# EDCS IN WASTEWATER: WHAT'S THE NEXT STEP?

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## ABSTRACT

The fact that many known and suspected endocrine disrupting chemicals (EDCs) are being found at environmentally significant concentrations in the effluent of wastewater treatment plants (WWTPs) is receiving increasing attention in public and regulatory arenas. The public is concerned about the safety of consuming trace amounts of EDCs in drinking water, though the only confirmed negative effects from EDC exposure have involved wildlife health.

Ample research opportunity exists for the scientific community on this topic: most EDCs have not been identified and/or studied, analytical methods for many identified EDCs have yet to be developed, and the levels of toxicological significance or impact must be established. Additional work must also be done to determine the potential for (1) interactive toxicological effects in EDC mixtures and (2) the formation of undesirable byproducts through treatment. It is likely that the EPA will not consider regulating EDCs until more research has been completed.

Research shows that complete biodegradation of many chemicals of concern can be achieved with adequate SRT and/or HRT in the activated sludge system. When contaminants are persistent or if extremely low effluent concentrations are required, however, higher level removal technology may be needed. Several advanced technologies, such as activated carbon adsorption, ozonation, AOPs, and NF/RO, have successfully removed potential EDCs from water. Most of these technologies, however, are expensive to implement and to operate. Optimization of the activated sludge process could be a less costly option. Issues of by-product formation and EDC additive effects will be important considerations in the design of any treatment strategy.

Long-term facility planning should allow for design flexibility to accommodate possible future EDC regulations. Potential treatment strategies can be incorporated into existing layouts, and room should be left for new equipment. Process selection criteria such as space requirements, byproduct issues, and compatibility with existing facilities must be considered. Planning should favor processes and management strategies that will address not only the concern for EDCs, but other water quality goals as well, so that capital expenditures will cover more than the single, somewhat unclear EDC issue.

Based on current information, it seems logical that a major focus for EDC and PPCP removal should be at the WWTP. Removal of these pollutants from WWTP effluent may solve much of the apparent endocrine disruption problem in the water environment, in addition to providing a cleaner source for drinking water.

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## KEYWORDS

Wastewater treatment, endocrine disruption, pharmaceuticals, personal care products.

## INTRODUCTION

The endocrine system is one of the two main regulatory systems in humans and other organisms. It consists of glands that secrete hormones which are transported in the bloodstream to different parts of the body. These hormones act to control body functions, including reproduction, growth, and development.

Simply stated, an endocrine disrupter is an exogenous substance that changes the function of the endocrine system, affecting the way an organism or its progeny reproduces, grows, or develops. Though most research to date has focused on the disruptive effects on reproduction and development, more recent efforts are examining the effects of disruption on thyroid function and the immune system (McCann, 2004).

Endocrine disrupting chemicals (EDCs) and pharmaceutical and personal care products (PPCPs) are ubiquitous in the environment because of their seemingly endless number of uses and origins in residential, industrial, and agricultural applications. EDCs are derived from both anthropogenic and natural sources; the USEPA is in the process of defining exactly what an EDC is, and those chemicals that meet the toxicity definition will be classified as such in the coming years. The term PPCPs refers to chemicals that enter the environment through use of human and veterinary pharmaceuticals and myriad other products such as antibiotics, analgesics, fragrances, sunscreen, mouthwash, bug spray, and cosmetics. Some PPCPs are suspected of being EDCs, but the terms are not interchangeable and the toxicity concerns associated with the two different groups can be very different. Though the potential hazards associated with some EDCs and PPCPs, such as DDT and DES, have been known for decades, the environmental and health effects of these chemicals in general are only beginning to gain worldwide attention in public and regulatory arenas. Hundreds of compounds are now listed as suspected EDCs; some of these, along with their primary sources, are presented in Figure 1.

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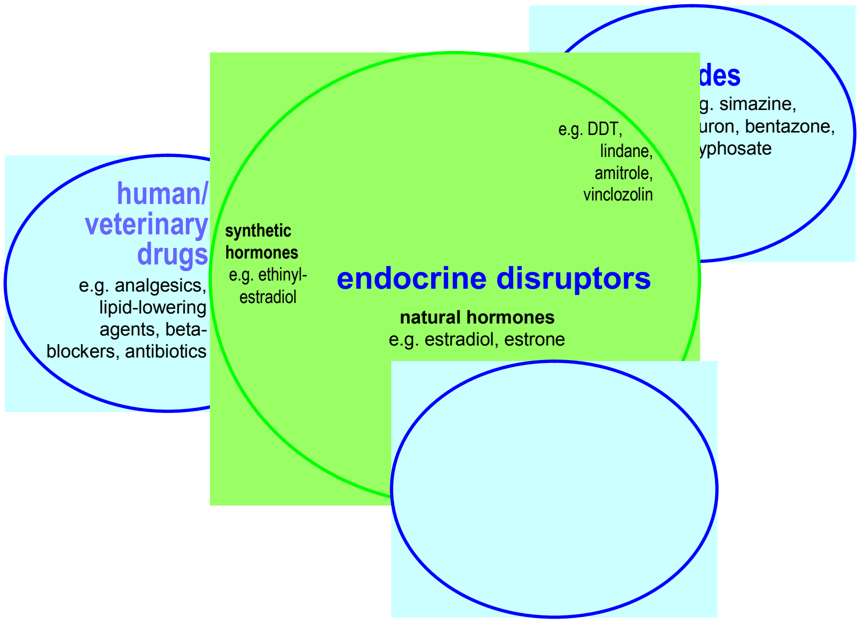

Figure 1. Example of Some EDCs from Various Sources.

There are various pathways by which organisms can be exposed to EDCs and PPCPs; of these, contamination of the water cycle is especially important. EDCs and PPCPs enter the water environment largely through treated wastewater effluent and inputs to water bodies from agricultural or feedlot operations. Agricultural inputs are significant in some areas, and controlling them will be quite a challenge for many reasons. Wastewater treatment plant (WWTP) effluent can be a source for various types and amounts of EDCs and PPCPs, depending on service area characteristics, because most of the WWTPs in service today have not been designed to remove them. Thus, some micropollutants will not be completely degraded or removed through the wastewater treatment process. Aquatic organisms and other wildlife are exposed to EDCs and PPCPs through direct contact in the water environment. Numerous researchers in various countries have reported on the negative effects of WWTP effluent on the reproductive systems of aquatic organisms living in the vicinity of WWTP outfalls. For example, sexual disruption of fish has been linked to estrogenic substances in treated WWTP effluent (Purdom et al., 1994; Jobling et al., 1998; Pickering and Sumpter, 2003). Such effects on wildlife have led to concerns about adverse health consequences in humans, as it is possible that humans can be exposed to EDCs and PPCPs through their drinking water and food.

## STATE OF CURRENT EDC KNOWLEDGE BASE

### Regulatory and Research Efforts

There is ample research opportunity for the scientific community on the topic of EDCs: most EDCs have not been identified and/or studied, analytical methods for many of the identified EDCs have yet to be developed, and the levels of toxicological significance or impact must be

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established. Beyond identifying EDCs, additional work must also be done to determine the potential for (1) interactive toxicological effects in EDC mixtures and (2) formation of treatment byproducts that are more dangerous than the parent compounds that were targeted for removal. The U. S. Environmental Protection Agency (EPA) is not likely to consider regulating EDCs until more research has been completed, though long-term facility planning should take into account that some EDCs may be regulated in the future.

