Special Edition

CAROLLO ENGINEERS

research SOLUTIONS

A Publication of the Carollo Research Group

Endocrine Disrupting Chemicals and Pharmaceuticals and Personal Care Products:

A Closer Look at Occurrence, Relevance, and Treatment

In this **Issue**

INTRODUCTION

Endocrine Disrupting Chemicals and Pharmaceuticals and Personal Care Products:

A Closer Look at Occurrence, Relevance, and Treatment

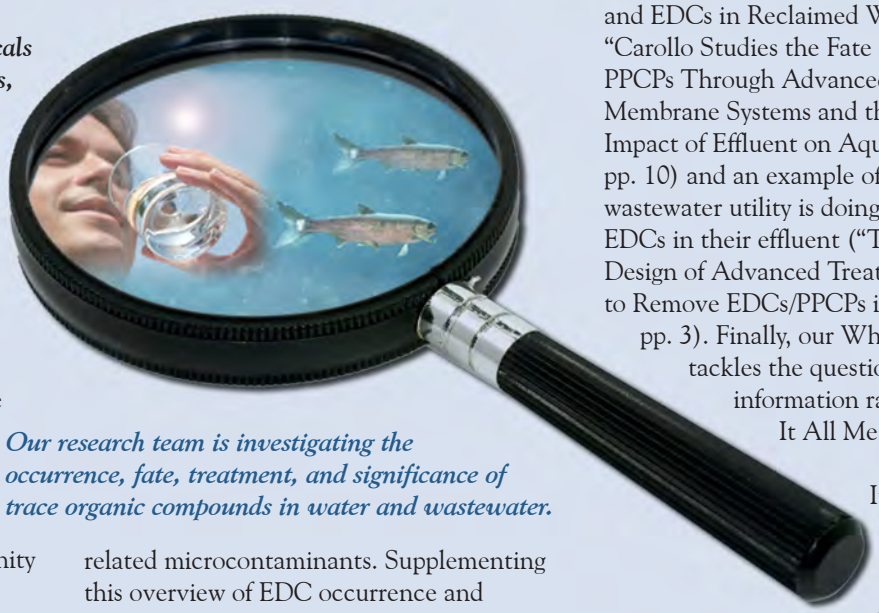
***March 9th, 2008** - The Associated Press reports that a vast array of pharmaceuticals — including antibiotics, anti-convulsants, mood stabilizers, and sex hormones — have been detected in the drinking water supplies of at least 41 million Americans.*

Recent press releases and media attention have raised substantial concerns among the public, politicians, and regulators about the potential implications of the presence of endocrine disrupting chemicals (EDCs) and pharmaceuticals and personal care products (PPCPs) in water for human health and the environment. As a result, the water and wastewater community faces many questions, including:

1. Are there potential human health and/or aquatic impacts from these trace contaminants?
2. What is the appropriate level of treatment?
3. What technologies can we use?
4. How much will it cost?

This special edition of *Research Solutions* was put together by Carollo, working with some of its research partners in emerging contaminants, to provide water and wastewater utility managers with up-to-date information on the important issues of occurrence, impact, and treatment of these chemicals as it pertains to drinking water, wastewater, and reclaimed water.

In this issue's Feature Story ("Occurrence and Impact of Endocrine Disrupting Chemicals in Water and Wastewater," pp. 6), Dr. Shane Snyder distills and simplifies the confusing mountain of data published on EDCs to help readers better understand where they have been observed, what we know and don't know about their effects on the environment and in humans, and how Federal and State regulators currently approach managing EDCs and related microcontaminants. Supplementing this overview of EDC occurrence and effects are four Project Updates highlighting some current Carollo-led projects that are advancing our understanding of how best to mitigate EDCs in water, wastewater, and reuse water. This includes both on-going research into the efficacy of various treatment methods ("Removal of NDMA, EDCs, and PPCPs from South Delta Water," pp. 4; "Filtration and Destruction of PPCPs and EDCs in Reclaimed Water," pp. 5; and "Carollo Studies the Fate of EDCs and PPCPs Through Advanced Wastewater Membrane Systems and the Biological Impact of Effluent on Aquatic Organisms," pp. 10) and an example of what one wastewater utility is doing to manage EDCs in their effluent ("Testing and Design of Advanced Treatment Facilities to Remove EDCs/PPCPs in Park City, UT," pp. 3). Finally, our What's New section tackles the question all this new information raises: "What Does It All Mean?," pp. 12.



Our research team is investigating the occurrence, fate, treatment, and significance of trace organic compounds in water and wastewater.

It is our intent that this Special Edition of *Research Solutions* provides you with a snapshot of some of the latest information on EDCs that may be useful for water and wastewater utility managers as they go forward in assessing whether or not this is an issue for your utility, and if so, how you might approach managing it.

— Andrew Salveson, *Special Editor*

What are "pharmaceutically active compounds"?

Pharmaceuticals include human and veterinary drugs, both prescription and over-the-counter medications. Examples of anthropogenic pharmaceuticals that have been measured in the environment include antibiotics, antidepressants, heart medications, anti-convulsants, pain relievers, oral contraceptives, and x-ray contrast media. The term "pharmaceutically-active compounds" (PhACs) is sometimes used in recognition of the fact that both the parent compounds and/or their biologically active degradation products may be found in the environment.

PhACs and personal care products (PCPs) are often grouped together under the acronym "PPCPs." Most PPCPs are active ingredients or preservatives in cosmetics, toiletries, fragrances, sunscreen lotions, and insect repellents. Endocrine disrupting chemicals (EDCs) are substances that can interfere with the actions of hormones in the body. Hundreds of chemicals have been identified to be EDCs or potential EDCs, including industrial chemicals, pesticides, PPCPs, combustion byproducts (natural and anthropogenic), metals, inorganic ions, organic substances naturally found in plants and fungi, and hormones excreted by humans and other animals.

