Quality Assurance of Exposure Models

for Environmental Risk Assessment of Substances

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Summary

Environmental risk assessment of chemical substances in the European Union is based on a harmonised scheme. The required models and parameters are laid down in the Technical Guidance Document (TGD) and are implemented in the EUSES software. Although the results may have a considerable ecological and economic impact, guidance is rarely given on the applicability of the framework. To fill this gap, an evaluation study of the TGD exposure models was carried out. In particular, the models for estimating chemical intake by humans were investigated. These models, which are a key component in risk assessment, involve a quantification of human contact with environmental contamination in various media of exposure through various exposure pathways. The objective of this study was two-fold: firstly, to develop an evaluation methodology, since no appropriate approach is available in the scientific literature. Secondly, to elaborate applicability and limitations of the models and to provide proposals for their improvement.

The principles of model evaluation in terms of quality assurance, model validation and software evaluation were elaborated and a suitable evaluation protocol for chemical risk assessment models was developed. Since scientific theories and the mathematical models embedded therein cannot be proved as true, a pragmatic meaning of validation is required, of which the primary purpose is to increase the level of confidence placed in the model. The accuracy of the model outcome is a necessary, but insufficient criterion for the quality assurance of models. A wider approach is required which examines the scientific inference that can be made about models with regard to their intended purpose. By reviewing the literature on the validation problem, it was found that all the facets of validation can be assigned to generic (internal) and task-specific (external) properties of a model. In this context, sensitivity and uncertainty analyses are essential to tackle the issues of uncertainty. Sensitivity analysis aims to ascertain how a given model depends upon the information fed into it. Uncertainty analysis aims to quantify the uncertainty regarding what comes out of the model. It was argued that targeted uncertainty analysis and sensitivity analysis, as a part of it, is capable of reducing critical uncertainties and represents an essential contribution for assuring the quality of a model. Appropriate and detailed quality criteria for fate and exposure assessment software were developed. These are based on common standards for software supplemented by specific requirements for application in risk assessment. Altogether, quality assurance of a model includes internal and external validation, and addresses the evaluation of the respective software. It should focus not only on the predictive capability of a model, but also on the strength of the theoretical underpinnings, evidence supporting the model's conceptualisation, the database and the software.

The evaluation protocol was subsequently processed and applied to the TGD human exposure models. External validation was performed using a set of reference substances with different physico-chemical properties and use patterns. Substances of interest were PCDD, PCB, DEHP, HHCB, LAS, EDTA, benzene and 1,2-dichloroethane. By using different scenarios, model calculations were carried out and the results were compared with monitoring data and experimentally determined values. The comparison was carried out for single submodels on the one hand and for the

entire system on the other. For the latter, two scenarios were applied: for the default parameter set of EUSES and for a parameter set representing the German State of North Rhine-Westphalia.

From a theoretical point of view, it was shown that the models strongly depend on the lipophilicity of the substance, that the underlying assumptions drastically limit the applicability, and that realistic concentrations may seldom be expected. If the models are applied without adjustment, high uncertainties must inevitably be expected. In several cases, considerable (explicable) deviations from the measured values were found. This affects extremely lipophilic substances or substances with degradation. Altogether, the comparison to measured real field data shows that for the test chemicals, an accuracy within a factor of ten is rarely achieved. It was shown that the concentrations are overestimated by up to two orders of magnitude for the aquatic environment. For superlipophilic and persistent chemicals, higher uncertainties emerge and measured concentrations may also be underestimated. The deviations are caused by unrealistic bioconcentration factors or metabolism on the one hand and by neglecting biomagnification on the other. The biotransfer model for meat and milk represents a conservative estimation. The overestimation is most significant for non-persistent or superlipophilic substances with more than two orders of magnitude. A lack of steady state, metabolism and/or reduced resorption were presumed to be the reasons. The model for describing uptake by plants often leads to an underestimation of the measured concentrations because the model considers chemical uptake from air only via gas exchange. The calculated total daily dose was compared with alternative estimations available from the literature. For several chemicals it corresponds with deviations within two orders of magnitude (for chemicals without a lack of data) when applying more realistic intake values. It was found that low deviations are sometimes caused by an equalising effect of overestimations and underestimations in the submodels. The sensitivity analysis revealed that the total daily dose is sensitive to the majority of parameters if a variety of chemicals is investigated. However, there is a set of parameters with negligible impact. Few of the sensitive parameters show extremely sensitive values and should be treated with caution. In order to assign sensitive parameters to substance classes, it is sufficient to distinguish between lipophilic, waterborne and airborne substances. Taking the distribution of input parameters into account, the result only depends on a relatively small subset of parameters. Depending on the substance, up to a quarter of all parameters are important. The uncertainties are high for chemicals ingested via the food chain and lower for those ingested directly via air or drinking water. Only the parameters of the exposure module are important for the former, and parameters from all submodels are important for the latter.

Regarding the software, it was found that EUSES basically fulfils the postulated quality criteria. Particularly with regard to correctness and stability, (almost) no errors were found. EUSES contains some innovative features. However, numerous alterations are necessary. High complexity, low modularity, and incomplete documentation result in a lack of transparency and are emphasised as major points of criticism. To overcome the inadequacies a more modular design is proposed.

All in all, the overall system was classified as a good compromise between complexity and practicability. But several chemicals and classes of chemicals, respectively, with several restrictions were revealed: The investigated models used to assess indirect exposure to humans are in parts currently not applicable for dissociating compounds, very polar compounds, very lipophilic com-

pounds, ions, some surfactants, and compounds in which metabolites provide the problems and mixtures. In a strict sense, the method is only applicable for persistent, non-dissociating chemicals of intermediate lipophilicity. Further limitations may exist. Finally, recommendations for improvements and maintenance of the risk assessment methodology were presented. Relevant processes which were not included should be considered, several new and simpler concepts should be added, and the relevancy of certain exposure pathways has to be refined urgently.

Keywords

Risk assessment, TGD, EUSES, quality assurance, model validation, software evaluation, fate and exposure models, uncertainty analysis, sensitivity analysis, scenario analysis, assumptions, limitations.

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Abbreviations

ASCII	American Standard Code for Information Interchange
BCF	Bioconcentration factor
BTF	Biotransfer factor
CR	Carryover
DSS	Decision Support System
ECB	European Chemicals Bureau
EU	European Union
EUSES	European Union System for the Evaluation of Substances
FG	Fresh weight (= wet weight for fish)
HEDSET	Harmonised Electronic Data Set
IC	Industry Category
LG	Lipid fraction
MC	Main Category
MCI	Molecular Connectivity Index
n	Sample size
NRW	North Rhine-Westphalia (State in Germany)
NG	Detection limit (German: Nachweisgrenze)
OoM	Orders of magnitude
PEC	Predicted Environmental Concentration
PNEC	Predicted No Effect Concentration
QSAR	Quantitative Structure-Activity Relationship
QSPR	Quantitative Structure-Property Relationship
QPPR	Quantitative Property-Property Relationship
SNIF	Structured Notified Interchange Format
STP	Sewage Treatment Plant
TG	Dry weight
TGD	Technical Guidance Documents
TOC	Total Organic Carbon Content
UC	Use Category
WWW	World Wide Web

Preface

Risk assessment of chemicals requires the application of mathematical models. The European Union risk assessment scheme provides a framework including software in the form of the *Technical Guidance Document* (TGD) and the *European Union System for the Evaluation of Substances* (EUSES). Nevertheless, an evaluation of the entire system regarding its applicability and limitations is lacking.

Neither a standard nor a consensus on how to evaluate such models exists in the scientific theory. Thus, the development of an appropriate methodology was required, which is presented in Chapters 2 and 3. The methodology is also useful for evaluating similar models in the context of chemical fate and exposure assessment. Chapters 4 and 5 present the models and underlying database. Presentation of the results is to be found in Chapters 6 to 11. An evaluation of the entire system including proposals for improving it is given in a concluding chapter. It is intended to contribute to a forthcoming update of models and software.

Both the entire system and individual models are investigated. The paper should also be viewed as a reference book to support the user. The nomenclature corresponds with the TGD (EC 1996A) and EUSES documentation (EC 1996B), respectively. In order to assure lucidity, names of variables were sometimes abbreviated.

This paper is one out of two parts of a superior validation study. It focuses on the food chain part of the TGD and on the software evaluation. The regional distribution model was validated by BERDING (2000).

Osnabrück, June 2000

1 Introduction

The risk posed by existing and new notified chemical substances to humans and the environment is to be evaluated within the framework of the implementation of European chemicals legislation. The EU member states put forward procedures for a harmonised risk assessment of chemicals in the *Technical Guidance Document (TGD) on directive 93/67/EEC and regulation (EC) 1488/94* (EC 1996A). With the *EUSES (European Union System for the Evaluation of Substances)* software a computer programme was developed which contains the mathematical models and calculation processes described in the TGD (EC 1996B).

The risk assessment methodology is based on a four-step procedure (NRC 1983) consisting of hazard identification, exposure assessment and dose-response assessment as key components. The risk characterisation as the last step culminates in a so-called PEC/PNEC approach for ecosystems: *Predicted Environmental Concentrations (PECs)* and *Predicted No Effect Concentrations (PNECs)* are determined to characterise risk by computing the ratio of both concentrations. For human populations the total daily intake of a chemical is compared to the *No-* or *Lowest-Observed-Adverse-Effect-Level (N(L)OAEL)* to specify a *Margin of Safety (MOS)*.

The PECs and the total daily intake are estimated by a combination of mathematical models, leading to a relatively large and complex system. The models have been developed to estimate emissions, environmental distribution, fate and exposure, and to guide the assessment of potential human and ecological risks in situations where measurements have not been made or would be impossible or impractical to make. In order to establish their effective use, however, there is a need to establish the magnitude and sources of uncertainty associated with model predictions to achieve a better understanding of environmental systems, to increase the reliability of models predictions, and to define realistic values that should be used in subsequent risk assessment. The need for this task was strikingly pointed out by GLAZE (1998):

> *Reliance on models is essential, but the beauty (or deception) of models is that their output can be so impressive even if there is almost no validation beneath. This is particularly troubling when models are used by persons who do not understand their limitations.*

To solve this problem, studies intending to improve the validation status and to elucidate model limitations are needed. GOBAS ET AL. (1998) stated several reasons limiting the applicability of fate and exposure models and emphasised the lack of validation studies and the poorly characterised uncertainty inherent to models.

For the European Union risk assessment approach, there are only a few papers dealing with this problem. Some provide statements on the validity of individual exposure models, while others provide statements on the general approach: JAGER (1995) pointed out where validation efforts are required. DIDERICH (1997) and TRAPP AND SCHWARTZ (2000) discussed future needs and made

proposals for the European Union risk assessment methodology in general. They emphasised the need for a detailed evaluation of the models and parameters. In the scope of the development of the EUSES precursor *USES (Uniform System for the Evaluation of Substances)*, a comparison of measured concentrations with modelling results was undertaken by TOET ET AL. (1991). The regression equations for estimating bioaccumulation in fish were evaluated by JAGER AND HAMERS (1997) and ECETOC (1998). Validation studies of the biotransfer model for cattle are shown in DOUBEN ET AL. (1997). The generic one-compartment model for plants was validated by TRAPP ET AL. (1994), TRAPP AND MATTHIES (1995), JAGER AND HAMERS (1997), POLDER ET AL. (1998). SCHWARTZ (1997) investigated the food chain part and provided a framework for a comprehensive validation study. An inventory of experiences and validation activities of EUSES can be found in JAGER (1998), who subsumed "...the user should be aware of the degree of accuracy and precision to facilitate interpretation of the model results." All in all, the need for more reliability and certainty of the model calculations was emphasised. Despite all the efforts dealing with individual models, a holistic validation study on the overall system, including the role of interaction between the models, is still lacking. Furthermore, in most of the investigations single processes or partition coefficients were considered under laboratory conditions, not real-life situations.

To summarise, the current EU risk assessment scheme tends toward a large complex system that has not been rigorously validated and that lacks comprehensive uncertainty analyses, based principally on the belief that holistic correctness justifies predictive extrapolation. In particular, validation studies that lead to statements on accuracy and applicability of exposure models are scarcely available. But since model predictions are used as a basis for important decisions, it is essential to evaluate their reliability. Expression of the state (or lack) of knowledge about uncertain model predictions is also necessary for both public and scientific credibility and can identify the most important areas for further research. Furthermore, over the last several years it has become obvious that our increased understanding of chemicals' behaviour require the improvement of current methods as well as an implementation of new approaches.

To fill these gaps, the overall objective of this paper is to carry out a comprehensive evaluation of the models for indirect exposure of humans via the environment (i.e. the models to calculate the total daily dose). To accomplish this task, a central question first has to be answered: How can the models of the TGD be evaluated? Often, a procedure termed as validation and realised by a comparison of model outcomes with observed data is applied. But how can one evaluate models that will be applied to new chemicals on the market, i.e. chemicals for which – by definition – no observed data are available? An appropriate methodology has not yet been developed and, thus, is derived in the first chapters. This includes a collation of techniques that are capable of evaluating exposure models. As a preparatory task, the investigated models have to be presented and, with the intention of covering a wide range of chemical properties, various substances have to be selected. Before dealing with model calculations, the general applicability of the models and their agreement to scientific theory has to be revealed. This makes an elaboration of scientific knowledge about relevant physical and chemical processes necessary. The accuracy of the models is addressed by two distinct but complementary approaches: (1) analysis of the uncertainty associated with the predictions and (2) tests of model predictions against measurements. A comparison of the models

laid down in the TGD with alternative models concludes the overall study. Finally, the results are combined to evaluate the applicability of the models and the accuracy of their predictions.
Again, the general goal of this paper is to evaluate the models. Dividing this goals into details reveals several questions that have to be answered. For instance:

- What are the underlying assumptions of the models?
- What are the limitations of the models?
- Are the models formally correct?
- What is the quality of the software?
- Which classes of chemicals will cause problems?
- Do the models correspond to monitoring and experimental data (for chemicals already on the market) and what is the accuracy of the predictions?
- What is the effect of changing an exposure scenario?
- How can the default parameters be evaluated?
- What are the sensitive parameters?
- What are the uncertain parameters?
- What is the impact of a certain parameter or a group of parameters on the result?
- What is the ratio of important to unimportant parameters?
- Do the models offer the best compromise between simplicity and complexity?

Altogether, this paper should provide an overview of the context in which the TGD exposure models may be employed and of what degree of accuracy may be expected. It aims to be a contribution to elaborate the scientific basis and underlying model theory and to provide recommendations for improving the current methods. With the intention of answering questions on general applicability, accuracy of the results, sensitive parameters and all other relevant aspects for exposure assessments, it should also be a contribution for users of the models and software, respectively.

2 Evaluation of models

"We do indeed have a problem with validation", BECK AND CHEN (2000) articulated, pointing out a profound problem which arise when given the task to evaluate a model. In this chapter the background of model evaluation is investigated. The objective is to understand the meaning of validation, to compile a methodology, and finally, to derive a protocol for evaluating the environmental exposure models as laid down in the TGD.

2.1 Assuring the quality of models

2.1.1 The validation problem

The construction and use of mathematical models for exposure assessment are crucial in the context of environmental risk assessment for chemical substances (LEEUWEN AND VAN HERMENS 1995). After the development (or synthesis) of a model, questions concerning its applicability emerge: is my model applicable to the class of chemicals under consideration? Can I justify a carry-over of the model from one chemical to another? How accurate are the predicted results? Does the conceptual structure of the model reflect that of the real phenomena? Given a certain task, is my model better than another one? To recapitulate: *should I use the model?*

In any case, a concept termed as *validation* (from *validus* (lat.)) is used to answer these questions. But in the scientific community the concept of validation is debatable, it is defined inconsistently and has led into an intellectual impasse (BECK AND CHEN 2000). Confusions arise from the philosophical question to what extent, if at all, models or more generally scientific theories can be validated. Not only commonly accepted fundamental works of POPPER (1963, 1959) show that the truth of a scientific theory cannot be proved, at best it can only be invalidated. Despite this, the public has its own understanding of what the word validation implies and is misled by this expression (BREDEHOEFT AND KONIKOW 1993). Even among modellers, who deem validation as a kind of *confirmation*, there is no clear and uniform concept and many expressions circulate. Confusion appears with such concepts as validation, verification, credibility, capability, adequacy, reliability, to name just a few. Despite their plethora and variety, all of these phrases emphasise the applicability of a model to perform a designated task. Against this background, papers have been written to place all encountered terms into an ordered context and to abolish the discords on validation (GAYLER 1999, BECK ET AL. 1997, RYKIEL 1995, ORESKES ET AL. 1994, SARGENT 1993). Nevertheless, the debate continues.

2.1.2 External and internal validation and software evaluation

Predicting the concentration of chemicals in a strict sense poses problems: Since the ideal of achieving, or even approximating truth in predicting novel behaviour of natural systems, is unattainable (BECK ET AL. 1997), a more practicable understanding of the concept of validation is required. Proposals emerged to renounce the word validation and to replace it with *evaluation* (KONIKOW AND

BREDEHOEFT 1992) or to broaden the discussion of validation into one of *quality assurance* (BECK AND CHEN 2000). For this reason, the meaning of validation should be specified precisely.

The historical, but constantly widely accepted understanding of validation is a comparison of model results with numerical data independently derived from experience or observations of the environment, which is indeed insufficient for environmental exposure models. The application of these models in the field of environmental risk assessment for new notified chemical substances exposes this insufficiency. In a pragmatic manner validation of a (mathematical) model can be realised as a rudimental part of the quality assurance of the entire model. The relevance of software quality was stressed by GAYLER (1999), who discussed the evaluation of a computer-based model in terms of adequacy, reliability, accuracy and software quality. Then, the entire model not only includes the mathematical model, but also the software (Fig. 2.1).

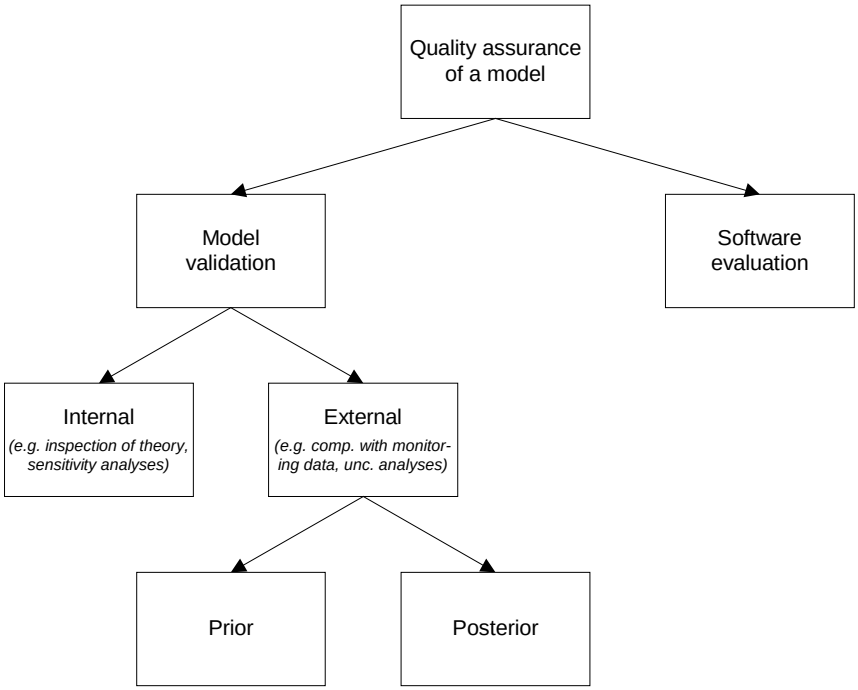


Fig. 2.1 Model validation and software evaluation as parts of the quality assurance.

In the literature validation mainly consists of two aspects: The first is commonly referred to as *conceptual* (SARGENT 1993. ROBINSON 1999), *conceptual & functional* (JAGER 1995), *compositional* or *internal* (BECK ET AL. 1997) validation, and addresses the behaviour, structure and principle application of the model under consideration. Questions of concern are: Do the underlying assumptions allow an application? Are all obviously relevant processes considered? Does the model conform to expert judgement? What are the most critical parameters in the design of the model?

The second aspect is described by terms like *empirical* (SARGENT 1993), *operational & numerical* (JAGER 1995), *experimentation & solution & white/black-box* (ROBINSON 1999), *performance* or *external* (BECK ET AL. 1997) validation and focuses on task-specific properties. This aspect aims to answer questions such as: What are the most critical parameters in the design of the model with respect to successful achievement of the particular task? Are there alternative models providing

more accurate results by comparison with observed data? How strong are the deviations to a given monitoring study?

It is crucial to distinguish between task-specific properties of a model and its task-irrespective or generic properties. Following BECK ET AL. (1997), it is proposed to classify the validation of a mathematical model into an *internal* and an *external* part. The internal part addresses all generic properties of the model, while the external one represents all task-specific properties of a model. An external validation is possible before calibrating the model, i.e. fitting the generic model to a given task, or after its calibration. These possibilities are termed as *prior* and *posterior* external validation.

The external validation also comprises the evaluation of the used data, because statements on external validity are primarily limited by the nature, amount and quality of the available data. These characteristics can vary considerably from the investigated circumstances and define the boundaries of what can be achieved by the validation. It is therefore important that considerable effort is made to ensure that the data are as accurate and representative as possible.

