficiency	Location	Source	Treatment Type
PBDE-47	102	14	86%	Ontario	Song et al., 2006	Activated Sludge
PBDE-99	121	14	88%	Ontario	Song et al., 2006	Activated Sludge
PBDE-100	19	2.8	85%	Ontario	Song et al., 2006	Activated Sludge
PBDE-153	11	1.6	85%	Ontario	Song et al., 2006	Activated Sludge
PBDE-154	7.6	1.1	85%	Ontario	Song et al., 2006	Activated Sludge
PBDE-183	1.7	-	-	Ontario	Song et al., 2006	Activated Sludge
Total PBDE	265	36	86%	Ontario	Song et al., 2006	Activated Sludge
Total PBDE	-	29	-	California	North 2004	Activate Sludge with gravity filter
PBDE-47	3.4	0.1	97%	China	Peng et al., 2009	Anoxic, anaerobic, aerobic biological treatment (A2O)
PBDE-99	3.4	0.09	97%	China	Peng et al., 2009	A2O

All of the wastewater treatment plants documented above have a removal efficiency ranging from 85 to 97% with respect to PBDEs. This indicates that PBDEs are consistently at efficiencies higher than 80% by activated sludge processes.

1.6 Endocrine Active Compounds

Endocrine active chemicals (EACs) are natural and synthetic molecules that can interfere with hormone systems in the bodies of animals. Recognition and regulation of adverse impacts by estrogenic pesticides and pharmaceuticals (xenoestrogens) in wildlife, domestic animals, and humans began in the mid 1900s, but ample evidence of endocrine disruption of reproduction related to Nano quantities (parts per billion and parts per trillion) of human-based xenoestrogens in wastewater effluents appeared in the late 1980s and early 1990s [12-13]. Numerous studies have documented disruption of sex determination and sex ratios, feminization or demasculinization of adult males, and alterations in reproductive behaviors, as well as contraceptive-like actions in both male and female fishes. Similar disruptions are reported for amphibians, reptiles, birds, and mammals exposed via various routes. Studies of accidental exposures as well as correlative studies of exposure to estrogenic chemicals present in diets, plastics, personal care products, etc., have appeared for cultured cells, rodents, and humans. Developing animals, including the human fetus, are much more sensitive to exogenous estrogenic chemicals than are adults [14]. Observations in rodents and humans show that a single exposure to an estrogenic chemical during development not only affects that generation but also induces permanent changes that are passed to the next and the next and the next generation without additional exposures [15].

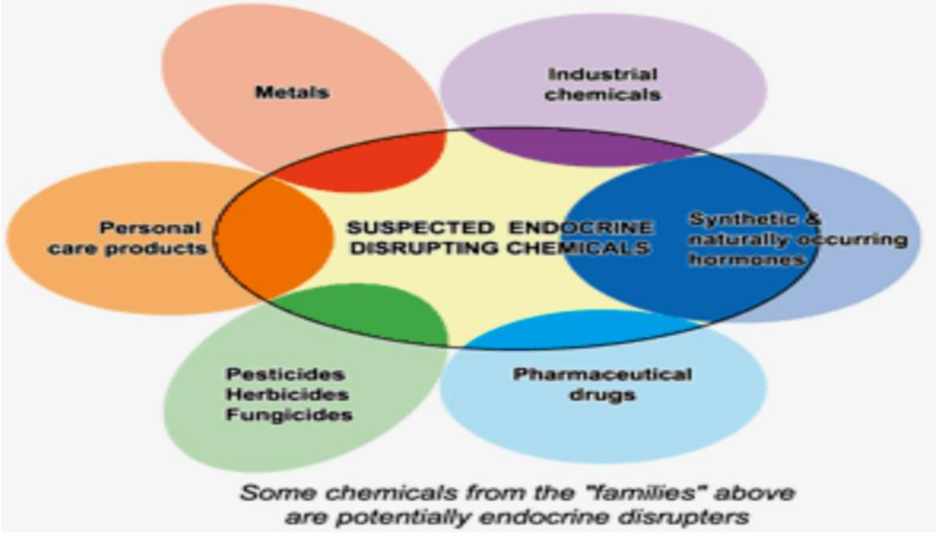


Figure 1: Typical form of endocrine disrupters

1.7 Antibiotics

Antibiotics are a class of drug used to kill or slow the growth of bacteria. They are available by prescription from a doctor and are used to treat a variety of bacterial infections. The following table outlines concentrations of various antibiotics in municipal wastewater plants.

