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# Model selection and evaluation for risk assessment of dioxin contaminated sites

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## ABSTRACT

The general European population has a total intake of dioxins and dioxin-like chemicals near the limit recommended by the European Union, making additional exposure above background levels undesirable. For populations living near dioxin contaminated sites additional exposure may occur by intake of locally produced food, inhalation of particles, dermal contact with soils, or by other exposure pathways. Risk assessment tools are required to estimate risks associated with contaminated sites and to set priorities for site remediation. Here, we review several multimedia models that can be applied as tools to support risk assessment. We then and present a strategy to select, apply, evaluate and adapt a model to address a specific situation. The case study we consider is a risk assessment of generic background dioxin exposure in Sweden, and we compare the predictions with environmental observations and exposure data from Sweden. Arguments are presented for selecting the CalTOX model for this case study. We demonstrate the application, evaluation and adaptation of the model, and discuss the requirements for extending the analysis to conduct risk assessment for subpopulations living near dioxin contaminated sites.

## INTRODUCTION

In the process of assessing risk and setting priorities for site remediation it is beneficial to find and further develop site-specific risk assessment tools that are

flexible and predictive. These tools should also be able to generate information on the relative significance of exposure routes. In this paper we identify and review several multimedia models that can be used as decision-support tools for risk assessment at contaminated sites. As part of a risk assessment project with the goal to compare Swedish background dioxin exposure with exposure occurring at a dioxin contaminated site, we present arguments for selecting of one of the models (CalTOX) for the actual investigation. Finally, we demonstrate a case study where CalTOX was applied, evaluated and adapted to describe a dioxin exposure scenario at a generic Swedish background site.

We used the acronym PCDD/F to represent two groups of chemical compounds, polychlorinated-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). PCDD/Fs are identified as Persistent Organic Pollutants (POPs) under the Stockholm Convention, and thus are internationally acknowledged to resist degradation in the environment for long periods, to become widely distributed geographically and to accumulate in living organisms. Among the recognized POPs, PCDD/Fs are distinguished by extremely high toxicity through a specific mode of toxic action-binding to the aryl hydrocarbon or Ah receptor (1, 2). Health organizations have assigned high cancer potencies to these compounds (1). The most toxic single chemical among the PCDD/Fs is 2,3,7,8-tetrachloro-*p*-dioxin (TCDD). The effective toxicity of a mixture of PCDD/Fs is often expressed as TCDD toxic equivalents (TEQs) (2). In this work, we used the Toxic Equivalence Factors (TEFs) established by the World Health Organization (WHO) in 1998 to quantify toxicity using what we refer to as WHO-TEQs (3). These TEFs were recently reevaluated and revised slightly (4). However, TEQ-data that were used for comparative purposes in

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this study were based on the original WHO-TEFs, and therefore we also used the original factors.

Minimizing PCDD/F contamination of the environment has been a goal of the public and governments for several decades. In the 1980s, PCDD/F emissions from combustion processes and the pulp and paper industry were a major focus, and strong measures were taken to reduce these primary sources. Today, secondary sources, such as mobilization from previously contaminated soils and sediments are receiving increasing attention. There are approximately 500 PCDD/F contaminated sites in Sweden (5), most of which are former sites of wood preservation activities. The chlorophenol agents that were used to protect the wood contained PCDD/F impurities.

Although chlorophenol agents were banned for use in Sweden in the late 1970s, the current content of PCDD/Fs in soils at saw-mills and wood preservation sites has been estimated to be 2-50 kg TEQ (5). This can be compared to total emissions of PCDD/Fs to air in Europe, which have been estimated to be approximately 6 kg TEQ per year in 2000 (6). PCDD/Fs have low vapor pressure, low water solubility and high affinity for particles. They are therefore not easily mobilized from contaminated sites and will remain in the soil for decades or longer. While a typical background soil contains a few ng TEQ/kg dry weight (dw), levels up to several ten thousands ng TEQ/kg dw are not unusual at contaminated sites (7) and levels as high as several hundreds of thousands ng TEQ/kg dw have been found (8).

The dominant exposure pathway for human exposure to PCDD/Fs is intake via food. The European Commission estimates that food is responsible for at least 95% of total intake (9). It has been shown that the exposure of humans may be of concern even at background levels (10). The tolerable weekly intake (TWI) of PCDD/Fs and chemicals with dioxin-like toxicity (dioxin-like PCBs) is 14 pg TEQ per kg

bodyweight (bw) as recommended by the European Union (11), and this recommendation is commonly referred to as a tolerable daily intake (TDI) of 2 pg TEQ per kg bw. The average intake of PCDD/Fs and dioxin-like PCBs by adults has been estimated to be 1.2-3.0 pg TEQ/kg bw/day for European countries, and the average daily intake by the Swedish population was estimated to be in the same range (average 1.4 pg TEQ per kg bw; $95^{th}$ percentile 3.0 pg TEQ per kg bw) (12). Thus, even when considering only background PCDD/F contamination, a considerable proportion of the European and Swedish populations have a daily intake close to or above the recommended limit.

Based on the information above, it is apparent that additional PCDD/F exposure above background levels can pose a health risk. For populations living near contaminated sites, additional exposure may occur by intake of locally produced food, inhalation of particles, dermal contact with soils, or by other exposure pathways. Remediation of all of the chlorophenol contaminated sites in Sweden would eliminate these exposures; however the estimated costs significantly exceed available resources. Therefore, informative risk-benefit analyses are needed to prioritize actions for mitigating the elevated risk at specific sites through actions to remediate the site or block exposure pathways. In this process, models can provide a quantitative framework to support decision making. The goals of this paper are to identify and select an appropriate model and to demonstrate how model evaluation can be used to build confidence that the selected model is a useful tool to support decision making.

## MODELING TOOLS

We conducted search to identify frequently used and easily available models appropriate for regional- or local-scale risk assessment that could be applied to

contaminated sites. This search identified five models that have been applied in Europe in the past. In this section, we provide a brief summary of these models and compare and contrast their features. All models discussed here are multimedia box models, which calculate the distribution of a chemical among various environmental compartments on the basis of chemical partitioning (13). To carry out these calculations, physicochemical properties of the compound and properties of the environment are used. Another common feature of the models is that human exposure is modeled by considering a number of potential exposure pathways. A comparison of the direct and indirect pathways included in each model is presented in Table 1. Direct exposure pathways describe exposures to the substance in a medium to which it was first released. Indirect exposure pathways are those for which there is at least one intermedia transfer, or intermediate biological transfer step between the source and the points of exposure. Below is a short review of the five models. Each has its advantages and disadvantages, and the choice of model depends on the aim of the user.