Through the Safe Drinking Water Act (SDWA), the EPA currently regulates a number of possible EDCs such as atrazine, chlordane, DDT, dioxin, cadmium, lead, and mercury. But the maximum contaminant levels for these chemicals are defined by their toxic/cancer-causing effects rather than endocrine disruption. EDCs have not been mentioned specifically in U.S. legislation until 1995, when amendments to the SDWA and the Food Quality Protection Act mandated screening of all chemicals and formulations for potential endocrine activity prior to their use or manufacture where they could cause contamination of drinking water or food. To develop a comprehensive screening program, the EPA established the Endocrine Disruptor Screening and Testing Advisory Committee (EDSTAC). In its final report in 1998, the EDSTAC recommended consideration of: (1) both human and wildlife effects; (2) examination of estrogen, androgen, and thyroid endpoints; (3) a plan for assessing an estimated 87,000 chemicals; and (4) evaluation of six specific classes of mixtures in addition to discrete chemicals. In 2001, the Endocrine Disruptor Methods Validation Subcommittee (EDMVS) was formed to evaluate and validate methods for standardization of EDC testing. Once this work is completed, we should be able to definitively identify which chemicals are indeed EDCs (Snyder et al., 2003b).

In 1999 and 2000, the United States Geological Survey (USGS) sampled 139 streams across 30 states in the U.S. as the first nationwide reconnaissance of the occurrence of PPCPs and potential EDCs. The survey included sampling for 95 constituents from a wide variety of origins, and found that contamination was generally prevalent and widespread (Koplin et al., 2002). While the authors noted that contaminant concentrations tended to be low and rarely exceeded guidelines for drinking water quality, few federal guidelines or regulations exist concerning EDC or PPCP contamination of our drinking or natural waters. Additional studies must be conducted at relevant concentrations of these substances to identify their toxicologically significant levels and to establish reasonable regulations, if any are required. The state of California is considering regulations for EDCs and PPCPs in indirect potable reuse applications, prompting some practitioners of indirect potable reuse to establish monitoring programs now. Since California is a leader in water reuse, this move may stimulate similar actions in other programs around the world.

The EPA is establishing a reference dose for perchlorate, which may become the first pollutant to be regulated in the U.S. for endocrine disrupting toxicity (Snyder, 2003). Several European countries and Japan, however, already have begun phasing out or limiting the use of a few specific EDCs. Besides the U.S., Europe, and Japan, Australia and Canada also see EDC and PPCP contamination as a priority issue and have research programs in place. Both the European Union (EU) and the United Nations (UN) have launched plans for elimination of priority hazardous substances (European Commission Report, 2001; Stockholm Convention, 2001). The current U.S. administration has pledged support of the UN effort.

In addition, a model has been developed to estimate the concentrations of active pharmaceutical ingredients (API) in U. S. surface waters that result from human consumption. Using a mass balance approach, the P*h*ATE (Pharmaceutical Assessment and Transport Evaluation) model predicted the environmental concentrations of several APIs and the results were compared with measured values at 40 locations. In general, the P*h*ATE model was able to estimate concentrations to within a factor of ten of measured values, indicating that it may have value as a screening tool for estimating the presence of human pharmaceuticals in watersheds nationwide (Anderson et al., 2004).

## Human and Wildlife Health Effects

Regarding the effect of EDCs on human health, it has been primarily fear of the unknown rather than fear of the known that has fueled widespread public concern. Excluding specific cases of "high dose response" exposure, results of studies involving population and health trends are inconsistent and do not establish an irrefutable link between low-level exposure to EDCs and adverse consequences to human health. It is the opinion of some scientists, such as Snyder (2003), that the amount of estrogenic chemicals in drinking water is not likely responsible for adverse human health effects because the estrogenic content in water is minute compared with the amount in foods. In addition, exposure to EDCs for humans is completely different from that for fish or other aquatic organisms, so the same response should not be expected. New findings released last year at the ECOHAZARD conference in Germany indicate that it is nearly certain that human exposure to EDCs through drinking water is not significant (McCann, 2004). The scientific community is far from consensus on the topic, though. The issue is far from closed, and scientists, along with environmental and industry groups, are likely to continue to debate it for years to come.

Research into the health effects of EDCs on wildlife is far from exhaustive, but there is more evidence linking EDCs with adverse impacts on wildlife health than on human health. Numerous studies over the past 70 years have demonstrated endocrine disruption in a variety of organisms, including gulls, marine gastropods, frogs, fish, and alligators, as a result of exposure to pesticides, steroids, surfactants, plasticizers, and other synthetic chemicals (Snyder et al., 2003b). New research indicates that there are over 200 species with known or suspected adverse reactions to endocrine disruptors (McCann, 2004).

## Identifying the Most Hazardous Chemicals

While debate over what actually defines an EDC is still ongoing, it is generally accepted that the three main classes of endocrine disruption endpoints are estrogenic (natural estrogen blocked or mimicked), androgenic (natural testosterone blocked or mimicked), and thyroidal (thyroid function affected directly or indirectly). The majority of research to date has focused on estrogenic compounds, though disruption of androgen or thyroid function may prove to be of equal or greater importance biologically (Snyder et al., 2003b). Currently, the scientific community is drawing conclusions about the relative hazards of potential EDCs based on collective results of batch, pilot, and full-scale experiments and studies from around the world. In an excellent summary of research involving fate of EDCs and PPCPs in WWTPs, Johnson and Sumpter (2001) conclude that estrone (E1) and estradiol (E2) would be the EDCs of greatest concern based on *in vitro* potency, while ethinylestradiol (EE2) and the alkylphenols OP and NP

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would be more important based on the more relevant *in vivo* potencies. They state that this latter group could account for as much as 90% of the estrogenicity in a typical WWTP effluent. Discharge concentrations, magnitude of in-stream dilution, and type(s) of species involved are also important factors in considering the impact of the estrogens. It is important to remember, though, that much of the research to date has focused on estrogens, since that is where most wildlife effects have been observed, so conclusions may change as our data base broadens. It is also likely that as research in this area proceeds and analytical technologies advance, scientists will only discover more hazardous chemicals and/or degradation products at even lower concentrations, so this list may prove to be constantly evolving.

Pinpointing the effects of EDC exposure in humans and wildlife is very difficult, since environmental exposure is at very low levels and the perceived effects of endocrine disruption can be subtle and their manifestation may take years. Confusing the matter is the fact that research centers in different countries may use different EDC testing and screening procedures, so they may not agree upon the endocrine disrupting properties of a given substance found in the environment. And without unbiased internationally agreed-upon testing procedures, any unified international response to EDC contamination may be difficult (McCann, 2004).

## EFFECT OF WASTEWATER TREATMENT ON EDCS

### General

Though WWTPs have been shown to remove substantial amounts of many EDCs from the influent wastewater, low concentrations in the effluent may still lead to in-stream concentrations that are of significance to fish and other aquatic species (Johnson and Sumpter, 2001). Levels of toxicological significance are still being investigated, though research has shown estrogenic effects in rainbow trout at E2 and EE2 concentrations as low as 10 and 0.5 ng/L, respectively (Purdom et al., 1994). The actual concentration seen by aquatic organisms depends on the quantity of water available for dilution in the receiving stream. In population-dense, water-poor areas, high pollutant concentrations in the final effluent are of obvious concern.