Table 6: Selected Literature Concentration Values for Antibiotics in WWTPs

Compound	Influent (ng/L)	Effluent (ng/L)	Removal Efficiency	Location	Source	Treatment Type
Ciprofloxacin	150	60	60%	Wisconsin	Karthikeyan and Meyer 2005	Activate Sludge
Sulfamethoxazole	300	200	33%	Wisconsin	Karthikeyan and Meyer 2005	Activate Sludge
Trimethoprim	330	170	48%	Wisconsin	Karthikeyan and Meyer 2005	Activate Sludge
Erythromycin	3900	1100	71%	Wisconsin	Karthikeyan and Meyer 2005	Activate Sludge
Ciprofloxacin	3800	640	83%	Australia	Watkinson et al., 2007	Activate Sludge
Sulfamethoxazole	360	270	25%	Australia	Watkinson et al., 2007	Activate Sludge
Trimethoprim	340	50	85%	Australia	Watkinson et al., 2007	Activate Sludge
Sulfamethoxazole	390	310	20%	Mexico	Brown et al., 2006	Activate Sludge
Trimethoprim	590	180	69%	Mexico	Brown et al., 2006	Activate Sludge

The papers above report similar removal rates for respective compounds across locations ranging from 21 to 33% for Sulfamethoxazole, 60 to 83% for Ciprofloxacin, and 48 to 85% for Trimethoprim.

1.8 Estrogenic Compounds (Natural/Industrial)

Estrogenic compounds are substances that interact with the human body's hormone system. They include natural compounds such as estrogen and synthetic compounds such as Bisphenol A and nonylphenol. The following table outlines the concentrations of some estrogenic compounds found in municipal wastewater treatment plants.

Table 7: Selected Literature Concentration Values for Estrogens in WWTPs

Compound	Influent (ng/L)	Effluent (ng/L)	Removal Efficiency	Location	Source	Treatment Type
Esterone	30	13	56%	Ontario	Lishman et al., 2006	Activated Sludge
Estradiol	8	-	-	Ontario	Lishman et al., 2006	Activated Sludge
Estrone	54.8	8.1	85%	Australia	Braga et al., 2005	Activated Sludge with Reverse osmosis
a-estradiol	22	0.95	95%	Australia	Braga et al., 2005	Activated Sludge with Reverse osmosis
Bisphenol A	1600	900	43%	Greece	Gatidou et al., 2006	Not specified
Bisphenol A	140	86	38%	Australia	Tan et al., 2007	Anaerobic/ aerobic with activated sludge
Nonylphenol	3070	335	89%	Australia	Tan et al., 2007	Anaerobic/ aerobic with activated sludge
Octylphenol	229	23	89%	Australia	Tan et al., 2007	Anaerobic/ aerobic with activated sludge

The wastewater treatment plants as reported above have estrogenic compound removal efficiencies ranging from 40 to 90% with the highest removal efficiencies found in locations with tertiary, reverse osmosis treatment systems.

1.9 Perflourinated Compounds

Perfluorinated compounds are organofluorine compounds with hydrogens replaced with fluorine on the carbon chain. They are used in a large variety of products for properties such as water, oil, and stain resistance. PFOA is used; for example, to make Teflon while PFOS is used in the semiconductor industry,

Table 8: Selected Literature Concentration Values for Perfluorinated Compounds in WWTPs

Compound	Influent (ng/L)	Effluent (ng/L)	Removal Efficiency	Location	Source	Treatment Type
PFOA	100	122	0%	Kentucky	Loganathan et., al 2007	Activated Sludge
PFOA	50	53	0%	Georgia	Loganathan et al., 2007	Activated Sludge
PFOS	16	13	18%	Kentucky	Loganathan et al., 2007	Activated Sludge
PFOS	7.8	9.3	0%	Georgia	Loganathan et al., 2007	Activated Sludge
PFOS	20	24	0%	Oregon	Schultz et al., 2006	Trickling filter and activated sludge
PFOA	15	11	26%	Oregon	Schultz et al., 2006	Trickling filter and activated sludge
PFOA	16.3	24.3	0%	Singapore	Yu et al., 2009	Activated Sludge and Membrane Bioreactor
PFOS	13.9	12.6	10%	Singapore	Yu et al., 2009	Activated Sludge and Membrane Bioreactor

The removal efficiencies reported in the table above range from -49% to 20%. These low rates are reported across all of the papers and locations indicating per fluorinated compounds are not effectively removed from wastewater streams by activated sludge, membrane bioreactor, or trickling filter systems.

1.10 Pathogens

Bacterial growth in water distribution systems has been investigated for several decades. For example, Baylis (1930) reported coliform growth in sediments accumulating in water distribution pipes [16]. Researchers have found that mycobacterial numbers were substantially higher in the water distribution systems (on average 25,000-fold) than those collected immediately downstream from the water treatment facilities, indicating that mycobacteria grow in the distribution system [17]. In recent years, there has been great concern about the presence of emerging pathogens such as Legionella spp., Mycobacterium spp., and Aeromonas spp. and other opportunistic pathogens in water distribution pipes and home plumbing systems. It should be noted that both Legionnaires' disease (a serious, life-threatening pneumonia) and Pontiac fever (a mild, flue like illness) are caused by members of the genus Legionella.