*The Swedish Model (NV):* The Swedish model for general and site specific guideline values for soils was developed at the request of the Swedish Environmental Protection Agency (14). The calculations are based on soil contamination and partitioning of the chemical between the soil, pore-water and pore-air. From these compartments, the contaminants can be transported to other compartments (groundwater, surface water and air), and concentrations in receiving compartments are calculated. Human exposure is then calculated from these environmental concentrations and dilution factors via selected exposure routes (Figure 1, Table 1). The model is static, and does not include prediction of future changes, such as declining levels due to degradation and transport out of the model world.

*CLEA:* The Contaminated Land Exposure Assessment Model (CLEA) was developed for the British Environmental Agency (15). It was created for the purpose of estimating soil guideline values and site specific assessment criteria. The model world consists of soil and air, and the only contamination source is secondary emissions from soil. It is a static model since contaminant degradation over time is not considered. There are 10 exposure routes (Table 1) and the calculated exposure is compared to certain health criteria such as tolerable daily soil intake. In the case of site-specific assessment, recommendations are given on whether further actions are needed.

*CSOIL:* This model was developed by the National Institute for Health and the Environment for the Ministry of the Environment, the Netherlands (16-18). It was designed to assess soil and ground-water quality and is a progenitor of the Swedish model. It calculates the distribution of contaminant between soil, pore-water and pore-air. Contaminants may be transported to groundwater, outdoor air and indoor air. Similar to the Swedish model and CLEA, the model is designed to handle a pool of contaminants present in the soil, but no other primary or secondary sources are considered, and neither are loss processes; it is thus also a static model. CLEA accounts for potential human exposure through seven different pathways (Table 1). It can be applied in a site-specific way to determine remediation urgency.

*EUSES:* The European System for the Evaluation of Substances (EUSES) has been developed within the European Union for risk assessment of new and existing substances and biocides (19). This model is intended to be used as a screening tool at a personal scale (for consumers and workers), at the local scale (for human populations and ecosystems near point sources) and at regional scale (for human populations and ecosystems exposed as a result of releases in a region). It has two

modules; one that estimates the distribution and transport of the chemical in environmental compartments (Predicted Environmental Concentrations; PECs), and one that calculates the human and environmental exposures from this scenario. EUSES includes air, surface water, ground water, soils and sediment compartments. It can accommodate continuous primary as well as secondary source emissions. However, the model is not designed for initial soil concentrations. EUSES is a steady state model (mass fluxes are constant over time) that considers contaminant degradation and other outflows. Most of the human exposure pathways that are considered are indirect pathways via food (Table 1).

*CalTOX:* CalTOX is a multimedia mass balance model that was developed originally by McKone (20) for the California Environmental Protection Agency to assess risks in connection with hazardous waste sites. Similar to EUSES, it integrates a local/regional multicompartment fate model with a multi-pathway human exposure model. The user can specify either continuous emissions to air, soil or surface water, or a one-time initial concentration in soil representing in-place contamination. Unlike the other models reviewed here, CalTOX can be used for calculating human exposure from contaminated soil in which the concentrations decline over time. It is, however, not a fully dynamic model since emission incidents can not be introduced other than at the initial stage. The exposure pathways include most conceivable exposure routes (Table 1).

## THE PCDD/F BACKGROUND SCENARIO FOR SWEDEN: AN EXAMPLE OF MODEL SELECTION, APPLICATION AND EVALUATION

In this section we describe the selection of the most suitable model, and the requirements for building confidence in the model. Using our selected model, we

demonstrate a process of model evaluation and adaptation to achieve satisfactory model performance for the scenario of human exposure at a Swedish background site.

## MODEL SELECTION

The first task in our investigation was to select the most appropriate model for risk assessment of PCDD/F exposure for populations living at or near contaminated land as compared to populations living at background sites. We carried out this process with the recognition that all models have inherent capabilities and limitations. But it was also important that the model selected for the PCDD/F risk assessment contain a minimum set of capabilities that we identified as necessary to capture both the potential magnitude and variation of exposures near contaminated lands as well as background exposure. Based on the aims of our risk assessment project, it was desirable that the selected model should support:

i) continuous emissions to air and water

ii) initial concentrations in soil and sediment

iii) estimation of changes over time (dynamic model)

iv) estimation of human exposure via all pathways that are potentially relevant for PCDD/Fs in situations with elevated soil concentrations, i.e. via direct as well as indirect exposure routes.

Even though CSOIL, CLEA and the Swedish model were all designed to support decisions in risk assessments of contaminated land, none of these models were designed to handle a complete range of multiple transport and/or exposure pathways as a consequence of a given source strength in the environment. Among EUSES and CalTOX, only CalTOX provided a possibility to treat both emissions to air and water as well as an initial soil concentration. Another factor that favored the

use of CalTOX was its ability to fully address long-term mass balance gains and losses by chemical degradation processes in each compartment and by transport to and removal with soil erosion, surface water outflow, and ground water movement. Although CalTOX is not a truly dynamic model, it takes degradation into consideration and initial concentrations in soil are depleted over time due to transport and transformation processes.

The models also differed in the exposure pathways that are considered (Table 1). EUSES does not focus on direct exposure pathways, which are of importance when considering exposure from contaminated soil, and the NV, CSOIL and CLEA models do not consider some food chain pathways that are of importance for estimating PCDD/F exposure (i.e. intake of eggs and cow's milk). In summary, CalTOX was the model that offered the best fit to the requirements that were set for our investigation.

However, CalTOX also has limitations. Although it allows dynamic modeling in soil layers, it does not allow for dynamic mass balance in air, surface water, and sediments and it does not allow for dynamic incidents other than initial loadings and continuous emissions. Another limitation in CalTOX (as well as the other models) is the inability to introduce an initial pool of contaminants in sediments. Such a secondary source is likely to be significant in many real-world PCDD/F contamination situations where land areas for former industrial activities are in close proximity to surface waters.