Testing and Design of Advanced Treatment Facilities to Remove EDCs/PPCPs in Park City, UT

Background and Design Objectives

Snyderville Basin Water Reclamation District (SBWRD) in Park City, UT, operates two wastewater treatment facilities, East Canyon and Silver Creek. Measurable concentrations of EDCs and PPCPs were found in both the influent and effluent from these facilities; SBWRD is concerned about the potential hormonal impacts of treated effluent on sensitive fish species. As part of a design project currently underway to upgrade and expand both facilities, Carollo was asked to look for ways to incorporate EDC/PPCP treatment.

Carollo conducted bench- and pilot-scale studies of three EDC/PPCP treatment technologies: granular activated carbon (GAC) adsorption, ozone/peroxide advanced oxidation, and ultraviolet light (UV)/peroxide advanced oxidation. Reverse osmosis membranes were not considered due to budget and concentrate disposal limitations. Carollo evaluated the treatment effectiveness of each technology and estimated the cost of full-scale implementation.

Expansion plans for East Canyon are to increase plant capacity from 4 to 7.2 mgd. The plant currently uses both biological and chemical processes to meet a total phosphorous limit of 0.1 mg/L. Chemical phosphorous removal is accomplished by dosing coagulant upstream of a tertiary filter. In order to improve the reliability of this filtration process and treat to the eventual total maximum daily load (TMDL) target of 0.05 mg/L, MF membranes will replace the existing granular media filters. Rather than demolishing the existing filters, Carollo proposed replacing filter media with GAC and converting them to contactors for EDC/PPCP removal. This method, coupled with the membrane filtration and existing UV disinfection, may be the most cost effective means of incorporating EDC/PPCP treatment into the current project. Silver Creek will be expanded from 2 to 3.7 mgd and upgraded to match the process used at East Canyon.

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Results and Project Direction

Bench- and pilot-scale testing of GAC contactors was conducted at East Canyon using tertiary effluent from the existing granular media filters. Three GAC columns were dosed at a filter loading rate of 3.5 gpm/ft^2, which matches the rate of the full-scale filters, and sampled once a week over a 6-week study period. Samples were also sent to labs for bench-scale testing of advanced oxidation processes (ozone, ozone/peroxide, UV, and UV/peroxide). Applied Process Technology, Inc. (Pleasant Hill, CA) performed testing on the plant effluents using three different ozone doses (5, 10, 15 ppm) and two ozone/peroxide doses (peroxide:ozone molar ratios of 0.35 and 0.70 at an ozone dose of 5 ppm). UV and UV/peroxide testing was performed by Dr. Karl Linden at Duke University (Durham, NC) [he is now at University of Colorado-Boulder]. Plant effluent was treated at a UV fluence of 80, 200, and 400 mJ/cm^2 and for each fluence three different doses of peroxide (2, 5, 10 ppm) were added.

Pilot-scale carbon filters for EDC/PPCP removal.

For each removal technique, sampling of treated and untreated effluent was designed to quantify and compare EDC concentrations, estrogenic activity (by E-Screen bioassay), and water quality parameters (biological oxygen demand [BOD], total suspended solids [TSS], total organic carbon [TOC], ultraviolet transmittance at 254 nm [UVT], and/or coliform counts). In an attempt to reduce the cost of analytical chemistry and focus on some of the EDC compounds that may contribute most to the potential estrogenicity of fish, only the first phase of an analytical quantification method (USGS Method 2, LC/MS APCI positive-ion mode only) was used for pilot and bench-scale analyticals. This narrowed the spectrum of possible EDC/PPCP tests to just six compounds: Carbamazepine, Estrone, Estradiol, Ethinyl Estradiol-17 alpha, Progesterone, and Testosterone. Results are summarized as follows:

- Plant effluent concentrations for the six EDCs of interest ranged from 146 to <1 ng/L.
- Estrogenic activity of the plant effluent as measured by E-screen is typically 1 ng/L of estradiol equivalents.
- EDCs and estrogenic activity in GAC treated effluent were below detection or were reduced by an order of magnitude.
- The lowest ozone dose of 5 ppm, without any peroxide addition, reduced estrogenic activity below the detection limit.
- Although there were some inconsistencies in the data, a trend of estrogenic activity removal was seen at a UV fluence of 400 mJ/cm^2 with peroxide addition.

Ozone oxidation was recommended as the best EDC treatment technology based on life-cycle cost, even though it required the highest initial capital investment. However, given the results for GAC that show it is an effective treatment method and the ability to use existing infrastructure, SBWRD has indicated that they will exchange filter media and begin treating for EDCs/PPCPs at the completion of the expansion project.

NF pilot skid at Contra Costa Water District, CA.

Removal of NDMA, EDCs, and PPCPs from South Delta Water

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Background and Research Objectives

Various trace organic compounds are finding their way into the nation's water supply. The Sacramento-San Joaquin River Delta System (Delta) is one example where the presence of these compounds is of growing concern. The Delta is a vital source of drinking water for more than 23 million Californians. Supplies from the south Delta are conveyed through the South Bay Aqueduct and the Contra Costa Canal, serving more than 2.5 million people in the Bay Area, much of the time as an unblended source of drinking water.

Deterioration of the Delta source water quality due to xenobiotic inputs (*e.g.*, agricultural drains and wastewater effluents), and increased freshwater diversions is a growing concern for drinking water users. In particular, users must consider the possibility of significantly deteriorated water quality during severe drought events. As such, Delta water utilities must be prepared for both the presence of existing contaminants (*e.g.*, pesticides and nutrients) at significantly greater concentrations than currently observed, as well as contaminants of emerging concern (*e.g.*, EDCs and PPCPs). An additional concern with treatment of Delta waters is the formation of disinfection byproducts (DBPs) including trihalomethanes (THMs), haloacetic acids (HAAs), N-Nitrosodimethylamine (NDMA), and other nitrosamines. Delta utilities need to understand the treatment effectiveness of existing processes as well as advanced treatment processes that may be implemented in the future to remove these chemicals. To meet this need, a diverse research team, including: the Contra Costa Water District, Carollo Engineers, Trent University, the University of Colorado at Boulder, and the University of Toronto, as well as numerous specialists, is evaluating the performance of advanced water treatment technologies with respect to the destruction/removal of trace organic compounds and the formation of DBPs. The California Department of Public Health and the California Department of Water Resources are providing funding and oversight for the project.

