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Evaluation of a novel high throughput screening tool for relative emissions of industrial chemicals used in chemical products

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ABSTRACT

Tens of thousands of chemicals are currently marketed worldwide, but only a small number of these compounds has been measured in effluents or the environment to date. The need for screening methodologies to select candidates for environmental monitoring is therefore significant. To meet this need, the Swedish Chemicals Agency developed the Exposure Index (EI), a model for ranking emissions to a number of environmental matrices based on chemical quantity used and use pattern. Here we evaluate the EI. Data on measured concentrations of organic chemicals in sewage treatment plants, one of the recipients considered in the EI model, were compiled from the literature, and the correlation between predicted emission levels and observed concentrations was assessed by linear regression analysis. The adequacy of the parameters employed in the EI was further explored by calibration of the model to measured concentrations. The EI was found to be of limited use for ranking contaminant levels in STPs; the r^2 values for the regressions between predicted and observed values ranged from 0.02 ($p = 0.243$) to 0.14 ($p = 0.007$) depending on the dataset. The calibrated version of the model produced only slightly better predictions although it was fitted to the experimental data. However, the model is a valuable first step in developing a high throughput screening tool for organic contaminants, and there is potential for improving the EI algorithm.

1. Introduction

Tens of thousands of chemicals are currently marketed worldwide. For the majority of these compounds, the emissions and environmental fate are unknown due to lack of environmental monitoring data (Muir and Howard, 2006). Rational approaches to discover new undetected environmental contaminants are being developed (see e.g. Muir and Howard, 2006; Brown and Wania, 2008). These screening methodologies are designed to identify chemicals with properties that make them persistent, bioaccumulative, toxic, or susceptible to long range transport. They do not, however, quantitatively estimate or rank the actual emissions of chemicals. Such estimates are required for moving forward with the output of any property based screening system and for the assessment of historical, present and future environmental contamination.

Emission inventories have been conducted for only a few organic pollutants to date (Breivik et al., 2004; Denier van der Gon et al., 2007), and the uncertainties in such estimates are large (Breivik et al., 2002). The inputs required in the existing release models (e.g. the Mass Flow Tool of Environment Canada or the PCB mass balance model developed by Breivik et al. (2002)), such as data on annual production, use pattern, disposal, accidental release and emission factors for each use/disposal/accidental release category, are chemical and article specific. This information, if available, must be collected from different sources for each chemical (e.g. pollutant release and transfer registers, or other data that manufacturers/importers report to authorities). This laborious process is not conducive to high throughput screening.

To our knowledge, high throughput screening tools for generic emission estimates are currently lacking. The exception is an emission ranking tool, named the Exposure Index (EI), which was developed by the Swedish Chemicals Agency at the request of the Swedish EPA in 2005 (Fischer et al., 2005, 2006). This tool utilizes data on product use patterns and chemical content (i.e. quantities) registered in the Swedish Product Register (PR) to rank regional emissions of the ~15 000 compounds recorded in this register. Low, medium and high emission factors are associated with various function and branch categories, as previously suggested by Health Canada (2002). These categories resemble the use/industry categories defined in the EU Technical Guidance Document (TGD) (EC European Commission, 2008)). High or low emission factors are also associated with hazard labeling and consumer availability

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of chemical products. The model outputs are chemical specific indices ranking emission levels to various environmental matrices. The EI is currently one of the tools employed to select candidate chemicals for the Swedish Environmental Screening Program.

In this study, we present an evaluation of this first attempt to employ chemical use patterns and quantities in a high throughput screening tool for emission ranking. Data on measurements of organic contaminants in Swedish sewage treatment plants (STPs), one of the recipients considered in the model, were compiled from various published sources. The predictive power of the EI to rank emissions to STPs was then evaluated by comparing measured levels and predicted EIs using linear regression analysis. The adequacy of the model setup was further tested by assessing the potential for model improvement by calibration to the measured data.

2. Methods

2.1. The Exposure Index

The Exposure Index (EI) is described in detail in (Fischer et al., 2005, 2006). A brief summary is given below.

The EI ranks the diffuse emissions to a number of recipients (“surface water”, “soil”, “air”, “sewage treatment plant (STP)” and “human”) based on data registered in the Product Register (PR). The PR is a register of all chemical products manufactured in or imported to Sweden in a quantity >100 kg, and has been updated annually since 1992. A chemical product is defined as either a substance or a preparation of chemical substances. Articles are not registered in the PR. The raw materials for articles are, however, generally chemical products. Around 2000 companies have registered data for 65 000 chemical products and 15 000 compounds in the PR. Pharmaceuticals, cosmetics and food are not covered by the register. The raw materials for medicines and cosmetics are, however, included. The PR includes information regarding the chemical products’ technical functions, the branches where they are used, private consumer availability, hazard symbols, chemical composition (weight% in each product) and annual quantities. These data are combined in the EI algorithm to enable emission based ranking of the substances present in the PR.