Depending on their physicochemical properties, EDCs may be removed through adsorption, biological degradation and transformation, chemical degradation, or volatilization (Birkett and Lester, 2003). Findings reported in the literature indicate that removal efficiency through wastewater treatment varies considerably depending on the type of compound and removal process. The latest research into WWTP reduction capabilities indicates that “endocrine active substances” in the influent from primarily domestic sources were more susceptible to breakdown and removal. With other types of contaminants, very little reduction may occur through the WWTP. If these more intractable chemicals must be removed, application of advanced wastewater treatment technologies like membranes or ozonation may be needed (McCann, 2004). Thus, the technology applied at any given plant must be based on a thorough understanding of wastewater constituents.

Table 1 is an example of general information that can be found in the literature concerning removal of some EDCs and PPCPs through various wastewater treatment processes. It is important to note that the removals are discussed in terms of percentages; since initial concentrations are not provided, it is impossible to know how realistic the removal rates are or

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what can be expected for effluent quality in a given situation, especially given the fact that some performance studies are done using influent spiked with high contaminant concentrations. This is important, since some compounds may affect the aquatic environment at very low concentrations, and must therefore be reduced to extremely low effluent concentrations through wastewater treatment.

**Table 1. Treatment Types and Removal Efficiencies for Selected EDCs***

| **Compound** | **Process Type** | **Removal Efficiency** |
|---|---|---|
| PCB (polychlorinated biphenyls) | Biofiltration | 90% |
| | Activated sludge | 96% |
| | Biofiltration/activated sludge | 99% |
| NP (nonylphenol) | High loading/non-nitrifying | 37% |
| | Low loading/nitrifying | 77% |
| $NP_1EO$** | High loading/non-nitrifying | -3% produced as degradation product |
| | Low loading/nitrifying | 31% |
| $NP_2EO$** | High loading/non-nitrifying | -5% produced as degradation product |
| | Low loading/nitrifying | 91% |
| $NP_6EO$** | High loading/non-nitrifying | 78% |
| | Low loading/nitrifying | 98% |
| 17β-estradiol/17α-ethinylestradiol | Filtration – Sand/microfiltration | 70% |
| | Advanced treatment - Reverse osmosis | 95% |
| Organotins | Primary effluent | 73% |
| | Secondary effluent | 90% |
| | Tertiary effluent | 98% |
| Triazines | Conventional two-stage | <40% |

*Taken from Birkett and Lester (2003).

**$NP_nEO$ = Nonylphenol ethoxylate, where n = specific number of EO groups

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Table 1 indicates that several compounds undergo significant degradation through biological treatment, particularly in nitrifying systems with longer SRTs. While sand filtration or microfiltration appear to remove 17β-estradiol and/or 17α-ethinylestradiol with decent efficiency, removal rates for other contaminants will be higher or lower depending on their association with colloidal or particulate matter. The more advanced membrane treatment option shown, reverse osmosis, provides a significantly higher removal rate, though it is important to realize with this technology that the contaminants removed from the main waste stream are concentrated in a smaller reject stream which may require further treatment and must be disposed of properly.

Depending on the type of contaminant involved, coagulant addition, as is practiced for various reasons at many WWTPs, might help to remove some EDCs and PPCPs, particularly those associated with colloidal or particulate matter. However, many of the EDCs and PPCPs of concern are relatively polar with log $K_{ow}$ values of less than three, so a high degree of removal by partitioning onto particles is not expected. In general, research has not shown that coagulation and flocculation with alum and ferric is particularly effective for removal of PPCPs and pesticides (El-Dib and Aly, 1977; Adams et al., 2002; Yoon et al., 2002). It should also be noted that if a coagulant was used as an adsorbent for a particular EDC or PPCP, the resulting sludge could be hazardous and may require special handling.

## Estrogenic Chemicals and Biological Treatment

Though there are many chemicals released into the water environment that are potential EDCs, most work reported to date has focused on xenobiotic estrogens of the alkylphenol group and steroid estrogens, since these two groups of chemicals have demonstrated estrogenic effects in fish. Thus, most of the information presented in this section will pertain to these particular groups of contaminants.

The parent compounds of these two groups, alkylphenol polyethoxylates (APEs) and estrogen conjugates, are not particularly estrogenic; the potentially hazardous estrogenic intermediates are formed because the parent compounds are only partially broken down through wastewater treatment. APEs are nonionic surfactants used in a variety of industrial and household applications, and breakdown into nonlyphenols, octylphenols, and a wide variety of other intermediates during wastewater treatment. Humans excrete natural and synthetic steroid estrogens in inactive forms, which are converted to active hormones, such as estrone (E1), estradiol (E2), ethinylestradiol (EE2), and estriol (E3), in the sewer and through treatment (Johnson and Sumpter, 2001). (EE2 is excreted only when birth control pills are used.)

Many different researchers have reported on the presence of EDCs and/or PPCPs in wastewater and their fate through the biological wastewater treatment process. Studies from research efforts around the world include work by Belfroid et al., 1999; Ternes et al., 1999; Baronti et al, 2000; Korner et al., 2001; Svenson et al., 2002; D'Ascenzo et al., 2002; Lee and Peart, 2002; Andersen et al., 2003; Giger et al., 2003; and Huang, Y., 2003. These authors report a range of removal results for a variety of chemicals, though effluent concentrations of the estrogenic compounds were often found to be in the lower ng/L range, and below detection limits in some cases. In comparing between different studies on the fate of various chemicals through wastewater treatment, it is important to keep several important facts in mind. First, treatment conditions and

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objectives, such as HRT, SRT, temperature, pH, nitrification, denitrification, and bio-P, are often not sufficiently described by researchers. These factors can have a significant impact on EDC removal rate at any given plant. Second, sampling strategy and analysis can dramatically affect results. Third, spiking the influent to a biological process with high concentrations of contaminant may select for an adapted population of microorganisms that would not normally develop (Johnson and Sumpter, 2001).

This section focuses on the impact of biological treatment design on EDC removal, since that is the key component of a conventional WWTP for EDC/PPCP removal. A recent study in England showed particularly dramatic benefits of adding a biological step. Simply adding a short secondary treatment stage of fine bubble aeration to a domestic WWTP that previously had only primary settlement produced a sudden and sustained reversal in feminization trends in downstream fish (McCann, 2004).

Not all types of biological treatment provide the same degree of benefit. For example, Ternes et al. (1999) and Korner et al. (2001) both observed that trickling filters (TF) were less efficient at reducing the estrogenic content of influent wastewater than activated sludge. More recently, two WWTP in the southwestern U.S. were observed. Both plants have primary clarification and effluent filtration, but the biological process of one plant is a Bardenpho BNR activated sludge system with a solids retention time (SRT) of 10-13 days, and the other is a TF system. Both plants receive primarily domestic influent and operate at an average temperature of about 20 degrees C. A comparison of the effluent concentrations of several potential EDCs and PPCPs, a few of which are the known estrogenic compounds, are shown in Figures 2 and 3.