CalTOX has been examined by experts in the field, and it has been applied in several scientific studies (21-26). The design of the model is shown in Figure 2 and a compilation of the processes that are considered is shown in Table 2. The model can be used for any substance for which partition coefficients, biotransfer factors,

bioavailability, and degradability are known. Deterministic calculations can be carried out using single-value inputs producing single-value exposure estimates, and probabilistic results (mean and variance of exposures) can be obtained when one or more of the model inputs are specified using a range of values selected from a distribution of input values. The model is structured to carry out parameter sensitivity analysis and uncertainty importance ranking. CalTOX includes support for Monte Carlo uncertainty analysis using the software Crystal Ball® (Decisioneering, Inc., Denver, CO, USA), but there are several options for conducting sensitivity and uncertainty analyses.

There are three categories of input data that are needed for CalTOX: physico-chemical data ($n$=35), landscape parameters ($n$=58) and population exposure-factor parameters ($n$=54). Since CalTOX has a default option of estimating many parameter values from other parameters, not all of the input data has to be specified by the user. For example, many of the bioconcentration factors in CalTOX can be estimated using the octanol-water partition coefficient; hydrologic parameters, such as evapo-transpiration and runoff, can be estimated from rainfall; and many of the human-activity parameters can be set at default values that are representative of norms in industrialized countries.

## BUILDING CONFIDENCE IN A MODEL AND ITS RESULTS

Identifying a model that meets the user's selection criteria is only the first step in the process, and is not adequate to assure that the model will provide useful information for risk assessment decisions. Any model, even one that is widely used and commonly cited, should be subjected to a performance evaluation that builds confidence in the appropriateness of the model to support a specific decision. For

example, Bonnard (26) reviewed the use of CalTOX in French risk assessment investigations, and showed that in spite of the many advantages of the model and its value for assessing waste sites in France, there are many common errors that have been made in applying this model. These errors cause loss of confidence in risk assessments based on models. Bonnard suggests that many of the errors are the result of using the model as a "black box", and that this problem can be avoided with a model evaluation process.

Here, we divide model evaluation into three steps: 1) obtaining appropriate input data, 2) evaluation of model output values by comparing with measurement data, and 3) adapting the model and re-evaluating input data to achieve better agreement with real world observations. Deviations between the model and observations can be due to applying an inappropriate model or lack of reliability in measurements and input data. In the sections below, we consider some aspects of the performance evaluation that can increase or decrease the confidence about the results obtained from a model.

*Selecting input data.* Reliable model performance can only be expected if input data are accurate. In particular, selecting physicochemical properties values to describe the chemical may be challenging since there is generally a range of measured and calculated values in the literature (27). In many modeling studies, property values are selected on the basis of expert recommendations. But to increase confidence in the model output values, it is advisable to select chemical properties data using methods that exploit the relationships between partition coefficients to identify values that may be in error, and in some cases to calculate a single set of values that are internally consistent (28, 29). For our case study we reviewed the literature and evaluated the internal consistency of our selected partitioning properties. However, there are large

uncertainties about the true values of the partitioning properties of the PCDD/Fs due to wide variability in reported values and a degree of expert judgment is required to select appropriate values.

*Comparison with measurement data.* Environmental fate models are incomplete descriptions of open systems consisting of the chemical and the environment (30). Therefore they cannot be validated using methods that work for models that describe completely closed, controlled systems. However, in any risk assessment it is important to evaluate the performance of the model by comparing predicted environmental concentrations and exposures with available data. Ideally, a model evaluation and adaptation process is conducted for a site specific scenario for which recent environmental measurements have been conducted. However, even if the temporal and spatial scale is not always a perfect match, this kind of evaluation is necessary and can help to find weaknesses. It is also advantageous for the model evaluation to include chemicals with a range of physico-chemical properties.

*Adapting the model and re-evaluating input data to address differences between model output and measurements.* An important step in the model evaluation process is to scrutinize the model output values that do not agree with relevant measurement data that is believed to be reliable. Quality of input data as well as usefulness of algorithms must be evaluated. In this process, it is advantageous to start with sensitivity and uncertainty analyses. These procedures help to identify parameters with a large impact on the results. A complete description of these methods is beyond the scope of this summary paper, but the widely cited text by Morgan and Henrion (31) can be consulted for a summary and suggestions. Once the critical parameters and model algorithms are identified, efforts can be focused on refining the input data and model algorithms as much as possible.

## MODEL APPLICATION, EVALUATION AND ADAPTATION

After the model selection, we created a background scenario for Swedish conditions and modeled human exposure by using the selected model (CalTOX). This scenario is intended to be the base case, which can be compared to a similar scenario in which a highly contaminated soil is introduced. Our case study considered six tetra- through octa-chlorinated dioxin and furan congeners (2,3,7,8-TCDD, 2,3,4,7,8-PeCDF, 1,2,3,6,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF and OCDD). The congeners were selected to represent i) a broad range of physicochemical properties of PCDD/Fs (2,3,7,8-TCDD and OCDD), ii) congeners that commonly contribute significantly to the TEQ-value at contaminated sites (1,2,3,6,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDD and 1,2,3,4,6,7,8-HpCDF), and iii) a congener that is relatively abundant in food (2,3,4,7,8-PeCDF).

We modeled a land area of 10,000 $m^2$, and assumed that there was surface water with an average depth of 3 m covering 1000 $m^2$ of this area. The modeled area reflects the typical size of a contaminated site in Sweden. The mixing height of the atmosphere over the modeled area is assumed to be 9 m and the residence time of air is 100 seconds. Climate data was selected to represent southern Sweden and other landscape parameters were matched as much as possible to available data from Swedish and Norwegian investigations. The modeled soil system consists of three layers (from top to bottom); the surface layer (2 cm), the root layer (1 m) and the vadoze zone (1 m), and the organic carbon content for each layer (from top to bottom) were set to 3.6%, 3.6% and 0.4 %, respectively. The thickness of the sediment layer was set to 5 cm and its organic carbon content to 2.9%.

It is generally believed that the most important current PCDD/F releases are air emissions from current activities and/or reservoir sources (32). As a result, our background scenario was based on continuous air emission and an in-place reservoir in soils as PCDD/F sources. The emission parameters were adjusted to levels at which the calculated air and surface soil concentrations were in good agreement with average concentrations measured at background sites in southern Sweden since the year 2000 (33, 34).

