Emerging compounds: A concern for water and wastewater utilities

Water utilities have come under increasing scrutiny as the agencies responsible for controlling human exposure to pharmaceuticals, endocrine-disrupting chemicals, and other compounds of emerging concern (CECs) in drinking water. For example, a recent investigation by the Associated Press (Donn et al., 2008) reported on the presence of pharmaceuticals in the drinking water of 24 major US metropolitan areas. This report prompted public concern and put additional pressure on water utilities to respond to what was presented as a threat to public health.

Most CECs are likely benign to humans at the concentrations detected in the environment and in drinking water. They continue to be of concern, however, because some of the compounds that have been detected are endocrine disrupting chemicals (EDCs). EDCs can have effects on the human endocrine system at extremely low concentrations, although no risk to humans has been demonstrated from exposure to EDCs in drinking water. EDCs have, however, been implicated in adverse effects in aquatic organisms that are exposed to wastewater in the environment. There is also evidence that some CECs that are not EDCs demonstrate toxicity with more traditional endpoints toward aquatic organisms (such as growth inhibition or change in behavior), even at environmentally relevant concentrations. There is also evidence that some CECs that are not EDCs demonstrate toxicity with more traditional endpoints toward aquatic organisms, even at environmentally relevant concentrations.
Although more research needs to be conducted regarding the effects of CECs on human and aquatic health, it is likely that these compounds represent the next realm of regulatory concern and that their removal will drive the research agenda and the selection of water and wastewater treatment processes in the future.

**CECs ARE UBQUITOUS DOWNSTREAM OF WASTEWATER DISCHARGES**

Although many CECs have been present in the environment for decades, concern about their possible effects on humans and wildlife is being driven by improved analytical techniques that are able to detect them at increasingly lower concentrations. In general, these compounds are present at trace concentrations (i.e., parts per trillion or less) in complex mixtures.

CECs encompass a large number of compounds (Table 1). Compounds can be grouped either by their intended use—such as pharmaceutical products or surfactants—or by their potential environmental or human health effects. For example EDCs, which interfere with human or animal hormonal function, sometimes even at very low levels, comprise trace constituent classes such as pharmaceuticals, personal care products, detergent metabolites, plasticizers, brominated flame retardants, and pesticides. Additionally, individual compounds within a class can have significantly different toxicities and removal rates within wastewater treatment plants (WWTPs) and in the environment.

CECs have been detected in surface waters and groundwater downstream or downgradient of wastewater discharges. Much of the early occurrence survey work was conducted in Europe—particularly Germany and Switzerland—where high population densities and low per capita water use result in relatively high concentrations of CECs in wastewater and receiving waters. Even illegal drugs such as cocaine are detectable in some surface waters that are affected by wastewater (Zuccato et al, 2005).

There has been, and continues to be, a significant amount of investigation into the presence of CECs in the environment in North America. For example, Kolpin and colleagues (2002) at the US Geological Survey (USGS) performed a survey of trace organic contaminants in US surface waters and detected 82 of 95 individual CECs on their analyze list, with low levels detected in almost all samples. As a testament to the attention this topic has been attracting, the resulting article was the most downloaded article ever published in Environmental Science and Technology (Kolpin et al, 2002).

David Sedlak, a professor at the University of California at Berkeley and the principle investigator of an Awwa Research Foundation-sponsored occurrence survey of pharmaceuticals in wastewater effluent, has been researching the fate and transport of CECs over the past decade. “At this point, it’s not whether we can find them. We see them everywhere. The question now is whether they’re having adverse effects on wildlife populations, and what can we do about it,” Sedlak said.

**CECS IN WASTEWATER ADVERSELY AFFECT AQUATIC ORGANISMS**

There has been growing attention during the past several years from the public and the scientific and wastewater communities on the potential ecological effects of trace constituents. For example, during the past two years, significant media coverage was given to reports that up to 100% of the male smallmouth bass in some sections of the Chesapeake Bay had measurable levels of CECs. The presence of CECs and their potential effects on wildlife thus present a significant concern for aquatic organism health, as well as for the ecosystem as a whole.