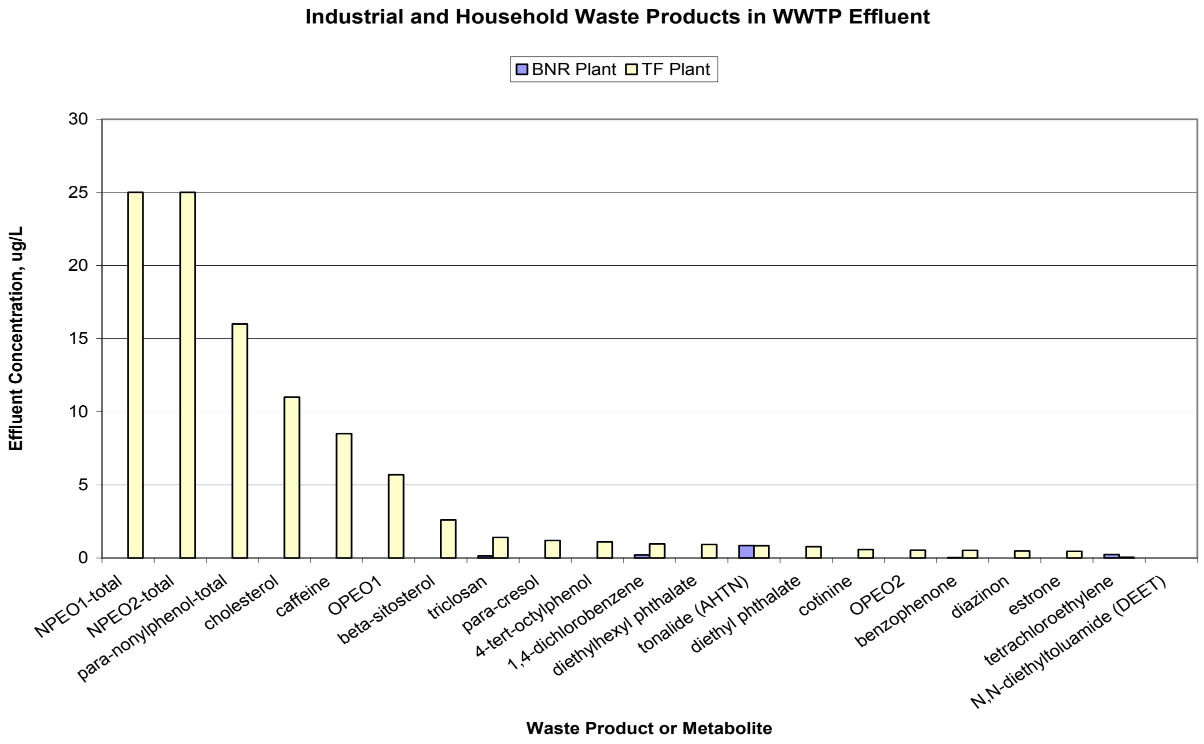

Figure 2. Comparison of Waste Products in BNR and TF WWTP Effluent.

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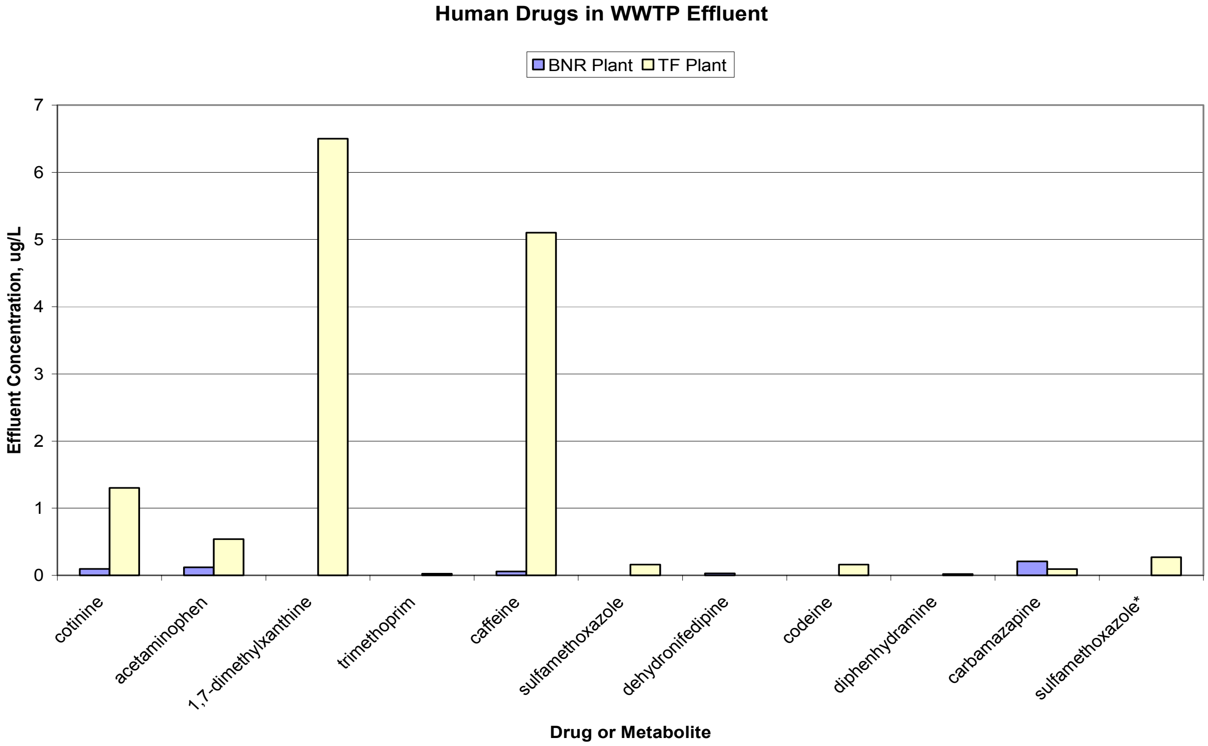

Figure 3. Comparison of Human Drug Concentrations in BNR and TF WWTP Effluent.

From Figures 2 and 3, it can be seen that the activated sludge system does a generally superior job of micropollutant removal as compared to the TF system. Though the more recent studies demonstrate analytical capabilities for measuring EDCs and PPCPs down to the nanogram per liter level, the concentrations shown here in micrograms per liter still provide an excellent comparison of process capability.

In activated sludge systems, hydraulic residence time (HRT) and/or SRT seem to be especially important factors in EDC removal. The longer the HRT, the longer the time available for biodegradation. The HRT of most European activated sludge systems is between 4 and 14 hours (Johnson and Sumpter, 2001), which would explain why this type of treatment would provide better performance than a TF, which might have an HRT of less than one hour. An increase in SRT may enhance the biodegradative and sorptive capacity of the activated sludge. The longer SRT could lead to a more specialized microbial population that can adapt to removal of EDCs and PPCPs. SRT also influences the hydrophobic or hydrophilic properties of the flocs and their ability to act as sorbents.

