New England Water Treatment Technology Assistance Center
University of New Hampshire • Durham, New Hampshire
PROJECT SUMMARY REPORT

Antibiotic Removal in Slow Sand Filtration

Objectives

This research study focused on removal of antimicrobial contaminants in roughing and slow sand (multi-stage) filtration drinking water treatment; methods commonly used in rural regions and developing nations.

The specific objectives of the study were as follows:

1. Review of potential human and environmental health risks from antimicrobial contaminants and resistant bacteria introduced into the aquatic environment from diffuse pollution pathways;

2. Development of a simple predictive model for removal of antimicrobial contaminants in slow sand filters;

3. Characterization of antimicrobial contaminant removal in pilot roughing and slow sand filters;

4. Calculation of biodegradation coefficients and half-lives for fluoroquinolones exposed to bacteria cultured from slow sand filter schmutzdecke biomass.

Methodology

Laboratory experiments were conducted to investigate antimicrobial sorption on schmutzdecke and sand media phases of the treatment process. A mathematical model of antimicrobial removal was developed for slow sand filtration considering media characteristics, source water organics concentration, headloss, and equilibrium photolysis and sorption coefficients. This model may be used to assess ionizable organic contaminant removal in slow sand filtration and evaluate risk to water treatment systems using contaminated source water.

Antimicrobial removal efficiency studies were performed at the slow sand filter research facility of the City of Salem, Oregon. Representative antimicrobials include the sulfonamide, macrolide, tetracycline, fluoroquinolone, lincosamide, and diaminopyrimidine classes of compounds. Each of the antimicrobial compounds was applied to pilot-scale roughing and slow sand filters, and removal of aqueous concentrations was assessed for at least eighteen pore volume detention cycles using an HPLC MS/MS method developed for this research project.

Results

Although terraccumulation and diffuse pathways contribute to antimicrobial residues found in surface and groundwater, recent human and aquatic organism risk assessments rarely include this phenomenon.

A 60-day slow sand filtration simulation using the slow sand filter computer model developed to investigate the behavior of ionizable antimicrobial contaminants suggested greater than 3-log removal from 1 µg/L influent concentrations within the top 40 cm of the sand column for the tetracycline, quinolone, and macrolide classes of antimicrobials.

Antimicrobial removal efficiency of the filters was calculated using HPLC MS/MS, and sorption coefficients of several antimicrobials (Kd, Koc, Kom) were calculated for schmutzdecke collected from a mature filter column. Sulfonamides had low sorption coefficients and were largely unaffected by multi-stage filtration. Lincomycin, trimethoprim, and tylosin exhibited higher sorption coefficients and limited mobility within the slow sand filter column. Tylosin was primarily sorbed within the top 40 cm of the sand filter column and these results supported earlier findings of laboratory-scale soil column studies.

At the end of a 14-day filtration period, slow sand filtration exhibited less than 25% removal of lincomycin and less than 4% removal of the sulfonamide class of antimicrobials from spiked river water. These results were consistent with the low schmutzdecke sorption coefficients, and multi-stage filtration is regarded as an ineffective treatment method for lincomycin and sulfonamides.

The lack of a significant increase in overall antimicrobial removal efficiency during the challenge studies indicated biodegradation is less significant than sorption in multi-stage filtration.

Enrofloxacin introduced to a pilot roughing filter at a 100 µg/L concentration was not detected after the second filter compartment in calcite-amended roughing filter pretreatment, and oxytetracycline exhibited > 50% removal from influent concentrations.

Enrofloxacin and oxytetracycline were significantly removed in slow sand filtration, and concentrations dropped below the analytical detection limit within the top 50-cm of the sand column.

Schmutzdecke biodegradation coefficients (k) calculated from first-order kinetics of enrofloxacin, ciprofloxacin, and norfloxacin were 10-1.17, 10-1.25, and 10-1.53 day-1, respectively, and half-life calculations (t1/2) for enrofloxacin, ciprofloxacin, and norfloxacin exposed to aerobic bacteria cultured from schmutzdecke biomass were 10.3, 12.3, and 23.5 days, respectively.