Measurements of PCDD/Fs are expensive and often excluded from national environmental monitoring programs. For evaluating our background scenario, we were obliged to resort to relatively scattered data from the peer-reviewed literature, Swedish reports and also some unpublished (in-house) values. As described below, Figures 3 and 4 show monitoring data for PCDD/Fs from background sites in and around Sweden that we used to evaluate the model results. The modeled data in Figures 3 and 4 is the final result obtained after model adaptation and re-evaluation of input data. However, in our initial model evaluation based on our first selected input data and the default model algorithms, the model underestimated PCDD/F concentrations in vegetation by approximately an order of magnitude relative to measurement data, and aquatic and the terrestrial food chain transfers of the PCDD/Fs were found to be out of agreement with measurement data by up to several orders of magnitude. The unsatisfactory agreement between modeled and measured environmental concentrations and exposures was evidence of inappropriate input data and model limitations that required actions to improve model performance. Here, we summarize our experience, which provides an example of how to overcome initial prediction errors.

*Concentrations of PCDD/Fs in vegetation*. With its default parameterization, the CalTOX model systematically underestimated concentrations of PCDD/Fs in vegetation leaves as compared to available data for grass gathered at a background site in Sweden. Analysis of the model results indicated that the calculated concentration in whole leaves was being controlled by the assumed rate of cuticle erosion, which transfers PCDD/F to the soil from the plant surface. The half-time for this process used by default in the model is 14 days (35), which is an estimate based on a consensus opinion of experts. However, more recent experiments that studied the uptake of semi-volatile organic chemicals by grass from air indicate that at least six weeks is required to approach steady-state conditions (36). We therefore changed the model input value for half-time for cuticle erosion to 42 days, which dramatically improved agreement between the model and measurement data.

*The terrestrial food chain transfer of PCDD/Fs*. Critical parameters for meat, cow's milk and egg concentrations are the various biotransfer factors (BTFs), which describe the degree of uptake of the contaminant in animals and the rate of transfer from feed to animal and further to food products (egg, milk). At an early stage of the modeling, we noted that the CalTOX model overestimated the meat and cow's milk exposure routes for highly chlorinated PCDD/Fs. The default algorithms for these pathways are based on work by Travis and Arms (37), which assume linear relationships between the logarithm of biotransfer factors and the logarithm of the octanol-water partition coefficients ($K_{ow}$). However, the bioavailability for super hydrophobic compounds (log $K_{ow}$>6) is substantially limited. Thus the highly chlorinated PCDD/Fs are less available for uptake, and the BTFs decline with increasing hydrophobicity for highly chlorinated congeners. For meat and cow's milk, a better agreement between predicted and measured values was achieved by using

experimentally determined BTFs (38) rather than default algorithms in CalTOX. For eggs, no experimental BTFs could be found in the literature, and we were therefore relied on the default algorithms. The congener patterns and absolute levels of the modeled meat and milk exposures were in good agreement with those found in the food survey, in both cases within a factor of 4 of reported exposures for 7 out of 12 comparisons and in all cases within a factor of 8, except for OCDD in meat (prediction 24 times below observed value; Figure 4). Egg exposure was clearly under-predicted for the lower chlorinated congeners, but agreement for the higher chlorinated congeners was within a factor of 3.

*The aquatic food chain transfer of PCDD/Fs.* The limited bioavailability of super hydrophobic chemicals is also not considered in the default description of the bioconcentration process from water to fish in CalTOX. Similar to the terrestrial food chain transfer, a better agreement between predicted and measured values was achieved if the CalTOX default BCFs were replaced by measured bioconcentration factors (BCFs), and the best fit was obtained by using BCFs published by Govers and Krop (39) (Figure 4). Analysis of fish from Swedish lakes with background levels generally showed PCDD/F concentrations below the limit of detection, and therefore a comparison was made with fish from a lake that is located in an urbanized region in Sweden. The average value of six composite samples of salmon (*Salmo salar*), trout (*Salmo trutta*) and char (*Salvelinus alpinus*) from Lake Vättern was used (two samples per species; 7-10 specimens in each composite) (40). The individual fish weights ranged from 0.5 g to 3.7 kg (average 1.9 kg) and the lipid weights from 1.3 to 4.6% (average 2.9%). The observed values were significantly higher than the predicted (Figure 4). The large deviation between observed and predicted values is partly attributable to the difficulties in finding relevant comparison data, but primarily

to the use of the water-concentration BCF product in CalTOX to calculate contaminant concentrations in fish. This approach neglects the bioaccumulation of contaminants by high-trophic-level fish via intake of lower trophic level prey. For high trophic level fish it is more appropriate to use bioaccumulation factors (BAFs). The use of BAFs observed for adult lake trout (*Salvelinus namaycush*) from Lake Ontario (41) instead of using BCFs resulted in good agreement between predicted and measured values. The lipid fraction of the Lake Ontario trout was 8%, and therefore the BAF-values shown in Figure 4 were down-adjusted to match a lipid fraction of 3% in order to better compare with the Lake Vättern data.

The predicted values in the CalTOX model are on wet weight basis, and there is no option in the model to distinguish between lean and fat fish. In light of all these issues, it is clear that modeling “average” fish concentrations is a challenge. A step towards better predictions is to expand the fish model so that differentiated information on the fish that is consumed can be used. From the fish concentration modeling perspective, it would also be beneficial to adjust the model so that it allows initial dioxin concentration in the sediment. The sediments often act as significant secondary sources to the water column and to the benthic food chain due to historically higher emissions, thereby also influencing the fish levels and the exposure from fish ingestion.

As stated earlier, a more complete and comprehensive model evaluation would be possible with consistent site-specific measurements and modeling. However the findings described above are an example of the benefit of conducting model evaluation even for generic scenarios. Our experience also highlights the advantage of including congeners with a range of physico-chemical properties for the evaluation of a modeled scenario.

## PERFORMANCE OF THE ADJUSTED MODEL

In Figure 3, we compare the final, adapted model output for concentrations in vegetation, surface water, and wet and dry atmospheric deposition to Swedish reference data for three of the congeners (2,3,7,8-TCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDF). Figure 3 also shows the background concentrations in air and surface soil that were fitted as emission parameters. The model output and observed concentrations/rates in most cases agreed within a factor of 4. The exception is the atmospheric deposition rate of 2,3,7,8-TCDD, which the model underestimates by an order of magnitude. The extent of gas-particle partitioning of 2,3,7,8-TCDD is sensitive to partitioning properties, and the observed discrepancy could be attributable to errors in the specific values selected for the study. However, considered as a whole, Figure 3 indicates that the model results are in very good agreement with observations, especially considering that the monitoring data has been collected from various sites around Sweden.