Several researchers have noted improved EDC removal with increased SRT (Ternes et al., 1999; Holbrook et al., 2002; Andersen et al., 2003). Saino et al. (2004) even specify that SRTs of at least 10 to 12.5 days are required for the organisms that decompose E2 and E1 to grow. In existing WWTP where it may not be possible to adequately increase the SRT because of expense or site constraints, MBR could offer advantages of more flexibility to operate at higher SRTs in a smaller footprint. While microfiltration membranes themselves will not provide an enhanced

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degree of EDC removal, it has been suggested that EDC adsorption to particulate matter that is retained by the membrane would reduce EDC concentration in the effluent. Ivashechkin et al. (2004) operated conventional activated sludge and MBR pilot units in parallel, operating both for denitrification at two different SRTs (12 and 25 days), and applying the same influent wastewater and sludge loading rate to each system. They did not find an appreciable difference in removal of nonlyphenol (NP), bisphenol A (BPA), and 17α-ethinylestradiol (EE2) between the two systems. The authors determined that EDC removal was due primarily to biodegradation; removed EDCs were not simply sorbed onto sludge particles, nor were they retained in the membrane material or the membrane biofilm. Other researchers, however, have found that microfiltration membranes are able to display some retention of smaller particles or colloidal material onto which EDCs may adsorb (Holbrook et al., 2003; Wintgens et al., 2004). Since pore sizing of membrane material is not uniform between manufacturers, it is possible that a difference in membrane material may explain some of the discrepancies in colloid retention. Differences in limits of detection also likely play a role.

Influent and effluent EDC and PPCP data was also collected from a BNR WWTP in the western U.S. that operates at an average SRT of six days and a temperature of 25 degrees C (Snyder et al., 2003). A pilot MBR was also run in parallel at a much higher SRT. The differences in removal rates for some chemicals are shown in Figure 4. Hormones E1, E2, EE2, E3, progesterone, testosterone, and androstenedione were removed to below detection limits (10-25 ng/L) in both systems. It is likely that the increased removal efficiency of the MBR for some compounds was due to the higher SRT, though it is possible that the filtering action of the membrane contributed.

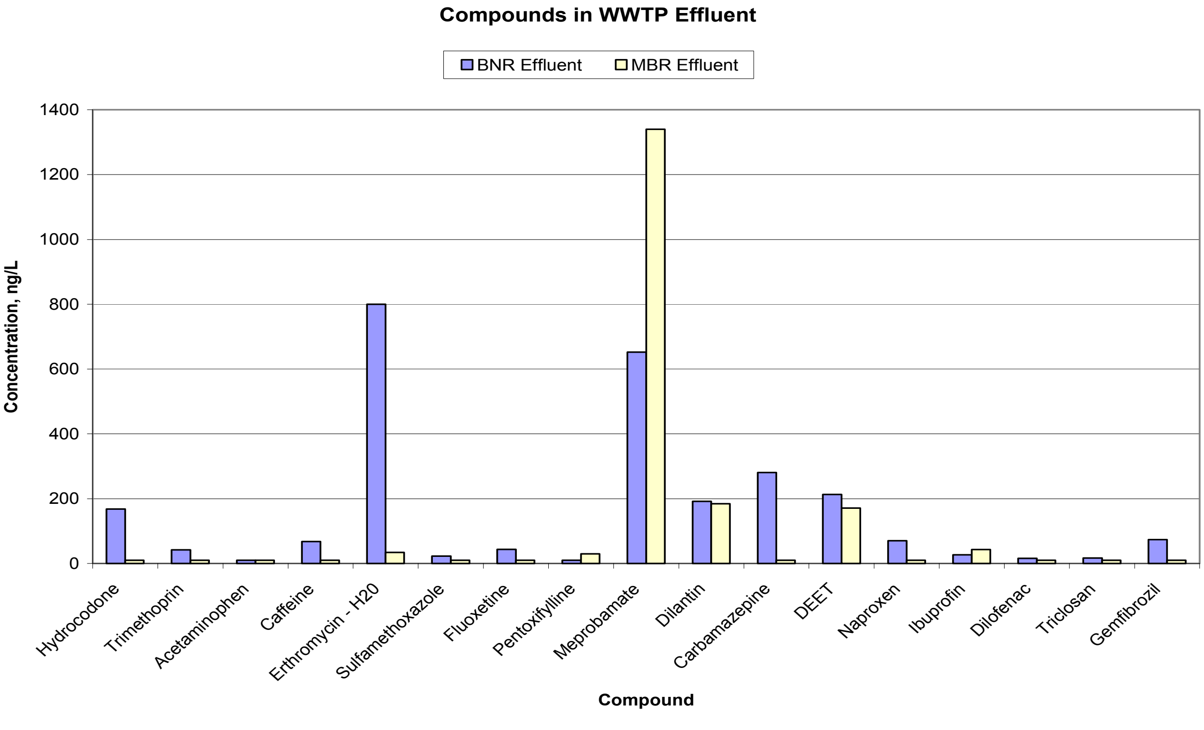

Figure 4. Comparison of Removal Rates Between High SRT MBR System and BNR System.

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Johnson and Darton (2003) state that E1, E2, EE2, and NP are all "inherently biodegradable and so in theory should not present an intractable problem." A drastic increase in the SRT or HRT of existing WWTPs to allow more complete biodegradation would be both cost and space prohibitive, but application of advanced tertiary treatment technologies for many communities would be far too costly. The alternative approach that they propose is to locally increase the amount of biomass sorbent by providing a carrier material within the activated sludge basin onto which a biofilm can develop. A wide range of mild to strongly hydrophobic organic contaminants would be intercepted by the bacterial surfaces and biodegraded. They propose a fixed surface rather than mobile carrier particles to ensure contact of influent wastewater with the biofilms. The fixed matrices would be located toward the front end of an aeration tank (some degree of plug flow is desired) and would be laid out in several packed zones. Laboratory scale tests have shown that almost all steroid estrogens can be removed by this process at a modest extra cost to existing facilities.

With reference to pollutant adsorption onto activated sludge, many EDCs or PPCPs of concern tend to be hydrophilic, though a few of the estrogenic compounds discussed in this section, like octylphenol and EE2 to some extent, are more hydrophobic (Yoon et al., 2003). Such chemicals can adsorb to and concentrate in activated sludge, and may survive anaerobic digestion. Thus, land application of biosolids is another route of exposure for some EDCs to enter the environment, though the ecotoxicological significance of this is presently unknown (Johnson and Darton, 2003).

In summary, it has been shown that some WWTPs are capable of removing most if not all estrogenic activity, with secondary biological treatment being the key process (Pickering and Sumpter, 2003). These facilities should be studied to determine the reasons behind their success. Where it is not possible to increase SRT and/or HRT at an existing WWTP exhibiting less than optimal performance, addition of advanced tertiary treatment may be the only option if ultra low concentrations of EDCs are eventually required. However, it makes sense that we should first thoroughly research optimization of the activated sludge process as a cost effective treatment process that does not generate additional side streams requiring further treatment and disposal.

## Formation of Disinfection By-Products in Wastewater Treatment

All forms of typical wastewater disinfection practiced today will generate disinfection by-products (DBPs) to some degree (White, 1999). The EDSTAC has recommended that DBPs be evaluated for potential endocrine disruptive effects, as it has been suggested that DBPs formed though wastewater disinfection can act as EDCs. The latest research from Japan (Itoh et al, 2004) indicates that chlorination as performed at many WWTPs increases the estrogenic effect of waters containing natural organic matter (NOM). Though chlorination increases the estrogenic effect of NOM and a few other substances, many individual compounds are decomposed by chlorine, drastically decreasing the overall estrogenic effect. For this reason, the authors stress that the *overall* estrogenic effect be evaluated as the sum of increased and decreased activity by chlorination. Because DBPs are suspected to have toxic properties and are generally present in much higher concentrations in WWTP effluent than EDCs or PPCPs, efforts to control EDCs or PPCPs by oxidation may be counterproductive since additional DBPs may be produced (Snyder et al., 2003b). The two main ways that DBP production can be controlled are to 1) control the precursors that react with the disinfectant/oxidant or 2) allow the DBPs to form and then use a

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separate process to remove them (Marhaba, 2000). Various strategies are being evaluated to determine the best approach.