The treatment technology combinations under investigation include: ozone/biologically active filtration (BAF) [biologically active sand and granular activated carbon media], ozone/peroxide/BAF, and nanofiltration (NF) at Contra Costa Water District's Bollman Water Treatment Plant. The evaluation of ozone followed by BAF will provide a baseline understanding of the effectiveness of the existing full-scale treatment processes (Phase 1). Then the effectiveness of advanced oxidation with ozonation and peroxide will be investigated (Phase 2). Finally, NF following full-scale ozonation and BAF will be evaluated (Phase 3). In all test phases EDCs, PPCPs, and salts (bromide, iodide, sulfate, and chloride) will be added to the pilot feed water. Removal efficiencies for target EDCs and PPCPs will be quantified in the various treatment trains. Subsequent tests for DBP formation will be performed in bench-scale reactors for various ultraviolet (UV) treatment and chlorination schemes (dosing free chlorine or chloramines).

Results and Implications

Pilot testing was completed in July 2008. The data gathered from this study will provide information on the effectiveness of the various treatment trains for removing select trace organic compounds from Delta water. Results from the bench-scale DBP tests will provide side-by-side comparisons of expected DBP formation from different combinations of UV, free chlorine, and chloramination treatments. These results will be used for planning, design, and operation of existing and future disinfection systems.

Ozone pilot skid at Contra Costa Water District, CA.

Filtration and Destruction of PPCPs and EDCs in Reclaimed Water

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Background and Research Objectives

Although some microconstituents can persist through wastewater treatment (Stackelberg *et al.*, 2004; Gomez *et al.*, 2007), current research suggests that advanced treatment technologies can effectively remove a range of PPCPs (Tang *et al.*, 2006) to concentrations below human health risk levels (Snyder *et. al,* 2007). In addition, some research shows that advanced treatment technologies following conventional wastewater treatment can significantly reduce the risk to aquatic organisms (Schwatter *et al.*, 2007).

To better understand the most efficient ways to destroy EDCs and PPCPs in wastewater effluent, the WateReuse Foundation, the Southwest Florida Water Management District (SWFWMD), and the U.S. Bureau of Reclamation funded a research team from Carollo, Duke University, and the U.S. Department of Agriculture to conduct WRF 02-009, *Innovative Treatment Technologies for Reclaimed Water*. The project included detailed bench- and pilot-scale investigations to find and demonstrate emerging (market-ready) reclaimed water treatment technologies that could robustly destroy pathogens and PPCPs at a cost substantially below that of reverse osmosis (RO) (Wade *et al.*, 2008). Technologies investigated included media and membrane filtration, ozone, ozone/peroxide, peracetic acid/ultraviolet light (UV), hydrogen peroxide/UV, and titanium dioxide/UV.

Results and Implications

Significant results include:

- Substantial removal of EDCs and PPCPs by full-scale microfiltration (MF) and sand filtration, including 50 to 80% (or greater) reduction of 4-nonylphenol, bisphenol-A, and triclosan, and limited removal of DEET.
- Ozone after media filtration and after MF provided ≥90% destruction of the majority of the target compounds and hormonal activity (as measured by estradiol equivalency [EEQ]) at low ozone doses (<6 mg/L), while meeting the stringent disinfection criteria for reclaimed water for various states (*e.g.*, CA).
- Peracetic acid (PAA), when combined with medium-pressure UV, demonstrated robust removal of many trace organic compounds.
- UV disinfection at doses of 80 to 100 mJ/cm^2, when combined with hydrogen peroxide, showed ≥50% reduction of many of the target compounds.
- The TiO_2/UV (Photo-Cat) process in the range of power tested (0.1 to 0.6 kW per gpm) effectively oxidized dissolved organic compounds, provided complete disinfection of coliforms (~5-log removal) and MS-2 virus (>6-log reduction), destroyed trace organic chemicals such as triclosan and carbamazepine, and substantially reduced estradiol activity in the water based upon *in-vitro* bioassays.
- The TiO_2/UV process effluent had a particle count equivalent to distilled water and a UV transmittance at 254 nm (UVT) of nearly 90% (increasing from an influent of 67%).
- Most compelling, this work demonstrated that the use of MF or UF membranes with low-dose ozone

