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Developing and applying metamodels of high resolution process-based simulations for high throughput exposure assessment of organic chemicals in riverine ecosystems

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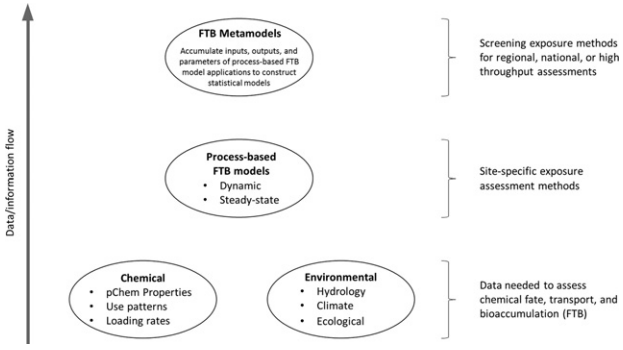
HIGHLIGHTS

- Integrated fate, transport, and bioaccumulation modeling of chemicals in rivers
- Developing metamodels for high throughput and screening exposure assessments
- Corroborating metamodels with flame retardants concentrations in EU rivers
- Using metamodels for model sensitivity analysis and interpretation of field data

GRAPHICAL ABSTRACT



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ABSTRACT

As defined by Wikipedia (https://en.wikipedia.org/wiki/Metamodeling), "(a) metamodel or surrogate model is a model of a model, and metamodeling is the process of generating such metamodels." The goals of metamodeling include, but are not limited to (1) developing functional or statistical relationships between a model's input and output variables for model analysis, interpretation, or information consumption by users' clients; (2) quantifying a model's sensitivity to alternative or uncertain forcing functions, initial conditions, or parameters; and (3) characterizing the model's response or state space. Using five models developed by the US Environmental Protection Agency, we generate a metamodeling database of the expected environmental and biological concentrations of 644 organic chemicals released into nine US rivers from wastewater treatment works (WTWs) assuming multiple loading rates and sizes of populations serviced. The chemicals of interest have log n-octanol/water partition coefficients ($\log K_{OW}$) ranging from 3 to 14, and the rivers of concern have mean annual discharges ranging from 1.09 to 3240 m^3/s. Log-linear regression models are derived to predict mean annual dissolved and total water concentrations and total sediment concentrations of chemicals of concern based on their $\log K_{OW}$, Henry's Law Constant, and WTW loading rate and on the mean annual discharges of the receiving rivers. Metamodels are also derived to predict mean annual chemical concentrations in fish, invertebrates, and periphyton. We corroborate a subset of these metamodels using field studies focused on brominated flame retardants and discuss their application for high throughput screening of exposures to human and ecological populations and for analysis and interpretation of field data.

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1. Introduction

Under various federal laws [i.e., the Federal Food, Drug and Cosmetic Act (FFDCA);the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA); the Food Quality Protection Act (FQPA); the Toxic Substance Control Act (TSCA), etc.], the US Environmental Protection Agency (USEPA) assesses the safety of new and existing chemicals released into the environment using a risk assessment paradigm that couples the inherent toxicity (hazard) of chemicals to their expected environmental fate, transport, and bioaccumulation (exposure). Based on such assessments, the Agency may restrict a chemical's manufacture, use, or disposal (GAO, 2009). Conducting such assessments, however, is a daunting task due to the sheer number of chemicals that must be evaluated annually by the Agency. For example, under TSCA, the USEPA receives annually as many as 1200 Pre-manufacture Notifications (PMNs) for new industrial chemicals which must be reviewed within 90 days. Similarly, under FIFRA, the Agency must annually review both new pesticides and existing pesticides that have been registered for no more than 15 years. In fiscal year 2016 alone, almost 25,000 pesticide products were subject to reregistration review (https://www.epa.gov/pesticide-reevaluation/reregistration-and-other-review-programs-predating-pesticide-registration). Consequently, there is a real need to have tools (i.e., calculators and models) that can facilitate high throughput (HT) risk assessments.

Intuitively, the most likely tools for such assessments would be could be simple exposure and hazard models that require a limited number of input parameters. Such "screening" level models would include both empirical and simple process models. For example, chemicals that are currently or expected to be released from point sources into rivers or streams might be screened as follows. First, if the chemical's physicochemical (pchem) properties are unknown, they could be estimated using an existing pchem calculator, such as the EPI Suite™ software (USEPA, 2011). Using these properties and empirical Quantitative Structure Activity Relationships (QSARs), the chemical's expected transformation rates (e.g., biodegradation rates), accumulation factors (e.g., BAFs), and lethal exposure concentrations (e.g., LC_{50}s) could then be estimated to assess its expected environmental persistence, bioaccumulation, and toxicity, respectively. The chemical's steady-state water concentrations could then be forecasted using expected environmental release rates estimated by ChemSTEER (USEPA, 2013) and a simple dilution model like E-FAST (USEPA, 2007). Finally, predicted water concentrations could be compared to the estimated LC_{50}s to assess hazards to aquatic biota and combined with estimated BAFs to predict dietary exposures to humans and wildlife. Because such screening models would be expected to have simplified or limited representations for all but the major physical, chemical, and biological processes that determine the fate, transport, and bioaccumulation of chemicals in real world environments and ecosystems, such screening assessments would be expected provide only a general picture of chemical risks to human and ecological populations whose uncertainty and variability would be difficult to quantify.

