EMERGING CONTAMINANT REMOVAL

IN WASTEWATER TREATMENT TRAINS UNDER CANADIAN CONDITIONS

WAYNE PARKER, UNIVERSITY OF WATERLOO

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Canadian Water Network

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RESEARCH BACKGROUND

Emerging Contaminants (ECs) are chemicals that occur at low concentrations in wastewaters. In general, ECs are unlikely to have lethal effects. However, the specific mechanisms of action of some ECs (e.g., endocrine disruption) have the potential for synergistic/antagonistic effects that may result in other serious effects following prolonged exposure.

Linking the presence of ECs to changes in biological responses is challenging. There are many different types of ECs in municipal effluents, including pharmaceuticals, personal care products and alkylphenols, and many ways in which they may affect organisms. There is a lack of evidence linking biochemical responses that occur during exposure and whole organism effects (e.g., growth and reproduction). It is also not known whether removing individual compounds will result in the attenuation of whole organism effects.

In Ontario, the minimum level of treatment required at a mechanical plant is conventional activated sludge (CAS). At smaller facilities lagoons are commonly employed for wastewater treatment. Other options for treatment include either nitrifying activated sludge (CAS-N, e.g. reduces ammonia levels) or biological nutrient removal (BNR, e.g. reduces both ammonia and phosphorus levels). The latter technologies typically have higher costs and it is not clear whether these upgrades will eliminate impacts of ECs on aquatic biota.

This nation-wide project expands the knowledge base on removal of ECs in these common Canadian wastewater treatment processes over a range of climatic and seasonal conditions. A significant contribution of this work is the concurrent analysis of a suite of aquatic toxicological endpoints, ranging from gene expression to whole organism responses. An analysis of the chemical composition of treated effluents was also conducted. This combined analysis was used to determine whether ECs in treated wastewaters will have harmful environmental effects on aquatic organisms.

The scope of the project included:

- → Comprehensive assessment of the relationship between the removal of emerging contaminants in wastewaters and the impacts of treated wastewater on aquatic species
- → Assessment of the impact of treatment process configuration on these responses
- → Assessment of the impact of climatic conditions on these responses
- → Recommendations for decision-makers regarding ECs in wastewater treatment

Emerging contaminants in wastewater or sewage include a suite of chemicals including pharmaceuticals and personal care products. The emerging contaminants measured in this research included gemfibrozil, sulfamethoxazole, trimethoprim, ibuprofen, carbamazepine, triclosan, naproxen, nonylphenol, HHCB (galaxolide) and AHTN (tonalide).

GLOSSARY OF ACRONYMS:

- ECs – emerging contaminants
- CAS – conventional activated sludge
- CAS-N – nitrifying activated sludge
- BNR – biological nutrient removal
- SRTs – solids residence times
- HRTs – hydraulic resistance times
- PPCPs – pharmaceuticals and personal care products
- MEBPR – membrane enhanced biological phosphorus removal process
- YES – Yeast Estrogen Screen
- C-Fin – cultured tail fin biopsy assay
- MBR – membrane bioreactor
- MLSS – mixed liquor suspended solids

RESEARCH METHODS

Wastewater treatment operations were carried out at four nodes across the country. Samples were characterized by chemical analysis and an array of biological assays (Table 1).

SUMMARY OF EFFLUENT TESTING

	NODES				
TESTS	Burlington P1	Burlington P2	UBC	Guelph	Trent
Chemistry analysis	X	X	X	X	X
Receptor assays	X	X		X	
Rainbow trout gene expression assay	X	X			
C-fin assay	X	X		X	
Zebra fish	X	X			
FHM Lifecycle	X				

Table 1: Summary of Effluent Testing

COMPARISON OF CAS, CAS-N, BNR WASTEWATER TREATMENT PROCESSES

The performance of conventional activated sludge (CAS), nitrifying activated sludge (CAS-N) and biological nutrient removal (BNR) processes was compared at pilot scale at Environment Canada's Wastewater Technology Centre. Testing was conducted at controlled temperature, solids residence times (SRTs) and hydraulic residence times (HRTs) to reduce variability in the effluent characteristics in two phases to emulate cold and warm weather conditions.

COMPARISON OF MEMBRANE BIOREACTOR (MBR) WITH ACTIVATED SLUDGE

At the Guelph node the removal of selected ECs in a pilot MBR plant and two full scale treatment plants was assessed. Two treatment trains were selected for the study; one with primary sedimentation and secondary nitrifying conventional activated sludge treatment units, while the other was an extended aeration configuration. In the MBR pilot plant, raw wastewater from the Guelph wastewater treatment plant was filtered prior to being fed to the aeration tank.

LAGOON STUDY

The removal of selected pharmaceuticals and personal care products (PPCPs) in a sewage lagoon serving Lakefield, Ontario was characterized. The sewage lagoon is split into two ponds, with the south pond aerated for biological treatment of the sewage and the north pond operated as a settling pond. Removals of ECs were evaluated in the summer and fall of 2010 and in the winter of 2011. PPCPs were monitored using passive samplers deployed at various locations in the sewage lagoon.