2.1.3 The importance of the model's purpose

It follows from the applied view of validation that a judgement about the validity of a model must be based on the – previously defined – purpose of the model, including statements on undesirable outcomes. Indeed, CASWELL (1976) also argued that a judgement about the validity of a model cannot be made in the absence of its purpose. He identified gaining an insight into the system's structure and the prediction of its future behaviour as the two possible basic purposes of a model. By taking this as a framework, purposes (or design tasks) of exposure models can be itemised. Exposure models to be used in a regulatory context are not so much a tool to gain insight into any system's structure, but rather they are applied in risk assessment for new notified and existing, but, with respect to their exposure, relatively unknown substances and, therefore, have a predictive character (EC 1996A). Examples of design tasks of fate and exposure models are estimations of median partition coefficients (e.g. by using regression equations) or of mean or worse-case exposure concentrations. But also the identification of the need for more detailed information is an imaginable purpose. All these purposes do not imply providing a model result which is as faithful as possible regarding the "true" behaviour of the substance. The goal of validation is rather to understand the realism of the model relative to its intended purpose. Or in the sense of the well-known saying "All models are wrong, but some are useful", the validation of models for exposure assessment means providing a confirmation of the underlying theory and statements on the degree of the accuracy to fulfil a given task.

2.2 Model validation methodology

The question remains as to how way the two aspects of model validation can be dealt with. This section reveals essential methods and derives from these a suitable protocol as a contribution to assuring the quality of environmental exposure models.

2.2.1 Internal validation

Model formalism: To deal with the generic properties of the model, the formal correctness has to be checked. The formalism of the mathematical model must be mechanically and logically correct, i.e. it has to be proven if all equations are adopted correctly from the original literature and if all mechanisms (e.g. the use of techniques to solve an equation) are free of errors. Together with the formal correctness of the computer programme this method is usually constituted as verification (RYKIEL 1995).

Model concept: There are no formal methods for validating the conceptual model (ROBINSON 1999), i.e. the underlying theory. However, the specification of relevant processes and their comparison with the underlying model assumptions is a useful device. A visualisation of the model complexity by depicting the parameters and their interdependence helps us to understand its behaviour, provides transparency and, therefore, greatly facilitates the validation study. It is also necessary to acquire an in-depth understanding of the environmental processes and chemical properties involved. With risk assessment models one often has to extrapolate outside current conditions, rendering a purely data-oriented approach invalid. As a consequence, implicit model assumptions and the relevance of implemented processes must be evaluated to justify the extrapolations.

Additionally, the time and cost of running the model and analysing its results should also be considered. All these methods contribute to the internal validation and may also be termed as an inspection of the underlying theory.

2.2.2 External validation

Parameter behaviour: Exposure modelling needs to make extrapolations from the knowledge gained for some chemicals to those with no or very limited field measurements. The release pattern and the environmental conditions that are appropriate for some substances are often substantially different for other chemicals. In predicting the fate of novel substances released into the environment – by definition – no monitoring data are available to be matched to the model results. In spite of this background a comparison of measured against predicted concentrations using surrogate chemicals may be helpful by analogies. But this inference is only appropriate if, simultaneously, all critical parameters are known, which lead to a completely different model response. In a recent work of BECK AND CHEN (2000) the distinction of key parameters in the model from those that are redundant to the task was introduced as a suitable method for the external model validation. They pointed out that a valid model is maximally relevant to its task. In this context “relevance” is defined as the ratio of key/redundant parameters, a property notably independent of the size of the model. A model is of poor relevance for a given task if it contains many input factors whose value does not drive variation in the output being sought for the task. They introduced these terms for models with a task which is defined by constraints (e.g. a predicted concentration must be below a maximal permissible level). However, if the task is merely to predict “most realistic” concentrations (without having further constraints), the proportion of key and redundant parameters is nevertheless valuable.

Accuracy of the results: When comparing the observed with predicted data the degree of *accuracy* becomes important. Validity and accuracy are related but separate concepts. As illustrated by ROBINSON (1999), a model can be valid but inaccurate. Agreement between the simulated and observed data in accordance with some pre-defined criteria is considered to be the accuracy of the model. It can be dealt with by using statistical measures or visual techniques. A compilation of visual as well as statistical methods can be found in GAYLER (1999). Although the application of statistical methods may often seem obvious, they focus on a purely quantitative comparison of calculated versus observed data and, as demonstrated in GAYLER (1999), different statistical measures may lead to differing results. Using statistics in this case is not, as it may seem, an objective method to determine the accuracy, because restraints arising from (1) the quality of monitoring data, (2) the selection of the statistical measure and (3) the subjectivity of the predefined criteria. In addition, due to the fact that a quantitative agreement of generic exposure models with monitoring data cannot be expected, we avoid using statistics for the evaluation of generic results.

Furthermore, default input values and other data provided together with the model and software have to be investigated. Where possible, actual values should replace default values selected for input.

2.2.3 Both aspects of validation

Uncertainties: The uncertainty inherent to all model calculations should be investigated and serves, depending on their usage, both validation aspects. For example, taking the proportion of key and redundant parameters as a measure for model performance the role of sensitivity analyses becomes a cornerstone in the external model validation. Due to their central role, sensitivity and uncertainty analyses require further elaboration and will be elaborated in a chapter nine.

Alternative models: As an alternative to the comparison of predicted against observed data, the model's results can be compared to both simpler and more complex models. A comparison to simpler models can reveal a too complex model and a comparison with a more complex model can indicate where the investigated model can be improved. One way to obtain an impression of the model's behaviour in a certain situation despite the lack of field data is to apply models with a different structure to identical problems and to compare the results (RAGAS ET AL. 1999). The range of results can be used as a measure for both aspects of validation resulting from different model assumptions and structures.

Expert judgement: This method, which has a qualitative nature, can also be used to extrapolate into an area of uncertainty. An expert's opinion covers knowledge based on both former internal and external validation efforts.

2.3 Software evaluation methodology

The following section describes the aim of the software evaluation and gives a brief overview of general quality requirements for software products. Quality requirements for software products are not a novelty, but they need to be specified in more detail for software dealing with the risk assessment of chemicals. The international quality standard for software products is also taken into consideration.

2.3.1 Quality testing of software

Software testing is a process in which compliance with quality criteria is monitored. These quality criteria are formulated in the software specifications and are realised by a defined development process. Software quality can be achieved (directly) by a systematic development process (KNÖLL ET AL. 1996). The aim of software testing is to discover the errors and weaknesses of the programme under consideration and hence to assist software developers in the improvement of the software. By declaring that software is to be tested immediately after its development, it could be possible to encourage developers to produce faultless software, thus influencing the stipulated development quality (indirect influence on the quality).

Two methods are basically available to test software: firstly, a dynamic test using the programme can be undertaken (*test*). Errors can be recognised by testing and simultaneously recording the results. These errors are limited to certain mistakes in the software's properties (e.g. the acceptance of nonsensical input data).

Secondly, the source code and documentation can be reviewed (*review*). This entails reviewing targets and valid guidelines with the aim of bringing errors and weaknesses to light, but this also serves to acknowledge positive features. Unlike the tests, the reviews represent a static process. Both methods were used to test EUSES. Since the source code was not available, it could not be reviewed.

2.3.2 Quality requirements regarding ISO/IEC 12119

The certification of software products according to international standards is a current issue: in 1994 the international standard ISO/IEC 12119 "Information technology - Software packages - Quality requirements and testing" was published. This standard describes quality requirements and testing conditions for user programmes, in particular in the field of science and technology. With software products, the accompanying documentation and product description are almost as important as the software products themselves. This standard demands the fulfilment of certain quality requirements for the following three components of the software product (KNORR 1997):

According to the standard, products need to be described. The aim of a *product description* is to provide details about the supplier, the task of the product, the hard- and software requirements, and the form and extent of the delivery. Also required is information about whether maintenance is offered, and the scope of such maintenance. Details concerning the specific knowledge required to operate the programme (e.g. specialist knowledge) are also significant. All provided details must be correct and verifiable.

Quality requirements are also given for the *user documentation*, which must contain all necessary details for the use of the programme and must describe all functions that can be called up in a complete and apt manner. Furthermore, general documentation guidelines (layout, construction, etc.) also have to be complied with.

The third component is the *programme itself and the accompanying data*. All functions listed in the documentation must be executable. All other details given in the documentation must also correspond completely to the programme. The functions also have to be operated correctly. The system must not get into an uncontrollable condition and must be prevented from falsifying or eliminating data, even when used incorrectly. No demands are made regarding efficiency, alterability and transferability.

2.3.3 Quality requirements for risk assessment programmes

Good Laboratory Practice (GLP) deals with the organisational development and the conditions under which laboratory checks are planned, carried out, and monitored, as well as the recording and reporting of the tests (KAYSER AND SCHLOTTMANN 1991). A similar approach is desirable for the generation of computer programmes for risk assessment, for which *Good Modelling Practice (GMoP)* should also be developed and established. The basis for this are quality criteria for software for exposure and risk assessment, which as yet can only be found in WAGNER AND MATTHIES (1996), VEERKAMP AND WOLFF (1996) and TRAPP AND MATTHIES (1998). According to these and the general quality requirements for software products, the following ten aspects were found to be essential for the software evaluation:

(1) *Product description:* The product description with the software tested here is not as important as for standard software. However, it should still be available in order to clarify technical queries and areas of application before purchase. Particularly important for software products that deal with chemical risk assessment are an exact indication of the version, changes with regard to previous versions, system requirements for use, scope of built-in evaluation functions, support and possible interfaces with other products.

(2) *Documentation:* The documentation should contain both technical references (installation, operation, etc.) and specialist references (description of the models and theory). It is advisable to provide these details in printed and in online form. The documentation should contain the following features:

- *Correctness:* Are all of the equations in the documentation identical to those in the original literature and to those implemented into the programme? Were the targets complied with (e.g. TGD equations)?
- *Completeness:* All details required for the use of the software product must be included. All functions need to be completely described and all error messages need to be explained. If the software is to be installed by the user, complete (correct) installation instructions need to be

provided. If users are intended to maintain the software, a maintenance manual is required. Tutorials often tend to complement the documentation of many programmes.

- *Consistency* of the various different user documents and the product description must be guaranteed (also with respect to the programme).
- *Comprehensibility:* Comprehensible choice of terms and graphics according to the user group. The use of such terms must be consistent throughout.
- *Clarity:* Logical structure of the user documentation, in which connections can be recognised (including a list of contents and key words).
- *Applicability:* List of the ranges and quality of regressions, the basic substance classes, list of the validation studies undertaken to date, etc.

(3) *Technical requirements:*

- *Installation and system requirements:* The installation of the programme must be possible according to the directions and without previous knowledge. The hard- and software requirements should not be more extensive than necessary for the type of problem. It should be possible to uninstall the programme without difficulty and whenever required.
- *Stability and reliability:* The programme should be stable and controllable at all times. In practice, however, errors can occur, especially when dealing with rather complex programmes, which could possibly cause the programme to ”crash”. It needs to be examined when such errors occur (e.g. input of extreme parameters) and what effects they have. Under no circumstance may data be falsified or eliminated.
- *State-of-the-art:* Current programming standards should be used. Furthermore, the functions provided by the operating system should also be adopted in the software and not newly developed. Examples of requirements for programmes based on Windows 95/NT are (1) a programme to install and uninstall the software, (2) saving of configuration settings in the system’s database (registry), (3) use of the dialog window provided by Windows, (4) input of long file names
- *Network support:* Due to the increased networking of computers it would be appropriate to install the software on a network server. This would save costs and administrative time. The presently examined software should also carry these features. Even with locally installed programmes a minimal amount of network support would be sensible to enable at least the resulting data to be stored in a central database.

(4) *Correctness of calculations:* The programme must compute correctly. All of the functions contained in the product description and user documentation must operate as described.

(5) *User interface and operability:* The most important aspects among the models discussed here are correctness of calculations and applicability. Despite this, software should also be tested to see

if its ”external appearance” lives up to the present standard of technology and to examine what is required of its users. Is the interface ordered ergonomically? Is redundant information given?

- *Programme control:* The control of the programme by the user and the reaction of the programme (messages, masks, lists, etc.) should be uniformly constructed. It must be apparent to users at all times which function is being carried out at that moment in time.
- *Flexibility:* All programme settings and especially the entering of parameters should not be subject to unnecessary limitations. This gives the programme a wide range of applications and makes it operable for different substances and environmental segments. In literature this criteria is also denoted by the term “generic” (Meyer 1988).
- *Output:* Queries, messages, and results of programme calculations should be comprehensible (clear choice of terms, graphic representations, background information, help function). The issuing of information should be easily perceptible and easy to read. When a message appears on the screen, users should be able to recognise immediately if it is an acknowledgement, an inquiry, a warning, or an error message.
- *Error messages* should contain sufficient information about the cause of the error and how to eradicate it (or at least refer users to the manual/documentation).

(6) *Transparency:* It must be clear to users at all times which calculations are being carried out and how individual models can be linked together. This transparency is achieved by free insight into equations and the logical structure of the models. The transparency of the models is a basic requirement for the acceptance of the software.

- *Free insight:* With uncertainties about computational steps taken by the programme, users should be able to comprehend the model calculations ”by hand”. Besides disclosing all model calculations, an exact description is also required. In particular, all variables, including the units used, must be explained and relationships between the individual models must be comprehensible. Complete transparency requires the insight into the source text, which is not the norm with commercial programmes. However, this is the only way to verify the result of a model calculation, since the documentation represents a further potential source of error.
- *Modularity:* A significant concept of software engineering is modularity, which allows for stability and reliability, and also enables programme parts to be reused and freely exchanged (Meyer 1988). For the programmes tested here, this means modularity of the individual models as well as the purely technical functions. This is particularly interesting for users of the software, since they can then recognise the connections between the various models. Data exchange between the individual modules occurs with clearly defined and disclosed interfaces. In REYNOLDS AND ACOCK (1997) modularity is explicitly elucidated and considered as one of the substantial quality criteria.
- *Complexity:* The programme should not be more complex than necessary. If the number of parameters used and their relationships and other conditions are kept low, the whole pro-

gramme is easier to understand, thus contributing considerably to its transparency. Low complexity is not necessarily a contradiction to the demand for flexibility: even a low complex model may offer high flexibility. A comprehensive discussion on complexity can be found in BROOKS AND TOBIAS (1996).

(7) *Features:* Because of its purpose as a DSS for experts, a certain amount of specialist knowledge is required to operate these programmes. But even experts can make typing errors, or may not know all ranges of each parameter. For this reason it is also important with a programme such as EUSES to support users when entering data and applying the models. Important operational requirements within the framework of quality control are:

- *Messages:* If implausible data are entered or if with a regression model the regression range is exited, the programme should deliver the appropriate message. A two-step process is suitable to test plausibility: first of all it is tested whether an entered value is realistic (e.g. molar mass < 1000 g/mol?). The value is accepted, but a warning may appear. In the second step it is tested whether the value is at all physically possible (e.g. concentration > 0 mg/l?). If this second test fails, the value has to be rejected by the programme.
- *Relationships and dependencies between parameters* should be monitored (e.g. is the melting point > boiling point possible?). Dependencies arise from estimated values. If changes are made to the original value, then the estimated value must also be updated automatically.
- *Variable units:* Errors often occur with the conversion of units (e.g. kg/kg to mg/g). The programme should be able to accept different units and convert them internally. If this is not possible, standards should at least be complied with (e.g. SI units).
- *Comments* on input data enable information on data sources and descriptions to be saved. Details on the user and date of input can be automatically recorded. It is important to ensure that comments are updated after input values have been modified. This could occur with the automatic appearance of a comment window after modification of a value.

(8) *Cooperation with other programmes:* Exposure models usually require a multitude of physico-chemical data, emission data, among others, which are often saved in the programme's own database. The results produced (e.g. the development of a concentration of a substance in a river dependent on time and place) may be further processed with visualisation or statistics programmes or, increasingly, with geographic information systems (GIS) (MATTHIES ET AL. 1997). This situation ensures the flexibility of the programme with regard to data input and output.

In order to cooperate with other programmes, appropriate interfaces must be evolved. With large data stocks, an interface to an (external) relational database system (e.g. Access®, Oracle®, etc.) would be suitable.

The problem here is that with the definition of these import and export interfaces, often only "raw data" are transmitted. But the transmission of all information contained in the programme (e.g. de-

pendencies between parameters, estimation functions used, comments on data, etc.) is also important.

(9) *Uncertainty analyses capability:* It is unreasonable to expect that no uncertainty will attach to a model and the predictions it generates. Users may often ask themselves how reliable or uncertain the computed results are. The facilitation of an uncertainty analysis hence represents a possibility to ensure quality. It needs to be tested to what extent the programme is supported by an uncertainty analysis or cooperates with special programmes such as Crystal Ball®, @Risk®, MCSim, etc.

(10) *Support:* The use of software often leads to technical problems (e.g. Why can't I install the software on my computer as described in the documentation?), to questions of a scientific nature (e.g. Is model *X* applicable to chemical *Y*?) or to the stage of the programme development (e.g. Is the programme version at hand the most up-to-date one? Do updates exist?). For this reason, technical and scientific support is interesting for users. Furthermore, there should be an information source which informs users about the present status of the programme development.

In order to realise such support, further information sources are required alongside the documentation. Examples are (a) postal contact, or contact by phone, fax or e-mail with the developers and/or contact persons or (b) information on the programme through Internet services (e.g. World Wide Web).

2.4 Discussion

The objective of this chapter was neither to elucidate all published concepts of validation, nor to develop a new one. The issue was rather to compile some of the major and most accepted concepts to establish a terminology for use in the field of predictive exposure modelling and assessment.

The concept of validation applied here focuses on the quality of the model. Herein, the terms model validation and software evaluation are the basis of the superior quality assurance task. Against the background of many published papers on validation, the concept responses to the "modern" view of validation, which broadens the validation task into a quality assurance procedure and which is closely related to the purpose of the model. Considering validation as a foundation of quality assurance seems to be pertinent, because a validation study assures quality in the sense that the model conforms to the user's requirements and the results are sufficiently accurate. What it does not determine is the extent of accuracy actually required by the user. Indeed, ROBINSON (1999) stressed that the manner in which a validation study is performed is more important in forming a user's quality perception than the quality (or validity) of the model and its results. Subdividing validation into an internal and external aspect is simple, but concise. It is expected that this terminology is pragmatic and able to minimise misunderstandings. Circulating terms of validity can be allocated to one of both aspects.

Additionally, the meaning of validation implies that the validation task is not necessarily cast in terms of predicted concentrations versus monitoring data. If models cannot be validated in a traditional sense, i.e. the comparison of predicted with measured values, which is the fact for novel substances, it becomes a major task to obtain a picture of the behaviour of the parameters involved. Following this approach, validation has an objective and a subjective element. Whenever possible, statements on, for instance, the quantity of uncertainty propagation have to be made in an objective sense. On the contrary, problems that arise from the assessment of new notified substances in complex environmental systems must be handled in a more subjective manner, i.e. evaluation of the model performance on the basis of expert knowledge.

The papers of RYKIEL (1995) and ROBINSON (1999) explicitly stress the validation of data. In this study data validation plays an important role, too, but it is a part of the external validation where provided model parameters and monitoring data flow into the quality assurance task. It is noteworthy that observed data as well as model results should be considered as an approximation towards reality and not as reality in itself, due to the averaging and generic character of exposure models.

The presented methodology should be considered as a selection which can be supplemented if more appropriate methods become apparent. Especially for the validation of the mathematical model methods cannot be instructed, since validation depends on the purpose of the model. More precise instructions can be derived for the evaluation of the software, because here the meaning of high quality is internationally standardised. The compilation of methods is a contribution to establishing a Good Modelling Practice in the field of environmental risk assessment models and is a tutorial for assuring the quality of models.

2.5 Conclusions

After reviewing the literature it became obvious that there is no general validity, i.e. validity is only meaningful with respect to the purpose of a model. Furthermore, the term validation is misleading, because it implies an affirmative result. One should rather speak of quality assurance, which is interpreted in a pragmatic manner. Since there are often no representative observed data available for a comparison with the model results, validation is more than comparing model results with monitoring data. The concept of validation rather underlines that the validity of the (mathematical) model is a necessary but insufficient condition for the acceptability of the whole system, which encompasses the mathematical model and the software. Furthermore, a valid model represents the existing processes and completes other methods of an internal validation successfully.

There is insufficient time to validate and evaluate, respectively, everything and the heaviness of the quality assurance task increases with the model's complexity, but the general rule is: the more the better. To assure essential needs the following protocol is recommended:

(A) Prerequisites, i.e. presentation of the

1. model's structure and its equations,
2. model's purpose,
3. substances and database.

(B) Model validation by

1. inspection of the underlying theory (particularly, model verification and evaluation of implicit assumptions),
2. sensitivity analyses,
3. scenario analyses and comparison with observed data,
4. uncertainty analyses,
5. comparison with alternative models,
6. evaluation of the used data.

(C) Software evaluation with respect to

1. product description,
2. documentation,
3. technical requirements,
4. correctness of calculations,
5. user interface and operability,
6. transparency,
7. features,
8. cooperation with other programmes,
9. uncertainty analyses capability,
10. support.

(D) Concluding statements on model and software and recommendations.

2.6 Summary

The principles of model evaluation in terms of quality assurance, model validation and software evaluation were elaborated and discussed with the intention to develop a suitable evaluation protocol.