## RESEARCH INTO ADVANCED TECHNOLOGIES FOR EDC REMOVAL

Biological processes are usually the most cost effective means of removing organics from wastewater, but when these organics are toxic or non-biodegradable, physical and/or chemical methods must be used. These methods include adsorption, chemical oxidation, and membrane processes that have more typically been used for water treatment. Research into advanced EDC/PPCP removal strategies is being conducted worldwide. The following is a sampling of new and traditional technologies that appear to have good potential for full-scale application if ultra low EDC/PPCP concentration limits are imposed. It is not suggested that any of these technologies be incorporated into current upgrade/expansion designs at WWTPs, but rather that the potential for EDC/PPCP regulation be recognized by designing flexibility into any long-range upgrade/ expansion plans.

### Activated Carbon Adsorption

Activated carbon has been shown to remove many different types of EDCs and PPCPs to varying degrees. Adsorption will depend on the properties of both the sorbent and the contaminant. Activated carbon efficiently removes hydrophobic organic compounds, but can remove some polar ones as well depending on the strength of polar interactions (Snyder et al., 2003b). NOM also competes for adsorption, so lower NOM content in the water will lead to more efficient use of carbon.

Activated carbon is generally applied in one of two forms: 1) powdered activated carbon (PAC) is added to a sedimentation or contact basin, contacted with water for a few hours, and removed through settling and/or filtration, and 2) granular activated carbon (GAC) is in the form of adsorptive packed beds or filters with continuous flow and short (< 30 minutes) contact times, and can stay in operation for months or years (Snyder et al., 2003b). Adsorbents are very effective for achieving a high degree of removal and low effluent concentrations of contaminant by removing the contaminant from the liquid phase onto the activated carbon. Once exhausted, the adsorbent must be either disposed of or regenerated. The former option merely transfers the pollutant from liquid to solid phase, and the contaminant-rich activated carbon may require further treatment prior to disposal. The latter option can be very costly. Brown et al. (2004) are conducting studies to develop a non-porous adsorbent that can be regenerated in a quick and cost effective manner.

PAC has been shown to achieve over 90 percent removal of E2, EE2, and other potential EDCs from distilled water (Yoon et al, 2002). Wintgens et al. (2004), however, examined use of GAC following MBR treatment of landfill leachate and found that performance was relatively poor for removal of BPA, with only 1.3 g/d of an influent 3.4 g/d being adsorbed. Adams et al. (2002) studied the removal of seven antibiotics from both distilled water and river water using common PAC dosages. They found no statistical difference between the removal results from the two different waters, and concluded that PAC was a viable means of providing treatment for the pharmaceuticals studied. Miltner et al., (1989) also showed that GAC represented a cost

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effective means of controlling several pesticides. Full-scale information on use of activated carbon for EDC/PPCP removal is not available at this time.

## Ozonation

Ozone is a powerful, but selective oxidant. During ozonation, molecular ozone and hydroxyl radicals, to some extent, may transform EDCs and PPCPs (Yoon et al., 2002). While ozone has been commonly used in water treatment, its application for EDC/PPCP removal at WWTP is only now being studied. Wintgens et al. (2004) performed ozonation on a BNR effluent to determine whether trace levels of NP and BPA could be removed. Very low effluent pollutant concentrations were measured for ozone doses of 8, 10, and 15 g $O_3/m^3$, with no appreciable increase in removal rate with dose. In a German pilot unit, application of ozone to BNR effluent resulted in some removal of over 50 trace organic pollutants that are typically found in wastewater effluent, with removal efficiencies frequently higher than 90% (Ried et al., 2004). Three important EDCs – E1, E2, and EE2 – were effectively oxidized or degraded by ozone, and the authors suggest that they lose most of their estrogenic potency in the process. In addition, antibiotics were no longer detected in the effluent. Ozone was not particularly effective in oxidizing iodinated contrast media compounds, and AOP combinations with ozone did not significantly enhance removal rates.

The nature or concentration of ozonation by-products were not discussed in either study. Formation of DBPs with ozone is an important consideration since some amount of NOM will be present in wastewater effluent. Bromate and brominated organic compounds are of particular concern when waters being treated contain bromide.

## Advanced Oxidation Processes (AOPs)

Combinations such as UV plus hydrogen peroxide, ozone plus hydrogen peroxide, and UV plus ozone are powerful oxidation processes that effectively oxidize contaminants. These combinations are designed specifically to increase the concentration of hydroxyl radicals formed, since hydroxyl radicals have less selectivity as oxidants. Substances that are difficult to biodegrade and not removed are oxidized, and the oxidized byproducts may be more amenable to biodegradation. AOPs can be followed by a biological process to further degrade the byproducts, or natural purification processes may be relied upon for treatment, depending on the situation (Ried and Mielcke, 2003). As with ozonation, the hazard potential of the byproducts formed through treatment is also a topic of investigation.

Ried et al. (2004) estimated costs of low pressure UV, ozone, and three AOPs. This information was converted into U.S. units and is presented in Figure 5. As another point of comparison, Ried et al. (2004) reference the total cost for a membrane step at an equivalent of \$1.8 - \$2.2 per thousand gallons.

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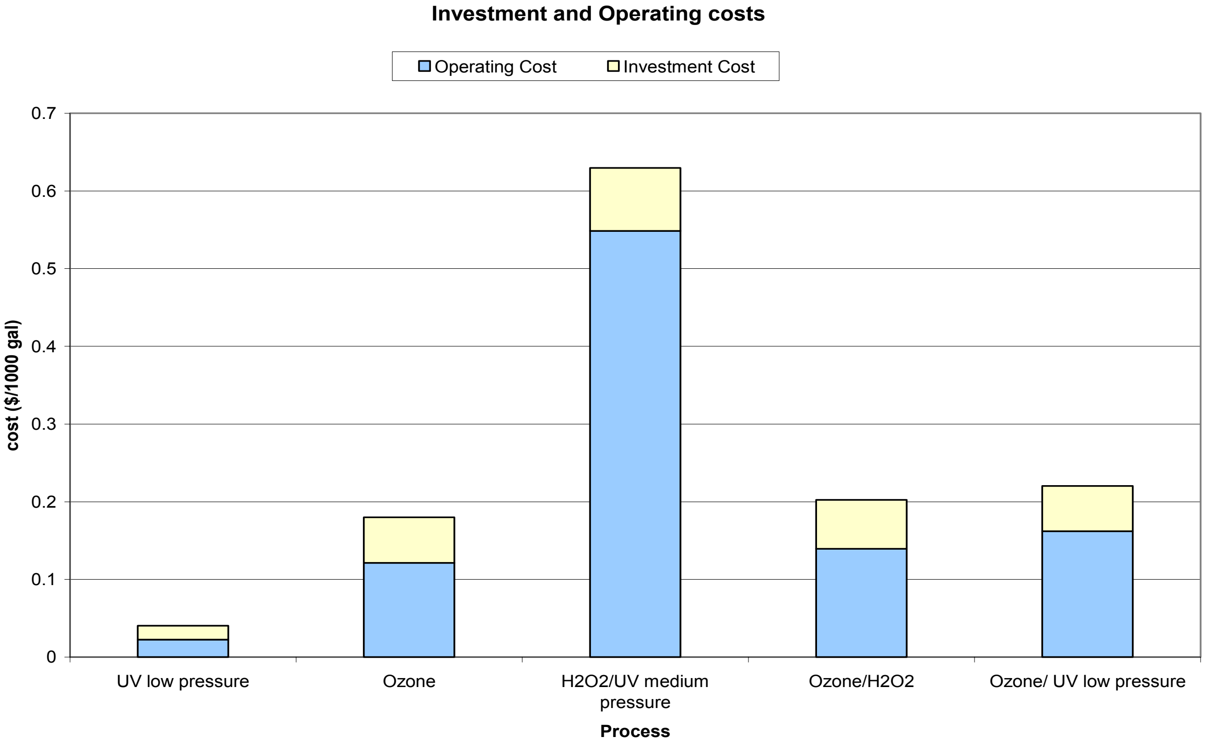