The corresponding exposure modeling was based on average food consumption values compiled by the National Food Administration in Sweden (12, 42). We used the default intake rates for an adult population and modeled the PCDD/F exposure near the background site described above. The predicted TEQ-values of direct and indirect exposures from the adjusted model are shown in Table 3, and exposures via specific food pathways are shown in Figure 4. Predicted direct exposure contributed only 1.5% of the total. This prediction is near the estimations from European measurement data, which indicate that direct exposure generally accounts for only 2-5% of the total exposure (9).

The indirect PCDD/F exposure (intake via food, including PCDD/Fs and no other dioxin-like compounds) for the Swedish population has been estimated to be 0.05-12 pg TEQ/kg bw per day (min and max values) and on average 0.8 pg TEQ/kg bw per day by analyzing a typical Swedish “food basket” (12). The comparison between the modeled food exposures in Table 3 and these food basket values is problematic since in the food basket study, all of the seventeen 2,3,7,8-chlorinated congeners were included, and in the modeled TEQ-value only six of these highly toxic congeners were considered. We estimated that these limitations will lead to an underestimation of TEQ intake by a factor of 2. Thus, in a more complete comparison with the national food intake value, the modeled value should be doubled. By considering this, the predicted value (0.12 pg TEQ/kg bw per day) is within the range estimated in the food basket survey, and 7 times lower than the Swedish average value (Table 3). We view this as reasonable given that the model scenario represents a background site and does not include exposure from consumption of fish with elevated concentrations, which is known to be an important exposure pathway for the general population of Sweden (12). It should also be noted that food-basket surveys also are subject to uncertainties and variability in their estimates of population exposure.

## DISCUSSION AND CONCLUSIONS

The case study described here clearly indicates that model selection is only the beginning of a successful and informative modeling process. Once a model is selected, a rigorous process of model evaluation is needed to build confidence in the performance of the model in the context of the specific issue under consideration. For the case study, satisfactory agreement between modeled values and available

measurement data was demonstrated after the model evaluation and adjustment process. However, it also became clear that site specific measurements and renewed evaluations and adjustments are required in order to establish sufficient confidence in the model to function as a reliable risk assessment tool for human PCDD/F exposure near contaminated sites.

The case study presented here supports our selection of the CalTOX model as a tool that can quantitatively inform risk assessment when applied in conjunction with appropriate monitoring data that enables model evaluation. Our experience provides several instructive lessons about the capabilities and limitations of a model-based site-specific risk assessment. In particular, risk assessors must be aware that confidence in model results can only be built based on successful model evaluation for cases similar to the one of interest. This requires a foundation of reliable input data for chemical properties and landscape parameters, and comparison of the model results with monitoring data appropriate to the space and time scales of the assessment. In our case study, we found that property data, landscape parameters and the default model algorithms can all introduce significant uncertainties in the assessment. To achieve satisfactory agreement between the model and monitoring data, careful selection of input data and adjustments to the model algorithms were required, namely using updated physiological parameters for cuticle erosion, using experimentally determined bioconcentration factors or BAFs derived from field studies rather than default algorithms in CalTOX, and using experimentally determined biotransfer factors rather than default algorithms for calculation of PCDD/F concentrations in cow's milk and beef.

This work shows that models should not be used blindly as a "black-box" source of information in risk assessments. There is a strong incentive to develop and

implement models with high predictive power, especially since quantitative risk assessment facilitates the formulation of risk management strategies. However, applying and interpreting chemical fate and exposure models requires expertise and experience. Decision makers should not expect models to provide unequivocal answers, and sound management strategies must be based on a combination of quantitative modeling and expert judgment. In the case study presented here, we provide an example of a sequential process that includes model selection, input data quality evaluation, collection and interpretation of observations, model evaluation, and model adaptation. Our results show that this process offers insight and knowledge that neither a model nor environmental data alone can provide.

Table 1. Exposure pathways included in selected risk assessment modeling tools (modified from Rikken *et al.* (17); consumption of egg and supplementary details for Swedish model (NV) were added).

| | | EUSES | NV | CSOIL | CLEA | CalTOX |
|---|---|---|---|---|---|---|
| **Direct exposure** | Ingestion of contaminated soil particles (outdoor) | | x | x | x | x |
| | Ingestion of contaminated soil particles / dust (indoor) | | x | x | x | |
| | Dermal contact with soil contaminants (outdoor) | | x | x | x | x |
| | Dermal contact with soil contaminants / dust (indoor) | | x* | x | x | |
| | Inhalation of contaminated soil particles / dust (outdoor) | | x | x | x | x |
| | Inhalation of contaminated soil particles / dust (indoor) | | x | x | x | x |
| | Inhalation of vapors from contaminants (outdoor) | x | x | x | x | x |
| | Inhalation of vapors from contaminants (indoor) | | x | x | x | x |
| **Indirect exposure** | Consumption of crops | x | x | x | x | x |
| | Consumption of soil attached to crops | | | | x | x |
| | Consumption of fish | x | x | | | x |
| | Consumption of meat | x | | | | x |
| | Consumption of egg | | | | | x |
| | Consumption of milk | x | | | | x |
| | Consumption of contaminants via mothers' milk | | | | | x |
| | Consumption of contaminants via drinking water | x | x | x | | x |
| | Ingestion of contaminants via bathing water | | | | | x |
| | Inhalation of vapors from drinking water from shower | | | x | | x |
| | Dermal contact with drinking water from shower/bath | | | x | | x |
| | Dermal contact with contaminants via bathing water | | | | | x |

x: Exposure route available; x*: Exposure route available, but not as a separate pathway**; EUSES**: European System for the Evaluation of Substances The Contaminated Land Exposure Assessment Model (19); **NV**: The Swedish model (14); **CSOIL**: Model developed by the National Institute for Public Health and the Environment, the Netherlands (16-18): **CLEA**: Contaminated Land Exposure Assessment Model, U.K. (15); **CalTOX:** Model developed for the California Environmnetal Protection Agency (20)

Table 2. Summary of the processes by which contaminants are exchanged and lost among seven CalTOX compartments (20).