Since scientific theories and mathematical models for exposure assessment embedded therein cannot be proved as true, a pragmatic meaning of validation is required, of which the primary purpose is to increase the level of confidence that is placed in the model. The accuracy of the model outcome is a necessary, but insufficient criterion for the quality assurance of models. A wider approach is required which examines the scientific inference that can be made about models relative to their intended purpose. By reviewing the literature on the validation problem, it was found that all the facets of validation can be assigned to generic (internal) and task-specific (external) properties of a model. Appropriate and detailed quality criteria for environmental risk assessment software

were not found in the scientific literature and, thus, they were developed. They are based on common standards, on available publications, and on newly established standards. Hence, a compilation of quality criteria emerged which can serve as a basis for the development and evaluation of programmes in the field of environmental risk assessment software.

Altogether, quality assurance of a model includes internal and external validation and addresses evaluation of the respective software. It should focus not only on the predictive capability of a model, but also on the strength of the theoretical underpinnings, the evidence supporting the model conceptualisation, the database and the software.

3 Handling Uncertainties

Heterogeneity in human behaviour and environmental characteristics as well as an inadequate model structure and measurement errors lead to inevitable uncertainties adherent to the model's outcome. In the preceding chapter the assessment and analysis of these uncertainties were introduced as crucial parts in order to evaluate exposure models. The common approach to handle uncertainties is to investigate diverse exposure scenarios and to represent them in terms of probability distributions (probabilistic exposure assessment). This chapter reviews the underlying theory of uncertainty analyses and develops a methodology as a framework for the TGD evaluation. The database used is presented in a later chapter.

3.1 Types of uncertainty

3.1.1 Uncertainties in exposure assessment

To obtain an impression of the amount of possible contributing sources, the overall uncertainty in exposure or risk can be split up into several parts. As depicted in Fig. 3.1, the US EPA (EPA 1997c) classified the sources of uncertainty in exposure assessment into (1) uncertainty regarding parameters (*parameter uncertainty*), (2) uncertainty regarding missing or incomplete information needed to fully define exposure and dose (*scenario uncertainty*) and (3) uncertainty regarding gaps in scientific theory required to make predictions on the basis of causal inferences (*model uncertainty*).

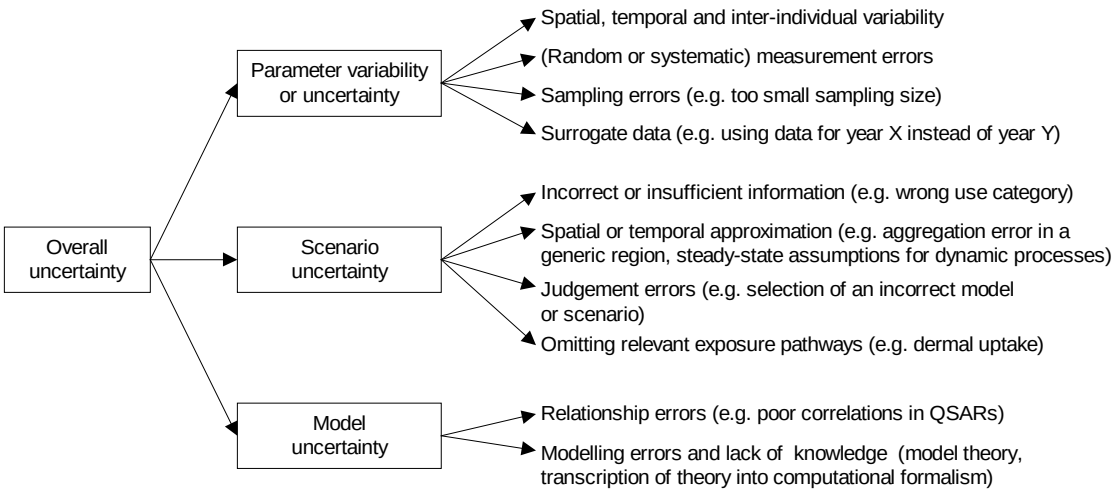


Fig. 3.1 Classification of uncertainty and associated sources.

Input parameters are uncertain for several reasons: variability or errors in measurement, sampling or exertion of data. Scenario uncertainty includes uncertainties resulting from false or incomplete information, such as description, aggregation or judgement errors or an incomplete analysis. Finally, due to lack of knowledge or errors in modelling and integrated relationships the structure of the

model (i.e. the model in respect of the mathematical expressions of its hypothetical relationships) can also be uncertain.

Alternative terms exist, although the classification scheme behind them is the same. Noteworthy are the terms *operational* and *fundamental* uncertainty as used by RAGAS ET AL. (1999), because they correspond to the internal and external aspect of validation: Operational uncertainty results from quantifiable uncertainties in the input propagated through the model equations into the output parameters (parameter plus scenario uncertainty) and can be assessed by quantifying the uncertainties in the input. Fundamental uncertainty stems from the assumptions underlying the model structure and equations (model uncertainty) and can be assessed by expert judgement. By comparing operational and fundamental uncertainty for the TGD regional distribution model, RAGAS ET AL. (1999) stressed that the fundamental uncertainty perceived by experts exceeds the operational uncertainty calculated by means of Monte-Carlo simulations. This finding emphasises the importance of considering fundamental uncertainty within a model validation.

The identification of the sources of uncertainty in an exposure assessment is important, because it represents the first step in determining how to reduce uncertainty (EPA 1997C). Once identified, the uncertainties can be dealt with using appropriate methods.

3.1.2 True parameter uncertainty and parameter variability

In the context of an uncertainty analysis a distinction between *true uncertainty* and *variability* is commonly claimed. True uncertainty (also called *type B* uncertainty) represents a lack of knowledge or partial ignorance about factors affecting exposure or risk, whereas variability (also called *type A* uncertainty) arises from true heterogeneity across people, places or time (EPA 1997C). Both together contribute to the overall parameter uncertainty, whereas the true uncertainty is a gap in one's knowledge that can be discerned from the overall uncertainty. In case of uncertain model parameters, it is useful to distinguish between both types because with respect to the interpretation of the model's result it is valuable to know the contribution of inevitable variance on the one hand and the reducible true uncertainty on the other hand. Secondly, with respect to the consequences of an exposure assessment it is difficult, due to the nature of variability, to constitute acceptable concentrations (e.g. 90%-ile vs. 95%-ile). Reducing uncertainty may help to constitute such values. If the difference between both types of uncertainty is ignored, it becomes difficult to draw useful insights. The fact that a certain parameter is both uncertain and variable aggravates the analysis. However, the overall uncertainty in the parameters can be described using the same formula (e.g. probabilistic distribution functions), although uncertainty and variability are conceptually diverse. If uncertainty dominates an exposure assessment, then one needs to intensify research in order to obtain better parameter values. If variability dominates, one may be able to stratify the variability for sensitive cases. When both true uncertainty and variability are negligible, one truly has a deterministic result. If true uncertainty is negligible relative to variability, then a variability analysis simply represents the expected statistical variation in the outcome. If neither variability nor uncertainty are negligible, for practical reasons the distribution function representing variability cannot be given precisely. Methods for analysing uncertain variability distribution are the subject of current research (PRICE ET AL. 1996).

3.2 Sensitivity analyses

3.2.1 Background and benefit

While analysis of the overall uncertainty involves the determination of variation in an output function based on the collective variability and true uncertainty of model inputs, the sensitivity analysis, in contrast, involves the determination of changes in model response as a result in individual model parameters. An investigation into sensitivity may be carried out beforehand or after an uncertainty analysis. Doing it beforehand helps to identify influential parameters with the intention of reducing costs and effort, since those without impact may be left as deterministic. Applying sensitivity analysis after working with uncertainties may confirm the reliability of the previous work or may reveal further need for research (FINLEY AND PAUSTENBACH 1994, HAIMES ET AL. 1994).

However, the usual approach is to carry out sensitivity studies to assess the effect of varying inputs on the overall output (COX AND BAYBUTT 1981). Also in this work the objective of a sensitivity analysis is to find out those parameters with the strongest impact on the models' results.

3.2.2 Methodology

Different approaches for conducting sensitivity analyses exist, including methods which operate on one variable at a time (e.g. differential sensitivity analysis, HAMBY 1994) or those which handle many variables simultaneously (e.g. Spearman rank order correlation, as implemented in Crystal Ball®, Decisioneering 1999). No consensus exists as to a best approach. However, the differential sensitivity approach (COX AND BAYBUTT 1981, MORGAN AND HENRION 1990, HAMBY 1994) always results in the same sensitivity indices, irrespective of the number of investigated variables, and is easily reproducible without further software. For that reason, this method is applied. It defines a sensitivity function $S(X_i)$ with respect to input parameter X_i by taking the partial derivatives.

$$S(X_i) = \frac{\partial Y}{\partial X_i} \cdot \frac{X_i}{Y}$$

The quotient X_i / Y is introduced to normalise the coefficient by removing the effect of units. The effort in solving this equation can be quite intensive and, instead, the derivatives can be approximated as a finite difference by replacing the denominator of the partial derivative by $X_i \pm$ n %.

3.3 Scenario analyses

3.3.1 Point estimates

It remains to find out by which means uncertainty can be reflected when a developed model is given. One way is to take a deterministic model and to carry out point estimates for various exposure scenarios and assumptions (*scenario analysis*). Each scenario used is a hypothetical construct, based on a set of facts, assumptions and inferences about how exposure takes place, which as-

sists in the estimation of uncertain exposures. For example, the EU risk assessment scheme deliberately creates a *standard scenario* in the TGD which is a conservative point estimate, i.e. it should protect public health. According to FINLEY AND PAUSTENBACH (1994), this approach is most useful as a screening approach which approximates a remote, yet plausible, worst-case situation for some subpopulation of potentially exposed persons. In addition, the calculation of point estimates is a desirable first step which should subsequently be followed by a probabilistic risk assessment (BURMASTER AND ANDERSON 1994).

The deviations between different scenarios can then be characterised by orders of magnitudes. This approach, which is presented in the form of range/confidence estimates and uncertainty indices by RICHARDS AND ROWE (1999), is useful for certain classes of problems: (1) as a mean to provide screening for uncertainties, (2) when data are insufficient for more comprehensive treatment, (3) when the data are from widely different sources with different degrees of precision, and (4) when safety factors are used to provide margins of safety (i.e. the ratio of the effect assessment results to the total daily dose).

3.3.2 Limitations of the approach

The intention of a scenario analysis is to cover a broad range of possible outcomes. However, averaged values are used for each scenario. Such an estimate is then interpreted as a reasonable case. To be on the "safe side" for the protection of human health and environmental damage, worst-case assumptions are commonly applied. But the more parameters are described by worst-case assumptions, the more unrealistic the result is likely to be. For example, applying worst-case assumptions (e.g. the 99%-tile) to both parameters of the simple multiplicative model $f(x,y) = x \cdot y$ leads, according to the laws of probability theory, to a resulting probability $P = (1\text{-}0.99)^2 = 0.0001$. Using 90%-iles as input for those at minimum more than 20 multiplicatively connected parameters of the TGD plant model (EC 1996A) would lead to P = 1E-20. This phenomenon is greater, the more parameters and models are combined and, thus, can be noted as a cumulative worst case. For example, COPELAND ET AL. (1994) showed using the example of a case study, that the Californian point estimate method results in estimates greater than the 99.99^{th} percentile. PRICE ET AL. (1996) calculated lifetime average daily dose rates for individuals exposed to 2,3,7,8-TCDD and found that predicting exposures from indirect exposure pathways may considerably overestimate the intakes for typical and high-end individuals. For those reasons, the point estimate should not be misconstrued as realistic. As stated in the guidelines for exposure assessment (EPA 1992), a point estimate cannot be used to make a determination that a pathway is significant, and it certainly cannot be used to estimate actual exposure. In the case of a scenario analysis, information on uncertainty is restricted to a qualitative statement of confidence in the results. For instance, uncertainty in the point estimate is less than one order of magnitude. Unfortunately, these qualitative statements are difficult to assess, particularly when the assessment involves potential exposure to several contaminants transferred via a number of different pathways (HOFFMAN AND HAMMONDS 1994).

3.4 Probabilistic analyses

A possibility to overcome the limitations of the previous section is to perform a quantitative analysis of uncertainty using probabilistic techniques to propagate uncertainty in models into an assessment of uncertainty in the exposure. The aim of the probabilistic assessment is then to quantify the probability of the model's outcome and to develop a ranking of input parameters concerning their contribution to the overall uncertainty.

3.4.1 Background

To assess uncertainty one can think of a model as producing an output Y, such as a PEC, that is a function of several input variables X_i i.e. $Y = f(X_1,...,X_k)$. Describing uncertainty in a predicted dose or concentration involves the quantification of the range of Y, e.g. by the arithmetic mean and standard deviation of Y, and upper and lower percentile values such as 10% lower bound and 90% upper bound. To characterise the uncertainty of a parameter with a measure independent of the parameter value, the coefficient of variation (standard deviation divided by mean) is stated whenever possible. It ranges typically between 0 and 1 and might exceed unity in cases where the standard deviation is very high. Convenient tools for presenting such information are *the probability density function (PDF)* and the *cumulative distribution function (CDF)* for Y. However, the PDF or CDF of Y can often be obtained only when meaningful estimations of the probability distributions of the input parameters X_i are available. If this information is missing or incomplete, the PDF or CDF for Y can still be constructed, but they should be characterised as screening distributions for parameter uncertainty rather than realistic representations of the uncertainty (McKone and Bogen 1991).

Several papers have identified, compared and evaluated probabilistic approaches for assessing uncertainty in exposure models: The subject of uncertainty analysis as a whole was discussed in a the fundamental work of Morgan and Henrion (1990). It was stressed that the probabilistic approach is a suitable tool for evaluating the uncertainty in the parameters, but not for handling the model or scenario uncertainties. McKone and Ryan (1989) investigated sources and the impact of uncertainty in simple compartment models for human exposure assessment. Case studies for organic chemicals were, for instance, provided by the estimation of the tetrachloroethylene cancer potency from uptake of water to characterise uncertainty in human exposure models (McKone and Bogen 1991), by human exposure assessments to hexachlorbenzene and benzo(a)pyrene through home-grown food to determine the relative contribution of uncertainty and variability (McKone 1994) or by the oral uptake of PAH via drinking water and other sources (Ihme and Wichmann 1996). The majority of publications deal with relatively simple multiplicative models for human health risk assessment. But some papers also exist for regional mass balance models (Mackay and Paterson 1984, Scott et al. 1998). Also Ragas et al. (1999) estimated uncertainties in the multi-media fate model SimpleBox by comparing the model calculations with independently derived environmental quality objectives for air and water. For a set of diverse organic chemicals by using the CalTOX™ (DTSC 1993) system, Hertwich et al. (1999) evaluated the variance in the calculated dose which can be attributed to the uncertainty in chemical-specific parameters as well as the variability in exposure factors and landscape properties for the state of California.

Using the example of two chemicals JAGER ET AL. (2000) have carried out the only probabilistic risk assessment with an EUSES equivalent system so far. Like many other scientists, they emphasised the gain of information.

3.4.2 Methodological survey

Probabilistic exposure assessments can be carried out by means of different methods. In an analytical manner, HELTON (1994) and KLEPPER (1997) dealt with methods for handling uncertainty in complex systems. COX AND BAYBUTT (1981) as well as IMAN AND HELTON (1988) considered analytic and numerical techniques, including Monte-Carlo simulations, response surface approaches, differential sensitivity techniques and evaluation by means of classical statistical confidence bounds. They concluded that some approaches are sufficiently general and flexible for use as overall methods of uncertainty analysis, and others may be very useful for particular problems. Recently, decision trees were used to characterise uncertainty and probability distributions to incorporate variability in a human exposure dose (SIELKEN AND VALDEZ-FLORES 1999). By using the TGD calculation for the local PEC in water as an example, SLOB (1994) has shown that analytical methods may be mathematically an elegant way of identifying uncertainty for multiplicative models with lognormal distributed parameters. However, they limit the assessment by constraints (e.g. requirements regarding the type of distribution functions). Furthermore, the models laid down in the TGD consist of a great amount of parameters, they are not linear and show discontinuities in their behaviour. Numerical methods for uncertainty analyses have proved to be useful for such large and complex models. One of these methods is the well-established Monte-Carlo analysis.

3.4.3 Benefits

Probabilistic approaches, particularly due their versatile applicability, have been identified as a valuable contribution to handling uncertainties in risk assessment. The benefit of the probabilistic approach has been elaborated by several authors: FINKEL (1994) emphasised in his didactical work the gain of information and perspective, which is not available in any less complete descriptions. THOMPSON ET AL. (1992), COPELAND ET AL. (1994) and FINLEY AND PAUSTENBACH (1994) have shown that the outcome of the probabilistic approach is considerably lower than the point estimates of a deterministic worst-case approach. Even for simple exposure scenarios the upper percentiles are overstated by a factor of 3 to 5. Looking at more complex assessments, deviations of up to 2 log units may occur. For example, after comparing case-studies for dioxins and volatile chemicals, FINLEY AND PAUSTENBACH (1994) pointed out that as the number of exposure pathways and variables growths, the difference between the point estimate and the 95th percentile of exposure increases and almost always becomes significant when secondary exposure pathways are considered: the 95th percentile of a probabilistic assessment which requires the consideration of multiple *direct* pathways is usually 3-5-fold less than the point estimate. Considering *indirect* pathways of exposure, the percentile is often as much as an order of magnitude less. Altogether, all these studies reveal that a probabilistic approach to uncertainty basically has the following three advantages:

- *More realism*: The complete distribution is considered instead of some single values. This extends information and perspective concerning the exposure.

- *More scientific* due to the separation of risk assessment and risk management: it becomes obsolete to constitute criteria for the different endpoints (e.g. 99th percentile as worst-case) within the scientific part of the risk analysis.
- *More robust:* It was shown that the probabilistic approach is more robust regarding changes in one single exposure variable.

3.4.4 Monte-Carlo analyses

In a Monte-Carlo analysis, one of two sampling schemes are generally employed (EPA 1997A): S*imple random sampling* or *Latin hypercube sampling*. In the basic form of a Monte-Carlo analysis the model's outcome is calculated directly from empirical probability distributions of the input parameters. Each input parameter is expressed by a probability distribution that defines both the range of values and the likelihood of each value in the range. Simple random sampling is used to select each member of the input parameter set. Arguing with the strong law of large numbers it follows, with high probability, that the outcome provides a good representation of the true output distribution. Latin hypercube sampling may be viewed as a stratified sampling scheme designed to ensure that the upper or lower ends of the distributions used in the analysis are well represented. It is considered to be more efficient than simple random sampling, that is, it requires fewer simulations to produce the same level of precision. Latin hypercube sampling is generally recommended over simple random sampling when the model is complex or when time and resource constraints are an issue. Advantageous is the fact that the inputs do not necessarily have to be stochastically independent (COX AND BAYBUTT 1981). Furthermore, there is no restriction on the form of the joint input distribution or on the nature of the relationship between input and output. A further advantage of this method is that the model can be used in its original form. Any error-prone re-formulations of the model, as needed for analytical methods, are not necessary. In addition, confidence intervals for calculated quantities can easily be developed. Several methods for ranking uncertainty exist, such as correlation coefficients and rank correlations (DECISIONEERING 1999). The disadvantage of the Monte-Carlo method is the huge amount of effort required to carry out calculations. Reliable results require a certain amount of simulations, so-called *shots*. According to MORGAN AND HENRION (1990), the number of shots can be estimated by $p(1-p)\ (2/\Delta p)^2$, where p denotes the percentile value which is achieved with 95% certainty and a deviation of $\pm\Delta p$. All assessments should be carried out following the principles of good practice for Monte-Carlo techniques proposed by BURMASTER AND ANDERSON (1994).

During a Monte-Carlo analysis, it is easy to generate a *rank correlation*: The calculated input and output parameter values of each shot are saved in lists. The lists are sorted and the values are replaced with a numerical ranking starting at 1 for the lowest value in the list and ending with n (the number of shots) for the highest value in the list. A correlation is then computed for each pair of lists and, thus, one obtains the strength of the relationship between each varied parameter and the result. An advantage is the possibility that after a normalisation according to

$$v_j = \frac{r_j^2}{\sum_i r_i^2}$$

a correlation coefficient (r_i) can be expressed as the contribution to the result's variance (v_i) in relation to all other parameters.

3.4.5 Probability distributions

The validity of any analysis is contingent upon the validity of its inputs. Characterising the type of distributions for input parameters is a major task, because Monte-Carlo simulations will transmit the input information directly to the final result, making its distribution appropriately sensitive to the influence of badly chosen distribution functions. But what is decisive for the assignment of distributions? HAIMES ET AL. (1994) pointed out that distributions should represent the state of knowledge. FINLEY ET AL. (1994) stressed the importance of physically meaningful distribution functions in contrast to the relevance of using mathematically elegant models. Additionally, they derived from various case studies that the type of distribution is often less important than the validity and applicability of the database. If the assumption is made that the uncertainty in the model's outcome is the result of many multiplicative factors, it follows from the *Central Limit Theorem* that the result will tend to be lognormally distributed. Since most exposure model parameters are the result of multiplicative factors, also in the literature most parameters are represented by a lognormal distribution. Also for most physico-chemical parameters, there are strong theoretical and empirical arguments to assume lognormal uncertainty distributions (SLOB 1994, SEILER AND ALVAREZ 1996).

Tab. 3.1 Probability distributions used in this study.