**TABLE 1** Trace constituents in wastewater

<table>
<thead>
<tr>
<th>Class of Compound</th>
<th>Typical Concentration in Effluent</th>
<th>Potential Ecological or Human Health Effects</th>
<th>Examples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pharmaceuticals</td>
<td>Up to 1 µg/L</td>
<td>Endocrine disrupting</td>
<td>Antibiotics, painkillers, caffeine, birth-control pill, anti-epileptics</td>
</tr>
<tr>
<td>Personal care products</td>
<td>Up to 1 µg/L</td>
<td>Bioaccumulative, endocrine disrupting</td>
<td>Soaps, fragrances, triclosan</td>
</tr>
<tr>
<td>Detergent metabolites</td>
<td>Up to 180 µg/L</td>
<td>Bioaccumulative, endocrine disrupting</td>
<td>Oxyphenol, nonylphenol</td>
</tr>
<tr>
<td>Plasticizers</td>
<td>Up to 10 µg/L</td>
<td>Weakly endocrine disrupting</td>
<td>Phthalate esters, bisphenol A</td>
</tr>
<tr>
<td>Perfluorooctane surfactants</td>
<td>Up to 1 µg/L</td>
<td>None at environmentally relevant concentrations</td>
<td>Stain-resistant coating for clothing and furniture</td>
</tr>
<tr>
<td>Brominated flame retardants</td>
<td>Up to 30 mg/L</td>
<td>Bioaccumulative, suspected endocrine disruptors</td>
<td>Polybrominated diphenyl ethers</td>
</tr>
<tr>
<td>Disinfection by-products</td>
<td>Up to 1 µg/L</td>
<td>Carcinogenic</td>
<td>N-nitrosodimethylamine</td>
</tr>
</tbody>
</table>

2008 © American Water Works Association
peake Bay watershed are intersex (Associated Press, 2006).

Wildlife are exposed to CECs and EDCs in effluent-dominated streams or by eating plants or animals in which these substances have bioaccumulated. Aquatic organisms have a greater risk than humans because they have greater exposure. Although many of these compounds undergo natural attenuation in the environment because they are being constantly discharged, organisms that are exposed to them experience a “pseudopersistence.”

Recent research has examined the effects of CECs and EDCs on wildlife both in the lab and in the field. In many species, this research has shown a causal relationship between exposure to human and synthetic hormones, plasticizers, and detergent metabolites and the induction of an egg protein in male fish, for example. Intersex fish found downstream of WWTPs have been linked to estrogenciety in wastewater discharges (Harris et al, 1996).

Most significant, in a seven-year study in the Experimental Lakes Area in northern Ontario, the population of fathead minnow exposed to a low but constant concentration of the hormone found in birth control pills ultimately collapsed (Kidd et al, 2007). Although these specific results cannot be immediately extrapolated to other species and watersheds, there is cause for concern.

THE SPOTLIGHT ON WATER UTILITIES IS DRIVEN BY HUMAN HEALTH CONCERNS

Potential human health effects resulting from exposure to CECs are harder to detect than effects on wildlife. Humans are exposed to pharmaceuticals and endocrine-disrupting chemicals through various routes. For example, fat-soluble compounds such as flame retardants or PCBs can be consumed by eating aquatic animals in which they have bioaccumulated, and water-soluble compounds such as most pharmaceuticals for humans can be consumed in drinking water. This latter route is of concern to the drinking water industry.

Research is just beginning to address questions about effects of chronic low-level exposure, including possible synergism and other toxicological factors associated with these compounds. It is difficult to predict the effects of chronic exposure to extremely low concentrations of a contaminant; generally, tests are done with higher concentrations than occur in drinking water, using other mammalian species, and the effects are extrapolated to lower concentrations using a dose-response curve. A safety factor is then applied to account for the differences between humans and the test species. Some toxicity studies are conducted by exposing human tissue cultures in vitro to the compounds being examined. Neither of these types of experiments provides conclusive evidence about the human health effects of exposure to parts-per-trillion levels of pharmaceuticals in drinking water.

The constituents of greatest potential concern to humans are the EDCs mentioned previously, because of possible effects at very low concentrations. They also pose a disproportionate threat to fetal development and young children because they interfere with normal developmental chemical signals in the body. Therefore, it is not just the level of exposure of EDCs that cause concern, it is also the timing of the exposure during the development of humans and animals.

The potential human health effects of EDCs are being studied by many groups, including the US Environmental Protection Agency (USEPA). The endpoints of exposure to pharmaceuticals and EDCs in water that are being considered by researchers include endocrine system effects, neurological problems, reproductive and developmental abnormalities, and cancers.