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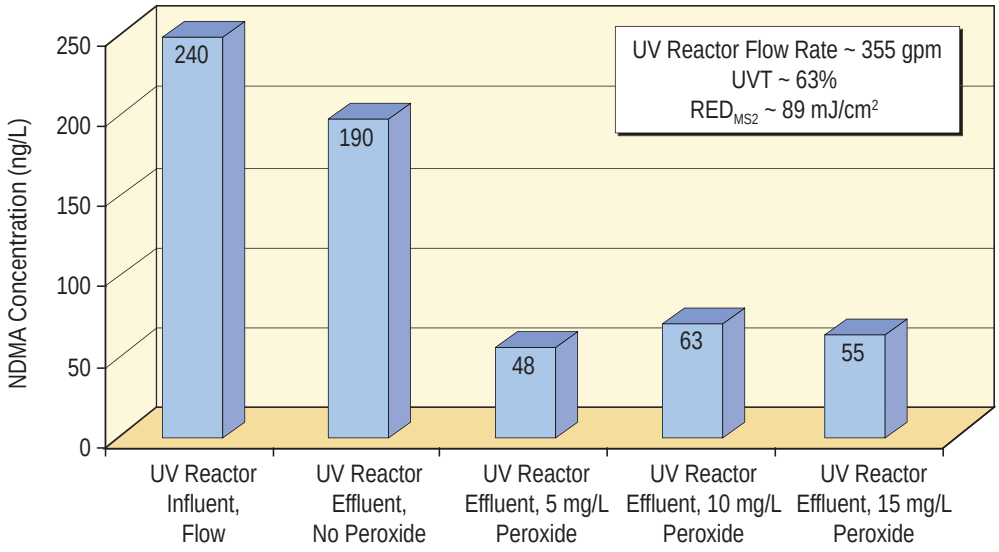
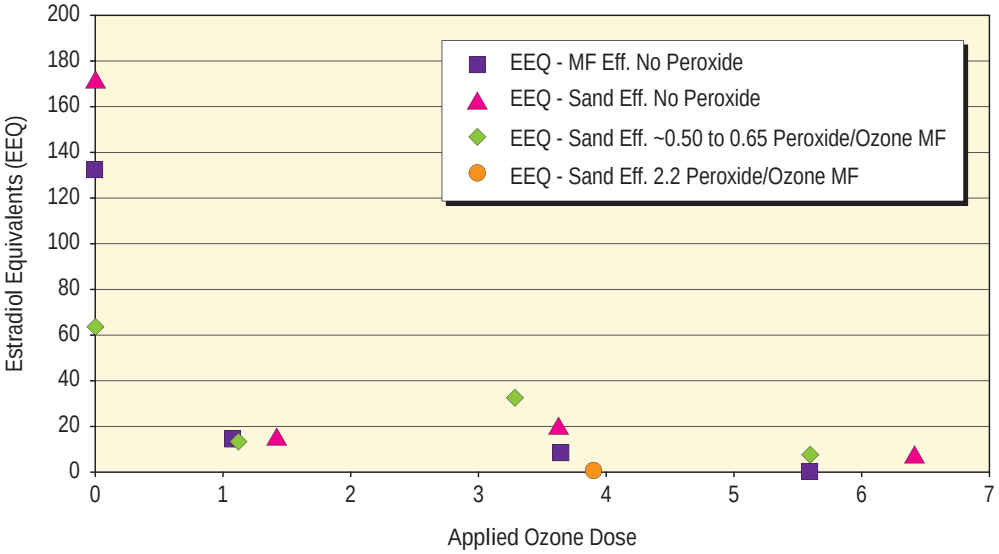


NDMA destruction by MP UV with hydrogen peroxide.



Destruction of hormonal equivalency by different treatment process combinations.

FEATURESTORY

Occurrence and Impact of Endocrine Disrupting Chemicals in Water and Wastewater

By Shane A. Snyder, Ph.D. [Total Environmental Solutions, Inc.]

Endocrine Disrupting Chemicals

EDCs are substances that interfere with the functioning of the endocrine system in humans or other animals. Alternative terms used to describe these chemicals include: 'endocrine disruptors,' 'hormonally active agents,' and 'endocrine-active substances.' Currently, there is no consensus among experts regarding the definition of an EDC or the criteria that should be used to determine whether a chemical is or is not an EDC. Some definitions require that an effect must be demonstrated *in vivo* (*i.e.*, in a live animal), while others stipulate only that the potential for an effect be demonstrated, such as through *in vitro* receptor binding (binding of test chemicals with estrogen receptors) or structure-activity relationships ([SARs] predictable impact based upon molecular structure). Other definitions seek to distinguish adverse effects from merely compensatory responses (non-adverse but measurable effects) (USEPA-EDSTAC, 1998; World Health Organization, 2002). This has also been a source of controversy.

Hundreds of chemicals have been implicated as potential EDCs based on a variety of criteria (Institute for Environment and Health, 2005). While screening-level evidence such as SARs, *in vitro* receptor binding activity, and certain short-term *in vivo* tests might suggest the potential for endocrine disruption, such effects are often not demonstrated in the more definitive *in vivo* tests, (*e.g.*, tests conducted on intact animals or on multiple generations of exposed animals). Standardized test methods are generally unavailable. At this time, only certain *in vivo* bioassays conducted with intact animals and using appropriate protocols (*e.g.*, encompassing susceptible life stages) provide data that are useful for risk assessment. Few chemicals have been subjected to this type of testing due to the cost and time required to conduct them. Most chemicals have not been tested for endocrine activity by any means.

The U.S. Environmental Protection Agency (USEPA) established the Endocrine Disruptor Screening Program (EDSP) to develop a battery of standardized toxicity tests that can be used to determine whether a particular chemical is an EDC by USEPA's definition. The program focuses exclusively on chemicals that act by interfering with estrogen, androgen, or thyroid action (USEPA-EDSP, 2008); these are the best-characterized modes of action. However, EDCs may also interfere with the functions of other hormones (World Health Organization, 2002). The EDSP will use a two-tiered testing strategy, with Tier 1 consisting of screening-level tests and Tier 2 consisting of *in vivo* bioassays that will generate data suitable for use in risk assessments (USEPA-EDSP, 2008). This process is not yet complete, but Tier 1 screening of an initial set of chemicals is expected in Fall 2008 (USEPA, 2007).

Sources and Occurrence in the Water Cycle

Known and potential EDCs encompass a wide variety of chemicals and a diversity of structures. They include both natural and synthetic chemicals (Table 1). EDCs arising from natural sources include hormones excreted by humans and other animals, substances found in plants (phytoestrogens, phytosterols) or fungi (mycoestrogens), metals, inorganic ions, and byproducts of natural combustion processes (*e.g.*, volcanic activity, forest fires) (Institute for Environment and Health, 2005; Myers, 2008; World Health Organization, 2002). Some of these EDCs occur normally in the environment or in dietary items, but their concentrations may be elevated due to human activities. For example, metals may be mobilized in the environment during mining (Wilkin, 2007), and endocrine-active phytosterols may be released to water in effluents from processing of forest products (MacLatchy *et al.*, 1997; Mellanen *et al.*, 1996). Synthetic EDCs include certain biocides and their degradates (pesticides, herbicides, and fungicides), PPCPs including veterinary and human drugs, industrial chemicals and intermediates or byproducts from their production and their environmental degradates, and combustion byproducts from human activities such as burning of fossil fuels and incineration of industrial and municipal waste (Institute for Environment and Health, 2005; Myers, 2008; World Health Organization, 2002).