| Compartment | Gains | Losses |
|---|---|---|
| **Air** (both the gas phase and particles of the troposphere) | diffusion from soil<br>diffusion from plants<br>diffusion from surface water<br>resuspension of deposited soil particles<br>***contaminant sources*** | diffusion to ground-surface soil<br>diffusion to surface water<br>diffusion to plants<br>washout by rainfall<br>convection losses<br>deposition to soil<br>deposition to plants<br>deposition to surface water<br>chemical/physical transformation |
| **Plants** (land vegetation) | deposition of particles from air<br>foliar uptake<br>root-uptake from root-zone soil | diffusion from leaf surfaces<br>wash off from leaf surfaces<br>chemical/physical transformation |
| **Ground-surface soil** | diffusion from air<br>diffusion from root-zone soil<br>washout from air by rainfall<br>dry deposition of air particles<br>***contaminant sources*** | diffusion to air<br>diffusion to root-zone soil<br>advection to root-zone soil<br>soil solution runoff<br>erosion (mineral runoff) to surface water<br>resuspension of soil particles<br>chemical/physical transformation |
| **Root-zone soil** | diffusion from ground-surface soil<br>advection from ground-surface soil<br>***contaminant sources*** | diffusion to ground-surface soil<br>infiltration (leaching) to vadose zone soil<br>chemical/physical transformation |
| **Vadose-zone soil** | infiltration from root-zone soil<br>***contaminant sources*** | infiltration to ground-water zone |
| **Surface water** | diffusion from air<br>washout by rainfall<br>deposition of atmospheric particles<br>soil solution runoff<br>erosion (mineral runoff)<br>diffusion from sediment<br>sediment resuspension<br>***contaminant sources*** | sediment deposition<br>diffusion of vapors to air<br>diffusion to sediment<br>surface-water outflow<br>chemical/physical transformation |
| **Sediment layer** | diffusion from surface water<br>sediment deposition (from surface water) | diffusion to surface water<br>sediment resuspension<br>chemical/physical transformation |

Table 3. Modeled and measured PCDD/F exposure for an adult population living at a background level site (pg WHO-TEQ/kg bodyweight and day).

| | Direct exposure | Indirect exposure | Total exposure |
|---|---|---|---|
| Modeled value including six of the seventeen 2,3,7,8-chlorine substituted PCDD/F congeners[a] | 0.0009 | 0.06 | 0.06 |
| Estimated value including all of the seventeen 2,3,7,8-chlorine substituted PCDD/F congeners[b] | 0.002 | 0.12 | 0.12 |
| Measured values, typical Swedish "food basket" (12) | | | |
| - range (min-max) | NA | 0.05-12 | NA |
| - average | | 0.8 | |

[a]Congeners included: 2,3,7,8-TCDD, 2,3,4,7,8-PeCDF, 1,2,3,6,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF and OCDD; [b]The value vas estimated by multiplying the modeled TEQ-value for six congeners by a factor of 2; NA: not analyzed

## Figure Legends

**Figure 1.** An overview of the structure of the Swedish EPA calculation model for site specific limit values for use of land.

**Figure 2.** The framework of the CalTOX model.

**Figure 3**. Modeled concentrations of A) 2,3,7,8-TCDD, B) 1,2,3,6,7,8-HxCDD and C) 1,2,3,4,6,7,8-HpCDF in background scenario as compared to measured concentrations of Swedish environmental compartments; air (ng/$m^3$) (33) ground surface soil (ng/kg wet weight) (34); total leaf (ng/kg dw) and surface water (ng/l; unpublished data, Umeå University, Sweden) and atmospheric wet and dry deposition (ng/day) (33).

**Figure 4.** Modeled and measured concentrations (mean and one standard deviation) of meat, diary products/cow's milk, egg (12) and fish (Lake Vättern; salmon, trout and char from the northern part; average of six pooled samples, two from each species) (40) in ng WHO-TEQ/kg wet weight. BTF: Biotransfer factor; BCF: Bioconcentration factor; BAF: Bioaccumulation factor. The modeled values were obtained by using BTFs from Birak *et al.* (38), BCFs from Govers and Krop (39) and BAFs from Morrison *et al.* (41). The modeled BAF-values were obtained by assuming a lipid weight of 3%.