Many researchers point out that the quantity of estrogens that may be consumed in reclaimed water is tiny compared with phytoestrogens and estrogenic hormones naturally present in the human diet. In fact, risk assessments that have been performed on drinking water to date
have not shown an unacceptable risk to humans (Snyder, 2007). However, most researchers currently agree that there are many unknowns regarding possible synergistic effects and long-term chronic exposure to low levels of complex mixtures of these compounds.

Although many of these compounds undergo natural attenuation in the environment, because they are being constantly discharged organisms that are exposed to them experience a “pseudopersistence.”

REGULATORY REQUIREMENTS FOR CECs ARE ON THE HORIZON

Regulations can control the input of CECs into the environment and drinking water in two ways:

- by restricting which chemicals can be marketed and therefore make their way into the waste stream and
- by setting wastewater effluent and drinking water concentration limits for individual compounds or bulk parameters such as estrogenicity.

So far, regulators have been more inclined to pursue the first option because toxic effects can more readily be shown in the parent product, rather than diluted in wastewater or drinking water.

European countries are further along than the United States in phasing out endocrine disruptors. Norway, for example, has banned the production, import, distribution, and most uses of nonylphenol and octylphenol ethoxylates. These are surfactants that have been shown to contribute significant estrogenicity to rivers downstream of industrial wastewater discharges. Additionally, the European Union requires the submission of Environmental Risk Assessments (ERAs) to gain market approval for new pharmaceuticals. These ERAs focus on the removal and chlordane. However, these compounds were banned because of their carcinogenic effects rather than their estrogenic effects. In 1996, Congress passed new legislation requiring the USEPA to determine whether certain substances may have an effect in humans that is similar to effects produced by a naturally occurring estrogen or other such endocrine effects. In response, USEPA developed the Endocrine Disrupter Screening and Testing Advisory Committee (EDSTAC), whose members include representatives of academia, industry, public health interests, water providers, and various state and federal agencies.

In its 1998 final report, EDSTAC recommended a priority-based tiered screening system to evaluate chemicals for endocrine-disrupting effects. However, this program has received little funding, and the past 10 years have seen little progress on the screening of suspected EDCs.

So far, neither the USEPA nor regulators in other countries have released any guidance about the effects of relevant levels of most trace constituents in drinking water. US regulatory action at the federal level has been delayed until more research has been done because most existing data on listed man-made chemicals focus on cancer risks, and CECs are generally present at concentrations too low to trigger concerns about carcinogenicity.

In response to the Associated Press article on pharmaceuticals in drinking water, a hearing on “Pharmaceuticals in the Nation’s Water: Assessing Potential Risks and Actions to Address the Issue” was held in April...
Of the advanced treatment technologies, ozonation removes the greatest percentage of compounds of emerging concern, for the lowest unit cost.

2008 by the Senate Subcommittee on Transportation Safety, Infrastructure Security, and Water Quality. Although not resulting in immediate regulatory action, this hearing demonstrates that the issue has caught the attention of US senators, which may provide impetus for further regulatory developments.

US state regulators are beginning to move ahead with adopting criteria, absent a federal mandate to do so. Several states, such as Massachusetts and California, have adopted water quality criteria for perchlorate, a CEC that is found in rocket fuel and other explosives that has been shown to interfere with fetal development at extremely low concentrations. Massachusetts intends to use the process it developed to regulate perchlorate to move ahead with examining possible limits for a list of 100 pharmaceuticals and personal care products.

Other agencies, although not yet ready to set drinking water limits for CECs, are beginning to require that they be monitored in anticipation of possible future requirements. For example, the California Department of Public Health (CDPH) updated its Groundwater Recharge Reuse Draft Regulations Criteria in August 2008 to include project-specific monitoring requirements for endocrine disruptors and pharmaceuticals as well as CEC indicator compounds or surrogates in recycled water and groundwater recharge projects.

The link between CECs in wastewater and adverse ecological effects is stronger than is the link between CECs in drinking water and human health effects and therefore has attracted more attention for possible regulation at the federal level. The USEPA (2008) recently released a draft white paper concerning the development of criteria for CECs. In this document, USEPA acknowledges the difficulty of adopting criteria for constituents with nontraditional endpoints related to endocrine disruption and begins to detail a framework to address this challenge.