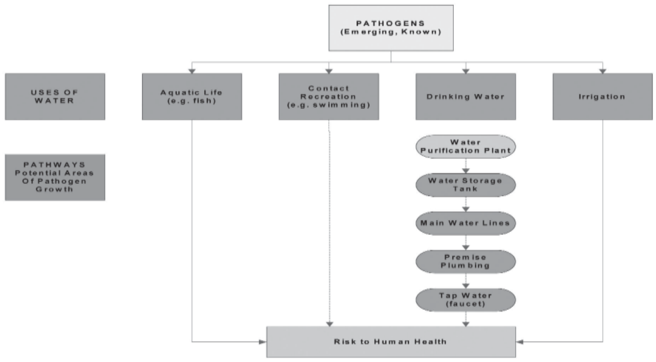


Figure 2: Uses of Water and Pathogen Pathways in Natural and Engineered Water Systems

2. LEGISLATION AND REGULATIONS

The lack of information on the fate, effects, and concentrations of emerging contaminants makes it difficult for governments to regulate their use as well as deal with the levels already existing in the environment. There are currently no laws or regulations outlining the maximum concentrations of emerging contaminants in wastewater effluent, drinking water or the environment. In the United States, the Environmental Protection Agency (EPA) maintains a list of compounds called the Contaminant Candidate List (CCL), which is a list of contaminants that are not subject to any drinking water regulations but are being monitored and may be included in future regulations. The most recent version, CCL3 was published in 2008 and includes several endocrine-disrupting compounds such as estrone, perfluoronated compounds such as PFOS, and some flame-retardants but no pharmaceuticals or personal care products [18]. Regulations outlining maximum allowable concentrations of these CCL3 compounds may be implemented in the future but have yet to be put into place. In the aquatic environment, the Food and Drug Administration currently does not require ecological testing of pharmaceuticals unless the environmental concentration exceeds 1ug/L [19]. This is a much greater than the ng/L level at which pharmaceuticals have been shown to interact with the aquatic environment [20-21]. In the Europe, the European Commission of the Environment called for the development of a comprehensive list of emerging contaminants in 2006 to comprehend the scope of contamination and develop steps to reduce contaminant levels [22-23]. Similarly to the United States, the commission implemented a monitoring strategy and in August 2011 published document outlining the preliminary approach to reduce the interference of EDCson humans and wildlife [24]. The document outlines a series of recommendations focusing on the reduction of endocrine disrupting compounds in consumer goods, food additives, and cosmetics but does not set any recommendations for maximum allowable concentrations in drinking water, wastewater, or the environment [25].

Table 9: Emerging Contaminant Regulations Summary

Country or Region	EC Regulations or Status
USA	-None Currently -Contaminant Candidate List for Monitoring Compound Occurrence Includes Endocrine Disrupting Compounds
European Union	-In Progress _monitoring EDCs -Recommends the Reduction of EDC as Editives in Consumer Goods
Canada	-None

In 2011, the World Health Organization published a report entitled Pharmaceuticals in Drinking-water which reviews the risks to human health associated with exposure to trace concentrations of pharmaceuticals in drinking-water. The report concludes that chlorination during the treatment of drinking water is sufficient to remove 50% of these compounds while advanced techniques such as ozonation and activated carbon can remove more than 99% of pharmaceutical molecule [26]. The authors suggest that pharmaceuticals in drinking water are unlikely to pose a risk to human health and that concern over pharmaceuticals should not divert resources away from bacterial, viral, and other chemical contaminants such as lead which are of a higher priority [27]. From the USA to the European Union, to the World Health Organization, the lack of knowledge regarding emerging contaminants is a major barrier for regulatory agencies responsible for drinking water and wastewater legislation. The burden of proof is on the scientific community to show emerging contaminants negatively affect the environment without which there is no motivation for governments to limit their use and exposure. With a more detailed and thorough look at the sources, fate, and transport of emerging contaminants, decision-makers are better able to implement regulations and design removal and mitigation strategies limiting their release into the natural environment.

3. PHYSICAL AND CHEMICAL PROPERTIES OF EMERGING CONTAMINATES

When examining the fate, pathways, and partitioning of emerging contaminants, it is important to consider the physical and chemical properties of each compound. After a chemical is created, the route that it takes between initial observation and final observations is referred to as a pathway. Common pathways include manufacture to initial use, initial used to disposal and initial use to release to the environment. The result of interactions between a chemical compound and its environment over a series of events and procedures is known as its fate. Thus the study of a compound's fate and pathways seeks to understand where a particular compound goes and how it is affected by different situations. By studying the physical and chemical properties of chemical compounds it is possible to predict their fate in some situations.