Type	**Characteristics**
Lognormal	SLOB (1994) concluded that it is a good strategy to choose the lognormal distribution as a default for data referring to non-negative physical entities, unless data clearly indicate that it fails to give an adequate description.
Triangular	For certain factors a triangular-shaped distribution is proposed. According to FINLEY ET AL (1994) and HAIMES ET AL. (1994), it can be viewed as a conservative estimate of an actual truncated normal or lognormal distribution that takes into account large amounts of uncertainty in the available data. In this case conservatism means that it will result in the more frequent selection of values in the extremes of the normal or lognormal distribution. Using such a distribution does not imply a triangular distributed parameter, it rather expresses that the triangular form is an acceptable way to represent the currently available data.
Uniform	As with triangular distribution, this type of distribution is used as conservative estimate if only an upper and a lower limit are known. This type should be used quite rarely, since physical processes will not show this type of behaviour (JAGER ET AL. 1997). SEILER AND ALVAREZ (1996) stressed that using a uniform distribution is often an indication that the knowledge available has not been used to its fullest extent.

In addition, to contribute to quality assurance the *Kolmogorov-Smirnov goodness-of-fit test* was applied to check if the assumption of lognormally distributed results can be justified. This test represents a measure of the largest vertical distance between two cumulative distributions. Generally, a value less than 0.03 indicates a good fit (DECISIONEERING 1999). The parameters which have a physical limit in value are modelled as truncated lognormals. Such parameters include fractions that cannot exceed unity or partition factors that, by theory and measurement, cannot exceed certain values. However, HAMED AND BEDIENT (1997) showed with restriction on the example of a multiplicative lifetime cancer model that the choice of distribution does not alter the *order of importance* of the basic uncertain variables.

Due to the fact that a distribution is a priori known or unknown, a procedure for selecting appropriate distributions can be derived. The procedure (Fig. 3.2) for a parameter is (1) to prove, based on the sensitivity analysis, if the parameter can be ignored. (2) If not, is there a known distribution or are there theoretical reasons to assign a certain distribution. (3) If this is not the case, are there adequate data to fit a distribution? If none of these three steps can be fulfilled, only surrogate data in combination with expert judgement have to be exploited. In this way the probability distribution is assigned on the basis of available data, combined with the judgement of experts.

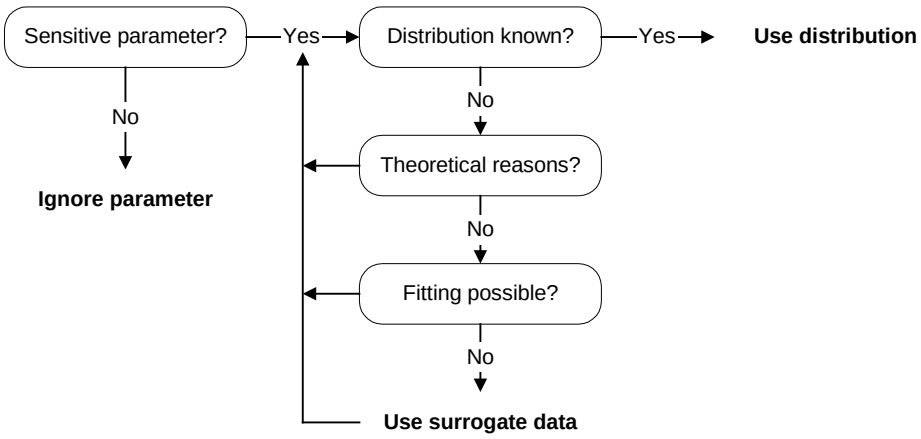


Fig. 3.2 Procedure to select appropriate probability distributions.

In assigning probability distributions the choice of the underlying database, reliability in extreme margins and correlations between parameters may cause problems and necessitate special attention:

Underlying database: Fitting distributions is possible by means of empirical data. But ANDERSON AND HATTIS (1999) stressed that distribution fitting is an overused and often pointless exercise, particularly if only a few data are available. Fitting a distribution to a non-representative dataset is, in fact, non-representative and may therefore be irrelevant to the assessment. In addition, a problem may occur when data originating from different studies are mixed (FINLEY AND PAUSTENBACH 1994) and when study design and study methods are incomparable. However, non-representative data is more the rule than the exception and the common problem of creating parameter distributions is the poor database for nearly all parameters. Thus, in practice the creation of distributions is less a question of statistical methods, it is more a question of expert judgement (ANDERSON AND HATTIS

1999). Lognormal distributions are applied as a default for all parameters, for instance, in CalTOX™ (DTSC 1993); and also in this investigation.

Extreme margins: The goodness of the distribution is often sufficient for the middle segments, but often unacceptable for the tails of the distribution curve (HAIMES ET AL. 1994). Indeed, characterising the extreme values in a distribution is a challenge, because an extreme percentile of a distribution can only be estimated with less certainty than a central percentile. A review of methods of how to handle extreme events and how to obtain appropriate distributions is provided by BIER ET AL. (1999). For a couple of distributions (e.g. lognormal) the precision of their parameters become important for a coefficient of variation of more than unity, while for low standard deviations only little difference in the extreme tails of distributions was shown (HAAS 1997). However, for many distributions there is little disagreement within the scientific community about appropriate values between the 10^{th} and the 90^{th} percentile (FINLEY ET AL. 1994, HAMED AND BEDIENT 1997). Therefore, to reduce uncertainty in the uncertainty analysis itself 10^{th} and 90^{th} percentiles are discussed in this study as most extreme values.

Correlations between input parameters (e.g. the interdependence of food intake rate and body weight) have an impact on the result. The effect of neglecting such correlations was studied in a theoretical manner by SMITH ET AL. (1992). They concluded that dependencies among the input variables must be considered in an uncertainty analysis. On the contrary, there are circumstances in which dependencies can be safely ignored, e.g. weak correlations, correlations (of any degree) among well-known variables and less sensitive parameters. For exposure assessments, two facilities may be used to handle the occurrence of correlations and to minimise errors. These are, firstly, the use of age-specific distribution functions (FINLEY AND PAUSTENBACH 1994) and, secondly, correlation factors (SMITH 1994). Hence, correlations between certain intake rates (e.g. intake of grass and soil for cattle) and between intake rates and body weight (e.g. intake of water and body weight for humans) are considered. Correlations between physico-chemical properties (e.g. between water solubility and K_{OW}) are not considered, because the underlying database allows no quantification of a correlation coefficient. However, possible impacts of such correlations are discussed in the corresponding sections of this study.

3.5 Standards for exposure assessments

A major step in an uncertainty analysis is the selection of appropriate probability distribution functions for the most sensitive and uncertain parameters. This requires a relatively broad database, which is not often available. For that reason this section reviews guidelines and reference values for dealing with uncertainty.

3.5.1 View on the US situation

Although there are no formal regulatory guidelines for conducting a probabilistic exposure assessment it has formed an essential part of the United States risk assessment methodology since the early 1990s. Principles for a practical use were elaborated in the *Guidelines for Exposure Assessment* (EPA 1992) and are also described in BURMASTER AND ANDERSON (1994). The proposed

strategy is to support within a tiered approach deterministic assessments, when needed (EPA 1997B). The definition of *Conditions for acceptance* focuses on quality assurance measures in the sense of a “good scientific practice” (EPA 1997A). An *Exposure Factors Handbook* (EPA 1997C) provides a comprehensive and valuable database. By using unified risk assessment tools that incorporate uncertainty, various states have already reached an advanced status in exposure assessment (e.g. California's CalTOX™, DTSC 1993).

3.5.2 View on the EU and German situation

Standards for exposure assessment are less developed in the European Union. With the *Technical Guidance Document* (EC 1996A), a conceptual framework with detailed formulations of mathematical models was developed. Reference values for an average adult in an EU *standard region* are given, but statements on variations as well as on uncertainty are lacking. Anyhow, if required, most European risk assessors wish to take a probabilistic approach also into account and, particularly, a comparison of the current deterministic TGD approach with a probabilistic measure is desired (JAGER 1998B). In Germany, the *Standards zur Expositionsabschätzung* (BAGS 1995) were initiated which can be used as a starting point to derive reference values for exposure assessments. Especially in the German public health services, it is commonly claimed to enrich current risk assessment by probabilistic methods (BAGS 1995). Recently, in human health risk assessment some effort was undertaken to promote probabilistic approaches (FEHR AND MEKEL 2000).

3.6 Discussion and conclusions

3.6.1 Methodology for handling the different types of uncertainties

Due to the fact that the source of uncertainty which arises from the model structure is essentially unquantifiable, this type of uncertainty is reduced only by stating and evaluating the underlying model assumptions. Even if expert judgement is employed to formalise and quantify model uncertainty, these efforts have a tentative and subjective character (RAGAS ET AL. 1999). New knowledge could be gained by applying a fuzzy approach to exposure assessment, since this methodology has shown to be able to utilise both quantitative and qualitative sources of information and has recently been applied to describe parameter uncertainties in chemical equilibrium calculations (SCHULZ ET AL. 1999). Quantitatively, this study addresses only the parameter and scenario uncertainty: The scenario uncertainty is handled by means of a scenario analysis. Parameter uncertainty is managed by Monte-Carlo simulations. The presented classification of the overall uncertainty is necessary, because the different types of uncertainty should be handled, whenever possible, in different ways. True uncertainty can be reduced by means of more data and investigations, variability can not. However, when variability differs by orders of magnitude, even relatively large datasets may be insufficient to pin down the mean with the desired degree of precision. Unfortunately, due to the poor underlying database, the desired distinction cannot be made in many cases and the probability distributions incorporate variability and true uncertainty.

3.6.2 Sensitivity analysis methodology

Due to the huge amount of parameters contained in complex models, it is advisable to identify those with the strongest impact on the modelling results. Therefore, before model calculations a sensitivity analysis has to be carried out. It is based on the partial derivatives and aims to ascertain how the model depends upon the information fed into it. Thus, the sensitivity is a measure for the dependence of an output on changes in the input, or, vice versa, a measure of the influence of changes in the input to the output. The sensitivity analysis as part of model validation is a necessary (though by no means sufficient) prerequisite for the evaluation of a model. The practical implication of this analysis is the ability to distinguish important versus unimportant parameters in terms of their sensitivity to the selected concentration or dose. It moreover provides a tool to monitor transparency, the relevance and robustness of a given model. Although an analysis of sensitivity reveals nothing directly about the reliability of the predictions, greatly differing predictions as a consequence of minor changes of its parameters points out suspected reliability. Parameter sensitivity may depend strongly on environmental and chemical data and must be determined for the specific problem. This is a drawback of the differential sensitivity analysis method, because the sensitivity measure depends on the chosen scenario. This localised behaviour may not be appropriate for other scenarios. Furthermore, the method is based on a linearised function and is invalid for non-linear models.

3.6.3 Probabilistic analysis methodology

In contrast to the United States where guidelines have been established, a standardised procedure for probabilistic exposure assessments or at least standards for parameter values, are lacking. For that reason, a procedure including the determination of appropriate parameters had to be developed. The crucial argument for the selection of an appropriate method for dealing with uncertainties is not so much the question of finding the “best” method, it is rather the question of the method which integrates all available information. Consequently, a combination of analytical and numerical techniques makes sense. This study favours the lognormal distribution, because it is the most appropriate representation of natural phenomena (e.g. there are no negative values). If there is a of lack of knowledge, triangular or uniform distributions are used in the sense of a conservative approach. Parameter values and their distributions are based on available data combined with the judgement of experts. Because available data are often scare, and expert opinions can vary widely, the assignment of probability distribution functions introduces an element of uncertainty into the uncertainty analysis itself. Such “uncertainties in the uncertainties” can be accounted for by means of sensitivity studies, in which distributional assumptions are varied and the effect on the overall analysis is calculated. Uncertainty in the uncertainty analysis itself is a point of concern that has yet been rarely quantified in the scientific literature.

Although almost complete agreement prevails in the scientific community that probabilistic methods represent a significant improvement in the exposure assessment process, practical risk assessment virtually relies on the traditional point estimate approach. Since the probabilistic approach is chiefly applicable to handle parameter uncertainties, it is as a consequence a reasonable complement to a scenario analysis. The aim of the probabilistic assessment is to quantify and evaluate the

degree of conservatism in the deterministic exposure assessment methodology by comparing the resulting point estimate of exposure with a probability-based exposure distribution generated by a Monte-Carlo analysis. Because of its general applicability and efficiency, the Monte-Carlo method with the Latin hypercube scheme was chosen to determine uncertainty in the models' outcomes. The advantage of the probabilistic approach is its statements on the likelihood of the exposure level (i.e. the broader scientific basis) and the methodological benefits (i.e. avoiding error propagation and increasing robustness). A probabilistic exposure assessment can integrate the available information and reflect the current state of knowledge including expert judgements, true uncertainty and variability. The disadvantage of the chosen procedure is the required database. The quality of a calculation depends on the availability of data. A lack of empirical data may be compensated by means of theoretical considerations.

3.6.4 The methodology in the context of model validation

The issues of model uncertainty relate closely to issues of model validation. Uncertainty analysis aims to quantify the uncertainty in what comes out of a model and it should be rated as a necessary and essential part of validation. It may also be argued that the validation is an option for assessing the extent of uncertainty in model predictions. This reveals that validation and uncertainty belong together. In this context a sensitivity analysis serves to detect the most important parameters and to reduce the effort in the uncertainty analysis. The first major part of the presented uncertainty analysis methodology is the assessment of scenarios for a wide array of situations which could occur, and, subsequently, to figure out the inherent uncertainties for one or more scenarios. The whole procedure corresponds to steps B.4 to B.6 (2.5) in the validation protocol. All in all, it can be concluded that the uncertainty analysis is an essential contribution to model validation, because it provides an important insight into the results, it may detect weaknesses of the models, and improve them. Accordingly, a more informed interpretation of the results is possible. Especially when the models are used with generic data, model sensitivities and uncertainties are very important considerations, because there are -by definition- no observed data to compare with.

3.7 Summary

When predicting environmental concentrations or exposures the question of their inherent uncertainties arises. In this chapter the emerging uncertainties were classified, a theoretical framework was elaborated and a methodology for dealing with the uncertainties was proposed and discussed. The methodology consists of a combination of a scenario analysis (using point estimates) with a probabilistic exposure assessment. Thereby, the possibly over-conservative character of point estimates arising from the transmission of uncertainty through multiplication was revealed. To overcome these limitations the probabilistic approach, which relies upon distributions for all key exposure parameters, was introduced. A prior sensitivity analysis is proposed to quantify the contribution of each uncertain model parameter to the uncertainty in the model results.

4 Exposure models

The food chain is a potential source of human exposure for many environmental contaminants. Particularly for lipophilic compounds, such as dioxins or polychlorinated biphenyls, exposure through food has been demonstrated to be the dominant contributor to the total dose within non-occupationally exposed populations (TRAVIS AND HESTER 1991). An assessment of such an indirect exposure via the environment requires first of all a transformation of emissions into environmental concentrations and subsequently into quantitative estimates of the amount of a chemical that comes into contact with an individual within an exposed population. The potential dose, expressed as the average daily dose, is thereby the amount of a chemical per unit of body weight per day that is ingested by an exposed individual and reaches the gastrointestinal tract. Thus, the indirect exposure models are used to link human intake to chemical concentrations in food and ambient air. The intention of this chapter is to introduce the significant exposure processes and to present the models laid down in the TGD, including a characterisation of their purpose.

4.1 Terminology

Exposure – or more precisely, internal human exposure or dose – is defined as the quantity of a substance reaching a receptor (e.g. the epithelium of the gastrointestinal tract in the case of ingestion). This is equivalent to the definition laid down in the TGD. In this context, *bioavailability* characterises the fraction of the uptake which is actually absorbed and able to interact with the biosystem of an organism. *Indirect exposure* addresses the amount of a substance that is ingested by humans via drinking water, air and food products (according to the TGD). *Environmental exposure* means the determination of emissions, pathways and rates of movement of a substance in the environment, and its transformation and degradation in order to predict environmental concentrations (PECs) to which ecological systems and humans are or may be exposed. This is the meaning of exposure as proposed by the OECD (UBA 1998).

Various expressions have been established to characterise the fate of contaminants in food webs: *Bioconcentration* describes a chemical's potential to accumulate in an organism related to an uptake via the ambient media alone. A *bioconcentration factor* is determined as the concentration of a parent substance in whole fish at steady-state divided by the mean concentration during the exposure period in the water phase; and/or as the ratio between the uptake and clearance rate constant, assuming first-order kinetics (ECETOC 1995A). *Biomagnification* is related to uptake via ingestion, resulting in an increase of the concentration in organisms to the successive trophic levels. Both contribute to *bioaccumulation* which considers all possible exposure pathways. A concrete measure for the bioaccumulation potential was introduced by *biotransfer* factors (TRAVIS AND ARMS 1988). These describe steady-state concentrations in, e.g. meat, divided by the daily intake. Thus, biotransfer factors cover different routes of exposure. In the context of persistent compounds, *carryover* is often spoken about. Carryover subsumes all processes that include the transfer of a substance from A to B. The affiliated rate is defined as the outflow (mass per time) via

B (e.g. milk) divided by the inflow via A (e.g. fodder). Consequently, it represents the fraction of the uptaken chemical that is excreted per time unit.

4.2 Types of models

Regression relationships and differential equations form the two principle types of exposure assessment models used. A regression equation means a statistical relationship between a descriptor and a biological or chemical endpoint. It is often termed as *QSAR (quantitative structure activity relationship)*, even if the relationship is not derived from the molecular structure. Often, the notion *QPPR (quantitative property property relationship)* would be more precise. A popular method is to take the partition coefficient between octanol and water, since this represents the tendency of a chemical to partition between these phases, which has been considered to be a good indicator of the bioaccumulation potential because partitioning into the octanol phase is assumed to mimic partitioning into fat. Interaction between both compartments and dynamic processes are represented by mass balance equations and described by means of analytically solved ordinary differential equations.

4.3 Description of the models' structure and equations

4.3.1 Overall system

The protection of humans against adverse effects is a major goal of risk assessment (EC 1996A). The TGD focuses on three protection goals: workers, consumers and humans exposed indirectly via the environment. Indirect exposure means uptake via air, water and the food chain. The exposure is calculated by a combination of various mathematical models, the linkage of which forms the overall system. Models forming one logical part are discussed in terms of modules. The overall system and its elements are depicted in Fig. 4.1 for four different resolutions. The modules for the PEC assessment describe the path of a chemical from its source to the human dose, comprising its fate in a sewage treatment plant (STP), the environmental distribution and the seven human intake pathways air, fish, meat, milk, plants, roots and drinking water.

I
Modules for
risk assessment

II
Modules for
PEC assessment

III
Distribution models
and exposure
modules

IV
Models for indirect
exposure of humans

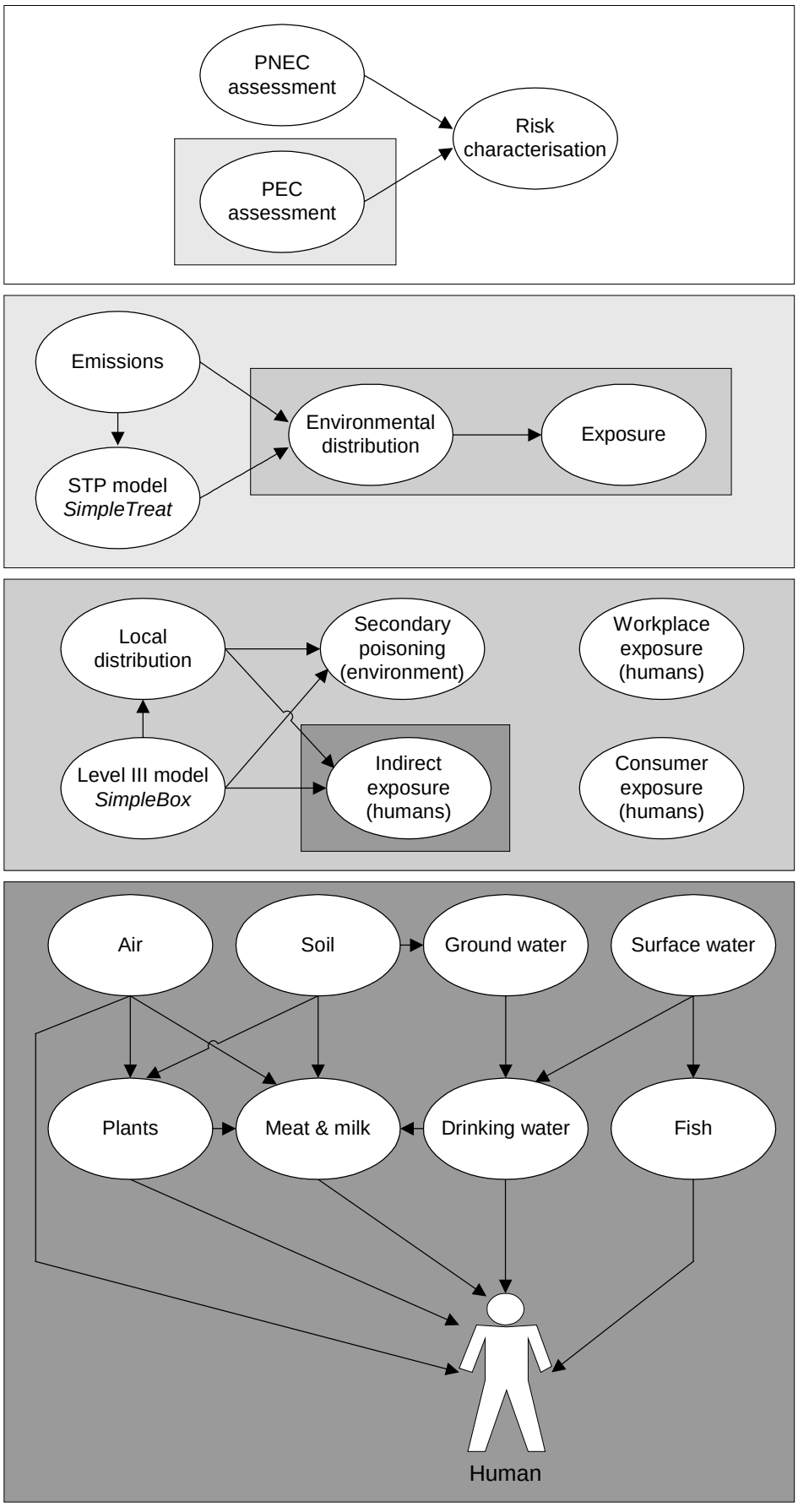


Fig. 4.1 Successive refinement of the overall system for assessing indirect exposure within the EU risk assessment scheme.