EDCs can originate from numerous sources and enter the environment by many routes. Effluents from municipal wastewater treatment plants (WWTPs) have been implicated as major contributors of EDCs to surface waters (Anderson, 2005). WWTPs receive EDCs from sources including plant material, plastics, items treated with fire retardants, cleaning products, pesticides, other household chemicals and consumer products, hormones excreted by humans, and PPCPs excreted or washed from the body and flushed to the sanitary sewer. WWTPs might also receive industrial or hospital effluents and stormwater runoff streams that contain EDCs. Although wastewater treatment processes can remove some EDCs, recalcitrant chemicals may remain at detectable levels in effluents discharged to surface water or in reclaimed water used for groundwater recharge. These contaminants may be diluted, sequestered (*e.g.*, in sediment), or degraded by physical or biological processes, but some are detected in the environment due to their persistence or relatively constant loading.

WWTP effluents and reclaimed water are not the only sources of EDCs to the environment. Other potential sources include private septic systems (Swartz *et al.*, 2006), untreated stormwater flows and urban runoff (Boyd *et al.*, 2004), industrial effluents (Kosaka *et al.*, 2007), landfill leachate (Coors *et al.*, 2003), discharges from fish hatcheries and dairy facilities (Kolodziej *et al.*, 2004), fish spawning in natural waters (Kolodziej *et al.*, 2004), runoff from agricultural fields and livestock enclosures (Orlando *et al.*, 2004), and land amended with biosolids or manure (Hanselman *et al.*, 2003; Khanal *et al.*, 2006).

Table 1. Examples of Known or Potential Endocrine Disrupting Chemicals

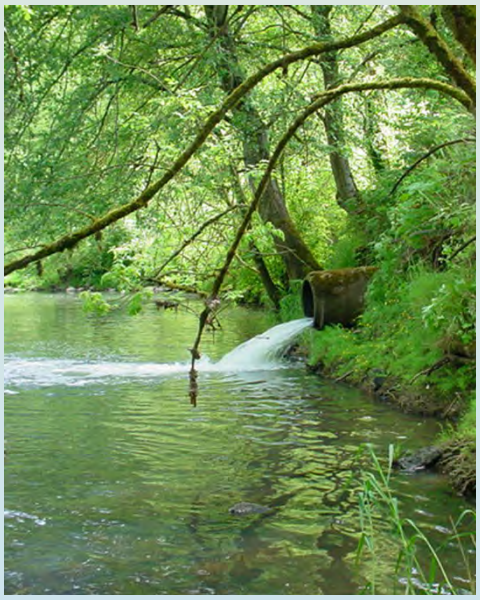
Chemical Class	Representative Chemicals
Naturally-occurring EDCs	
Hormones	Estradiol, estrone
Phytoestrogens and plant sterols	Genistein, ß-sitosterol
Mycoestrogens	Zearalenone
Metals	Arsenic, cadmium, lead, mercury
Inorganic ions	Perchlorate, thiocyanate
Combustion byproducts	Dioxins, certain PAHs
Synthetic EDCs	
Biocides or their degradates	Atrazine, DDT (or DDE), tributyltin
PPCPs	Ethynylestradiol, trenbolone
Industrial chemicals, intermediates, and degradates	PCBs, bisphenol-A, octylphenol
Combustion byproducts	Dioxins, certain PAHs

EDCs, endocrine disrupting chemicals; PAHs, polycyclic aromatic hydrocarbons; PCBs, polychlorinated biphenyls; PPCPs, pharmaceuticals and personal care products.

Various EDCs have been reported to occur in WWTP effluents, surface water, groundwater, reclaimed water, and drinking water, usually at concentrations in the ng/L (0.000000001 g/L) range. In general, EDCs are reported to occur with greater frequency and at higher levels in WWTP effluents than in reclaimed water or drinking water. Dilution and environmental degradation are responsible for smaller concentrations and less frequent detections in surface water. The limited data available to date indicate that EDCs originating from municipal WWTPs generally occur infrequently and at exceedingly small levels in finished drinking water because they are diluted and undergo degradation in the environment and then must survive drinking water treatment processes and distribution to remain in potable water at the tap.

Implications for Aquatic Life

There is a substantial and growing body of evidence indicating that EDCs at levels found in some WWTP effluents can cause endocrine disruption in fish and other aquatic life, with the literature suggesting that some EDCs at or above 0.1 ng/L can induce endocrine-mediated changes in aquatic life (Purdom *et al.*, 1994; Vanderford *et al.*, 2003). This issue first gained public attention when male fish collected downstream of WWTPs in the United Kingdom (U.K.) were found to have elevated levels of vitellogenin, a female-specific egg yolk protein, in their blood. Vitellogenin induction in male fish is a symptom of exposure to estrogens from external sources but generally is not considered to be an adverse effect. Later studies suggested a link between exposure to WWTP effluents and adverse or potentially harmful effects on the reproductive organs and fertility of fish (Jobling *et al.*, 2002; Jobling and Tyler, 2003). The findings in the U.K. studies spurred research in other European countries (Petrovic *et al.*, 2002; Diniz *et al.*, 2005), North America (Bevans *et al.*, 1996; Folmar *et al.*, 1996, 2001; Giesy *et al.*, 2003; Hemming *et al.*, 2004; Nichols *et al.*, 1999; Patiño *et al.*, 2003; Schoenfuss *et al.*, 2002; Snyder *et al.*, 2004; Woodling *et al.*, 2006), and elsewhere where WWTP effluents have been implicated in endocrine-related effects on fish.