One commonly used class of physical properties is partitioning coefficients. Partitioning refers to the tendency of a chemical to concentrate in one phase of a two-phase mixture at equilibrium. The mixture can be two liquid phases, a liquid and a solid phase, a liquid and a gas phase, or a combination thereof. A partitioning coefficient is the dimensionless ratio of concentrations present in the two different phases of a two-phase mixture.

The octanol-water partitioning coefficient is a measure of the partitioning between octanol and water, which describes the hydrophobicity of a compound and is inversely related to the solubility of a compound in water. Compounds with a high kow have been shown to preferentially adsorb to soil and sediment particles in water [28].

Similarly, a sludge adsorption coefficient or kd, is a ratio of the amount of compound adsorbed to sludge compared to the amount present in aqueous solution under the specific conditions the measurement was taken. In water and wastewater treatment, the sludge adsorption coefficient is commonly used to predict the extent to which a compound can be removed by physical adsorption to sludge particles in a primary or secondary clarification unit.

A commonly use chemical property is the acid dissociation constant of Ka. It is a measure of the strength of an acid in solution and is the concentration ratio of ionized to un-ionized species of a compound at equilibrium. The Ka of a compound enables the concentration of ionized or un-ionized versions of a chemical to be calculated for a given pH. Due to the large range in magnitudes of Kas, the logarithmic constant (pKa) is commonly used.

Studies investigating the removal of emerging contaminants from wastewater have been conducted at the lab bench scale as well as full wastewater treatment plant scale. The following table outlines the removal of emerging contaminants by selected treatment processes during lab scale experiments. Where not specified, removal in the above table refers to reduction to levels lower than the detection limit used in the experiment. Ozonation and powdered activated carbon show the potential to be effective removal methods for some classes of emerging contaminants however more data is needed at the pilot and WWTP scale.

4. OBJECTIVES

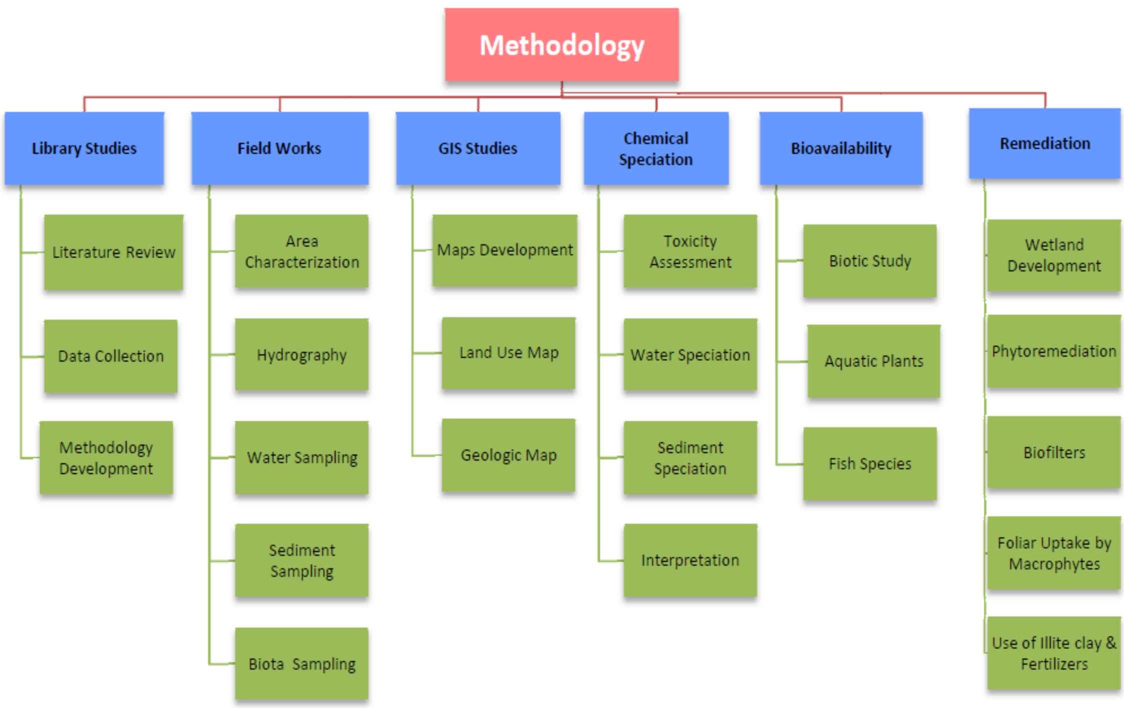
Study of emerging contaminants a new topic of research whose largely unknown environmental effects require the study of their sources, fate, and transport within the wastewater treatment system in an attempt to understand whether the need exists to abate their release. This study has the following objectives:

1. Monitor emerging contaminant concentrations at potential source locations such as hospitals, industrial areas, slaughterhouses and residential areas.
2. Monitor and compare emerging contaminant concentrations at multiple locations within the wastewater treatment plants of three municipalities to determine the fate and removal of these compounds in the wastewater treatment process.
3. To determine the fate and transport of contaminants (Organic and In-organics) in the waste-water, groundwater and soil at WWTP effluent disposal site in order to better understand the behavior, redistribution and sink of these contaminants and its ultimate impact on the terrestrial ecosystem such as soil, groundwater and biota
4. To develop new models for the determination of fate and transport of these contaminants in the groundwater and soil at WWTP effluent disposal site using information from field and laboratory studies.
5. To develop the possible methods or policies of remediation for contaminants and also study about their technical feasibility for implication in the area in order to protect further deterioration of the environment and to formulate recommendations suitable for socio-economic development. The study will enhance the indigenous capability for monitoring and analysis of contaminants and informed decision making concerning the control of such pollution and will build up constant and comparable time series of data relevant to the state of such sites, including the sources, amount, effect and future concern.
6. To explore the scenarios of water saving based on effluent impact assessment and their technical feasibility for implication to similar sites throughout the country. It will provide scientific and technical information needed by the Government and the related organizations to assess and for the necessary future plan and to contribute through such research project to the global environment monitoring systems.

5. METHODOLOGY

5.1 Research Methodology

The study will be performed in the form of an empirical study, which will follow both aspects, descriptive and inductive. The study will be based on primary data available for fate and transport, leading from field work to experimental analysis in order to come up with a general conclusion. Below is schematic diagram for the outline of complete methodology for the project.



5.2 Characterisation & GIS Mapping

A satellite image of potential sites (SPOT, 2009) interpretation and porgraphic data (Scale 1:500) will be used as a base map [29]. ArcGIS will use to develop final land-use map and bathymetric map from the base map where known points such as road junctions, bridges, milestones along trunk roads will serve as reference point. Locations of the reservoirs will be identified with reference to these known points with the help of GPS (Global positioning system) [30]. GPS is used to determine the actual coordinates of the surveying sites and to reconfirm the location of these sites during subsequent sampling periods. Hydrographical survey of the reservoirs will carry out on an engine boat by using echo-sounder and GPS while a tape and compass will be used to study the shapes and sizes of the reservoirs. Readings can be taken at 6 to 8 points in the central part of each reservoir to obtain the average depth. To study the thickness of the sediments depth, narrow, hollow borer will manually push into the sediments. Water discharges from the aquatic systems will be measured by current flow meter. A commonly applied methodology for measuring, and estimating, the discharge is based on a simplified form of the continuity equation [31-35]. The equation implies that for any incompressible fluid, such as liquid water, the discharge (Q) is equal to the product of the stream's cross-sectional area (A) and its mean velocity ($\bar{u}$), and is written as:

$$Q = A\bar{u}$$

Where

- Q is the discharge ([L^3T^{-1}]; m^3/s or ft^3/s)
- A is the cross-sectional area of the portion of the channel occupied by the flow ([L^2]; m^2 or ft^2)
- $\bar{u}$ is the average flow velocity ([LT^{-1}]; m/s or ft/s)

5.3 Field Survey

When choosing sample locations, glean as much information about the specific locations as possible before sampling starts. Useful information includes residential catchment area populations and specific hospital details.

The sample locations within each wastewater treatment plant were not consistent across the three municipalities. While they were similar enough to make some comparisons, identical locations would allow for more specific evaluations of the differences between WWTP performances. For a detailed description of WWTP sample locations, see the Appendix. Source sampling of hospitals was conducted at the sewage discharge point out side of each hospital. Some hospitals have multiple discharge points and thus sampling just one may give an inaccurate depiction of the overall hospital discharge especially if entire hospital units such as cancer or radiation suites are being released into an unmonitored discharge point. Since it is difficult to determine how specific hospital discharge systems are laid out, this is another potential source of experimental error. For Municipality A, two discharge points from the same hospital were monitored and the values averaged.

Funeral homes presented yet another challenge for sampling procedures. Partway through the project, it was discovered that some municipal staff in municipality C were presenting funeral home operators with bottles, which they then filled with the liquid discharge from the embalming process. In this way, data from Funeral Home C may not be indicative of an average funeral home effluent but rather describes funeral home discharge while an embalming is occurring.