4.3.2 Fish

The concentration of chemicals in fish is predicted by means of a bioconcentration factor BCF (related to fresh weight). Based on a two-compartment model (fish and surrounding water), the BCF is determined by the ratio of uptake and elimination rate (in steady state!). If experimentally determined BCF are unavailable, the octanol-water partition coefficient is used as a surrogate. It provides a measure of the extent of a chemical partitioning between octanol and water in equilibrium. The greater the K_{OW}, the more likely a chemical is to partition to octanol rather than to water. The K_{OW} is used to estimate the bioconcentration potential. The underlying regression equation is:

$$\log BCF_{Fish} = \begin{cases} 0.15 & \log K_{OW} \le 1 \\ 0.85 \cdot \log K_{OW} - 0.70 & 1 < \log K_{OW} \le 6 \\ -0.20 \cdot \log^2 K_{OW} + 2.74 \cdot \log K_{OW} - 4.72 & 6 < \log K_{OW} < 10 \\ 2.68 & \log K_{OW} \ge 10 \end{cases} \quad (1)$$

Multiplying the BCF with the (dissolved) concentration in water gives the concentration in fish. The bioconcentration is, therefore, a function of the substance's lipophilicity and the presence of sorption sites (i.e. lipid content).

4.3.3 Meat and milk

Also the concentration in meat and milk rests upon a correlation between lipophilicity and uptake potential of a substance. The biotransfer factors BTF, expressed as unit d/kg, are calculated for fresh meat and milk, respectively. A steady state is assumed (see Chapter 6). Based on biotransfer factors for 28 organic chemicals in milk and 36 chemicals in meat, TRAVIS AND ARMS (1988) developed the following geometric mean regression:

$$\log BTF_{Meat} = -7.6 + \log K_{OW} \quad (2)$$

$$\log BTF_{Milk} = -8.1 + \log K_{OW} \quad (3)$$

To obtain the respective concentrations, the BTF is multiplied by concentrations and uptake rates for grass, soil, air and drinking water.

4.3.4 Plants

Uptake to plants is calculated by means of a one-compartment model developed by TRAPP AND MATTHIES (1995, 1996). Uptake from soil porewater with subsequent translocation into upper plant parts and uptake from air by diffusive gas exchange form the sources of the model. Sinks are metabolism and photodegradation. In addition, growth leads to a dilution effect. The structure of the model, including the incorporated processes, is depicted in Fig. 4.2. The steady state concentration in the leaves (and in all other upper plant parts) is estimated according to equation (4) and comprises the parameters shown in Table 5.1.

$$C_{Leaf} = \frac{\alpha}{\beta} \quad (4)$$

where

$$\alpha = TSCF \cdot \frac{Q}{V_L} \cdot C_{PoreWater} + (1 - f_{Pa}) \cdot \frac{A_L}{V_L} \cdot g_L \cdot C_{Air}$$

$$\beta = \left(\frac{g_L}{K_{LA}} \frac{A_L}{V_L} + k_M + k_{Ph} + k_G \right) \cdot \rho_P$$

Tab. 4.1 Parameters of the plant model.

Parameter	Description	Unit
C_{Leaf}	Steady state concentration in upper plant parts	kg/kg (FW)
C_{Air}; $C_{PoreWater}$	Concentration in air and soil water	kg/m³
K_{PW}; K_{LA}	Partition coefficients plant/water and leaf/air	m³/m³
K_{OW}	Partition coefficient octanol/water	m³/m³
k_M; k_{Ph}; k_G	Rates for metabolism, photodegradation and growth	1/d
TSCF	Transpiration stream concentration factor	-
f_{Pa}	Particulate fraction of substance	-
Q	Transpiration stream	m³/d
A_L / V_L	Ratio area / volume of upper plant parts	m²/m³
f_A; f_W; f_{Li}	Air, water and lipid content of upper plant parts	m³/m³
g_L	Conductance (of diffusive gas exchange)	m/d
ρ_P	Plant density	kg/m³ (FW)
b	Correction for lipid/octanol difference	-

As in the previous models, the lipophilicity of chemicals affects their uptake. It is used to estimate the transpiration stream concentration factor (TSCF) using a relationship introduced by BRIGGS ET AL. (1987).

$$TSCF = \begin{cases} 0.09 & \log K_{OW} < -0.5 \\ 0.784 \cdot \exp\left(\frac{-(\log K_{OW} - 1.78)^2}{2.44}\right) & -0.5 \le \log K_{OW} \le 4.5 \\ 0.04 & \log K_{OW} > 4.5 \end{cases}$$

Plant-specific partition coefficients are calculated as follows:

$$K_{LA} = f_A + \frac{K_{PW}}{K_{AW}}, \quad K_{PW} = f_W + f_{Li} \cdot K_{OW}{}^b$$

The soil-to-root transfer is described by an equilibrium partitioning process. The concentration in roots is calculated by:

$$C_{Root} = \frac{K_{PW} \cdot C_{SW}}{\rho_P}$$

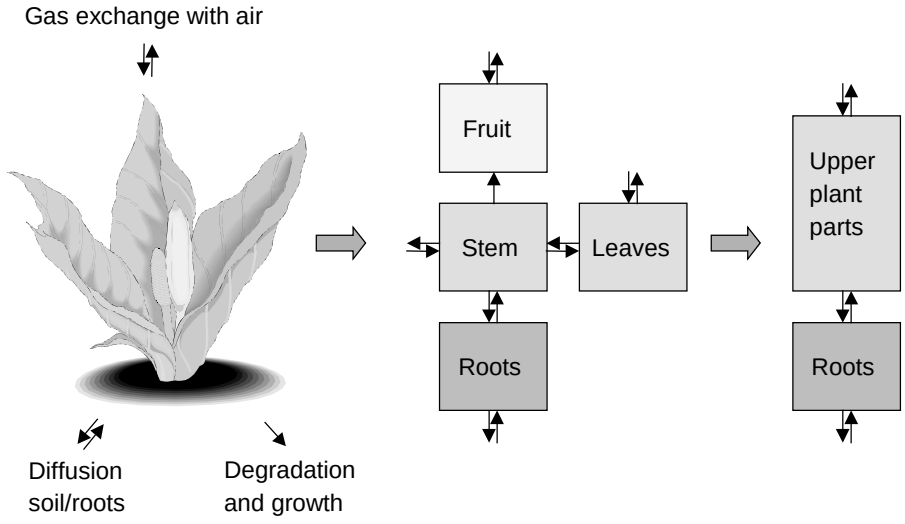


Fig. 4.2 From plant to model: Simplification and incorporated processes.

4.3.5 Drinking water

Drinking water is produced from surface water or ground water. Chemical's concentrations are estimated according to:

$$C_{Drw} = \max (C_{Water} \cdot F_{Pur}, C_{Grw}) \quad (6)$$

I.e., purification takes place only for surface water by using a purification factor F_{Pur}. A study was performed by HRUBEC AND TOET (1992) to investigate the predictability of organic pollutant removal by drinking water treatment on the basis of physico-chemical data. They found that the removal can be roughly predicted by the octanol-water partition coefficient, Henry's law constant and biodegradability. Depending on these data, F_{Pur} is assigned a discrete value ranging from 0.0625 to 1 (TGD, Table III-22).

4.3.6 Human exposure

The final model of the overall system transforms concentrations in the seven exposure media into the human dose under consideration of physiological data and human activity patterns. The time over which the exposure is averaged is one day. The daily human dose D is mathematically expressed by a sum of multiplicative models:

$$D = \frac{1}{BW} \sum_i B_i \cdot C_i \cdot IH_i$$

where i = {Air, Drinking water, Fish, Meat, Milk, Leaf, Roots}, B_i = bioavailability, IH_i = intake rate, C_i = concentration and BW = body weight. This dose represents the final result of the exposure assessment part and is, subsequently, combined with the margin of safety to characterise the risk.

4.4 Purpose of the models and software

The system under investigation can be discussed in terms of mathematical models and the appending software EUSES: As stated in the previous section the purpose of the models is to calculate the total daily intake for a broad range of new and existing chemical substances in accordance with the regulatory framework TGD. The resulting point exposure estimate is intended not to underestimate the true exposure and may significantly overstate it. Simultaneously, it should be as realistic as possible (EC 1996A, PART I: 31-33, EC 1996B, VI-3). These conditions are termed as the *reasonable worst-case*. Although the system is based on a generic environment and is thus not specifically designed for site-specific assessments, it offers the facility to adjust default data to yield an insight into specific local or regional situations and to reach more realistic assessments. In doing so, the system should be a tool for the initial (screening) and intermediate (refined) stage of risk assessment within the tiered approach proposed by OECD (1989).

According to the EC (1996B, VI-3) and JAGER ET AL. (1998), the European Union System for the Evaluation of Substances (EUSES) is a decision support system to facilitate quantitative risk assessment. Target groups are government authorities, research institutes, and chemical companies.

The software claims to be attuned to current chemical management policies and to be in accordance with the principles laid down in the TGD. Furthermore, it should be well documented, user-friendly, transparent and easy to perform. However, it is *not* intended to enfranchise the risk assessor from a comprehensive understanding of the quality of data used, of underlying model assumptions and limitations or of an interpretation of the results.

4.5 Probabilistic extension of the models

The investigation of uncertainties is a commonly claimed issue for conducting exposure assessments (Chapter 3) and forms an integral part of quality assurance (Chapter 2). However, it is not possible to carry out probabilistic assessments with the EUSES system. A theoretical discussion on how to integrate uncertainties into it was already put forward by JAGER AND SLOB (1995) and JAGER ET AL. (1997). ETIENNE ET AL. (1997) elaborated uncertainties of the regional distribution model. These papers provide suggestions on how to incorporate a probabilistic approach into the assessment. Since EUSES itself is not capable of performing probabilistic calculations, an Excel97® spreadsheet version was implemented. Various scenarios for different chemicals were investigated to verify that EUSES and the newly developed spreadsheet lead to the same results. Furthermore, the sensitivity analyses were performed for both implementations. The same results additionally confirm that both implementations are equivalent. Uncertainty analyses were carried out with special attention being paid to the most sensitive parameters using the spreadsheet version and the risk analysis tool Crystal Ball® 2000 (DECISIONEERING 1999). For each assessment, 5000 Monte-Carlo shots (Latin hypercube sampling) were carried out. According to section 3.4.4, the median is then achieved with approximately 95% certainty and an error of ±1.4%.

4.6 Discussion and conclusions

The investigated models offer a differentiating degree of complexity. The model to describe transfer into fish, meat and milk are the most simple, because they merely consist of a regression equation. In contrast, the model for chemical uptake by plants is relatively complex and requires a set of parameters which seems to be virtually unavailable for all edible plants. Obviously, each model depends strongly on the partition coefficient between octanol and water. Consequently, this parameter plays a central role and should be chosen carefully in the PEC assessment. Since “the interaction of biotic and abiotic materials within an ecosystem are so complex that they cannot be predicted (POWER AND MCCARTHY 1997)” it follows that the term PEC (Predicted Environmental Concentration) is somewhat misleading. In fact, in this context *predicted* means the determination of an environmental concentration for a certain substance and justifiable scenario with a certain range of uncertainty. To specify this range in both ways a quantitative and a qualitative is an important task in order to give the models credibility.

Most aspects of the model's purpose are described well in the validation protocol. However, a supplement for the validation task is necessary: Against the purpose of the software it has to be checked if all equations have been adopted correctly from the TGD. This is done within the soft-

ware evaluation. Finally, it should be checked if EUSES is an appropriate tool for the tiered approach of environmental risk assessment. This aspect is firstly investigated by using the standard scenario to obtain reasonable worst-case exposures. Subsequently, refined and more realistic scenarios are used with the intention of obtaining more site-specific exposures, which should be a better match to the observed data from that site.

4.7 Summary

The terminology used, the investigated models and the models' purpose have been briefly presented. It was shown in which way the models of the exposure module have been embedded into the overall system. The exposure module consists of models with differentiating complexity. The model for calculating the total daily intake is a sum of multiplicative models and considers seven exposure pathways. The concentrations in the intake media are calculated by a Level III multimedia model, several regression equations and a generic one-compartment plant model. Each individual model strongly depends on the K_{OW}.

5 Substances and parameters

The quality of the model's outcome depends upon the quality of the data used. Thus, inquiry and evaluation of the applied data plays a central role within the framework of quality assurance. A detailed listing of all parameters used can be found in Appendix A.1 to A.3. The aim of the investigated models is to estimate the exposure of chemical substances, also including chemicals that have not yet been emitted. However, an external validation can only be carried out for chemicals that have already been emitted. For those, the sources and properties have to be specified. The objective of this chapter is to introduce the investigated substances and to present the database used for both the scenario analyses and the probabilistic analyses. The selection should be representative for a broad variety of chemical substances, covering a variety of physico-chemical properties. The input parameters and their sources are dealt with and the strategy for deriving probability distribution functions against the background of the underlying database is presented. Finally, terms for describing the accuracy of the calculated parameters are defined.

5.1 Selected substances

Chemical substances from several classes were used. Among them are primarily air- and waterborne substances, but also ubiquitously occurring chemicals with a potential to accumulate in food webs. All of these chemicals show an acceptable database for at least one part of the food chain.

5.1.1 Polychlorinated dibenzo-p-dioxins (PCDD)

A total of 75 PCDD congeners exist. These are non-polar, rarely water-soluble and lipophilic compounds. Against the background of modelling a food chain, persistent substances are of interest. This applies to those congeners which are substituted in the 2,3,7,8 positions. 2,3,7,8-Tetrachlorodibenzo-*p*-dioxin (TCDD), the most toxic congener of this class, is the reference chemical for the toxicity equivalents (TE). To obtain the total 2,3,7,8-TCDD TE value for a given mixture, the concentration of each potent congener is multiplied by an appropriate factor, and then the TE are added. Such an approach is not feasible for the exposure assessment part, since different congeners behave differently. Consequently, each congener has to be treated separately. In this paper, the six best congeners investigated are therefore selected. Although PCDDs have never been produced intentionally on an industrial scale or for any commercial application, these chemicals are well investigated. They have been measured in practically all media, i.e. air, soil, meat, milk, fish, vegetation and human biological samples. A large amount of data is available on concentrations of PCDD in cows' milk. Likewise, there is relatively good coverage for dairy products, meats and fish. But only a few samples for cereals, fruits and vegetables are available. A further cause of uncertainty is the fact that exposure has declined in recent years. For Germany, the average daily intake was found to have declined by 45% from 127 pg I-TEQ in 1989 to 70 pg I-TEQ in 1995 (KING ET AL. 1999). Estimations by TRAVIS AND BLAYLOCK (1995) indicate that the food chain, especially meat and dairy products, contribute about 99% of the total daily intake of PCDD, while inhalation and ingestion of water, soil, eggs, and produce are not major exposure pathways.

5.1.2 Polychlorinated biphenyls (PCB)

Similar to PCDD, polychlorinated biphenyls (PCB) occur in mixtures. The term chlorinated biphenyl encompasses a chemical class of theoretically 209 isomers. BALLSCHMITER AND ZELL (1980) proposed a numbering system to identify the congeners, which is used in this study. The degree of chlorination determines the physico-chemical properties, as well as distribution and degradation behaviour. The higher the degree of chlorination, the lower the water solubility, volatility and transformation potential. On the contrary, lipophilicity and persistence increase. The relatively high production volume and a variety of application fields in combination with a high persistence lead, in particular for the higher chlorinated congeners, to a ubiquitous occurrence of the these chemicals in the environment (DFG 1988). They are widely versatile, synthetic compounds the manufacture of which was already prohibited in the seventies. Before then they were used as dye solvents, plasticisers, dielectric fluids, and hydraulic fluids. Also for these chemicals, the mixture is well investigated and provides a good database for the ubiquitously occurring congeners 28, 52, 101, 138, 153 and 180. However, congener-specific emission rates are not available. Human exposure to PCB also occurs primarily via food contamination.

5.1.3 Di-(2-ethylhexyhl)phthalate (DEHP)

Phthalatic acid esters (also simply termed phthalates) are ubiquitously occurring chemicals. The reason for this is their high production volume, a variety of use categories, their tendency to accumulate and a non-negligible persistence. Phthalates are predominantly used as plasticisers, but they were also used as solvents, lubricants, for paper production, in cosmetics and for dyes (NRW 1993). Approximately 50% of the production volume of phthalates can be apportioned to di-(2-ethylhexyl)phthalate (DEHP). DEHP was therefore chosen as representative of these chemicals. The published values for physico-chemical properties often cover several orders of magnitude, particularly regarding octanol-water partitioning (MACKAY ET AL. 1991-1997). However, numerous studies using reliable methods indicate that the log K_{OW} of DEHP is consistently in the 7.0-7.8 range (STAPLES ET AL. 1997). Photodegradation via free radical attack is expected to be the dominant atmospheric degradation pathway, and biodegradation is expected to be the dominant loss mechanism in surface waters, soils and sediments. The underlying database is good.

5.1.4 1,3,4,6,7,8-Hexahydro-4,6,6,7,8,8-hexamethyl-cyclopenta-[g]-2-benzopyrane (HHCB)

Polycyclic musk fragrances are an essential ingredient in numerous perfumes, cosmetics and cosmetic care products, soaps, detergents, and other cleaning agents and have been detected in various environmental media and organisms (RIMKUS AND WOLF 1996). Seven single compounds are involved, of which HHCB occurs with the highest concentrations in the environment. The applicability of exposure models was already demonstrated by SCHWARTZ ET AL. (1999, 2000) for describing the environmental fate and distribution of this chemical. With the exception of degradation in air (HHCB is suspected to be significantly degraded by photolysis), the physico-chemical data and concentration levels are comparable to those of PCBs. Samples show air concentrations in the range of pg/m³ for ambient air in urban areas and the level is approximately three log units higher

for contaminated indoor air (KALLENBORN ET AL. 1999). Uptake via skin through the use of consumer products was shown to be the most important exposure pathway for humans (FORD 1988).

5.1.5 Linear alkyl benzene sulfonates (LAS)

Linear alkyl benzene sulfonates are a group of anionic surfactants, characterised by possessing both a lipophilic and a hydrophilic group. LAS is used as a mixture consisting of different homologues and isomers. The fact that they are a major ingredient for synthetic detergents, and that their usage in both domestic and industrial applications, lead to high emission rates into wastewater and, subsequently, into rivers and on soils via sludge application. LAS degrades relatively quickly under aerobic conditions, but only very slightly or not at all under anaerobic conditions. Although LAS shows an increasing bioaccumulation potential with increasing length of the alkyl chain it is not suspected to accumulate in organisms (JENSEN 1999). In this study all substance properties are referred to an "average" homologue with 12 C-atoms. Data were taken from IPCS (1996).

5.1.6 Ethylendiaminetetra acetic acid (EDTA)

Due to its property as a chelating agent, EDTA has a broad field of applications. It is particularly used as a washing and cleaning agent, as a preservative and in the photographic industry. Due to its versatile applicability, high solubility in water, and low degradability, EDTA is one of the compounds with the highest concentrations in the aquatic environment, and has thus raised concern (ROSSKNECHT 1991). The danger of high EDTA concentrations in the environment is their ability to mobilise heavy metals. EDTA does not occur as an acid in the environment, just as a complex or a salt (BUA 1996).

5.1.7 1,2-Dichloroethane (EDC)

1,2-Dichloroethane, which is also known as ethylene dichloride (EDC), belongs to the chemical family of chlorinated alkanes. It is a synthetic, highly volatile solvent which is used predominantly as an intermediate in the synthesis of vinyl chloride. Releases to the environment are principally in emissions to ambient air and industrial effluents. High volatility and emissions lead to a region-wide distribution. The lack of detection of EDC in soil is due to its rapid removal into water and air. In the atmosphere, oxidation by hydroxyl radicals is the dominant loss process (BUA 1995). 1,2-dichloroethane is not expected to bioconcentrate in the food chain due to its low lipophilicity. The air is the major source of exposure with more than 75% (HUGHES ET AL. 1994).

5.1.8 Benzene (BENZ)

Benzene is the simplest aromatic hydrocarbon. It is used commercially as an intermediate in the production of many chemicals and is a by-product of various combustion processes, such as forest fires and the burning of wood, garbage, organic wastes, and cigarettes. Since benzene is not very lipophilic and readily biodegradable, it does not accumulate considerably in the food chain. Due to its volatility, benzene is typically airborne. Inhalation is therefore the primary human exposure pathway. The substance is well investigated (BUA 1988 and 1993). During the last three decades, several studies in Germany, the Netherlands and the US (WALLACE 1996) concerning benzene in

organisms and the environment have expanded knowledge of its occurrence. The average intake of benzene via food is exceeded by the average inhaled intake up to a factor of 500 and, thus, food can be deemed as an unimportant uptake pathway. Indoor concentrations exceed concentrations from outdoors. More than 99% of the total exposure is via air. However, the overwhelming source of benzene exposure in the case of smokers is mainstream cigarette smoke. For non-smokers, most benzene exposure is ultimately derived from traffic exhaust or petrol vapour emissions; only 40% result from outdoor intake (WALLACE 1996).