The USEPA has made the improvement of its chemical safety assessments a major priority (USEPA, 2015). Although many see increasing laboratory and field data to support such assessments as a major thrust (ICCA, 2011; NRC, 2012), improvements can also be achieved by using higher-tier models that add more process-based science to the exposure and risk assessments conducted by the Agency. Many such models have been developed and used by the Agency to obtain more robust representations of specific chemicals in targeted environments either as case studies or to support specific management decisions. Unfortunately, many of these models have not been widely used for risk assessments because they are considered too data-intensive or time-consuming to be used routinely.

In this paper, we demonstrate an approach for incorporating higher-tier process-based models into HT exposure assessments which circumvents the need to parameterize and execute the models of interest for every assessment. In this approach, technical experts in the use and the analysis of the process-based models of interest apply them to populations of chemicals and environments that span the range of chemical and environmental properties of concern to generate expandable databases of selected model outputs. These databases are then used to construct linear and nonlinear regression models which can evaluate the expected fate, transport, and bioaccumulation of non-database chemicals and environments for screening-level exposure assessments. Such metamodels not only capture the most salient features of the selected science models but also provide well-defined tools for conducting rapid exposure assessments. We demonstrate this approach for organic chemicals that are known to be released into rivers from point sources [e.g., treated discharges from publicly owned water treatment works (POTWs) or other wastewater treatment works (WTWs)]. For this demonstration, the chemicals of concern have log n-octanol/water partition coefficients ($\log K_{OW}$) ranging from 3 to 14, and the rivers of concern have mean annual discharges ranging from 1.09 to 3240 m^3/s. We use five existing USEPA models to generate the required databases. To estimate the pchem properties and loading rates of the chemicals of interest, we used the Chemical Transformation Simulator (CTS) (Wolfe et al., 2016) and the High Throughput Stochastic Human Exposure and Dose Simulation Model (SHEDS-HT) (Isaacs et al., 2014), respectively. Using these data, we then simulate the dissolved and total water concentrations, and total sediment concentrations for each chemical-loading combination in the rivers of interest using the Exposure Analysis Modeling System (EXAMS) (Burns, 2004). Lastly, we use the K_{OW}-based Aquatic BioAccumulation Model (KABAM) (Garber, 2009) and the Bioaccumulation and Aquatic System Simulator (BASS) (Barber, 2012) to estimate expected BCFs of periphyton and phytoplankton and BAFs of benthic invertebrates, zooplankton, and fish in the rivers of interest. Using these BCFs and BAFs, expected whole-body concentrations of exposed fish and invertebrates are calculated for each chemical-loading-river combination. Each of the aforementioned models, which are described in Section 1 of our Supplemental information, has been peer reviewed and used routinely by the USEPA to assess the fate, transport, and bioaccumulation of chemicals in aquatic ecosystems (USEPA, 2003; USEPA, 2008; USEPA, 2014).

2. Materials and methods

The overall metamodeling approach described in this paper is illustrated in Fig. 1. Existing USEPA models that predict chemical properties, consumer product exposures, environmental fate and transport, and bioaccumulation are used to create a database of chemical concentrations in water, sediments, and biota for multiple rivers. Regression methods are then used to develop metamodels for predicting environmental concentrations in water and sediment as functions of river hydrology, chemical loading rates, and chemical properties. Metamodels are also developed to predict BAFs for exposed biota as functions of the chemical's K_{OW} and the biota's ecology and life history; chemical concentrations in exposed biota are then predicted using these metamodels coupled to those developed for simulated dissolved water concentrations. The methods used to develop these metamodels are described below.

2.1. Chemicals of interest

For this demonstration, we selected four categories of chemicals that have been and continue to be of concern to the Agency at large. These are household products (e.g., personal care products, residential insecticides, etc.) currently included in the SHEDS-HT database; chemicals identified as or suspected of being endocrine disruptors (EDCs) [e.g., List 1 and List 2 EDCs in the Federal Register (78 Fed. Reg. 35,922)]; brominated flame retardants (BFRs) and their proposed substitutes; and medium-chain chloroparaffins [MCCPs, also known as polychlorinated alkanes (PCAs)] which are used as secondary plasticizers in polyvinyl