Reformulation of the original Exposure Index equation (Fischer et al., 2005, 2006) gives the following equation (see derivation in the Supporting information):

$$\mathrm{EI} = \log(9+P)\cdot\log\left(\sum_{i=1}^{P} F_i B_i Q_i K_i S_i\right) + C \qquad (1)$$

where P is the number of products containing the substance, F_i, B_i, K_i, and S_i are the function, branch, consumer and symbol number of product i, respectively, Q_i is the quantity of the substance in product i and C is a constant with a value of 13 (fitted to avoid negative EIs).

The function number (F) represents a low, medium or high exposure factor. These exposure factors have been assigned to each of the more than 800 function categories in the PR for each primary recipient based on expert judgment. Similarly, each of the ~120 branch categories has been assigned a low, medium or high exposure factor (represented by a branch number B) for each primary recipient. The low, medium and high levels of F and B are arbitrarily dimensioned with the factors 0.01, 1 and 100. The use function and branch for each chemical product are described by at least one function category and branch category. Examples of function categories are “pigments for paints and inks”, “drilling oils”, “detergents”, “cleaning agent for windows”, “solvent” and “hair spray”. The branch categories consist of a selection of the codes defined by the Swedish Standard Branch Classification which in turn is based on the European NACE code system. Examples of branch categories are “manufacture of dyes and pigments”, “manufacture of textiles”, “industrial cleaning” and “sale of motor vehicle parts and accessories”. Note that the function and branch categories are similar but not identical to the use/industry categories defined in the TGD (EC European Commission, 2008).

The Product Register also lists hazard labeling and (private) consumer availability of each chemical product. This information is translated into the symbol number (S) and the consumer availability number (K). The presence of a skull and crossbones label is assumed to be associated with more careful use and chemical products carrying this label are assigned an S of 1. Chemical products without this symbol are assigned an S of 10. Consumer availability, on the other hand, is assumed to increase emissions to all recipients. Available products are assigned a value of 10, and non-available chemical products a value of 1. Finally, the number of chemical products containing a substance, P, is included in the Exposure Index calculation. If a substance is present in several products, this is assumed to lead to a higher exposure to all recipients. The description of the parameters is summarized in Table S1 in the Supporting information.

2.2. Data compilation

Data on concentrations of organic compounds in sludge and wastewater were compiled mainly from the Swedish national database for environmental monitoring data (IVL, 2010). In autumn 2009 this database contained ~9800 data points for sludge and ~3700 data points for wastewater from various STPs in Sweden sampled between 1995 and 2008. Additional data were collected from the literature and reports: ~720 sludge data points and ~1030 wastewater data points judged suitable for the evaluation (see below) were compiled. In total, ~350 different chemicals (e.g. single compounds, compound classes, sums of congeners, etc.) and >15 000 measurements were included in the extended data base.

In the next step, data judged to be unsuitable for the Exposure Index evaluation were discarded. Compounds or mixtures of compounds lacking a CAS number were excluded as well as compounds with CAS numbers not present in the PR. In addition, certain compound classes were excluded due to likely underestimation of quantities in the PR, for example brominated flame retardants, chlorinated dioxins and polycyclic aromatic hydrocarbons (PAHs) which are either mainly imported in articles or formed via incomplete combustion. Overestimations of chemical quantities due to export of articles was not considered. However, the loss via export of articles containing a substance is in many cases compensated by import of similar articles. Samples collected close to point sources were also discarded, as well as data points defined as outliers (see the Supporting information). A more detailed description of the data compilation and selection is found in the Supporting information. The median concentration for each chemical (including all years and locations) and a weighted (based on samples year^{-1}) average EI for that chemical were used in the regression analysis.

The selection procedure yielded a dataset of 4508 data points for 143 different compounds measured in sludge, influent and/or effluent (see Table S1).

2.3. Estimation of STP fate and physical chemical properties

The diverse fate of the chemicals in STPs can distort the relationship between emission levels and concentrations in sludge and wastewater. Hence a mass balance model was employed to correct the measured concentrations for the transport and degradation processes occurring in the STPs. For the 143 chemicals in the evaluation dataset, the EPI Suite Version 4.0 STP-model (2009) was employed to estimate the fractions of chemical in the influent that were transferred to air, sludge and effluent as well as the fraction degraded. EPI Suite was also used to calculate the