Figure 1

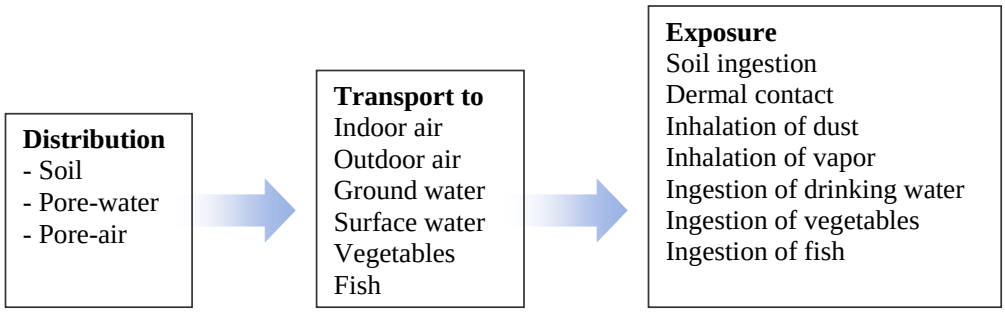
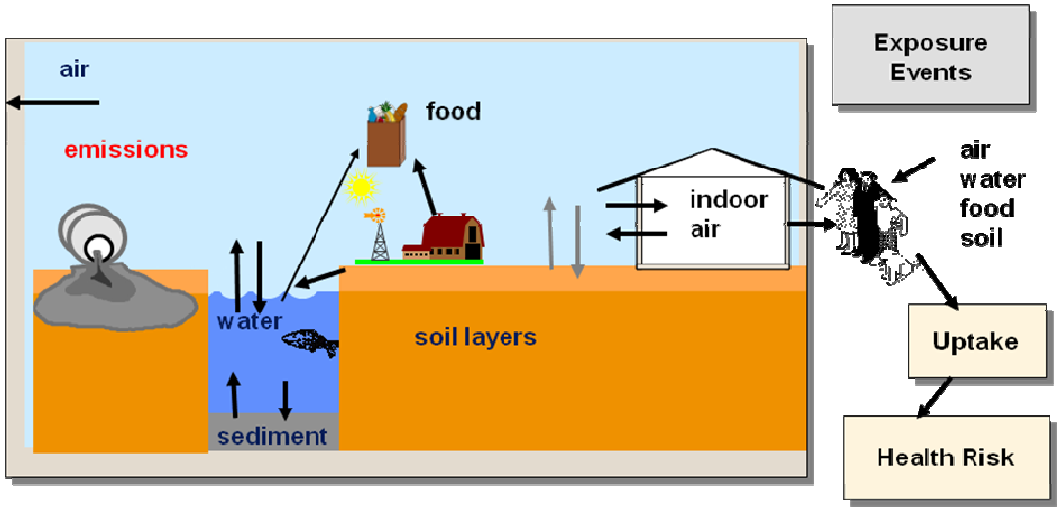
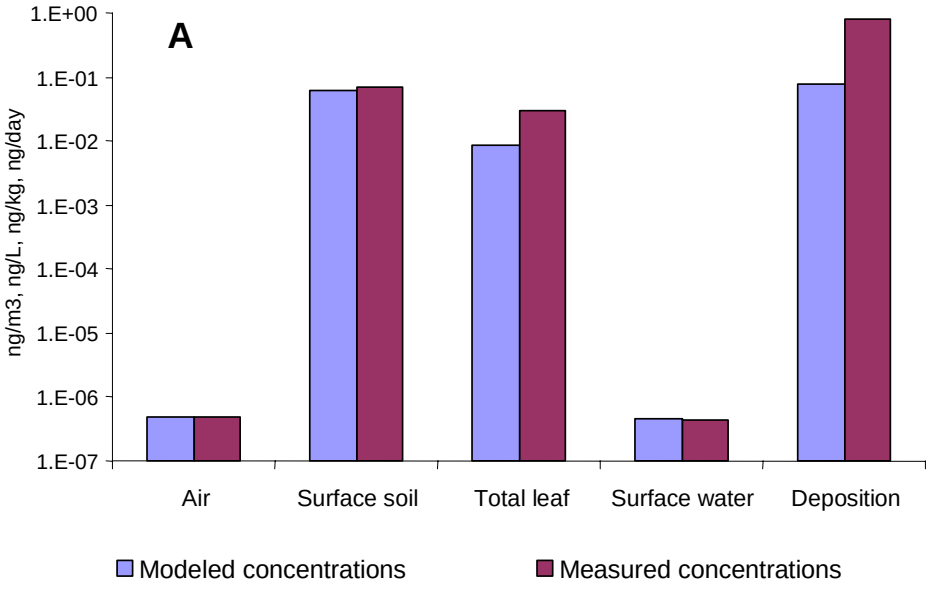


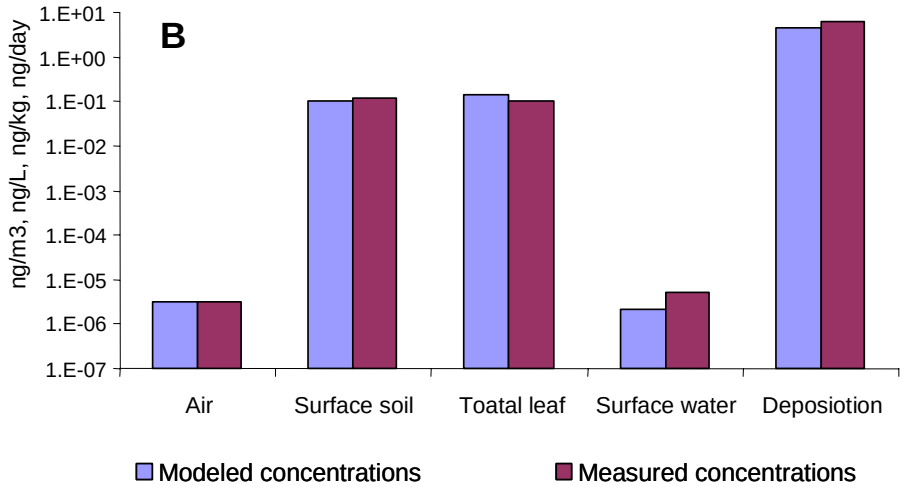

Figure 2



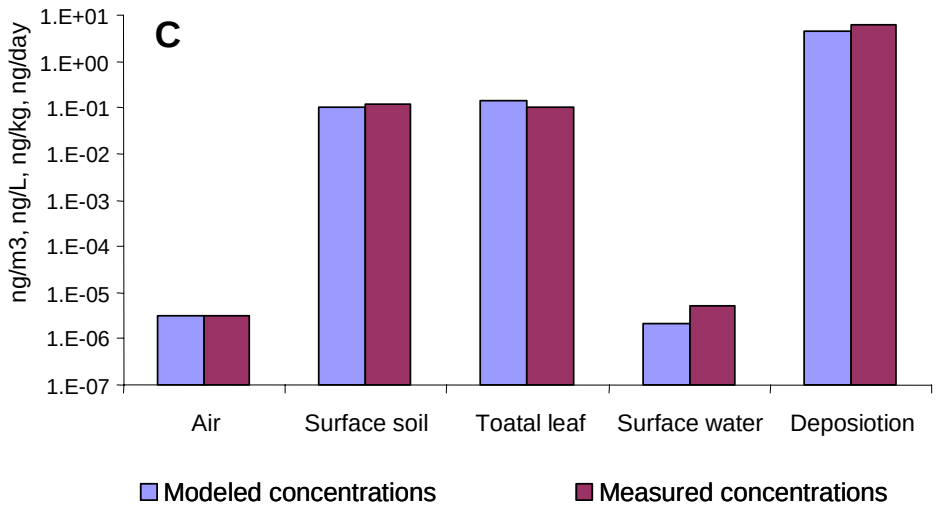
1 Figure 3


A
1.E+00
1.E-01
1.E-02
1.E-03
1.E-04
1.E-05
1.E-06
1.E-07
ng/m3, ng/L, ng/kg, ng/day
Air
Surface soil
Total leaf
Surface water
Deposition
Modeled concentrations
Measured concentrations