Figure 5. Comparison between UV, Ozone, and AOP Capital and Operating Costs (adapted from Ried et al., 2004).

## Reverse Osmosis (RO) and Tight Nanofiltration (NF) Systems

These types of membranes can reliably remove most EDCs and PPCPs, depending on compound size and membrane properties. Microfiltration is required as a primer step. Besides the advantage of effective removal of micropollutants, DBPs are not created in the treatment process. However, RO and NF systems are very expensive and produce a concentrated reject stream that requires further treatment.

Adams et al. (2002) used a low-pressure RO system to remove antibiotics from distilled and river water. Removal rates in both cases were about 90%. With two and three RO units in series, removal rates increased to 99 and 99.9 percent, respectively.

Wintgens et al. (2004) showed that concentrations of E1, E2, and EE2 in MBR effluent could be reduced to very low levels using NF and RO. The effluent hormone concentrations from RO were extremely low, but not zero, and effluent concentrations from NF were slightly higher. Consequently, the hormone concentrations in the reject stream from the membrane processes were extremely high. Depending on the level of hormone concentrations ultimately deemed “insignificant” in the water environment, either NF or RO could be useful as a polishing step, but the concentrated reject stream will pose a new treatment/disposal challenge. RO and NF systems are usually not an economical option at WTPs (Adams et al., 2002), and are therefore not expected to be economical at WWTPs either.

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## DESIGNING FOR FLEXIBILITY

Future regulatory requirements are unknown at this time, though it is possible that limits on some EDCs may be included in wastewater effluent discharge permits in the future. Several advanced technologies, such as AOPs and RO, have been shown to successfully remove potential EDCs and PPCPs from water. Most of these options, however, involve significant capital and operating expenses that may not be justifiable at this time, since clear regulatory guidance is not available.

In long-range design plans, flexibility should be included to accommodate possible EDC regulations. Potential treatment strategies could be incorporated into existing layouts, and it is important to leave room, both on the site and within the hydraulic profile, for new equipment. The conditions and waste characteristics at every WWTP are unique, so design of the most feasible or cost-effective EDC control strategy will be case-specific. Process selection criteria such as space requirements, byproduct issues, and compatibility with existing facilities must be discussed. Pilot trials will be essential for an optimized design and confirmation that treatment goals can be met. Planning should favor processes and management strategies that will address not only the concern for EDCs, but other water quality goals as well. In this way, capital expenditure will have a broader basis than resolving this one issue that has an unclear outcome.

One option for consideration is the multiple-barrier approach for the protection of public health. This approach includes additional equipment for multiple modes of defense against contaminants (i.e., biological oxidation, physical separation, and chemical oxidation). This could mean the use of activated sludge, filtration, and AOP, or MBR followed by RO and disinfection/oxidation. Incorporation of MBR or integrated fixed film activated sludge into existing biological treatment systems should be considered for enhanced EDC removal where site constraints exist. The higher-level technologies could be added as necessary to meet future treatment requirements. The formation of DBPs can be minimized by strategic positioning of any advanced technologies in the treatment train (e.g., oxidation following filtration).

Though there are several utilities, particularly in the western U.S., that are already considering use of higher-level technologies for EDC or DBP control, it is important to remember that regulations for EDCs and PPCPs are not yet in place. Some utilities are trying to stay ahead of the curve by considering treatment options based on where they think federal or state regulations are headed. In other cases, they may be responding to local demand brought about by public perception of water contamination.

### Example and Cost Estimate

The following example describes options that could be considered at a WWTP for enhanced reduction of potential EDCs. The first two options would involve upgrading the existing activated sludge basins to gain a significant amount of SRT; as mentioned previously, many chemicals of concern may be removed by providing adequate SRT. This amount of treatment might be sufficient to meet any new permit requirements. If a higher degree of treatment is required, however, one of several advanced treatment options may be added to suit the situation. Such higher-level tertiary treatment technologies use a powerful oxidation or straining step, and the associated expense is far greater. Table 2 shows a range of the equipment costs and operation and maintenance costs for each of the options, based on Black & Veatch design experience. The costs given for the AOPs do not match those shown on Figure 5, probably because of the differences in chemical and/or energy use and the equipment included. Additional considerations for each option follow Table 2.

**Table 2. Equipment and O&M Costs for EDC Removal Options.**

| **Process/Technology** | **Estimated Equipment Cost(1) ($/gal)** | **Estimated O&M Cost ($/1000 gal)** |
|---|---|---|
| MBR | 1.00 – 2.50 | (2) |
| IFAS | 0.20 – 0.30 | (2) |
| Peroxone | 0.40 – 0.80 | 0.40 -0.80 |
| UV/Peroxide | 0.40 -0 .60 | 0.30 – 0.50 |
| MF/RO | 1.65 - 3.74 | 0.60 - 1.00 |
| MF/RO followed by UV/Peroxide | 2.05– 4.34 | 0.90 – 1.50 |

(1) Does not include cost of construction.
(2) Separate costs not determined.

**MBR and IFAS:** These options maximize use of the existing facilities. Both have small footprints and can achieve high SRTs in small tank volumes. The consideration and use of MBR technology around the world is advancing rapidly, driven by the increasing need for high levels of treatment and/or small footprint technologies for both municipalities and industries. Most MBR installations are less than 10 years old; therefore, the design criteria for removing micropollutants using this technology are still evolving. Until recently, only a limited number of manufacturers have been offering this technology. Now, numerous MBR vendors offer systems with significantly different configurations, design approaches, and micropollutant removal efficiencies. In the event that a higher degree of treatment is needed in the future, MBR can also serve as the primer step for RO.

The IFAS process combines fixed-film and suspended activated sludge processes. Fixed film media is available from many manufacturers in the form of plastic elements, string systems, plastic webs, and sponges. Adding this media to existing aeration basins makes it possible to achieve nitrification and removal of micropollutants with less basin volume than would be required for a comparable single-stage activated sludge nitrification process. Further, the added media provides surface area for the growth of nitrifying bacteria without imposing excessive solids loadings on the final clarifiers, because the beneficial microbes remain attached to the media in the aeration basin.