BIOLOGICAL NUTRIENT REMOVAL TRANSIENT LOADING STUDY

The wastewater pilot plant at UBC is comprised of two parallel trains operating as membrane enhanced biological phosphorus removal processes (MEBPR). Tests evaluated the dynamic response to a slug input of ECs at two different SRTs. Samples were obtained from the influent (after the primary clarifier), the recycle flow from the aerobic zone, and the effluent (or permeate of the membrane), over a period of 48 hours.

RESEARCH FINDINGS

SUMMARY OF RESULTS

Activated Sludge Studies	
1.	In pilot studies CAS-N and BNR processes achieved lower effluent concentrations of a number of ECs than a CAS process.
2.	The BNR effluent had the lowest YES responses, while the CAS effluent had higher YES responses but with a large degree of variability. The CAS-N effluent differed between winter and summer phases.
3.	Consistent patterns in the expression of individual genes were not observed in either the rainbow trout or amphibians. A "meta" analysis revealed similar trends between activated sludge configurations for gene expression and ECs.
4.	CAS-N and BNR effluents had no effect on zebrafish egg production. The CAS effluent exposures of zebra-fish resulted in reductions of egg production.
5.	Exposure of fathead minnow to CAS effluent resulted in considerable mortality, reduced growth and reduced egg production. The CAS-N effluent also resulted in some mortality and reduced growth and egg production in the fathead minnows. By contrast, the BNR effluent had no effect on mortality, growth or egg production.
6.	An MBR (Membrane Bioreactor) pilot and the 2 full scale plants with activated sludge treatment, achieved similar EC removals. The similar performance of the differing plants in the different seasons was attributed to the relatively long SRTs (Solids Residence Time) that were employed for all of the plants.
7.	The temporal response of the concentrations of more hydrophobic ECs in the MLSS and final effluent of pilot activated sludge processes was affected by the SRT (and hence MLSS concentration).
Lagoon Study	
1.	Compounds that are susceptible to photodegradation (i.e., sulfamethoxazole and trimethoprim) showed greater removals in a lagoon in the summer season, while ibuprofen, which is susceptible to microbial degradation showed greater removals in the fall.
Internode Comparisons	
2.	There were substantial differences in concentrations of the target compounds in the raw wastewaters between the nodes while the estrogenicity of the raw wastewaters appeared to have similar values.
3.	With the exception of sulfamethoxazole all of the other overlapping ECs demonstrated removal efficiencies that were relatively consistent across the nodes.

COMPARISON OF CAS, CAS-N, BNR WASTEWATER TREATMENT PROCESSES

- The CAS-N and BNR processes achieved lower effluent concentrations of a number of ECs than the CAS process in both Phases of the project. In Phase 1 (warm weather) of the project the BNR process had more compounds with lower EC concentrations than the CAS-N process, while in Phase 2 (cold weather) the CAS-N process performed better in this regard.
- The CAS effluent had elevated concentrations of ammonia and ECs as compared to the other pilots. As the ammonia concentrations were considerably greater than the EC concentrations, it is likely that ammonia was responsible for many of the biological responses that were observed.
- The BNR effluent had the lowest Yeast Estrogen Screen (YES) responses, while the CAS effluent had higher responses but with a large degree of variability. The CAS-N effluent differed between winter and summer phases. The YES responses were somewhat inconsistent with the trends that were observed for ECs, likely because the YES values showed very weak estrogenicity and many of the ECs measured are not considered estrogens.
- Consistent patterns in the expression of individual genes were not observed in either the rainbow trout or C-Fin gene expression studies. A "meta" analysis of the rainbow trout gene expressions revealed that in Phase 1 there were fewer gene responses to the BNR effluent than to the other two pilot effluents. In Phase 2 there were considerably fewer gene expression responses than in Phase 1 in the rainbow trout. By comparison, the C-Fin test revealed fewer thyroid receptor gene responses in the BNR and CAS-N effluents than in the CAS in Phase 1. In Phase 2, the converse was true with greater thyroid receptor gene responses observed in the BNR compared to CAS-N and CAS effluents. Very few stress responses were observed overall in the C-fin assay. The trends in the gene expression "meta" analysis were generally consistent with the trends observed after categorizing EC concentrations in the effluent. However, these trends were not consistent when the individual genes were examined.
- Egg production bioassays conducted to examine the reproductive toxicity of the effluents to zebra-fish found no impacts of the CAS-N and BNR effluents on egg production. The CAS effluent exposures of zebra-fish resulted in reductions of egg production. However, a similar response was observed when exposures were conducted with ammonia. The zebra-fish tests did not identify any impacts of ECs on reproduction.