5.2 Input parameters

Collecting datasets is a critical issue since for any given exposure variable there may be several published estimates which differ widely in quality or credibility. These data sets must be well selected to minimise uncertainty in the results and to make sure, in the case of uncertainty analyses, that the estimates are not greater than values obtained using a point estimate. For various scenarios a database was collated and, according to the methodology presented in Section 3.4, reference values and reference probability distributions were compiled for several parameters (Appendix A.1 to A.3). Probability distributions were derived for all available data and expert judgement whenever needed. The stated coefficients of variation represent conservative values determined on the basis of collated values found in the scientific literature. In general, log-normal distributions were chosen. Unfortunately, insufficient databases restricted this procedure in many cases, and triangular and uniform distributions had to be taken (Tab. 5.1).

Tab. 5.1 Distribution types used.

Distribution	**Parameter (Crystal Ball®-compatible)**	**Abbreviation**
Log-normal	Mean, standard deviation	L (M, SD)
Triangular	Minimum, median, maximum	T (min, med, max)
Uniform	Minimum, maximum	U (min, max)

The motivation for choosing these types has already been explained in Section 3.4.5. In order to create probability distributions, the results of the sensitivity analyses were considered. The distributions were chosen according to the following procedure.

- *Log-normal distribution*: If sufficient data are available ($n \geq 25$), a log-normal distribution is derived from the mean and the standard deviation of the underlying data set.
- *Triangular distribution:* The triangular distribution is chosen if a poor database exists. Parameters of the distribution are the minimum, median and maximum of the available data. However, the distributions for emission and degradation rates require further elaboration: the database for these rates is too small to justify distributions. However, since these rates show a high impact on the results (see sensitivity analyses) and are highly uncertain and variable, the consideration of their inherent uncertainties is important. Thus, on the basis of the point estimate a triangular distribution was chosen with the point estimate as the median. The minimum of the

distribution is half of the median and the maximum is the fivefold of the median. It follows that the point estimate varies within one order of magnitude. The same procedure was applied to the half-lives of the chemicals.

- *Uniform distribution:* A uniform distribution is rarely used. This type was applied in the case of datasets with just two entries.

5.2.1 Parameters for the regional distribution model and its respective scenarios

Two basic scenarios are applied to calculate concentrations and doses, respectively: a *standard scenario* and a *realistic scenario*. The standard scenario uses all estimation functions and default values provided by EUSES. Besides this standard scenario, a realistic scenario is calculated for the German State of North Rhine-Westphalia. In this scenario, the estimated parameters, such as partition coefficients, bioconcentration factors, etc., are replaced by the measured values. EUSES estimates the emissions from a given tonnage using emission tables for a certain use category. In this investigation, the tonnage for a region is estimated in two different ways: For the default scenario it is estimated by EUSES according to the emission tables, whereby 10% of the European consumption rate is taken as a regional tonnage. For PCDD, for which the assignment of a use category is not justifiable, emission data reported in the literature are always used. For the realistic scenario, the tonnage is calculated on the basis of more realistic data reported in the literature. Regional parameters, which differ from the standard scenario, are summarised in Tab. 5.2.

Tab. 5.2 Characteristics of the investigated regions.

Parameter	**Standard** (EC 1996B)	**Realistic** (BERDING ET AL. 2000A)
Name of region	Standard region	North Rhine-Westphalia
Area of region [km²]	40,000	34,400
Average annual precipitation [mm/a]	700	679
Sewage treatment: BOD_{50} [g/d]	54	60
Sewage treatment: Mode of aeration	surface	bubble
Number of inhabitants discharging into one STP	10,000	17,226
Fraction connected to STP [-]	0.70	0.92
Depth of water [m]	3	3
Fraction of water area in region [-]	0.030	0.018
Fraction of water flow from continental scale to the region [-] (= Area of region / Area of EU · Depth of water)	0.034	0.029
Number of inhabitants in region	$2.00 \cdot 10^7$	$1.78 \cdot 10^7$
Estimated regional tonnage [t/a]	based on 10% rule	realistic data (see appendix)

Measured data for the regional distribution model parameters (volumetric, process and other model parameters) are rarely available and, hence, other sources were used: ETIENNE (1997) provided several proposals for distributions. Standard deviations for several parameters were taken from CalTOX (DTSC 1993). Volumetric parameters were chosen according to the procedure described

above. The regional area and the fraction for soil and water are very similar to realistic values for North Rhine-Westphalia. Thus, uncertainties are small. A uniform distribution derived using data from NRW (1998) was chosen for the area. For the parameters that describe fractions, data from the *Regierungsbezirke* of North Rhine-Westphalia were used. Since only a few data were available for the process parameters, mostly distributions proposed by ETIENNE (1997) or DTSC (1993) were used. Realistic data could only be applied for FRunoffSoil, Rainrate and wind speed. Distributions for transfer resistances are very problematic. ETIENNE (1997) used variation coefficients up to 1.8E7 without appropriate justification. However, since these are the only available information in the scientific literature, they were nevertheless used. For the resistances on the water-air interface, a regression based on wind velocity and molecular weight according to BRANDES ET AL. (1996) was used. All remaining parameters were chosen according the procedure mentioned above. It has to be emphasised that North Rhine-Westphalia was chosen as the region – a fact that leads to relatively small uncertainties. This holds in particular for the sensitive parameter Fconnect (the fraction connected to sewer systems). The coefficient of variability amounts to only 0.05 and, thus, causes merely a small impact. A detailed investigation of all of these parameters and examples of applying the regional distribution model to more discriminative regions are provided by BERDING (2000).

5.2.2 Parameters of the exposure module

Distributions are derived from measured data or are taken from the scientific literature. The problem of correlations is dealt with by means of age-specific distributions. Following a proposal by MCKONE (1994), correlation factors of +0.5 were used to express the likely moderate correlations between plant and soil consumption by cattle. Concentrations of many compounds in meat, milk and fish products are found to be usually log-normally distributed (BAFF AND BAM 1999).

Physiological data for plants are collected from DEER-ASCOUGH ET AL. (1993), DTSC (1993), TRAPP ET AL. (1997) and BÖHME ET AL. (1999). The considered values represent a variety of edible plants, such as lettuce, green cabbage, spinach or wheat. The standard plant parameters are conservative, e.g. for the ratio of the plant leaf area and the volume of upper plant parts, a value of 2,500 m^2/m^3 is assumed, which is even appreciably higher than that of lettuce (1,800 m^2/m^3). For the density of plants, five reported values were taken to derive the mean and standard deviation. The derived coefficients of variation are equal to those proposed in DTSC (1993). As shown later in the sensitivity analyses, the correction coefficient b is an important parameter: according to TRAPP AND MATTHIES (1995), exponent b for cut bean roots and stems was found to be 0.75, for barley roots it amounts to 0.77. For barley shoots 0.95 was found, and for isolated cuticles, it amounts to 0.97. The range will not exceed 0.5 to 1.5 (TRAPP 1999). The distribution used is based on the TGD point estimate (for the mean) and the data mentioned above (for the coefficient of variation). The plant's air content is viewed as constant, since this parameter is not sensitive. Water and lipid content were derived from measured data found in the literature. Correlations between these parameters were neglected because the lipid content is three orders of magnitude less than the water content and, therefore, this correlation was not expected to be relevant. Metabolic biotransformation rate constants are required by the plant model. Although biotransformation of most compounds occurs, common modelling practice sets these rate constants to zero because many bioaccumulative che-

micals have rates of metabolism that are so slow that they can be considered as non-metabolisable. Furthermore, measured rates of metabolism for nearly all bioaccumulative chemicals are unavailable. The assumption of no metabolism can be seen as a conservative approach and allows comparison of the results.

Even as early as the 1960s, several studies on modelling transport and accumulation of radionuclides in food chains were undertaken. Based on these efforts, the *International Commission on Radiological Protection* has proposed reference doses for man (ICRP 1975). HEIJNA-MERKUS AND HOF (1993) stressed the need for a harmonisation of parameters for chemical risk assessment and made proposals for their realisation. Default values are proposed for characteristics regarding various media, children and humans. The *Nationale Verzehrstudie* was established to collect information on the nutritional status in Germany from 1985-1988. The nutrient behaviour of all inhabitants was recorded in various regions of Germany. These data were then statistically adjusted to reflect the entire German population based on age, gender and location. Data, which are based on 7-day intake protocols for 23,209 individuals, have been published in ADOLF ET AL. (1995) and BAGS (1995). Recently, in human health risk assessment, proposals were made to establish standards for certain exposure groups (STUBENRAUCH 1999). The *Statistisches Bundesamt* has published a large amount of data concerning body weights of individuals in Germany. These data, taken from BAGS (1995), represent a comprehensive and reliable data set for body weight distributions in Germany. In order to derive parameters for the realistic scenario and, due to the need for harmonising reference parameters and distributions, an exposure factor handbook would be desirable, at least for the parameters of the TGD. A preliminary approach can be found in Appendix A.3

5.2.3 Concentrations

Current data on concentrations are available from various publications. These encompass publications issued by organisations and authorities (BML 1993, NRW 1993, NRW 1995, ECETOC 1992, WEIGERT ET AL. 1991), monitoring programmes (NRW 1991A and B, IKSR 1993, NRW 1994), monographs (BUA 1988, 1993, 1995, 1996, IPCS 1996, BALLSCHMITER 1996) and databases (RIPPEN 1995, KOCH 1995, HOWARD 1990, EC 1996C, EC 1999). In actual fact, compartments are not homogeneous. Air consists of a gaseous phase and a particulate phase. For the model to describe chemical uptake in plants, concentrations for the gaseous phase are required. Conversion between both phases may be attained using the *Junge-Pankow equation* (EC 1996A, PANKOW 1997), which is already implemented in EUSES. In order to calculate the particulate fraction (fPa), the chemical's sub-cooled liquid vapour pressure (VP_L), the melting point (M), the environmental temperature (T), the surface area of particles (SURFaer) and the Junge constant (CONjunge) are required.

$$fPa = \frac{CONjunge \cdot SURFaer}{VP_L + CONjunge \cdot SURFaer}$$

For liquid chemicals (i.e. M is less than T), VP_L is equal to VP, otherwise $VP_L = VP / e^{6.79 (1 - M / T)}$.

If not explicitly mentioned, the presented concentration will refer to the total phase. Water is also not homogeneous. It consists of a water phase and suspended matter. For most chemicals, water

concentrations are only available for infiltrated water, i.e. both the dissolved and the bounded fraction are measured. Since many chemicals are bounded to suspended matter, a conversion to the dissolved fraction is necessary since bioconcentration models are only applicable for the molecular dissolved fraction of a chemical. This fraction may be estimated as shown below.

$$\begin{aligned} C_T &= C_W + C_S \\ &= C_W + K_{OC} \cdot OC \cdot 10^{-6} \cdot X \cdot C_W \\ &= C_W + TOC \cdot K_{OC} \cdot 10^{-6} \cdot C_W \end{aligned}$$

where

C_T : Total concentration of the chemical in water body [mg/l water body]
C_W : Concentration of dissolved chemical [mg/l water body]
C_S : Concentration of chemical sorbed to suspended matter per volume [mg/l water body]
X : Content of suspended matter in water [mg/l]
OC : Organic carbon content (sorbed) [mg/mg]
TOC : Organic carbon content (total) [mg/l]

The partition coefficient between organic carbon and water (K_{OC}) may be estimated according to KARICKHOFF (1981). It follows for the dissolved concentration:

$$C_W = C_T / (1 + 0.411 \cdot K_{OW} \cdot 10^{-6} \cdot TOC)$$

Due to $f_W + f_S = 1$, the dissolved fraction is equal to

$$f_W = (1 + 0.411 \cdot K_{OW} \cdot 10^{-6} \cdot TOC)^{-1}$$

If the concentration sorbed to particles C_S' [mg substance/kg particles] is available, the dissolved concentration is:

$$\begin{aligned} C_W &= (C_W + C_S) / (1 + 0.411 \cdot K_{OW} \cdot 10^{-6} \cdot TOC) \\ &= C_S / (0.411 \cdot K_{OW} \cdot 10^{-6} \cdot TOC) \\ &= (C_S' \cdot X \cdot 10^{6}) / (0.411 \cdot K_{OW} \cdot TOC) \end{aligned}$$

Annotation: The organic carbon is assumed to be completely sorbed in these equations. It would be more accurate to consider that TOC also consists of a sorbed and a dissolved fraction. However, this fact has only a negligible impact on the dissolved fractions estimated in this study.

5.3 Evaluative terms for the external validation

For validating models externally, a comparison with monitoring data is applied and the deviations to monitoring data, i.e. the accuracy of the results, is estimated. The required accuracy is of course task-specific, but against the background of the fundamental task of risk assessment models, namely to assess risk by comparing the result of exposure models with toxicologically derived data, an evaluation of the accuracy in terms of “good” or “poor” results can be undertaken. This leads to

the question of appropriate cut-off criteria for the classification of such evaluative terms. Due to the PEC/PNEC approach, these criteria can be oriented by uncertainty inherent to the effect assessment part. This section presents the method by which uncertainty is dealt with for assessing a PNEC. On this basis, evaluative terms for an external validation are derived.

5.3.1 Accuracy and uncertainty in effect assessment

Uncertainty is also an integral component in the effect assessment part of risk assessment, and will always exist. It is mostly dealt with by use of so-called *safety factors*, if a threshold for toxicity is assumed to exist. For instance, these can involve adjusting a point estimate (e.g. an EC_{50} endpoint, i.e. the effective concentration at which 50% of a particular population is affected in a toxicity test) by a certain factor, typically 10, 100 or 1000) to estimate a safe concentration. According to the review and critical evaluation of this concept by CHAPMAN ET AL. (1998), the term safety factor[1] includes any means by which known data are extrapolated to deal with situations for which there are no data. It can be characterised as a conservative approach for dealing with uncertainty that has no or little relevance to acute uncertainty, but that is able to greatly reduce the probability of underestimating an effect. Safety factors are popular at the interface of science and policy, because they provide clear-cut answers (CHAPMAN ET AL. 1998). Drawbacks of safety factors are obvious: their use also greatly increases the probability of overestimating effects. Moreover, the selection of magnitude is more a policy decision than a scientific result, often caused by an insufficient database. Extrapolations involving safety factors are carried out on an ecosystem level and for human health risk assessment and include, theoretically, four basic areas: (1) inter- and intraspecies, (2) time (acute to chronic, subchronic to chronic), (3) lowest to no-observed effect concentration and (4) laboratory to field extrapolations. For the latter, it is recommended not to use the concept (CHAPMAN ET AL. 1998). For all other fields, a standard safety factor of 10 is commonly applied notwithstanding the fact that differences up to 4 log units in human health risk assessment (UBA 1998) and up to 5 log units on the ecosystem level were observed (CHAPMAN ET AL. 1998). However, safety factors range in general from 0 to 3 log units and the most commonly used factor is 100. An additional factor of 10 is recommended when further sources of uncertainty are taken into account. In the EU risk assessment, safety factors are applied only on the ecosystem level, and not yet for human effect assessment. The standard TGD factors are initially chosen to be conservative, but can be lowered by a certain factor for each extrapolation step if appropriate relevant information is added. If only a minimum of data is available (e.g. 3 LC_{50} values for the aquatic environment on a ecosystem level), then the standard assessment factor is set to 1000.

5.3.2 Definition of evaluative terms

In the context of characterising risk it becomes important to compare the magnitudes with those of the exposure assessment part. This implies the need to discriminate between levels of accuracy. ORESKES ET AL. (1994) stressed that a neutral language is needed for validation. A model can cer-

[1] Safety factors are also known as uncertainty or assessment factors. Though some jurisdictions differentiate between these terms, their underlying idea is identical. In contrast, the expression *extrapolation factor* refers in most studies exclusively to database-derived factors (UBA 1998).

tainly perform well with respect to observational data, in which case one can speak of the precision and accuracy of the fit. Evaluative terms such as good, fair and poor are useful because they invite, rather than discourage, contextual definition. The validity of a model is relative: a model may be considered to be good from one perspective but may not be considered good from another. Thus, criteria for valid models have to be specified. The safety factors described previously serve as an auxiliary herein. The results are termed to be *good* if the deviations are within one order of magnitude since they correspond to the minimum safety factor. The results are denoted as *fair* if the predictions deviate up to three orders of magnitude from the measured data. In the case of deviations that exceed the largest safety factor, the results are *poor*.

5.4 Summary

This chapter revealed the selected chemicals, model parameters and the respective sources. An appropriate terminology was derived for evaluating the results of the external validation. Various chemicals were chosen: Substances of interest are those used in numerous products or chemical processes or which are frequently found in the environment. In detail, congeners of polychlorinated dibenzo-*p*-dioxins (PCDD) and of polychlorinated biphenyls (PCB), DEHP, HHCB, EDTA, LAS benzene and 1,2-dichloroethane were chosen. Due to special monitoring programmes, the databases for PCDD and PCB were judged to be sufficient in nearly all environmental media. However, for other substances numerous measured values exist for certain media. All in all, selection should be deemed as a compromise between an acceptable database and a variety of chemical properties. Furthermore, the selection considers the different environmental compartments. The terminology for evaluating the results of a comparison of monitoring data with predictions is based on the safety factor concept used in the effect assessment part of the TGD. The terms good, fair and poor were derived from this.

6 Inspection of theory

All of the assumptions made during the construction of a model lead to limitations when using it. For instance, an implicit assumption for all submodels is that of a steady state. A steady state describes a time-independency of the model and represents a situation in which the input into a system is equal to the output. Each submodel contains numerous such assumptions. The goal of this chapter is to reveal the underlying theory of the models. The procedure comprises both statements on the verification of the models and a collation of underlying assumptions. As a consequence, we are able to make conclusions regarding the theoretical applicability of the models.

6.1 Verification

Erroneous calculations in a formal sense (e.g. the calculation of negative concentrations) could not be shown for the equations laid down in the TGD. Except in two cases, all of the equations are adopted correctly from the original literature: In the plant model (TGD, Appendix VII, page 208, Formula 3), the correction factor 'a' is missing, which is listed in the original literature (TRAPP AND MATTHIES 1996) as an erratum. In addition, the constants of the sewage treatment plant model (TGD, Appendix VII, page 213, Table 3) do not correspond to the original literature (HRUBEC AND TOET 1992). However, these differences between the original literature and the TGD models do not influence the calculations carried out in this paper.

6.2 Underlying assumptions

6.2.1 Fish

The bioconcentration of organic compounds in fish is a well-investigated process (CONNELL AND HAWKER 1988, NENDZA 1991, GEYER ET AL. 1994, JAGER AND HAMERS 1997, GEYER ET AL. 2000). Comments concerning the applicability of the TGD models can be found in EC (1996A), ECETOC (1998), JAGER AND HAMERS (1997) and JAGER (1997). BEEK ET AL. (2000) provided a comprehensive survey about limitations of applicability of simple bioconcentration models and presented criteria for a refinement of the models within the tiered process of risk assessment. The regression range of the K_{OW}/BCF model ($n = 55$, $r^2 = 0.90$) spans from log K_{OW} 1 to 6 (VEITH ET AL. 1979) and is based on experimental BCF values for 267 chemicals. Based on the values of CONNELL AND HAWKER (1988), a polynomial function ($n = 52$, $r^2 = 0.78$) is used for the range from log K_{OW} 6 to 10. The polynomial relationship is applied to fit the model more accurately to the very hydrophobic substances of the training set (exclusively PCDD). Tab. 6.1 lists the assumptions of this regression equation.

A *realistic* assessment of the bioaccumulation potential should not be founded on a K_{OW}-based approach alone since many chemicals do not obey the underlying correlation and the approach may be misleading. However, if all of the underlying assumptions hold, the model seems to deliver suitable results. If this is not the case, however, the models may still be considered as a *compromi-*

se for the screening phase of risk assessment. The best applicability is achieved for compounds with constant concentrations in surface waters, as long as diffusion into organic material is not hampered (e.g. some PCB and PCDD congeners). The considered organisms should be small and should belong to the beginning of the trophic chain.

Due to the weak database and relatively high experimental uncertainties, the model for fish should be used with caution for high K_{OW}-values. Particularly for chemicals with log K_{OW} > 6, high uncertainties must be expected. The model should not be applied for chemicals with a molecular weight of more than 700 g/mol because the underlying relationship does not consider such heavy molecules. The same holds for dissociating chemicals or chemicals with variable water concentrations. Furthermore, the relationship was elaborated for freshwater fish, forbidding its application to a marine environment.

Tab. 6.1 Assumptions for estimating bioconcentration for fish.