**Peroxone:** Peroxone, or ozone/peroxide, has been used for a number of years to remove trace pollutants from groundwater. It has also been installed as part of a multiple-barrier approach at numerous potable water treatment facilities. Because of the hydroxyl radicals formed, peroxone has been found to be very effective for removal of DBPs. Costs are site-specific, depending on the flow rate and type of pollutant being removed.

**UV/Peroxide:** UV/peroxide has been shown to remove trace pollutants such as DBPs, toxic organics, NDMA, EDCs, and 1,4-dioxane. It forms hydroxyl radicals to oxidize the various

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pollutants. Carbon dioxide and water are the products of complete oxidation of the various pollutants. For some specific trace pollutants, only UV may be needed, though the UV doses would have to be unreasonably high to obtain appreciable removal of most EDCs or PPCPs. Of the AOP options, UV/peroxide may result in the lowest DBP formation. Pilot testing should be conducted to confirm costs Equipment costs for this option are the lowest of the advanced technologies, shown at about $0.50 per gallon, but to put this in perspective, this means that the equipment cost for a 20 mgd facility may be as high as $10 million, which does not even include the cost of the building.

**MF/RO:** Microfiltration followed by reverse osmosis has been used to remove trace pollutants from potable water. Additional research is being conducted to increase the throughput capacity of membrane systems. These types of systems will generally remove DBPs, EDCs, and PPCPs that have a molecular size larger than the molecular cutoff of the membrane system.

**MF/RO plus UV/Peroxide:** MF/RO followed by UV/peroxide is an example of a multi-barrier approach. Both processes can independently remove a variety of DBPs, EDCs, and PPCPs. Any trace amount that may pass through the membrane process is oxidized by UV/peroxide. As shown in Table 2, UV/peroxide is the most expensive option; however, it is also the most complete barrier for removing pollutants.

Figure 6 provides an example of how these and other options might be designed into an existing wastewater treatment system.

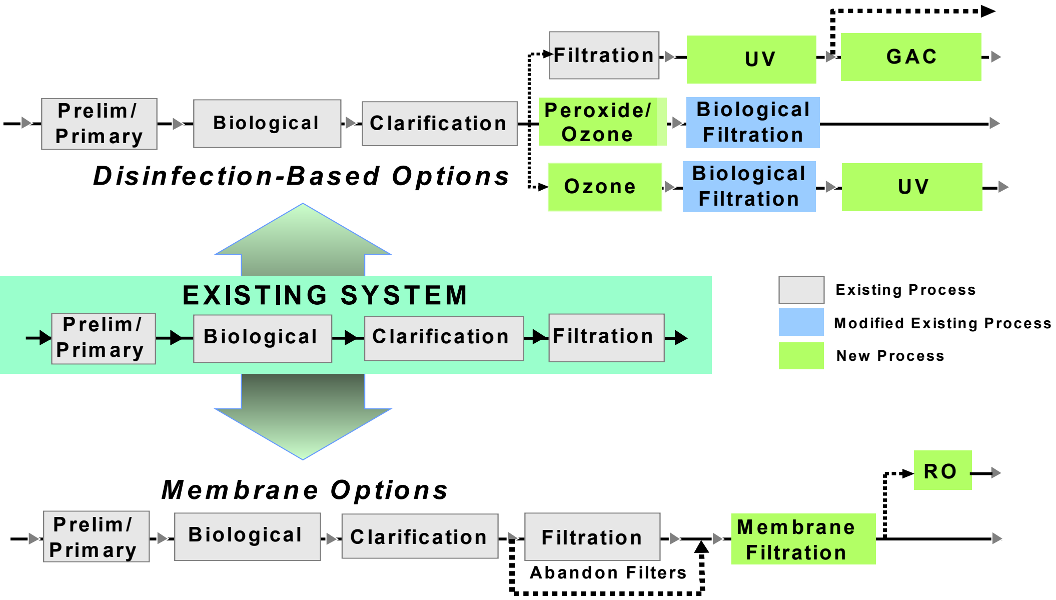

Figure 6. Designing for Flexibility at an Existing WWTP.

## WHERE SHOULD TREATMENT EFFORT BE FOCUSED?

Effects of endocrine disruption on wildlife exposed to estrogenic and other chemicals in the water environment have been demonstrated over the past several years. The public has expressed concern about its safety, because the public drinking water sources may contain trace amounts of chemicals that have been shown to cause adverse health effects in fish and other aquatic organisms. Effects on human health cannot be easily extrapolated from effects on aquatic organisms, however, because aquatic organisms are subjected to continuous exposure to these chemicals, whereas human exposure is generally limited to the amount of water consumed. Further, aside from DBP concerns resulting from disinfection, public drinking water supplies have yet to be proven to be causing adverse effects to human health. While it has been shown that high-tech methods such as reverse osmosis and various advanced oxidation processes can remove many suspected EDCs with impressive efficiency, these methods are generally costly and do not solve the problem of environmental pollution if they are installed at the water treatment plant.

It has been demonstrated through many studies, Johnson et al. (2000), Ternes et al. (1999), and Baronti et al. (2000), to name a few, that activated sludge systems have the potential to remove many suspected EDCs to a fairly high degree. The biological process can likely be optimized to achieve an even higher degree of treatment as researchers further study the effects of SRT, HRT, and other parameters. Attempts to achieve a higher level of treatment with activated sludge should be made before resorting to advanced technologies for EDC removal at WWTPs that may be cost-prohibitive for many communities.

This is not to suggest that the current efficiencies of our WTPs be relaxed; the importance of minimizing DBPs and the contaminants that make their way into water sources through runoff, leaching, and other means is recognized. But based on our current knowledge, it seems logical that a major focus of EDC and PPCP removal should be at the WWTP. Removal of these pollutants from WWTP effluent may solve much of the apparent endocrine disruption problem in the water environment, in addition to providing a cleaner source for drinking water. New data may indicate that tighter controls on industry and agriculture/livestock operations should be required as well to make a more significant difference. Once the scientific community has identified "safe" levels of exposure for the affected organisms, any WWTP effluent limits on contaminants of concern can be targeted to support the health of the water environment.

## CONCLUSIONS AND RECOMMENDATIONS

Though EDCs are currently not regulated in the U.S., the possibility exists that future regulations will be established for some EDCs in WWTP effluent. Processes are available to remove many, if not all, EDCs and PPCPs from wastewater. Since with adequate retention time, a biological treatment system may achieve complete biodegradation of many chemicals of concern, cost effective options for optimization of the activated sludge process should be explored before investing in advanced treatment technologies with high capital and O&M costs. The issue of by-product formation must be researched further, since chemical or biological oxidation can successfully eliminate a parent compound, only to produce potentially more hazardous breakdown products. The potential additive effects of some EDCs must also be considered in the design of any treatment system.

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It is recommended that some degree of flexibility be included in long-term WWTP design to take into account the potential for new regulations on EDCs. Before specific process components can be recommended for treatment of emerging contaminants, however, the scientific community must identify the hazardous contaminants, determine their acceptable concentrations (singly and in combination), and establish standardized analytical methods for their detection. Various conventional and advanced technologies can be assessed for their removal capabilities, and it can be determined whether any additional processes are required at WWTPs to achieve necessary removals.

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