5.4 Sampling

This study seeks to improve the knowledge of emerging contaminants' fate within the municipal wastewater system. Through a partnership Jeddah municipality, the source characteristics of hospitals, funeral homes, residential areas, and slaughterhouses will identify. By monitoring multiple locations within the wastewater treatment facilities of each municipality, the fate and transport of emerging contaminants inside WWTPs can be examined. To assemble a greater understanding of the potential pathways of emerging contaminants when entering the wastewater system, source sample locations will be chosen to monitor sites where ECs are hypothesized to be used. Within Jeddah Municipality, hospitals will be monitored due to the high use of pharmaceuticals, antibiotics, x-ray contrast media, disinfectants, and other emerging contaminants in the health care field. Industrial areas will also monitored to see if the chemicals used in the embalming process leads to high discharge rates of emerging contaminants. Disinfectants and pharmaceuticals in the effluent from a slaughterhouse will be monitored in order to determine the role of farming practices in the loading of ECs in wastewater. Finally, several residential neighborhoods will monitor to compare which contaminants come from general population use, and which ones are due to point sources. Within Hada wastewater treatment plant, multiple sample locations will selected to track the fate of each compound through the system. Samples will take from both the aqueous and sludge streams in order to calculate the removal efficiencies of each stage. The following diagram shows a basic schematic of a conventional wastewater treatment plant.

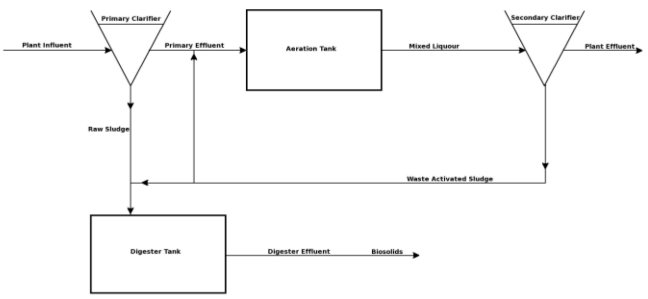


Figure 3: Conventional Wastewater Treatment Plant Diagram

Emerging contaminants enter the wastewater treatment plant at the Primary Clarifier where a portion physically adsorb to particulate matter and are settled out of suspension. The liquid discharge is sent to the aeration tank where aerobic bacteria work to break down the organic matter in the wastewater. After passing through another secondary clarifier, the aqueous phase is typically disinfected by chlorination prior to discharge to receiving waters. The sludge from both the primary and secondary clarifiers is sent to an anaerobic digester tank where bacteria further break down the sludge in the absence of oxygen. For each of the WWTPs in this study, samples will take after each of the major steps in the treatment process as well as overall plant influents and effluents.

Table 10: Typical Environmental Concentrations of Emerging Contaminants

Location	Concentration
WWTP Biosolids	mg/kg or ppm
Raw Sewage	ug/L or ppb
Treated Sewage	ng/L or ppt
Surface Waters	Low ng/L
Drinking Water	Very low ng/L or non detect

The above table shows the typical concentrations ranges for emerging contaminants found different locations. The lowest concentrations are found in drinking water with the greatest being observed in the biosolids from wastewater treatment plants.

5.5 General

A three-pronged sampling approach will perform that includ collection of liquid phase, suspended solids, and bed sediments at six to ten locations along Hada water treatment plant. Liquid samples (3L) will collect using pre-cleaned and muffled amber glass bottles and filtered within 24 hours of collection using 0.7 µm glass fiber filter membranes (Whatman). Bed sediments will collect proximate to the effluent outfall and at five additional locations downstream to Road. At each location, bed sediments will collect at two depths: 0-3 cm and 10-12 cm using pre-cleaned and muffled amber glass jars. Each sediment sample will be a composite composed of at least 2 replicates will obtain along a cross section of the channel at each location.

5.6 Sample Preparation/Extraction

All analytical work will perform in laboratory. Aqueous-phase samples sometimes require a degree of "cleaning" and analyte concentration, which can be carried out by solid phase extraction (SPE) and elution from the SPE resin in a stepwise methanol gradient. Compounds more hydrophobic than *p*-nonylphenol (log KOW ~ 4.5) tend to be retained on reverse phase resins, even through alcohol elution steps, and can be separated from the estrogens and estrogen mimics in this way. The technique is equally useful for androgen separations. Concentration factors >103 are conveniently obtained by processing initially large water volumes—on the order of a few liters. The *in vitro* endocrine disruption activity tests require an aqueous-phase sample, so that the methanol/water eluent must be evaporated before analytes are redissolved in water. Solid-phase samples like dried sludge or sediment/soil provide a more formidable challenge. Analytes will separate from bulk solids in an adaptation of microwave accelerated extraction (MAE), involving low heats/pressures during 30-min extractions in methanol. Extracts will dilute in ultrapure water, and the methanol water mixtures then processed using normal SPE procedures.