Future regulatory action is likely as research progresses, and, once officially identified, industrial chemicals and personal care products that are strong endocrine disruptors will probably be phased out of use. In the meantime, wastewater treatment facilities could be regulated for release of suspected endocrine disruptors to the environment to protect aquatic habitats. In the future, it is likely that drinking water criteria will be developed for some CECs. Therefore, master planning studies for both water and wastewater facilities would be advisable to address the possibility of new regulations when making long-term (10- to 20-year) plans.

SCIENTIFIC RESEARCH AND PILOT TESTING ARE LEADING TO CEC REMOVAL SOLUTIONS

Both water and wastewater treatment can provide a barrier that prevents the introduction of CECs into drinking water. However, for the protection of aquatic life, the preferred barrier is removal during wastewater treatment. Although most wastewater treatment plants are not specifically designed to remove trace constituents, the majority of these compounds are removed at least partially during conventional wastewater treatment.

One of the most effective ways of increasing the removal of trace constituents during biological wastewater treatment is to increase the sludge-retention time to at least 15 days or more. Under these conditions Clara and co-authors (2005) found that most pharmaceuticals, surfactants, and plasticizers are removed to below the limit of detection. Concentrations of human estrogens are reduced by 90-100% with high sludge-retention times. In fact, removal rates depend more on the sludge-retention time than on the treatment technology, because trace constituents are removed equally in an activated sludge process such as a membrane bioreactor. For treatment plants with excess capacity, sludge-retention times can be increased by changing operating procedures without additional capital investment.

Several advanced treatment technologies have been shown to be effective for removing trace constituents from wastewater. Filtration through granulated activated carbon (GAC),
advanced oxidation, and membrane treatment have all been studied to determine how well they remove trace constituents. Table 2 provides a summary of the removal efficiencies for CECs by different technologies.

An advanced oxidation technique that is being studied for removing trace constituents is the irradiation of filtered wastewater with ultraviolet (UV) light after hydrogen peroxide has been added. This process generates free hydroxyl radicals that react quickly and nonspecifically with organic constituents in wastewater. In a study by Rosenfeldt et al. (2004), two common hormones in wastewater were more than 95% removed from lab water with a concentration of 15 mg/L hydrogen peroxide and either a low- or medium-pressure UV lamp.

Ozonation is often put forth as a good technique for oxidizing trace contaminants in wastewater. Although ozone reacts preferentially with some compounds depending on their structure, it can also react with natural organic matter to form hydroxyl radicals and indirectly oxidize a greater number of constituents. Hormones are among the compounds that react well with ozone, as do most pharmaceuticals that have been tested. Ozone is effective at oxidizing some of the most frequently detected trace constituents, such as carbamazepine (an antiepileptic drug), caffeine, cotinine (a nicotine metabolite), and atrazine (a pesticide).

With respect to advanced oxidation, both UV/peroxide and ozonation are good treatment technologies for trace constituents. Both, also provide disinfection for wastewater, and improve its aesthetic qualities. However, several analyses have shown that ozone can provide removal at a lower cost (Ternes et al., 2007).

In general, trace constituent removal is poor during sand filtration. However, with chemical addition prior to sand filtration, Salveson et al. (2007) showed that the increased particle size can improve removal to 70% for some of the more hydrophobic compounds such as hormones. This can be an economical treatment strategy for WWTPs that already practice sand filtration.

### Table 2: Summary of CEC removal by a suite of treatment technologies

<table>
<thead>
<tr>
<th>Classification</th>
<th>AS</th>
<th>AC</th>
<th>BAC</th>
<th>O$_3$/AOPs</th>
<th>UV</th>
<th>Coagulation/Flotation</th>
<th>Softening/Metal Oxides</th>
<th>NF</th>
<th>RO</th>
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<td>Surfactants/detergents</td>
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Adapted from Snyder et al., 2003

AS—activated sludge, AC—activated carbon, BAC—biological activated carbon, CEC—compounds of emerging concern, NF—nanofiltration, O$_3$/AOP—Ozone/advanced oxidation processes, RO—reverse osmosis, UV—ultraviolet irradiation
In membrane filtration, water and wastewater are pushed through tiny pores at high pressure to reject particles that are not desired in the permeate. Microfiltration (MF) and nanofiltration (NF) have progressively decreasing pore sizes and require increasing pressure for operation, with reverse osmosis (RO) having the smallest pore size and the highest pressure.