	Assumption	**Evaluation**
1.	The chemical's concentration in the organism is in a steady state.	The application of a bioconcentration factor assumes a constant concentration of the substance in the water. This can only be justified for constant emission rates. To reach a steady state a certain period of time is needed, depending on the substance.
2.	The bioconcentration factor can only be derived from the lipophilicity of the substance.	This represents a very simple approach that does not consider important properties of the chemical. This problem will be further investigated within the context of comparing measured with experimental bioconcentration factors in Section 8.1.
3.	Substances are only enriched in the lipid fraction of the organism.	This assumption is a further limiting simplification because a deviating accumulation in different organs is observed (BEEK ET AL. 2000).
4.	Bioconcentration is the only relevant process.	The correlation between n-octanol and the water partition coefficient, and the bioconcentration factor has been proved to be weak for some types of chemicals. It cannot be expected that the coefficient is generally a sufficient model of the bioaccumulative behaviour of organic chemicals because it ignores several factors influencing bioaccumulation into organisms. Very lipophilic organic chemicals, such as dioxins and others, have the ability to biomagnify in food chains, and result in concentrations in the organisms of higher trophic levels that are much greater than those in the often smaller organisms. Thus, bioconcentration alone should not be considered as the only relevant process.
5.	Growth of the organism is neglected.	This assumption can only be justified if the steady state is attained sufficiently quickly .
6.	Degradation of the chemical in the organism is not considered explicitly.	Due to the fact that the regression equation used is predominantly based on relatively persistent chemicals, this assumption can lead to an overestimation of the bioconcentration

	Assumption	Evaluation
		factors for chemicals with significant metabolism.
7.	Dissociation does not take place.	Ions are principally hydrophilic. However, the bioconcentration factor describes a lipophilic relationship. Thus, for dissociating substances the bioconcentration model may only be applied to the fraction of neutral molecules.
8.	There is a polynomial K_{OW}/BCF relationship for super-lipophilic substances.	The polynomial relationship for super-lipophilic substances is debatable: Conceptual explanations of non-linearity mainly refer either to biotransformation, reduced membrane permeation kinetics or reduced biotic lipid solubility for large molecules. Other arguments consider experimental artefacts, such as a not reached equilibrium or a reduced bioavailability due to sorption to organic matter in the aqueous phase, which lead to an underestimation of the bioconcentration factors. Furthermore, substances with special structural features are known to have an increased potential of bioaccumulation, e.g. molecules containing amines or other nitrogens.

6.2.2 Meat and milk

In comparison to the uptake of chemicals in fish, their transfer into meat and milk has been investigated insufficiently. Although several monitoring data exist, only a few investigations reveal both the theory and a quantification of relevant processes (MCLACHLAN 1992, 1996, DOUBEN ET AL. 1997, THOMAS ET AL 1999A, 1999B).

The TGD applies the regression equations for biotransfer factors[2] by TRAVIS AND ARMS (1988A) for describing biotransfer into meat and milk (Section 4.3.3). The BTF for meat ($r^2 = 0.67$) is based on 36, the BTF for milk ($r^2 = 0.55$) on 28 organic chemicals. The regression is valid for a log K_{OW} ranging from 1.5 to 6.5 and from 3 to 6.5, respectively. According to the TGD, the model has to comprise *all* dairy products and makes the following assumptions.

The mathematical approach of both models (meat and milk) is the same. However, the inherent uncertainties are different: JAGER ET AL. (1997) emphasised that uncertainties in assessing concentrations in meat are greater than those for milk. Despite this, according to MCKONE (1989) the 95% confidence interval for both transfer factors shows a range of approximately two orders of magnitude. Uncertainties caused by unfulfilled assumptions must be taken into account in addition. All in all, against the background of available publications large uncertainties must be expected regarding the application of this model. It represents an extreme simplification of chemical uptake and ignores important toxico-kinetic processes, such as metabolism. Also, for this model the best applicability will be achieved for persistent and ubiquitously occurring compounds.

[2] Sometimes also termed bioconcentration factors, e.g. in the EUSES manual (EC 1996B).

Tab. 6.2 Assumptions for estimating biotransfer into meat and milk.

	Assumption	Evaluation
1.	The uptake time is sufficient for reaching a steady state between daily intake on the one hand and degradation and elimination on the other hand.	A sufficient uptake time can only be expected for constant concentrations in the major uptake media. This will only be the case for chemicals that are emitted diffusively and continuously. But problems may also occur for all other chemicals: e.g. a steady state for OCDD is never reached within the life time of an animal or human and, thus, the concentration is continuously increasing. But for cows the opposite is also possible because of lactating and the concentration is able to decrease.
2.	Grass is the only fodder.	Due to this assumption, the model calculations represents a conservative situation since the concentrations in any other fodder than grass may be significantly lower.
3.	A lipid content of 25% for meat and 3.68% for milk are assumed.	Because of the higher fat content of cheese and butter, this assumption forbids application of the model for these dairy products. This contradicts the model's claim of being applicable for all dairy products.
4.	The biotransfer factor is equal and constant for each uptake pathway.	Depending on the uptake pathway, the resorption of a chemical will be different in reality.
5.	Accumulation takes place only in the organism's lipid fraction.	See assumption 3 for fish (Section 6.2.1).
6.	Metabolism and dissociation are not explicitly considered.	See assumption 7 for fish (Section 6.2.1).

6.2.3 Plants

General investigations regarding the fate of chemicals in plants, including a quantification of concentrations, can be found in BRIGGS ET AL. (1987), HSU ET AL. (1990), TRAPP AND PUSSEMIER (1991), TRAPP ET AL. (1994), KAUPP (1996), TRAPP ET AL. (1997), TRAPP AND MATTHIES (1998), BÖHME ET AL. (1999), MCLACHLAN ET AL. (1999) and several other publications. Nevertheless, the assessment of concentration levels in plants poses several problems: The term 'plant' subsumes a variety of fruits, vegetables and cereals. Furthermore, different parts of a plant (roots, fruits, stem, leaves) are consumed. Chemical uptake into plants occurs differently by uptake from the pore water of the soil and diffusive exchange with the gaseous phase of the air, by deposition of particles on the leaves and subsequent absorption. Plants are often characterised by a high metabolic activity and fast growth (TRAPP AND MATTHIES 1998). All of these problems hamper the construction and application of mathematical plant models. In order to be able to assess chemical concentrations despite the many problems, the TGD make the following assumptions.

Tab. 6.3 Assumptions for estimating concentrations in plants.

	Assumption	Evaluation
1.	The substance is non-dissociating.	Numerous substances are able to dissociate. Acid-base reactions have an impact on the partitioning behaviour and thus affect the result. BRIGGS ET AL. (1987) suggested how to estimate uptake by roots and translocation to shoots for dissociating substances. Their results indicate that the entry of these substances into plant tissue is primarily caused by diffusion of the non-dissociated form. Correction of the respective partition coefficients is possible, but has not been implemented into the TGD. Another effect is also possible: Once inside a cell, dissociation occurs and the ions are unable to diffuse out, leading to the so-called ion-trapping effect and an increased accumulation, which is also not considered by the model.
2.	Exponential growth is assumed.	Exponential growth is only valid for plants that are harvested before maturation, e.g. lettuce.
3.	Plants are in steady state with annual average air concentration	An upper bound for the time after which 95% steady state is reached depends only on the growth rate of the plant. The time is given by $-\ln(0.005)/\alpha$, where α cannot be lower than the growth rate of the plant (TGD). This implies that 95% of steady state in the standard scenario will always be archived within 86 days, and justifies this assumption.
4.	Deposition of particles and resuspension of particles from soil are not considered significant.	Strongly adsorbing compounds such as dioxins are significantly or even exclusively bound? to particles. The transfer pathways air-particle-plant can be expected to be relevant in this case. All substances, including nearly immobile ones, can reach the leaves by resuspension of soil particles. This transfer pathway is of importance for deep-growing parts of plants that show a funnel-formed shape (e.g. lettuce) (TRAPP ET AL. 1997).
5.	A “typical” generic plant is assumed.	Another factor that can be expected to influence the uptake of chemicals into plants are the plant properties. There is only little and contradictory information available on this subject. BÖHME ET AL. (1999) reviewed existing studies and reported a 10-fold range in PCB concentrations on a dry weight basis in foliage which show interspecies variability. On the other hand, a quite small range with variations of a factor of 3-5 for organochlorine pesticides and PCB is reported. In their own study on the accumulation of airborne semi-volatile organic compounds, BÖHME ET AL. (1999) found that interspecies variability in the vegetation/gas-phase partition coefficient is larger than variability in the net gaseous and particle-bound deposition velocities, yielding a greater interspecies variability in plant levels for more volatile organic

	Assumption	Evaluation
		compounds. The variation of substances that are primarily accumulated by gaseous or particle-bound deposition was found to be less than a factor of 4. For more volatile substances, for which the plant levels were determined by equilibrium partitioning, the interspecies variability exceeded a factor of 30.
6.	There is only one soil type, i.e. there is no difference between surface soil and root-zone soil.	Various soil zones show different concentrations. Particularly for highly adsobing and mobile chemicals, concentrations in the upper layers of the soil will be higher in relation to the deeper zones.
7.	All upper plant parts are equal, i.e. they are treated as one compartment. Only a distinction between the root zone and the upper plant parts is made.	The investigated plants should be within an exponential process of growth in which the surface/volume ratio is approximately constant. The model does not have a fruit compartment. It cannot be used for fruits such as cereals.
8.	A constant environmental temperature averaged over the year is assumed (12°C in the standard scenario).	Due to the fact that plants grow mainly in spring and summer, this assumption is a source of additional uncertainty.
9.	The roots are assumed to be in direct equilibrium with the pore water.	Due to the large root surface area of several fine roots, it can be expected that the roots are in equilibrium with the pore water. However, a large root surface area is required, i.e. root crops may behave quite differently. It follows that the model is only applicable for fine roots, not for root vegetables or tubers.

Diffusive gas exchange is an important process for several compounds with a quantifiable vapour pressure. Uptake via pore water and a subsequent translocation may be important for low-chlorinated PCB and all other substances with relatively high water solubility. It is of minor importance for less soluble compounds, e.g. PCDD. The relevancy of certain transfer pathways is additionally determined by the height of the plant and the shape of the leaf (TRAPP ET AL. 1997, see also Section 12.1).

Since substances like PCDD or PCB are airborne substances, they can also reach leaves via air. This applies to important plants, such as grass, lettuce, leafy vegetables and leafy fodder. Other plant products, such as cereal, non-leafy vegetables, fruits, tubers, etc. are not considered. The model covers a critical exposure pathway for leafy vegetables regarding human PCDD intake, particularly due the to relevancy of the air-grass-meat/milk transfer pathway. The estimated concentration tends to be most accurate for plant leaves. Concentrations in fruits or other plant parts may vary significantly. Assumed equilibrium with the pore water requires a large root surface area, i.e. the model is not applicable for roots or carrots. Furthermore, equilibrium will not be reached for readily degradable or highly lipophilic compounds.

6.2.4 Drinking water

The model for estimating concentrations in drinking water is not a model for describing physico-chemical processes, but merely a table. The table determines a purification factor that depends on physico-chemical properties and the biodegradation rate. Complete removal of suspended matter is assumed. The estimated purification factor should represent a worst-case situation. Due to the poor underlying database, JAGER ET AL. (1997) argued that the application of a uniform factor of 0.15 would be more realistic. However, the goal of this paper is to evaluate the mathematical models and, hence, this part of the exposure module is not investigated further.

6.2.5 Human exposure

In contrast to the other models that transform releases or environmental concentrations, the human exposure models merely sum up the products of concentrations in exposure media and intake rates. However, this procedure also contains underlying assumptions:

Tab. 6.4 Assumptions for estimating the total daily intake.

	Assumption	**Evaluation**
1.	Averaged physiological data and consumption rates of an adult are used (“generic person”).	Averaging ignores the significantly different consumption rates in the various European regions and age groups.
2.	Only air, drinking water, upper plant parts, roots, fish, meat and dairy products are relevant for human exposure.	Some exposure pathways that may play a role for certain exposure groups or compounds are ignored. For instance, uptake of pollutants by soil ingestion was found to be significant for small children (FINLEY 1994). Another possibly significant exposure pathway is uptake from air or tap water via the skin (DTSC 1993). Uptake via the skin is in fact considered in the context of the TGD consumer exposure, but it is missing in the estimation of background concentrations.
3.	In the context of the estimated dose, a resorption of 100% is assumed. Only the dose from air assumes a default resorption rate of 75%.	This assumption represents a worst-case situation because complete resorption is seldom observed.
4.	Exposure is constant over the whole day.	Varying concentrations are ignored. In particular, mobility of a person may cause strongly deviating exposures. Furthermore, indoor exposure exceeds outdoor exposure for many substances (WALLACE 1996).
5.	Consumption of freshwater fish is assumed.	This is a quite unrealistic assumption, since seawater fish is more often consumed. Besides this, in general large fish are consumed, while the model is at best applicable for small organisms.
6.	All sorts of consumed meat are treated as beef.	This assumption ignores the different sorts of fodder. For example, fodder for cattle or fowl is different and, thus, the resulting chemical concentration in the meat is expected to be different.

This submodel is independent of the substance's properties. A resorption rate can only be entered for uptake from air. Regarding the substance properties, the model represents a worst-case situation. With respect to intake rates, good results are expected for substances that show constant concentrations in the separate food products (fish, meat, milk, plants). However, this will not hold for a single chemical, because of the heterogeneity of food products. Most realistic results are expected for chemicals taken up directly and exclusively from the air or drinking water.

6.3 Conclusions

Both direct uptake via air and drinking water and indirect uptake via the food chain are the most important exposure pathways for most chemicals. These pathways are considered by the exposure module. Exposure of persistent and ubiquitously occurring compounds is therefore well described. A few non-integrated exposure pathways remain, although they may became important in some situations.

Against the theoretical background, all underlying assumptions together drastically reduce the applicability of the models. Although the regression equations used sometimes show a wide regression range, the joint range (log K_{OW} from 3.0 to 4.6) is quite small. This means: for polar and very lipophilic chemicals, the regression equations result in uncertain and possibly misleading estimations. Fig. 6.1 presents the range of applicability of various regression equations and the resulting joint range of the human exposure model (solid lines). Values of the partition coefficients used are represented by dotted lines. None of the investigated substances fall into the joint range.

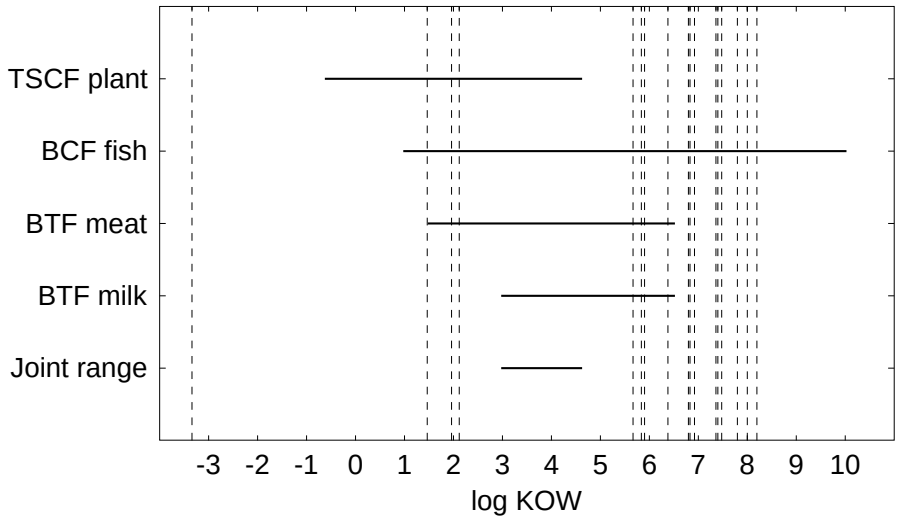


Fig. 6.1 Regression ranges in the exposure module. The dashed lines represent the log K_{OW}-values of the investigated chemicals.

Additional uncertainties occur: Measured partition coefficients have been observed to vary by as much as several orders of magnitude for the same compounds (MACKAY ET AL. 1991-1997). This source of uncertainty is propagated when predicting BCF or BTF. A quantification of this source of uncertainty is given in Section 10.2. K_{OW} represents an equilibrium partitioning between octanol

and water, which cannot be applied for substances that underlie dissociation or surfactants. In particular, the applicability of the system to EDTA (dissociating substance) and LAS (surfactant) is questionable. The partitioning theory is only applicable for the non-ionic fraction. It is equal to $(1+10^{a\cdot(pH-pKa)})^{-1}$ (TRAPP AND MATTHIES 1998), where 'a' is the correction factor in the case of acid-base reactions (a=1 for acids and -1 for bases), pH is the negative decadic logarithm of the H_3O^+-ion concentration in the compartment, and pKa is the negative decadic logarithm of the substance's dissociation constant. Fig. 6.2 depicts the impact of neglecting the effect of dissociation for both acids and bases. As can be seen in the figure, the greater the difference between the values of pH and pKa, the lower the fraction of neutral molecules.

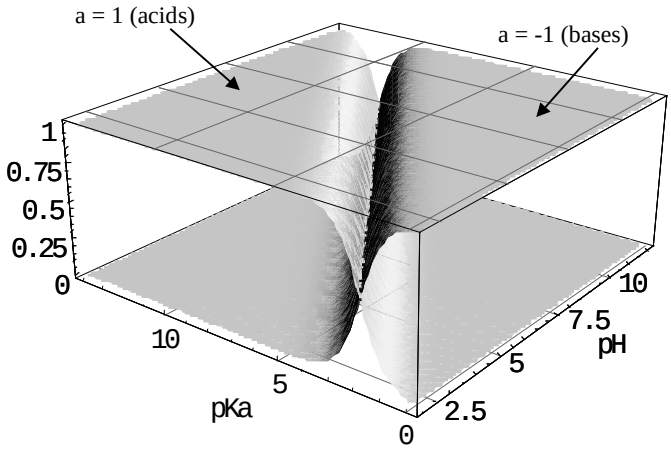


Fig. 6.2 Fraction of the non-dissociated form for both acids and bases.

All in all, it must be emphasised that none of the investigated substances fulfils all underlying assumptions – a fact that causes considerable uncertainties. The exposure part of the TGD is best applicable for chemicals with characteristics by which POPs are defined: Multiphase chemicals that, however, are long-lived with a preference for organic phases. It is disputable for short-lived substances, the fate of which is more controlled by the rates of transformation. Disputable are also the intake rates for plant consumption: It follows from the underlying assumptions that the model must not be applied for fruits, thick roots or tubers. Nevertheless, these assumptions form the basis of the intake rates defined by the TGD (TGD, Appendix VII, Page 216). For example, in a German region more than 80% of the mass of underground-growing parts of a plant are potatoes (BAGS 1995). Thus, potatoes are assigned to the upper plant parts in both realistic scenarios of this study. This is arguable, because potatoes are tubers and an uptake via deposition of particles is not considered in any case. Such a correction of the parameter values results in a leaf/root ratio of approximately 17 for children and 13 for adults, in contrast to 3 for the TGD values. Consequently, upper plant parts are more weighted, and roots are less weighted. With respect to the models investigated here, the protection goal of the European Union exposure assessment methodology is a weakly active and average adult, which is disputable against the background of a highly heterogeneous consumption behaviour.

Although the accuracy of various submodels has already been evaluated, evaluation of the entire system has yet to be carried out. This situation points out the need for a holistic validation study, because even if each submodel were shown to be accurate, this would not imply the entire system is sufficiently accurate. The individual errors for the submodels may accumulate and result in an unacceptable error for the whole model. The entire model must therefore also be quantitatively validated.

6.4 Summary

This chapter revealed the theoretical applicability of the models. Since all models are based on theories and assumptions that may be open to dispute, a compilation and discussion of the assumptions was presented. From a theoretical point of view, it was shown that the assumptions limit applicability drastically and that realistic concentrations may seldom be expected. None of the investigated substances complies with all assumptions. Applicability is therefore not given in a strict sense. If the models are applied without adjustment, high uncertainties must inevitably be expected.

7 Sensitivity analyses

In this chapter, the sensitivities of input parameters regarding the total daily intake are determined. Model parameters (classified into parameters of the sewage treatment plant (STP), the regional distribution model and the exposure module) as well as substance-specific parameters were thereby investigated. Firstly, by means of an analytic approach, statements were made that are valid for all chemicals. Subsequently, by taking the substances into account and using the differential quotient approach, chemical-dependent sensitivities were obtained. This latter substance-based investigation was carried out twice: (1) for the parameters of the overall system and (2) for those of the exposure module.

The objective of this chapter is three-fold: firstly, to develop sensitivity tables to guide the implementation of exposure assessments by other users; secondly, to prepare for an efficient uncertainty analysis; thirdly, to contribute to the evaluation of the overall system.

7.1 Analytic approach

The generalised formalism of the TGD-model for estimating the total daily intake D is (Section 4.3.6):

$$D = \frac{1}{BW} \sum_i B_i \cdot C_i \cdot IH_i$$

where i = {air, drinking water, fish, meat, milk, leaves, roots} and bioavailability $B_i = 0.75$ for i = air, otherwise $B_i = 1$. The body weight BW shows with

$$S(BW) = -\frac{\sum_i B_i \cdot C_i \cdot IH_i}{BW^2} \cdot \frac{BW}{D} = -1$$

a constant sensitivity S and, hence, is independent of the substance. The sensitivity of bioavailability Bj, concentration C_j and intake rate IH_j is equal to the contribution of each intake medium to the total dose:

$$S(B_j) = S(C_j) = S(IH_j) = \frac{B_j \cdot C_j \cdot IH_j}{BW \cdot D} = \frac{B_j \cdot C_j \cdot IH_j}{\sum_i B_i \cdot C_i \cdot IH_i}.$$

It follows that sensitivity is dominated by the major exposure pathway. In this pathway the parameters of the respective submodel become important (Tab. 7.1).

Tab. 7.1 Substance-independent statements on parameter sensitivities of the submodels.

