Fate of Pharmaceuticals and Personal Care Products in Wastewater Treatment

Pharmaceuticals and personal care products, or PPCPs, are broad classes of compounds marketed to improve human health or used as key components of cosmetics, toiletries, and household consumer products. Some of these compounds require extensive testing before registration and use. Although the human health effects of many of these compounds are understood within the context of their intended use, their environmental impacts to various wildlife and broader impacts from widespread environmental exposure are uncertain. The results from this study will assist the wastewater community to develop more efficient treatment strategies to detect and remove these compounds.

Municipal wastewater treatment facilities were developed to reduce the organic load of discharged effluent to minimize loss of oxygen in receiving waters, and were not specifically designed to remove compounds in pharmaceuticals and personal care products. The biological activated sludge process is the predominant form of secondary treatment, and ranges from short-duration, high-rate processes to biological nutrient removal and extended aeration activated sludge with long detention times. Although much is known about these processes, their ability to remove the PPCP compounds is not well understood.

The primary objective of this study was to evaluate, through collection and analysis of samples from a range of full-scale activated sludge treatment facilities, the fate of aqueous phase PPCP compounds as a function of process solids retention time (SRT). The study also included evaluation of PPCP removal through two different pilot-scale membrane bioreactors. The participating facilities and membrane bioreactors cover a 0.5-30 day range of SRT operating conditions, and the collected samples were analyzed for a target list of 20 PPCP compounds using gas chromatography and mass spectrometry with selective ion monitoring (GC-MS-SIM). The samples were collected as 24-hour time-weighted composites using refrigerated samplers equipped with stainless steel and Teflon tubing.

Occurrence and Removal Behavior

The aggregate data analysis sorted the target compounds into "bin" classifications on the basis of their frequency of occur-

Table 1. Descriptions of Participating Treatment Facilities

Facility	Primary Treatment	Secondary Treatment	Secondary Aeration	MLSS (mg/L)	SRT (days)	Filters	Disinfection
A	Polymer Ferric	High-purity O_2 Activated Sludge	Pure O_2	1,300-2,600	0.5–1.5	None	None
B	No chemicals	MLE[1] Nit./Denit.	Diffused Air	1,800-2,000	3–5	Deep bed[2]	Chlorine
C	No chemicals	Activated Sludge	Diffused Air	2,000-3,000	4–6	Deep bed[2]	UV
D	No chemicals	Nit./Denit.	Diffused Air	2,500-3,000	7–20	Granular MF/RO	Chlorine
E	None	Nit./Denit.	Diffused Air	2,100	11–16	None	UV
F	None	Ext. Aeration Nit./Denit.	Surface Air	4,000	20–30	Deep bed[2]	UV
MBR#1[3,4]	N/A	Nit./Denit.	N/A	14,000	14	N/A	N/A
MBR#2[5,6]	N/A	N/A	N/A	11,500	15	N/A	N/A

[1]MLE (Modified Ludzack Ettinger Process); [2]Granular Media; [3]Utilizes flat sheet membranes; [4]Located at plant E; [5]Utilizes free-end hollow fiber; [6]Located at a facility not listed above.

BENEFITS

- Provides treatment removal data for pharmaceuticals and personal care product compounds commonly used in the U.S.
- Informs wastewater treatment operators about specific compounds that are difficult to remove through secondary treatment.
- Suggests the possibility of seasonal impacts on SRT requirements for PPCP removal.
- Demonstrates the benefit of the longer SRT used for nitrogen removal in reducing aqueous phase PPCP compounds prior to effluent discharge.

RELATED PRODUCTS

Removal of Endocrine Disrupting Compounds in Water Reclamation Systems (01HHE20T)

Contributions of Household Chemicals to Sewage and their Relevance to Wastewater and the Environment (03CTS21UR)

Technical Brief: Endocrine Disrupting Compounds and Implications (04WEM6)

RELATED ONGOING RESEARCH

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rence in the secondary process influent and their degree of removal through the secondary treatment process. Compounds of greatest concern fall in the bin classification of frequent occurrence and poor removal. The target compounds distribute amongst the nine different bin combinations of occurrence frequency and treatment removal. Galaxolide, a fragrance used in many products such as musk perfumes and laundry detergents, is the only compound to fall into this classification category (Table 2).