MF rejects relatively fewer compounds than NF or RO because most trace constituents are smaller than the MF membrane's pore size, although some hydrophobic compounds are excluded through adsorption. NF performs better than MF with most compounds because NF membrane pores are small enough to reject many compounds based on size, but NF is expensive. RO removes most compounds with a very high efficiency except for NDMA, which is small, polar, and behaves like a water molecule. However, although RO removes most constituents below the limit of detection, its capital and operating costs are extremely high.

**SOURCE CONTROL IS AN ATTRACTIVE OPTION FOR REDUCING EDCs IN THE WATER CYCLE**

Cash-strapped utilities may not have the means to implement advanced treatment to remove unregulated constituents from their wastewater or drinking water. Additionally, with recent focus on climate change and carbon footprints, the cost of advanced treatment can be counted in carbon dioxide emissions as well as dollars (Jones et al, 2007). Therefore, source control is being looked at as an alternative for reducing the concentrations of CECs in the environment and in drinking water.

Although the potential for CEC reduction in wastewater is much smaller for some compounds through source control than through advanced treatment, it is an immediate step that can be taken by local governments and utilities that does not require facility upgrades or changes in operational procedures. Source control can include the following measures:

- Pharmaceutical take-back programs to prevent the practice of flushing unneeded or expired pills. It is estimated that a maximum of 8% of pharmaceuticals are flushed down the toilet and that removing this source would lead to an approximately 9% reduction in surface water loading (Tischler et al, 2007).
- Ecolabeling of household and personal care products to encourage consumers to choose products with nonpersistent, nontoxic ingredients. Regulatory oversight would be needed to allow products to claim ecofriendliness on their labels.
- Reduction of over- and unnecessary medication. This has already been recommended to prevent the development of antibiotic-resistant bacteria and could also help reduce concentrations of CECs.
- Requiring ERAs for new pharmaceutical products and phasing out persistent or toxic pharmaceuticals when there is another compound that could have the same benefit without a toxic effect.

Thus far, source control efforts have focused on pharmaceutical take-back programs. Several states have initiated such programs with great success—as measured by the quantity of unused drugs that have been recovered. Although most drugs enter the water cycle via human excretion, these programs can remove a portion of the loading to WWTPs and have additional benefits such as reducing accidental prescription drug poisoning and misuse.

Source control efforts require educational campaigns to inform and engage the public. Because the Southern Nevada Water Authority (SNWA) has led or been involved with much of the scientific work related to the low-level detection, treatment, and health effects associated with pharmaceuticals and EDCs in drinking water supplies, the agency has been at the forefront of communication about this topic. “By educating the public about the proper disposal of all types of chemicals—from household solvents and pesticides to unused pharmaceuticals—utilities are encouraging their customers to become stewards of both their water resources and the environment,” said J.C. Davis, the senior public information coordinator at SNWA.

Source separation is another alternative that could be considered to reduce CECs in wastewater. The most feasible means of achieving this goal is to provide onsite advanced treatment to hospital wastewater, which is a significant point source of pharmaceutical discharges to wastewater treatment plants.

**LOOKING AHEAD AND MOVING FORWARD**

Members of the public will continue to demand that the issue of CECs in the environment and in drinking water be addressed by their utilities, even in the absence of regulatory guidance. A coherent ap-
approach for treatment and control of CECs, as well as a communication strategy, is necessary to assure the public that the issue of pharmaceuticals in drinking water is being considered and addressed by their water purveyors and that aquatic life is being protected by wastewater dischargers.

Clearly, there is the need for more research and continued research funding until the risks from CECs are understood and addressed. Shane Snyder, a scientist at the SNWA, who has contributed significantly to knowledge about the treatment and effects of CECs, emphasizes the need for continued research. “As a scientist, I recommend we focus on research related to health effects from trace pharmaceuticals with a lesser emphasis on occurrence, in order to determine whether there is in fact a problem to solve. The critical question we must address is not ‘Do they exist?’ but rather, ‘At what concentration are these compounds harmful to human health?’ Only then can we make intelligent, rational decisions that protect the health of this country’s municipal water customers.”

CECs are an issue for both water and wastewater utilities. AWWA members and member utilities should encourage cooperation among water and wastewater organizations, such as AWWA and WEF, to provide leadership and dialog for the development of standards and mutual policies to prevent and treat CECs in the water cycle.

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REFERENCES