5.7 Endocrine Activity Assays

In both the yeast estrogen screen (YES) and yeast androgen screen (YAS) procedures, a genetically modified strain of *Saccharomyces cerevisiae* will use to detect and signal the presence of estrogen/androgen agonists and antagonists in environmental samples, wastewater, sludge, etc. A degree of sample preparation is required. The YES is a reporter-gene assay in which β-galactosidase is produced by the genetically modified yeast strain in the obligate presence of estrogenic compounds. The human *hER-α* gene will use to transform the yeast genome, where it is expressed constitutively. After an estrogen agonist or antagonist enters the yeast cell, it combines with the *hER*-α estrogen receptor protein, forming a complex that binds to the plasmid-borne estrogen receptor element (ERE) leading to transcription/translation of the reporter gene, here *β-gal*. β-galactosidase so produce is capable of cleaving chlorophenol red-β-galactopyranoside (CPRG) into chlorophenol red and galactose. The concentration of the red dye so produce will determined colorimetrically at λ = 570nm after a specified incubation period in the presence of CPRG and compared to a set of standards to determine whole-sample estrogenic activity. YAS procedures are entirely parallel. Differences between the tests arise from the nature of the genetic modifications to the test organism only. Anti-estrogen and anti-androgen activities can be determined via modest modification of the original procedures. All of these tests suffer from a singular shortcoming, however, in that each responds only to compounds that are capable of binding to respective steroidal hormone receptor proteins. Other forms of endocrine system disruption cannot be detected in this way.

6. CEC ANALYTICAL METHODS

6.1 Sample collection and preparation

All samples will collect in pre-cleaned and muffled amber glass bottles. Trace organics will be extracted within 24 hours. Samples will filter through 0.7 μm PALL glass fiber filters, deuterated internal standards will add and then the samples will extract using Waters HLB cartridges. HLB sorbents will condition with 5 ml of MeOH, 5 ml of MTBE and 5 ml of water. One-half g of EDTA will dissolve in one liter of each source water sample before it will load onto the SPE sorbent at 10 ml min-1. Sorbents will dry with N2 for 40 min before sorbates will sequentially elute with 3 ml of MeOH, 3 ml of 5% NH4OH in MeOH, 3ml of ACN and 3ml of MTBE. The combined eluents will evaporated to about 50 μl and re-dissolve in 1 ml 50% aqueous methanol for LCMS analysis.

6.2 Logistics and Data Assembly

Over the course of this research, samples will be obtained every two months for a period of twelve months. Three liters of sample at each location will be collected using a 24-hour composite auto sampler to ensure that the sample contained an average representation of the location's contents. The three liters will split into two amber glass bottles and one HDPE plastic bottle, and will store at 4^{o}C until analysis. The composition of emerging contaminants can be altered by prolonged exposure to light and thus dark amber bottles will be used to prevent the degradation of the samples. Similarly, for the source sampling within the sewer catchment areas, will set up 24-hour composite samplers in the sewage manholes outside of the target hospitals, slaughterhouses, and residential areas.

Once the samples from all locations of the municipality have collected, will transport to the laboratory where the majority of the analysis will undertake. Their advanced techniques and specialized equipment allow the detection of emerging contaminants down to the ng/L level required to accurately study EC presence in municipal wastewater as seen in Table 2. Once the analytical work will complete, the raw data will use for analysis and interpretation.

6.3 Emergent Contaminats Classes

The following table outlines the different categories of emerging contaminants investigated in this study and their status.