- Life-cycle fathead minnow tests demonstrated considerable differences between the effluents with respect to growth and reproduction. The CAS effluent exposures resulted in considerable mortality, reduced growth and reduced egg production that was likely due to the elevated ammonia concentrations. The CAS-N effluent that had much lower ammonia concentrations than the CAS effluent also resulted in some mortality and reduced growth and egg production in the fathead minnows. By contrast, the BNR effluent had no effect on mortality, growth or egg production.

COMPARISON OF MBR WITH ACTIVATED SLUDGE

- The MBR (Membrane Bioreactor) pilot and the 2 full scale plants with activated sludge treatment achieved similar EC removals. Only gemfibrozil appeared to be removed to a slightly greater extent in the MBR pilot than in the full scale systems. The similar performance of the differing plants in the different seasons was attributed to the relatively long SRTs (Solids Residence Time) that were employed for all of the plants.
- The C-Fin gene expression assay revealed that the normal thyroid hormone receptor alpha *(thra)* response to thyroid hormone was attenuated by influent, primary, and secondary effluent exposure from one of the full scale treatment trains. By contrast the thyroid hormone receptor *(thrb)* was not affected. An indicator of general stress, the heat shock protein *(hsp30)* mRNAs showed a noticeable increase in expression with exposures to influent and primary treated wastewater exposure but did not demonstrate a response to exposures to secondary treated wastewater.

LAGOON STUDY

- The removals of PPCPs in the lagoon were generally consistent with removals of these indicator compounds that have been reported in the literature for conventional mechanical wastewater treatment plants.
- Compounds that are susceptible to photodegradation (i.e., sulfamethoxazole and trimethoprim) showed greater removals in the lagoon in the summer, while ibuprofen, which is susceptible to microbial degradation showed greater removals in the fall. Carbamazepine, which is considered recalcitrant in wastewater treatment systems showed inconsistent results for removals in the summer, fall and winter. For Triclosan, HHCB and AHTN, higher removals were observed during the summer season. Gemfibrozil increased in concentration significantly as it passed through the lagoon. This increase was most likely caused by de-conjugation of the metabolites throughout the wastewater treatment process.
- A model that was developed in this study was able to adequately describe the fate of four ECs with differing susceptibility to fate processes in the summer and winter.

BIOLOGICAL NUTRIENT REMOVAL TRANSIENT LOADING STUDY

- The temporal response of the concentrations of more hydrophobic ECs in the MLSS and final effluent was affected by the SRT (and hence MLSS concentration) of the pilots. These responses were slower than that predicted from liquid-phase tracer testing.

INTERNODE COMPARISON

- There were substantial differences in concentrations of the target compounds in the raw wastewaters between the nodes. In general the concentrations at the Burlington and UBC nodes were higher than those at the other locations.
- Despite the differences in EC concentrations between the Burlington and Guelph nodes, the estrogenicity of the raw wastewaters appeared to have similar values. The results suggest that the target compounds employed in this study may have not been good indicators to compare the estrogenicity of the raw wastewaters.
- With the exception of sulfamethoxazole all of the other overlapping ECs demonstrated removal efficiencies that were relatively consistent across the nodes. Naproxen, ibuprofen and nonylphenol were consistently highly removed (>90%) at the Burlington, Trent and UBC nodes. Carbamazepine was not removed at both the Trent and Burlington nodes.
- Sulfamethoxazole was characterized at both the Burlington (activated sludge) and Trent (lagoon) nodes and its removal efficiencies differed substantially between the two sites. The improved performance of the lagoon treatment as compared to the activated sludge processes may be due to increased photodegradation in the former process.

RESEARCH APPLICATION

The results of this study provide guidance to designers, owners, operators and regulators of municipal wastewater treatment facilities when considering their capacity to reduce the harmful effects of ECs on the aquatic environment.

Wastewater treatment plants with improved nitrogen removal (nitrification and denitrification) can result in enhanced EC removal. There is evidence that this can reduce harmful effects, as evidenced by the YES receptor assay, gene expression

assays and whole organism tests. The extent of improvement appears to depend upon the operating conditions, as the performance of the treatment trains changed when the operation was switched from winter to summer operating conditions. Regional climatic conditions should be taken into account when considering the appropriate wastewater treatment technology for improved removal of ECs.

For selected compounds, similar removals of ECs were observed in activated sludge, membrane bioreactors and lagoon systems. The results suggest that municipalities have several technology options for reducing the impact of ECs on the aquatic environment. Further, appropriate operation of existing wastewater treatment processes (i.e. extending SRT when possible) can influence EC reduction. Lagoons appear to be particularly effective in removing ECs that are sensitive to photodegradation.

There was considerable inconsistency between the abiotic assays employed in this study. In many cases ammonia, rather than ECs, was likely responsible for many of the biological effects that were observed. Gene expression assays are potentially attractive tools for evaluating the effectiveness of wastewater treatment, as they provide insight into multiple stressor pathways. Additional development of these tests is required to address their apparent lack of reproducibility.

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