Compound removals within each treatment bin were further analyzed to assess their dependence on secondary treatment SRT values. A critical SRT value was defined for each compound as the minimum SRT at which greater than 80% removal was consistently observed (SRT80%). SRT80% was compound-dependent with most compounds consistently removed in systems with a SRT of 5 to 15 days.

Table 2. PPCPs of Concern Based on Occurrence and Treatment Bins

Occurrence \ Treatment	Bin T1 Good Removal	Bin T2 Moderate Removal	Bin T3 Poor Removal
Bin O1[1] Infrequent	Methyl-3-phenylpropionate	Octylphenol	TCEP triphenyl-phosphate
Bin O2 Intermediate		Ethyl-3-phenylpropionate	**BHA** **DEET** **Musk ketone**
Bin O3 Frequent	Caffeine Ibuprofen Oxybenzone Chloroxylenol methylparaben Benzyl salicylate 3-Phenylpropionate butylbenzyl Phthalate Octylmethoxycinnamate	**Triclosan** **Benzophenone**	**Galaxolide**

[1]For compounds in Bin O1 (infrequent occurrence), treatment classification is limited by insufficient data as they were seldom detected in the influent.
Good removal = MPR >80%; Moderate removal = MPR between 50% & 80%; Poor removal = MPR < 50%; MPR is the median of the percent removal across the 6 treatment plants and 2 MBR units.

Observations

Seasonal Variations and Compound Characteristics There was evidence of seasonal variation of PPCP concentration in secondary influent with the influent concentrations decreasing in cooler months for plants without primary treatment and the reverse trend of increasing for plants with primary treatment. Additional data is required to fully substantiate this observation.

Treatment Hydraulic Retention Time (HRT) Of the six plants observed in the study, several plants were operated at similar SRT values and dissimilar HRT values. The limited data demonstrated no impact of varying HRT on the removal of the PPCP compounds.

Filtration The overall media filtration results demonstrated no evidence of PPCP compound removal because of this process. Reverse osmosis treatment instead of media filtration, however, was very effective in removing these same compounds.

Treatment Performance and SRT Values Below 5 Day With the exception of three compounds, an SRT ≥ 5 days was required to achieve consistent compound removal above 80% (i.e., SRT80% = 5 days) for the compounds studied. Two facilities studied operated below 5 days, and much better removal was observed for the one facility that used high-purity oxygen.

Key Conclusions

The research team observed that an increase in SRT enhanced the removal of the majority of monitored PPCPs. SRT80% is compound-specific with many of the target compounds well removed by activated sludge processes with SRTs from 5 to 15 days. An SRT80% of more than 30 days was observed for Galaxolide, musk ketone, and TCEP. Activated sludge treatment can be effective in removing many PPCPs. However, to achieve good removal of most target compounds, a second treatment barrier might be necessary.

Future research efforts could include expanding the occurrence and treatment database by including information from additional wastewater treatment facilities, increasing the target list of PPCPs, and reducing the detection limits of the analytical methods used for quantifying these compounds.

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CONTRACTOR

Joan Oppenheimer, M.S.P.H.
Roger Stephenson, Ph.D., P.E., DEE
MWH

PROJECT TEAM

Arturo Burbano, Ph.D.
Li Liu, MSc.
Suvid Surendran, MSc.
MWH

Greg Loraine, Ph.D.
Dynaflow

Rolf Halden, Ph.D., P.E.
Johns Hopkins University

John T. Novak, Ph.D., P.E.
Virginia Tech

Neil J. Parke
Eli Lilly & Company

Bernard M. Sawyer
Metropolitan Water Reclamation District of Greater Chicago

David L. Sedlak, Ph.D.
University of California, Berkeley

PROJECT SUBCOMMITTEE

Kenneth E. Banks, Ph.D.
City of Denton, TX

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