CHEMICAL FATE PREDICTION FOR USE IN GEO-REFERENCED ENVIRONMENTAL EXPOSURE ASSESSMENT

VOORSPELLING VAN HET GEDRAG VAN CHEMICALIEN IN HET MILIEU MET HET OOG OP GEOGRAFISCH GEREFEREERDE BLOOTSTELLINGSBEOORDELING

door

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Summary

The work described in this thesis was conducted in the framework of the GREAT-ER project (Geography-referenced Regional Exposure Assessment Tool for European Rivers). The objective of this international project was to develop a tool to accurately predict chemical exposure in the aquatic environment, for use in environmental risk assessment. As the current generic 'unit world' techniques to assess regional exposure do not account for spatial and temporal variability and do not offer realistic predictions of actual concentrations, they are merely applicable on a screening level. In GREAT-ER, a software system was developed to calculate predicted concentrations of 'down-the-drain' chemicals (e.g. detergents) in surface waters, using a Geographic Information System (GIS) for data storage and visualization, combined with simple mathematical models for the prediction of chemical fate. In this thesis, the development of the simulation methodology used within GREAT-ER is described. Models to be used for the prediction of chemical fate are subsequently selected, adapted or newly developed. Also, new measurement methods are proposed and evaluated. Finally, a method to obtain spatially aggregated predicted environmental concentrations (PECs) for a catchment simulation was worked out.

Methodology

A geo-referenced simulation methodology for the prediction of aquatic exposure to individual 'down-the-drain' chemicals, which uses real-world data, was developed. By combining steady-state deterministic chemical fate models with a Monte Carlo simulation methodology, statistical frequency distributions of predicted concentrations in the aquatic environment are obtained. Emissions are predicted from chemical market data, and are further processed in sewer, treatment and river models. This results in geo-referenced predictions of chemical concentrations in the considered river systems. The practical applicability and the scale-independent character of this simulation methodology was illustrated by means of a hypothetical but realistic case study.

Measurement and Prediction of Chemical Fate

The standardized CAS (Continuous Activated Sludge) laboratory test system and the mathematical fate model *SimpleTreat* are used to routinely assess the elimination of substances in activated sludge waste water treatment plants. As these do not incorporate the effects of biological nutrient removal (BNR), which is rapidly gaining importance, a number of modifications were presented. The CAS test was adapted to include BNR processes. The performance of two modified CAS units, which were fed with an improved synthetic sewage, was monitored, and was also compared with model predictions. Next to this, primary removal of the surfactant LAS was also measured. It could be concluded that the proposed systems hold potential to complement the standard CAS. Similarly, the *SimpleTreat* model was modified to increase its applicability to BNR plants. The adaptations

focused on an improved description of sludge recycling and on the presence of different redox zones (anaerobic, anoxic, aerobic) in the biological reactor. Two updated models were applied to the modified CAS units, and tested using the LAS measurements. This illustrated the improved predictive power of the adapted models for the fate of readily biodegradable chemicals. However, further research is recommended for other substances.

For trickling filter waste water treatment plants, no standardized chemical fate model exists. A new fate model was developed, based on the approach used in *SimpleTreat* in combination with an existing biofilm model. To test the new model, a pilot-scale (200 *L*) trickling filter was built and operated. Using this setup, removal of LAS was measured under different well-characterized steady-state operating conditions. The new model could be fitted to LAS removal in the pilot-scale trickling filter as well as in two full-scale domestic filter plants. The same biodegradation rate coefficient (derived from activated sludge data) could be used in all cases, except for the laboratory experiments with higher LAS influent levels. As only the biodegradation and sorption aspects of the model could be confronted with measurements (because LAS was used as test substance), further research is needed to test the relevance of volatilization aspects. It is also recommended to focus additional research on the effect of the influent concentration on the biodegradation rate.

To predict the biodegradation of chemicals in rivers, a mathematical model was developed which considers both biofilm and suspended biomass activity. This model was calibrated for LAS using experimental data obtained in a small artificial river. Biofilm processes were shown to be by far the most significant removal mechanism in the considered system. The model was further tested by comparing its predictions to a field study in the Red Beck, a small Yorkshire river. The predicted overall LAS half-life (without any calibration using the field data) was within 25% of the in-stream removal measured in the field.

In a tentative in-sewer removal field experiment, no removal of LAS could be measured. This is in contradiction with literature information and with a modeling exercise based on consumption data. To gain a better understanding of chemical fate in the sewer system, further detailed research is recommended.

Calculation of Predicted Environmental Concentrations (PEC)

GREAT-ER simulations result in digital maps with predicted concentrations for individual river stretches. This output may contain too much local detail for practical risk assessment applications and decision making, for which a spatial aggregation of the results is desirable. Two spatially aggregated PEC types were developed: $PEC_{initial}$ (aggregation of river concentrations just downstream of waste water emissions) and $PEC_{catchment}$ (aggregation of all average stretch concentrations). Potential scale-dependencies and issues related to stretch selection were discussed and resolved. Testing for 2 case study catchments confirmed the theoretical considerations which led to the different definitions, and illustrated the need for appropriate weighting to resolve scale-dependencies.