Table 11: Summary Table for Analytical Tests

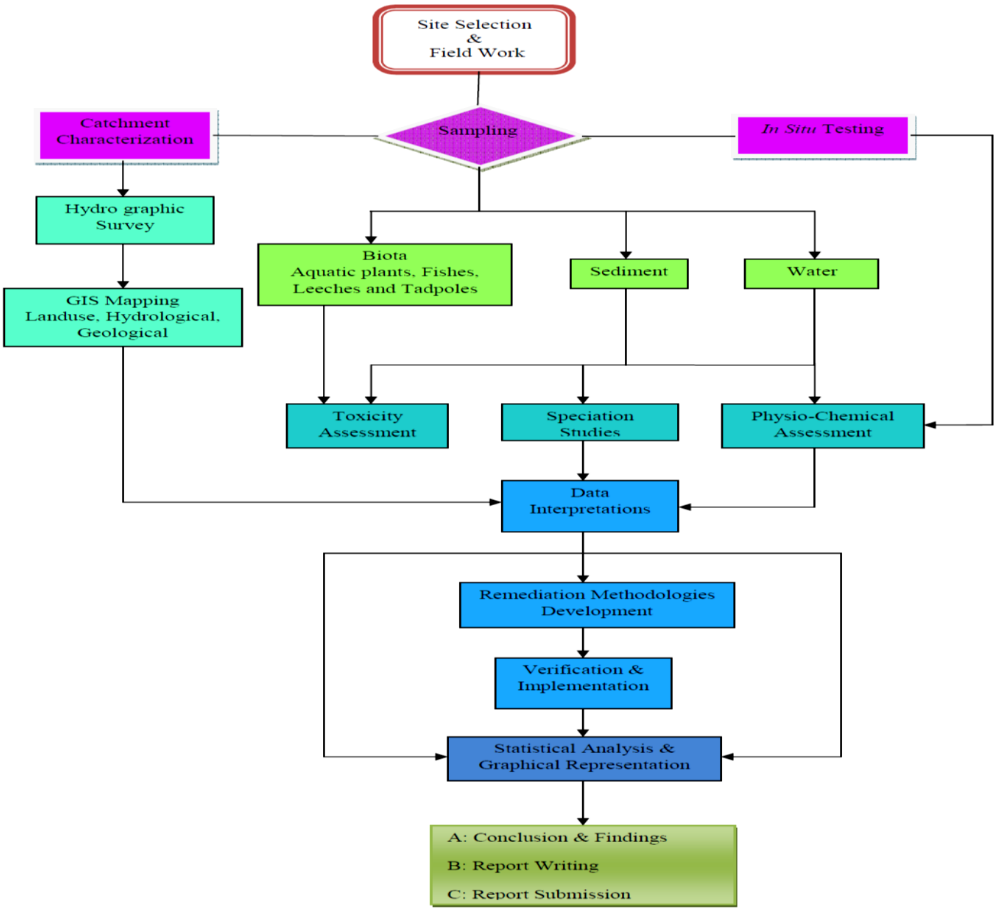
Test Type	Chemical Type	Compound
Chemical	Beta Blockers	Atenolol, Metoprolol, Propanolol,Sotolol
Chemical	Antidepressants	Citalopram desmthyl- citalopram, Venlafaxine desmthyl-venlafaxine
Chemical	Disinfectants	Triclosan, Triclocarban
Chemical	Antibiotics	Ciprofloxacin, Erythromycin, Sulfamethoxazole, Sulfapyridine, Trimethoprim
Chemical	Natural Estrogens	a-estradiol, a-ethinylestradiol, Estrone
Chemical	Industrial Estrogens	Nonylphenol, Octylphenol, Bisphenol A
Chemical	Musks	Celestolide, Phantolide, Tonalide, Traseolide, Cashmeran, Galaxolide, Musk Ketone, Musk Xylene
Chemical	Perflourinated Compounds	Perflourooctanoate, Perflourooctane, Sulfonate
Chemical	PBDEs	PBDE 28, PBDE 33, PBDE 47, PBDE 99, PBDE 100, PBDE 153, PBDE 154, PBDE 183, PBDE 209
Chemical	Acidic Pharmaceuticals	Acetaminophen, Gemfibrozil, Ibuprofen, Naproxen
Chemical	Basic/Natural Pharmaceuticals	Caffeine, Carbamazapine
Toxicity Tests		
Microbial Tests		

6.4 Remediation and Removal

Once a more complete picture describing the sources of emerging contaminants to municipal wastewater is obtained, reduction strategies can be investigated. If point sources for specific classes of emerging contaminants are identified, on-site pretreatment should be investigated. A small membrane bioreactor or similar on-site treatment system could remove contaminants from a concentrated effluent source such as a large hospital or slaughterhouse facility. On-site treatment may provide a viable option for contaminant removal if there are sources identified with high enough contaminant loadings to make practical and economical. If there are no specific point sources for the rest of the emerging contaminants studied, limiting the entry of these contaminants to the municipal wastewater system will be difficult. Removal strategies may have to be focused on enhancing the removal efficiencies of the wastewater treatment plants. Once the fate of the remaining emerging contaminant classes has been analyzed, methods to increase the removal capacity of treatment plants can be investigated. For compounds such as beta-blockers and disinfectants where biological degradation is thought to be the primary removal mechanism, further research should investigate the differences in operating conditions between the three different wastewater treatment plants to determine why there was found to be

differences in the removal efficiencies specifically around the aeration basin where aerobic degradation occurs. Increased removal capacity may also be added by constructing tertiary processes to existing facilities such as membrane bioreactors, sand filters, and rotating biological contactors. As scientists continue to investigate the prevalence of emerging contaminants in the environment and their effects on the aquatic ecosystem, there will be increased pressure to find solutions and methods in order to reduce the loading these contaminants into the natural world. By gaining a better understanding of the sources and fate of emerging contaminants in municipal wastewater, removal strategies can be developed mitigating the risk these compounds pose to both humans and the environment.

7. RESEARCH METHODOLOGY FLOWCHART



7.1 Expected Research Outputs

This research has a significant impact as the demonstrable contribution to society and the economy. Below is brief introduction of the potential range of impacts that can be generated from present research.

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