2

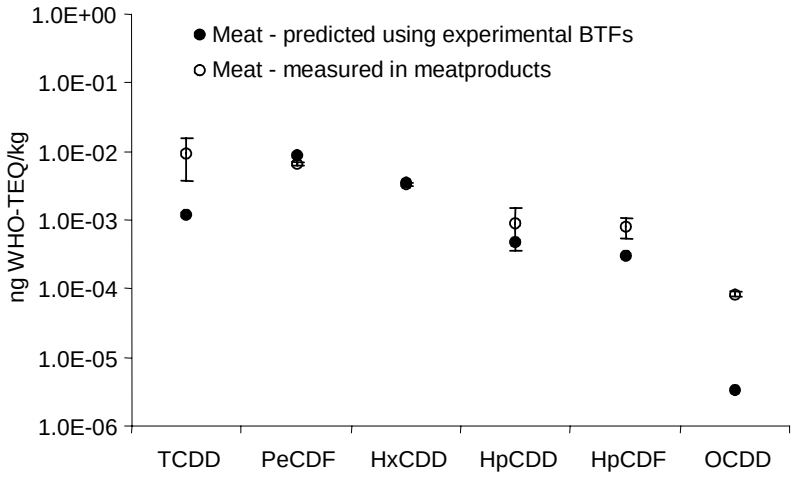

B
1.E+01
1.E+00
1.E-01
1.E-02
1.E-03
1.E-04
1.E-05
1.E-06
1.E-07
ng/m3, ng/L, ng/kg, ng/day
Air
Surface soil
Toatal leaf
Surface water
Deposiotion
Modeled concentrations
Measured concentrations

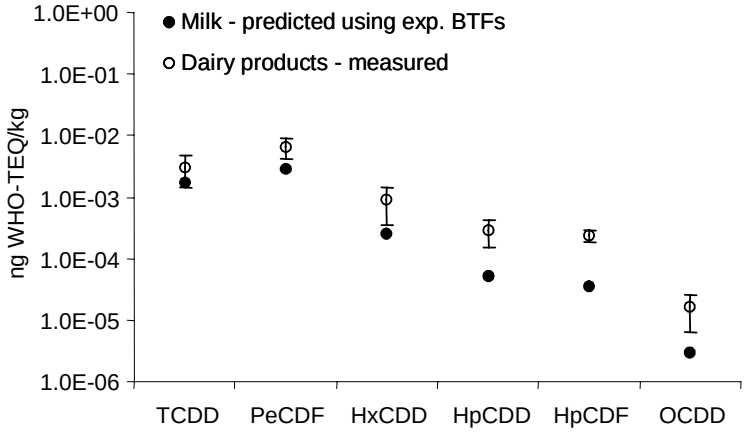
3

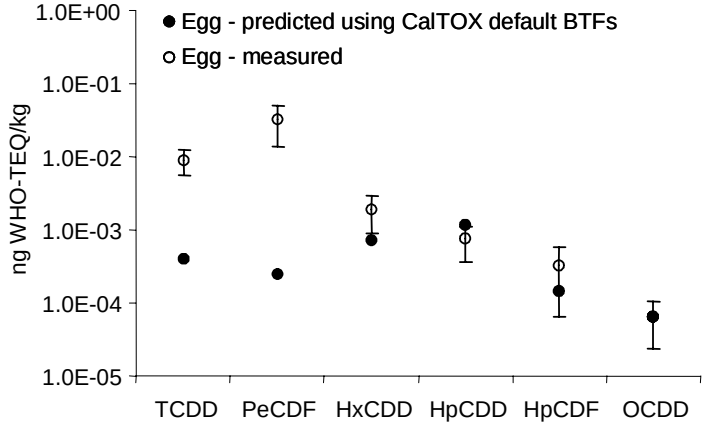

C
1.E+01
1.E+00
1.E-01
1.E-02
1.E-03
1.E-04
1.E-05
1.E-06
1.E-07
ng/m3, ng/L, ng/kg, ng/day
Air
Surface soil
Toatal leaf
Surface water
Deposiotion
Modeled concentrations
Measured concentrations

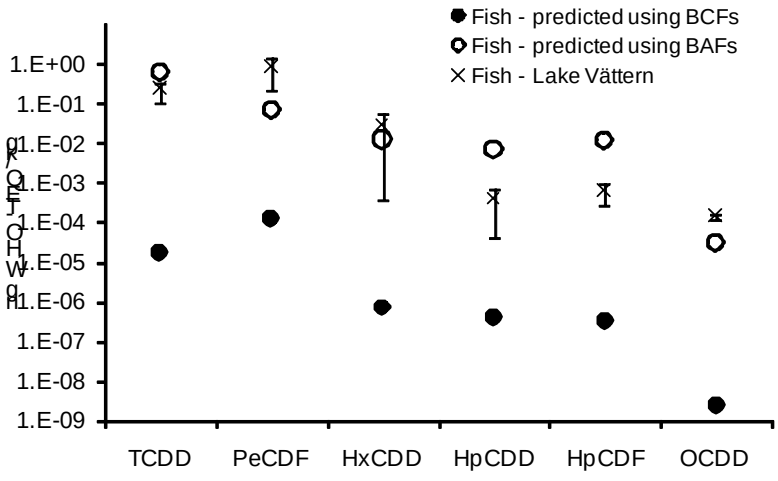
1
2

1 Figure 4


1.0E+00
1.0E-01
1.0E-02
1.0E-03
1.0E-04
1.0E-05
1.0E-06
ng WHO-TEQ/kg
Meat - predicted using experimental BTFs
Meat - measured in meatproducts
TCDD
PeCDF
HxCDD
HpCDD
HpCDF
OCDD


1.0E+00
1.0E-01
1.0E-02
1.0E-03
1.0E-04
1.0E-05
1.0E-06
ng WHO-TEQ/kg
Milk - predicted using exp. BTFs
Dairy products - measured
TCDD
PeCDF
HxCDD
HpCDD
HpCDF
OCDD


1.0E+00
1.0E-01
1.0E-02
1.0E-03
1.0E-04
1.0E-05
ng WHO-TEQ/kg
Egg - predicted using CalTOX default BTFs
Egg - measured
TCDD
PeCDF
HxCDD
HpCDD
HpCDF
OCDD


Fish - predicted using BCFs
Fish - predicted using BAFs
Fish - Lake Vättern
1.E+00
1.E-01
1.E-02
1.E-03
1.E-04
1.E-05
1.E-06
1.E-07
1.E-08
1.E-09
TCDD
PeCDF
HxCDD
HpCDD
HpCDF
OCDD

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