Outfall from a wastewater treatment plant can be a source of EDCs to the environment.

WWTP effluents contain a mixture of known or potential EDCs. In most cases researchers have been unable to pinpoint the specific chemicals responsible for effects indicating endocrine disruption in exposed fish. Estradiol, estrone, ethynylestradiol, nonylphenol, octylphenol, alkylphenol ethoxylates, and bisphenol A have been identified as likely causes (Purdom *et al.*, 1994; World Health Organization, 2002) based on their concentrations in wastewater effluents and their potency in laboratory studies. Natural hormones produced in the bodies of humans and other animals (*e.g.*, estradiol and estrone) and synthetic hormones intended to mimic the actions of endogenous hormones (*e.g.*, the oral contraceptive ingredient ethynylestradiol) are of particular concern because they are potent at very small concentrations and are commonly detected in WWTP effluents.

While hormonal disruption of aquatic life by wastewater-derived EDCs has clearly been demonstrated, limited information exists on the possibility of long-term effects on aquatic life populations. This is an area for further research.

Implications for Human Health

Although there are well substantiated links between environmental exposure to EDCs and effects in fish and wildlife, there is little evidence to suggest that typical low-level environmental exposures to EDCs (including EDCs in WWTP effluent, reclaimed water, and drinking water) have had any adverse effects on human health (World Health Organization, 2002). However, the science of endocrine disruption is relatively new, as is research into exposure to EDCs and the potential human health consequences.

Reports of endocrine disruption in fish collected from source waters for drinking water treatment plants (*e.g.*, Blazer *et al.*, 2007) and detection of wastewater-associated EDCs in these waters commonly trigger consumer concern. However, there are important differences in exposure to wastewater contaminants between fish and humans. Fish may be immersed in effluents

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at their point of entry into surface water, where concentrations are greatest. They can take up contaminants directly across their body surfaces, particularly the gills. Fish can also be exposed to EDCs and other effluent contaminants that accumulate in their food or that are associated with particulate material and sediments. In contrast, people tend to receive little direct exposure to EDCs in WWTP effluent, so concerns related to potential human health effects generally center around drinking water contamination. EDCs discharged in WWTP effluents or reclaimed water undergo dilution, environmental degradation, and water treatment processes that can substantially reduce their concentrations before they reach the tap.

Regulations

Although some chemicals that might be considered to be EDCs are regulated in WWTP effluent for the protection of aquatic organisms, these rules are not based on endocrine modes of action except to the extent that they are captured in effects on more traditional ecotoxicologic endpoints (*e.g.*, mortality or reproduction) (USEPA, 2005). Likewise, chemicals that might be classified as EDCs are federally regulated in drinking water, but not on the basis of their potential to cause endocrine disruption. In Massachusetts, the level of perchlorate in drinking water is regulated on the basis of its potential to act as an EDC (by interfering with thyroid function) (Massachusetts DEP, 2006), but to date no other state has regulated any drinking water contaminant as a putative EDC. For indirect potable reuse projects in California, the California Department of Public Health (CDPH) has mandated monitoring and reporting of EDCs and pharmaceuticals, but not their removal (California Department of Public Health, 2008).

Conclusion/Summary

Trace levels of EDCs and pharmaceuticals are ubiquitous in municipal wastewater effluents. In some cases, steroid hormones have been implicated as the primary source of estrogenicity in effluents; however, some synthetic organic chemicals have the ability to mimic endogenous hormones. Advanced water treatment processes can greatly reduce the concentration of organic contaminants in water. While analytical technology is capable of identifying and quantifying sub-ng/L levels of organic contaminants, it is prudent to determine the toxicological relevance of these emerging contaminants in order to establish meaningful treatment goals.

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Carollo Studies the Fate of EDCs and PPCPs Through Advanced Wastewater Membrane Systems and the Biological Impact of Effluent on Aquatic Organisms

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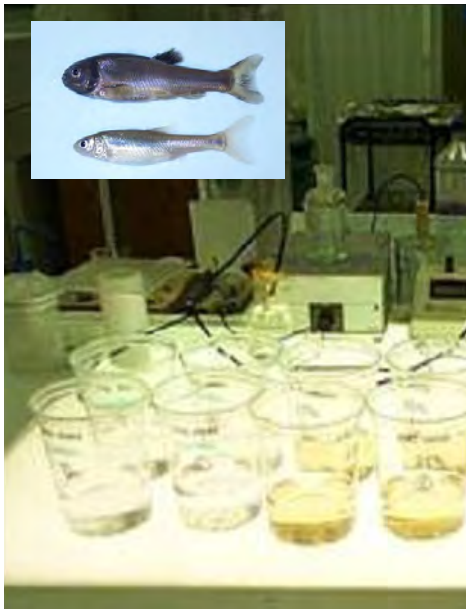
Background and Research Objectives

Population growth in South Florida has put pressure on valuable fresh water resources. Various utilities are investigating the use of advanced wastewater treatment processes to indirectly supplement existing potable water supplies. However, concerns have been raised by the public regarding presence of EDCs and PPCPs in wastewater effluents and the potential impact of these compounds on aquatic life and human health.

In response to some of these concerns, the WateReuse Foundation (WRF) and the South Florida Water Management District (SFWMD) funded WRF 06-019, *Monitoring for Microconstituents in an Advanced Wastewater Treatment (AWT) Facility and Modeling Discharge of Reclaimed Water to Surface Canals for Indirect Potable Use*. This research, led by Carollo and SFWMD, with help from the University of Florida at Gainesville, the City of Plantation, FL, Hazen and Sawyer, and others, monitored and evaluated water quality through advanced wastewater treatment facilities and modeled the fate and transport of various compounds through a canal system and into groundwater supply wells.



Fish biossays at the University of Florida at Gainesville.