Qualitative antibacterial resistance comparisons of microbial populations cultured from schmutzdecke collected before and after the pilot filtration period indicated exposure to low antimicrobial concentrations may enhance resistance formation among bacteria in schmutzdecke biomass.

Conclusions

Simulation using the mathematical model of antimicrobial removal confirmed the high removal rates observed in previous soil studies for the tetracycline, quinolone, and macrolide classes of antimicrobials.

Slow sand filtration exhibited antimicrobial removal efficiencies in the order of tylosi>trimethoprim> lincomycin>sulfamethoxazole>sulfamethazine, and trimethoprim and tylosin were almost 100% removed by this treatment method.

Sulfonamides, because of their greater relative persistence among pharmaceutical antimicrobial compounds, may be a suitable indicator for suspected veterinary and human pharmaceutical contamination of conventional and slow sand drinking water treatment processes.

Considering the antimicrobial contaminant behavior in microcosms containing bacteria cultured from a biologically-active slow sand filter, fluoroquinolone concentrations in schmutzdecke are expected to be persistent when the biomass is removed from the filter and applied to agricultural soils.

Recommendations

Further research is needed to examine the effects of complexation on antibacterial activity of trace antimicrobials in finished drinking water.

Continued research investigating bacterial resistance transfer in biological filtration systems and antimicrobial residue mobility in drinking water waste products is warranted for adequate environmental risk assessment.

Publications and Presentations

Peer-reviewed Journal Publications

Rooklidge S, Miner J, Kassim T, Nelson P. (In press). Antimicrobial Contaminant Removal by Multi-Stage Slow Sand Filtration, Journal of American Water Works Association.

Rooklidge S, Bennett J, Dolan M. (2005). Antimicrobial Contaminant Behavior in Slow Sand Filter Schmutzdecke Mixed Cultures, Research Journal of Chemistry and Environment, 9, 1: 5-11.

Rooklidge S, Burns E, Bolte J. (2005). Modeling Antimicrobial Contaminant Removal in Slow Sand Filtration, Water Research 39, 2-3: 331-339.

Rooklidge S. (2004). Environmental Antimicrobial Contamination from Terraccumulation and Diffuse Pollution Pathways, Science of the Total Environment, 325: 1-13.

Book Chapters

Rooklidge S. (In press). Slow Sand Filtration, The Encyclopedia of Water, Lehr J. ed., John Wiley & Sons, New York.

Rooklidge S. (In press). Multi-Stage Drinking Water Filtration, The Encyclopedia of Water, Lehr J. ed., John Wiley & Sons, New York.

Conference Presentations

Rooklidge S. (2005). Antibiotic Contaminant Removal in Slow Sand Filtration, Proceedings of the 124th Annual American Water Works Association Conference, June 12-16, San Francisco, CA.

Kassim T, Allen N, Brunson E, Nodolf A, Tokumoto V, Visitacion B, Rooklidge S. (2005). Biodegradability and Toxicity of Human and Veterinary Antibiotics in Natural and Engineered Environmental Systems of the Seattle Metropolitan Area. 15th Annual Northern California Society of Environmental Toxicology and Chemistry Meeting, May 3-4, Berkley, CA.

Kassim T, Rooklidge S, Nelson P, Simoneit B. (2004). Removal of Pharmaceuticals, Hormones and Endocrine Disrupting Chemicals by Multi-Stage Drinking Water Filtration. 4th World Congress of the Society of Environmental Toxicology and Chemistry, Nov. 14-18, Portland, Oregon.

Principal Investigators

Peter O. Nelson, Ph.D.
Stephen J. Rooklidge, Ph.D., P.E.
Department of Civil, Construction and Environmental Engineering
Oregon State University
Corvallis, Oregon 97331

M. Robin Collins, Ph.D., P.E.
New England Water Treatment Technology Assistance Center
University of New Hampshire
Durham, NH

Disclaimer

This project was funded by the U.S. Environmental Protection Agency grant number X827736-01-0. Mention of specific trade names herein does not imply endorsement on the part of the USEPA or the University of New Hampshire.