Results and Implications

The removal of select EDCs/PPCPs was tracked across two treatment trains: Train 1: membrane bioreactor (MBR) + reverse osmosis (RO); Train 2: activated sludge(AS) + denitrifying filtration (DNF) + ultrafiltration (UF) + and RO. Removal of EDCs/PPCPs by MBRs and UF processes was demonstrated. Significant removal of EDCs/PPCPs by RO was observed for all compounds, with most removed to levels below the analytical detection limits (1-25 ng/L). The RO performance data were in accordance with other published data (Drewes *et al.*, 2006).

Furthermore, the concentrations of PPCPs were correlated with toxicity tests and both tissue culture and live fish bioassays. The results indicated that the EDCs and PPCPs in the RO permeate caused no toxicity. Detailed results include:

- The toxicity tests were chronic definitive tests on the waterflea *Ceriodaphnia dubia* and the fathead minnow *Pimephales promelas* following EPA 821-R-02-013 Test Method 1002.0 and 1000.0, respectively. The results of tests on MBR and UF permeate indicated that there were no significant survival differences between the control (deionized water) and the treated samples for *P. promelas* and C. *dubia*, except that the survivability of C. *dubia* was low in MBR permeate in one of the five sampling events. These results suggest that the MBR permeate and UF permeate did not have significant toxic effects on the survival of *P. promelas* and C. *dubia*.
- Some toxic effects were observed in the RO permeate, but they did not appear to have been caused by PPCPs. The survival of *P. promelas* and C. *dubia* in the RO permeate was poor for one sampling event. Because PPCPs in the RO permeate were all non-detectable (<1 ng/L in most cases), the observed toxic effects were likely caused by other compounds added or generated in the RO permeate. These included ammonia, chloramine, and anti-scalant (used for maintaining the membrane system). Dechlorination (quenching) of chloramine with sodium thiosulfate reduced and delayed toxicity effects. Further experiments without ammonia, chloramine, and antiscalant eliminated the toxic effects in the RO permeate and significantly increased the survival of *P. promelas* and C. *dubia*. Similar results were found for the test with antiscalant alone, suggesting that antiscalant did not exert toxic effects on C. *dubia*. These results indicate that dechlorination or other quenching methods should be used to remove chloramines or ammonia in these AWT facilities to minimize their toxic effects.
- *In vitro* estrogen screen (E-Screen) bioassays were conducted to evaluate the endocrine disrupting potential of the treated permeate. In these bioassays, the estradiol equivalents (a measure of the hormone-inducing activity of a given solution) in all the RO permeate were below the detection limit (0.03 ng/L), although they were detected in secondary effluent (0.11 ng/L), DNF filtrate (0.12-0.23 ng/L), MBR permeate (0.12-0.18 ng/L), and UF permeate (0.14-0.21 ng/L). The results of the E-Screen bioassay indicated that the permeate did not produce a significant estrogenic response in MCF-7 cells, a breast cancer cell line that proliferates in response to estrogenic activity.
- The *in vitro* yeast estrogen screen (YES) bioassay was conducted to evaluate the endocrine disrupting potential in the treated effluents. In the YES bioassay, yeast cells are transformed to contain human estrogen receptors. The results

This Advanced Membrane Pilot was used for the WRF project in Plantation, FL.

showed that estradiol equivalents were not detected in MBR permeate, UF permeate, and RO permeate, although they were detected in the secondary effluent (0.22 ng/L) and DNF filtrate (0.23 ng/L). The results suggest that MBR permeate and RO permeate may not possess endocrine disrupting potential, though *in vivo* assays provide a higher measure of confidence for such a conclusion compared to *in vitro* assays.

- *In vivo* assays, such as fathead minnow vitellogenin assays and steroid immunoassays, were conducted to measure the estrogenic activity of the treated effluents. There was no plasma vitellogenin induction in fish exposed to the MBR permeate and RO permeate, suggesting that the MBR permeate and RO permeate were not estrogenic as assessed in this bioassay.

All of these results suggested that RO permeate did not exert endocrine disrupting potential to tissue cultures or live fish (Zhou *et al.*, 2008).

A hydrodynamic and water quality model was also developed to track the fate and transport of a range of potential PPCPs from a surface water discharge point to groundwater supply wells. The Plantation sub-model was extracted from the Broward County model including the surface and groundwater features since they have a direct hydraulic connection to the proposed discharge location, the East Holloway Canal. The model area was determined with both the surface water basin divides and the groundwater capture areas. Boundary conditions for the model area were extracted from the Broward model results for all of the groundwater and the surface water boundaries in the Plantation sub-model. In general, the groundwater results followed the observed data closely while the surface water results deviated somewhat from the observed data. Three representative PPCPs (sulfamethoxazole, phenol, triclosan) were selected for the water quality model based on their susceptibility to photodegradation, sorption, and biodegradation, as well as their occurrence.

Conclusions

The results suggest that RO is most effective among membrane technologies if utilities plan to reach maximum removal of microconstituents and their endocrine disrupting potential. Although MBR and UF could partially remove microconstituents, only RO completely removed microconstituents from wastewater. In addition, no hormonal threats were found in RO permeate to tissue cultures and live fish, while some hormonal responses were detected in MBR permeate and UF permeate.

The observed toxicity due to membranes pretreated with chemicals suggests that dechlorination equipment may be necessary to minimize the impact on aquatic organisms if similar RO processes are designed and operated.

Hydrodynamic and water quality models can help us evaluate the fate and transport of microconstituents in reclaimed water discharged to surface canals and therefore are useful tools for designing or upgrading advanced treatment facilities for future indirect potable use of reclaimed water.

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Table 1. Concentrations of Microconstituents

Microconstituents (ng/L)	Treatment Trains				
	MBR/RO		AS/DNF/UF/RO		
	RO Permeate Sampling Dates				
	10/29/07	11/26/07	1/14/08	1/31/08	2/21/08
2,6-di-tert-butylphenol	<10	<10	<10	<10	<10
4-Methylphenol	<25	<25	<25	<25	<25
4-Nonyl Phenol	<25	<25	<25	<25	<25
Acetaminophen	<1	<1	<1	<1	<1
Alpha Chlordane	<10	<10	<10	<10	<10
Bisphenol A (BPA)	<25	<25	<25	<25	57
Caffeine	<25	<25	<25	<25	<25
Carbamazepine	<5	<5	<5	<5	<5
Carbaryl	<50	<50	<50	<50	<50
Chlorpyrifos	<25	<25	<25	<25	<25
N,N-diethyl-m-toluamide	<25	<25	<25	<25	<25
Diazinon	<25	<25	<25	<25	<25
Dieldrin	<25	<25	<25	<25	<25
Estradiol	<1	<1	<1	<1	<1
Estrone	<1	<1	<1	<1	<1
Ethinyl Estradiol -17 alpha	<1	<5	<5	<5	<5
Fluoxetine	<1	<1	<1	<1	<1
Gemfibrozil	NA	<1	<1	NA	<1
Ibuprofen	<1	3.1	<1	<1	<1
Iopromide	<5	<5	<5	<5	<5
Methyl Parathion	<25	<25	<25	<25	<25
Phenol	<100	<100	<100	<100	<100
Progesterone	<1	<1	<1	<1	<1
Sulfamethoxazole	<1	<1	<1	<1	<1
Testosterone	<1	<1	<1	<1	<1
Triclosan	<50	<50	<50	<50	<50
Trimethoprim	<1	<1	<1	<1	<1
Triphenylphosphate	<25	<25	<25	<25	<25
Tris(1,3-dichloro-2-propyl) phosphate	<25	NA	<25	NA	<25
Tris(2-butoxyethyl) phosphate	<100	<100	<100	<100	<100
Tris(2-chloroethyl) phosphate	<25	<25	<25	<25	<25

WHAT'S**NEW**

What Does It All Mean?

Concentrations of EDCs in reclaimed water and surface water do not appear to pose a public health threat, but wastewater effluent can cause detrimental effects like endocrine disruption in aquatic life.

Economic analyses associated with the projects described in this publication show that advanced treatment technologies can be cost-effective for reducing EDCs and PPCPs in wastewater. Similar results are expected for drinking water. *The larger problem is that of perception.* Public concerns jumped after the recent U.S. Senate hearings on PhACs in water. Recent Associated Press articles led the general public to believe that ANY amount of pharmaceuticals in the water is toxic. "The dose makes the poison" logic goes unheard.

The solution to dealing with EDCs and PPCPs must be multi-faceted. Public health is paramount, but enhancing water treatment to eliminate very small amounts of EDCs and PPCPs will result in substantial cost and energy use. Increased levels of treatment must be scientifically and fiscally based. Balancing the improvement of water quality with the generation of greenhouse gases (due to energy-intensive advanced treatment) must be considered as must public concerns.

The technical work presented here is part of the solution, detailing EDC/PPCP occurrence, risk, and treatment. Integrating this knowledge base with a proactive public relations program is recommended.

PROJECT**UPDATES**

Filtration and Destruction

Continued from page 5

or UV (when combined with hydrogen peroxide, PAA, or TiO_2) can provide 90-99% reduction/destruction of various EDCs and PPCPs at a cost near that of conventional reclaimed water filtration and disinfection. Further, it showed that particle removal could result in removal of substantial amounts of particle-associated PhACs. Thus, optimizing the performance of sand filtration for particle removal can provide a significant barrier to EDCs and PPCPs without substantial capital cost.

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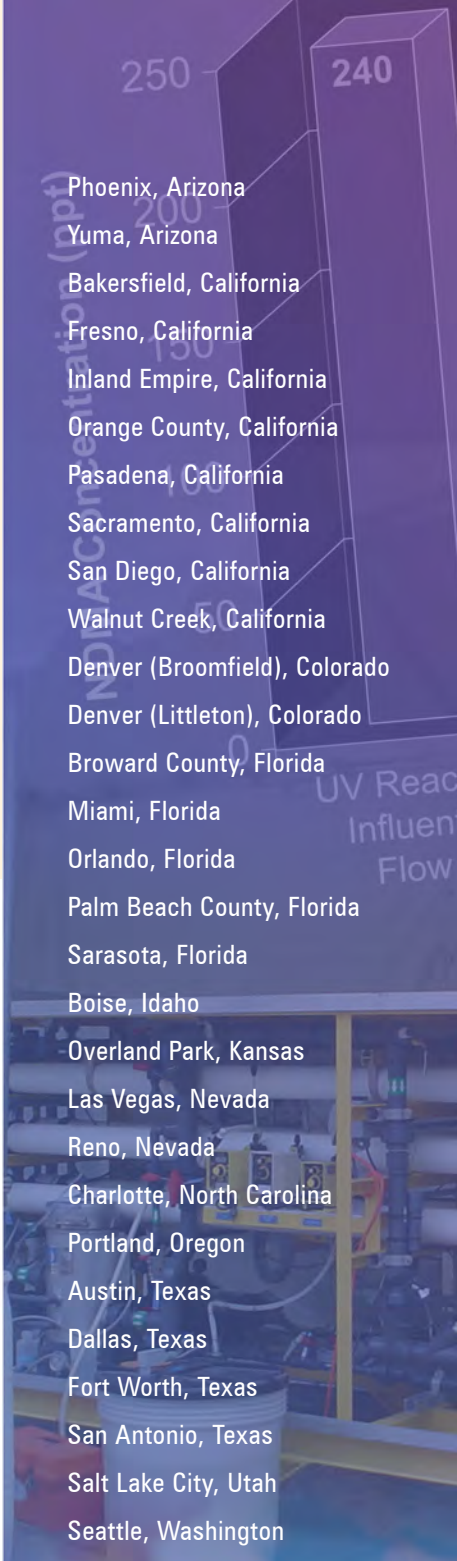

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