Pathway	Statements on sensitivity
Air (C_{Air})	Air is a compartment of the regional distribution model and, therefore, statements on sensitivity can only be made with respect to a substance.
Drinking Water (C_{Drw})	The concentration in drinking water equals either that of the groundwater or that of the surface water multiplied by a purification factor. This factor is constant for the substances investigated here and, thus, are not sensitive.
Fish (C_{Fish})	The sensitivity of both submodel parameters (C_{Water}, K_{OW}) does not depend on any other parameter. It results in $S(C_{Water}) = 1$ and $$S(K_{OW}) = \begin{cases} 0.85 & \text{for} \quad 1 < \log K_{OW} \leq 6 \\ 2.34 - 2.43 & \text{for} \quad 6 < \log K_{OW} < 10 \\ 0 & \text{otherwise} \end{cases}$$ Consequently, for less lipophilic substances the sensitivity of K_{OW} is lower than that of the water concentration. It becomes larger for highly lipophilic chemicals.
Meat and Milk (C_{Meat}, C_{Milk})	If K_{OW} falls into the regression range, it follows that $S(K_{OW}) = 1$, otherwise $S(K_{OW}) = 0$. Like the estimation of the total intake, the sensitivities of input concentrations C_j and intake rates IC_j can be described by the contribution of the exposure media i = {air, drinking water, leaves, soil}: $$S(C_j) = S(IC_j) = \frac{C_j \cdot IC_j}{\sum_i C_i \cdot IC_i}.$$
Plant (C_{Root}, C_{Leaves})	Sensitivity of the plant density with $S(RHO_{Plant}) = -1$ and the concentration in pore water with $S(C_{Porewater}) = 1$ with regard to C_{Root} do not depend on the substance. Also $S(RHO_{Plant})$ with regard to C_{Leaf} amounts to -1. Due to their multiplicative linkage, the sensitivity of gPlant and AREAplant will be equal. Statements on the impact of all other parameters can only be made by considering a certain substance.

All in all, seven highly sensitive parameters were found to be independent of the substance. The impact of intake rates, bioavailabilities and input concentrations equal the contributions of the exposure media.

7.2 Substance-based approach (overall system)

An analysis of the sensitivities of the overall system was carried out for those chemicals previously used for the investigation of the regional distribution model (i.e. PCBs were omitted due to the absence of emission data). In this section, the impact of the parameters of the overall system on the total daily dose is presented and discussed. The sensitivity values of the parameters for the individual doses (i.e. $DOSE_{air}$, $DOSE_{milk}$, etc.) are helpful to explain the results. These are listed in the appendix.

The sensitivity analysis reveals that for estimating the total daily intake nearly 40% of the overall system's input parameters (50 out of 128) always show a sensitivity of less than 0.05, and can be neglected. Taking all substances together, approximately the same quantity has a high impact (S(x) > 0.5). But one should distinguish between the individual substances: The total daily intake is only sensitive to STP-model parameters for LAS. The question arises as to why these parameters do not show the same impact for EDTA, because neither substances are volatile and both are highly water-soluble. The difference is the absence of biodegradability for EDTA and, thus, STP parameters irrelevant.

Tab. 7.2 Impact of all parameters from the overall system on the daily intake ($DOSE_{total}$).

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
Substance parameters	TempMelt	**1.1**	**2.4**	**0.7**	0.1							
	RRegfWasteWater			0.2	0.2	**0.6**	**1.0**	0.3		0.4	**0.8**	**1.0**
	kDegSoil		-0.3	**-0.8**	**-1.0**	**-1.0**	-0.1					**-0.5**
	BIOinh								**0.9**	**0.7**		
	K_{OW}	0.2	-0.1		0.1	0.1	**0.9**	-0.1		0.1		0.2
	ERegAir	**0.7**	**0.8**	**0.7**	**0.6**	0.3		**0.6**	0.2	0.3		
	EContAir	0.3	0.2	0.2	0.1	0.1		0.1	**0.6**	0.2		
	Vp	-0.1	**0.5**	0.1			**-0.6**	-0.2		-0.1		
	Molw	-0.2					**-0.6**	-0.2		-0.1		
	Sol	0.2					**0.6**	0.2		0.1		
	kDegWater										-0.1	-0.4
	kDegAir	-0.2						-0.3	-0.2			
	RContWasteWater									0.1	0.2	
	kDegSed							-0.1				
	RRegSurfaceWater								0.1			
	Negligible impact: kPlant, RContSurfaceWater, EContInd, ERegInd											
Exposure module parameters	b	**3.4**	**6.3**	**20.8**	**26.1**	**27.8**	**15.3**	**4.3**				
	BW	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**
	RHOplant	**-1.0**	**-0.9**	**-0.9**	**-1.0**	**-1.0**	**-0.9**	**-0.7**				**-0.6**
	FlipidPlant	0.2	0.3	**0.8**	**0.9**	**1.0**	**0.9**	0.3				
	IHroot		0.3	**0.8**	**0.9**	**1.0**	**0.9**					
	IHdrw									0.2	**0.9**	0.3
	IHair								**0.9**	**0.7**		
	kgrowthPlant	**-0.7**	**-0.6**	-0.2				-0.4				**-0.5**
	Vleaf	**-0.7**	**-0.6**	-0.2				-0.4				**-0.5**
	AREAplant	**0.7**	**0.6**	0.2				0.4				
	gPlant	**0.7**	**0.6**	0.2				0.4				
	ICgrass	**0.6**	0.4	0.1				**0.5**				
	Qtransp											**0.5**
	IHleaf	0.3	0.2	0.1				0.2				**0.5**
	IHmeat	0.4	0.3	0.1				0.3				
	IHfish		0.1	0.1			0.1	0.3	0.1	0.1	0.1	0.2
	IHmilk	0.2	0.2					0.2				
	Negligible impact: FairPlant, FwaterPlant, ICair, ICdrw, ICsoil											

(continued)

Tab. 7.2 Impact of all parameters from the overall system on the daily intake ($DOSE_{total}$) (continued)

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
Regional distribution module parameters	FconnectSTP				0.1	0.4	**0.8**	**-1.5**	-0.2	-0.2		**-1.2**
	AREAreg	-0.4	**-0.5**	**-0.6**	**-0.6**	**-0.8**	**-1.0**	**-0.7**	-0.2	**-0.5**	-0.2	**-1.0**
	RhoSolid		-0.3	**-0.8**	**-1.0**	**-1.0**	-0.1	-0.2				**-0.5**
	FsolidSoil		-0.3	**-0.8**	**-1.0**	**-1.0**	-0.1	-0.1				**-0.5**
	DepthAgric		-0.3	**-0.8**	**-0.9**	**-1.0**	-0.1					**-0.5**
	FocSoil		-0.3	**-0.8**	**-0.9**	**-1.0**	-0.1					**-0.5**
	FAgricReg		-0.1	-0.3	-0.3	**-0.6**	**-0.9**	-0.1			-0.1	**-0.6**
	HeightAir	**-0.9**	**-0.7**	**-0.5**	**-0.5**	-0.2		**-0.7**	**-0.9**	**-0.7**		
	RainRate			0.3	0.4	0.2	-0.2				**-0.9**	
	FrunoffSoil						-0.1				**-0.8**	
	Windspeed	**-0.7**	**-0.6**	**-0.5**	**-0.5**	-0.2		-0.3	**-0.7**	**-0.7**		
	AREA EU	-0.3	-0.2	-0.2	-0.1	-0.1		-0.1	-0.4	-0.2	**-0.7**	
	KaslSoilAir						**-0.6**					
	FFlowOutReg										**-0.5**	
	ConJunge	-0.1	**-0.5**	-0.1								
	SurfAer	-0.1	**-0.5**	-0.1								
	FwaterSed	0.1	0.3	0.2	0.1	0.1		**0.5**				
	FWaterReg						-0.1	-0.2	-0.1	-0.3		-0.4
	DepthWaterReg											-0.4
	CollEffAer			0.3	0.4	0.2						
	fNaturalCont		-0.1	-0.1	-0.1						-0.4	
	Qstp						-0.1	0.1		0.1	-0.1	-0.3
	FocSusp		-0.1	-0.1				-0.2				
	kawWater								-0.1	-0.2		
	Temp	-0.1	-0.2	-0.1								
	fAgricCont										-0.2	
	FInfSoil						-0.1					
	SuspEff						-0.1	0.1				
	fWaterCont										-0.1	
	SETTLEvelocity							-0.1				
	DepRateAer			0.1	0.1							
	depthSed							-0.1				
	SuspWater Reg							0.1				
	FWaterSoil											-0.1
	fIndCont										-0.1	
	fNaturalReg		-0.1								-0.1	
	Negligible impact: BIOwater, depthInd, depthNat, depthWaterCont, Erosion, FairSoil, fIndReg, FocSed, FSolidSed, kaslAir, kaslSoilWater, kawAir, kwsSed, kwsWater, NReg, NEU, OHconcAir, RHOair, RhoWater, SuspWaterCont											
STP- model parameters	FractionOCRawS											**0.5**
	InputSolidsIRawS											**0.5**
	SludgeLoadingRate											0.2
	CActivatedSludge											0.2
	BOD						0.1	-0.1				-0.2
	DensitySolidsPS											0.2
	DensitySolidsRaw											-0.2
	KwaterM									-0.1		
	DepthAerator									0.1		
	Negligible impact: AerationRate, DensitySolidsActivatedSludge, DensitySolidsSLS, DepthPS, DepthSLS, FactorBlackburn, FactorHsieh, FractionOCActivatedSludge, FractionOCSolidsPS, FractionOCSolidsSLS, HeightAirColumn, HRTPS, HRTSLS, KairM, KporewaterM, kGa/kLa, OxygenConcentration, Psi, t1/2Aeration, t1/2PS, t1/2SLS, TempSTP											

The group of exposure module parameters shows the highest impact. Particularly striking are the high values for b (correction coefficient between plant lipid and octanol). This phenomenon is explained by the use of b as an exponent in the model equations and is further investigated later in the plant model section. It is obvious that the relevant parameters are similar for the highly lipophilic substances: These are b, RHOPlant FlipidPlant and additionally, for the lower chlorinated dioxins and DEHP, the variables controlling the diffusive exchange with air. This arises from the fact that (according to the model (!)) the main exposure pathway of these chemicals is the plant. From the group of substance parameters, predominantly the emissions and degradation rates are important, but also physico-chemical properties may have an impact (e.g. for HHCB). From the group of regional distribution parameters, those variables controlling concentration in air and soil show a high impact. A detailed investigation of the impact of substance-specific and regional distribution model parameters on concentrations in air and soil can be found in BERDING (2000). For the airborne and waterborne substances, the sensitivity of important parameters is more balanced. The emission rates do make an impact, but the physico-chemical properties make only a negligible impact.

7.3 Substance-based approach (exposure module only)

This section illustrates the results from investigating the exposure module alone. For the input concentrations, values were used that correspond (by orders of magnitude) to measured concentrations (Appendix A.5). As can be seen in Tab. 7.3, seven parameters significantly influence the lipophilic substances. These are the intake rate of roots and various parameters of the plant submodel. For the volatile chemicals BENZ and EDC, concentration in air and the respective intake rate and bioavailability are important. For highly water-soluble chemicals, the total dose is highly sensitive to the concentration in drinking water and its intake rate. As previously discussed, the body weight always shows a constantly high sensitivity. It is remarkable that 18 parameters (i.e. 53%) have a negligible impact.

The high impact of certain parameters can be explained by the main exposure pathways: the more relevant a pathway is, the more relevant are the parameters of the respective submodel. The contributions of the pathways to the total dose (Tab. 7.4) were calculated (1) by entering the concentrations of the seven exposure pathways directly and (2) by estimating them using the indirect exposure module. The calculation shows a varying impact of the exposure pathways on the total dose, depending on the concentration used – whether they are measured or estimated. The deviation arises from the fact that the calculation of the concentrations is more or less accurate and does not always correspond to the measured values.

Tab. 7.3 Impact of all parameters from the exposure module on the daily intake ($DOSE_{total}$).

		TCDD	HxCDD	OCDD	PCB 52	PCB 138	PCB 180	HHCB	DEHP	BENZ	EDC	EDTA	LAS
Substance parameters	Cporewater	**0.9**	**1.0**	**1.0**	**0.9**	**0.9**	**1.0**	**0.9**	**1.0**				
	K_{ow}	**0.9**	**0.9**	**0.9**	**0.9**	**0.9**	**0.9**	**0.9**	**0.9**	0.1			0.3
	CDrw										0.1	**0.9**	**0.6**
	Cair					0.1				**0.8**	**0.8**		
	BIOInhal									**0.8**	**0.8**		
	BIOoral									**-0.8**	**-0.8**		
	CWater	0.1			0.1			0.1		0.2		0.1	0.4
	Negligible impact: Henry, kPlant, CSoil, fPa												
Model parameters	b	**13.8**	**16.9**	**17.8**	**11.0**	**13.7**	**15.9**	**12.2**	**16.1**				
	BW	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**
	FlipidPlant	**0.9**	**1.0**	**1.0**	**0.9**	**0.9**	**1.0**	**0.9**	**1.0**				
	IHroot	**0.9**	**1.0**	**1.0**	**0.8**	**0.9**	**1.0**	**0.9**	**1.0**				
	RHOPlant	**-0.9**	**-1.0**	**-1.0**	**-0.9**	**-1.0**	**-1.0**	**-0.9**	**-1.0**				
	IHdrw										0.1	**0.9**	**0.6**
	IHair									**0.8**	**0.8**		
	IHfish	0.1			0.1			0.1		0.2		0.1	0.4
	IHmilk				0.1								
	Negligible impact: AREAPlant, FairPlant, FwaterPlant, gPlant, ICair, ICdrw, ICgrass, ICsoil, IHleaf, IHmeat, kGrowthPlant, QTransp, Temp, Vleaf												

Tab. 7.4 Impact of the exposure pathways on the daily intake ($DOSE_{total}$) with (1) measured concentrations and (2) concentrations calculated by the indirect exposure module.

		TCDD	HxCDD	OCDD	PCB 52	PCB 138	PCB 180	HHCB	DEHP	BENZ	EDC	EDTA	LAS
(1) Measured concentrations	Plant roots	**0.5**	**1.0**	**1.0**		0.1	0.2	**1.0**	**1.0**			**0.9**	**1.0**
	Fish	0.4			**0.9**	**0.9**	**0.6**						
	Drinking w.										0.2	0.1	
	Air									**0.7**	**0.8**		
	Plant leaves	0.1								0.3			
	Meat				0.1		0.3						
	Milk												

		TCDD	HxCDD	OCDD	PCB 52	PCB 138	PCB 180	HHCB	DEHP	BENZ	EDC	EDTA	LAS
(2) Calculated concentrations	Plant roots	**0.9**	**1.0**	**1.0**	**0.1**	**0.9**	**1.0**	**0.9**	**1.0**				
	Fish	0.1			**0.8**			0.1				0.1	0.3
	Drinking w.										0.2	**0.9**	**0.7**
	Air									**1.0**	**0.8**		
	Plant leaves												
	Meat												
	Milk												

Meat, milk and plant leaves are never significant exposure pathways for the investigated substances. Air is the dominating pathway for the volatile substances. Drinking water and fish are important for hydrophilic chemicals and fish intake also plays an important role for PCBs. Intake via plant roots is (according to the model calculation!) relevant for PCDDs, PCBs, HHCB, DEHP and LAS. This shows that lipophilic substances reach the plant via the soil. Thus, the significant role of the

intake of LAS via roots in relation to EDTA is explained by LAS's relative high lipophilicity, which arises from the surface-active properties. Due to their high K_{OW} and a relatively low vapour pressure and water solubility, most of the substances investigated here show a high accumulation in plant roots. Root crops will result in a high total daily intake when using both calculated and measured concentrations for the intake media if they are multiplied by the daily intake. In any case, against the scientific background the significant contribution of the roots seems to be suspect and is later investigated in more detail (Section 8.4.2).

If a certain pathway becomes dominant it follows that the parameters of the respective submodel become relevant. A sensitivity analysis of the plant model yields the results presented in Tab. 7.5 and 1.6. All other submodels of the exposure module are less complex and the sensitivity of their parameters can be dealt with analytically (Section 7.1). The transfer of chemicals to plants depends on numerous parameters (Fig. 7.1). With the exception of the fraction of air in the plant (FairPlant), no parameter is always negligible or, vice versa, relevant in a certain situation. For the lipophilic substances all parameters, except FWaterPlant, of the root model have a high impact on the concentration in the roots. Correction exponent b is the most sensitive parameter for these chemicals. FWaterPlant plays a role for highly soluble substances. The impact of K_{OW} and the fraction of lipids in root tissue for LAS is understandable due to its amount, which is high in relation to EDTA.

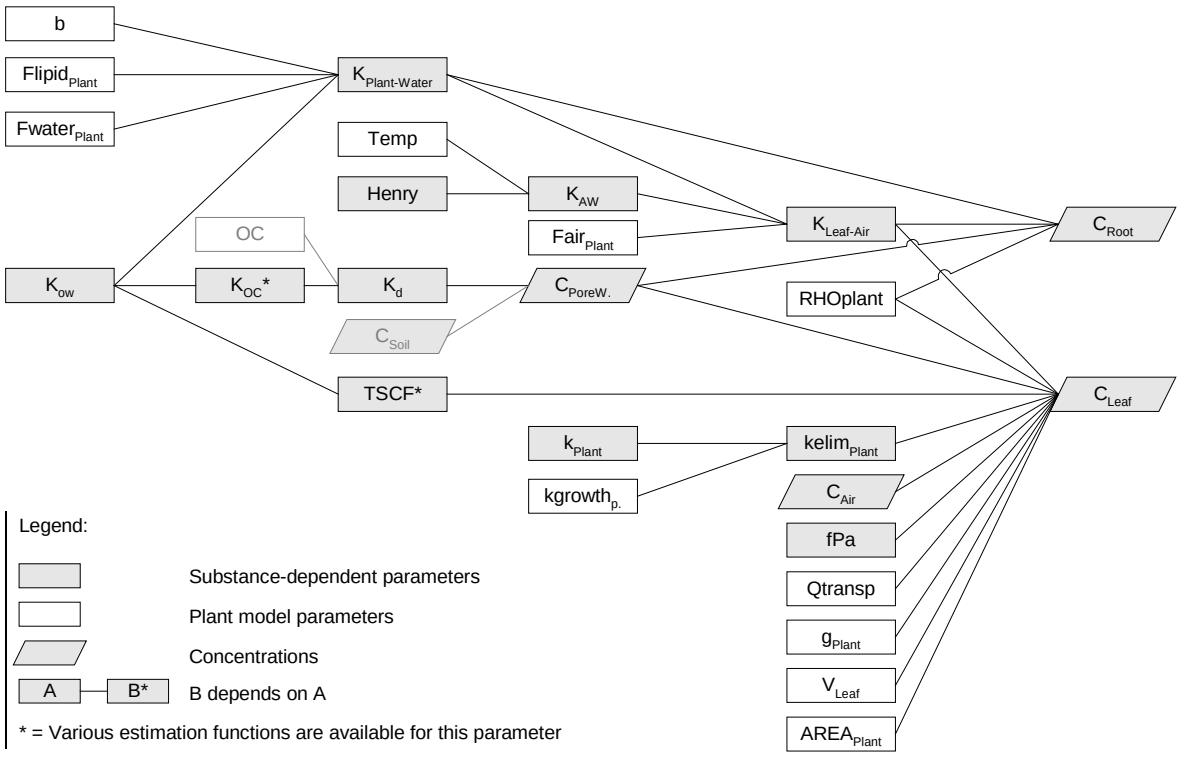


Fig. 7.1 Parameters and their connectivity in the plant model.

The concentration in the upper plant parts is sensitive to the degradation rate in plants when a significant degradation rate is assumed. BENZ and EDC in Tab. 7.6 seem to be exceptions, originating from the low K_{OA}. Besides the parameters mentioned in Section 7.1, which are always important, the Henry coefficient and temperature, but also the correction coefficient b (due to the mo-

derate lipophilicity of BENZ and EDC) play a role for volatile compounds. Concentrations of EDTA and LAS are sensitive to the concentration in soil pore water, the shoot volume and the transpiration stream. For lipophilic substances, such as PCBs, correction exponent b shows a high impact. In general, the following statement applies to all chemicals with a quantifiable vapour pressure: The higher the particulate fraction (fPa), the more sensitive it is and the more unimportant b is. In such a case, the gas exchange between air and plant is dominated by the particulate fraction. If it takes extreme values, its sensitivity will then also be extreme. On the other hand, changing a small particulate fraction simply leads to a relatively small impact and b chiefly controls the diffusive gas exchange.

Tab. 7.5 Sensitivity analysis of the plant model (C_{Rroot}).

		TCDD	HxCDD	OCDD	PCB 52	PCB 138	PCB 180	HHCB	DEHP	BENZ	EDC	EDTA	LAS
Substance parameters	CPorewater	**1**	**1**	**1**	**1**	**1**	**1**	**1**	**1**	**1**	**1**	**1**	**1**
	K_{OW}	**0.9**	**0.9**	**0.9**	**0.9**	**0.9**	**0.9**	**0.9**	**0.9**	**0.6**	0.3		**0.5**
	Negligible impact: None												
Model parameters	B	**14.9**	**17.1**	**17.9**	**12.8**	**14.9**	**16.1**	**12.9**	**16.4**	**2.8**	**0.9**		**2.3**
	RHOplant	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**
	FlipidPlant	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**0.6**	0.3		**0.5**
	FwaterPlant									0.4	**0.7**	**1.0**	**0.5**
	Negligible impact: None												

Tab. 7.6 Sensitivity analysis of the plant model (C_{Leaf}).

		TCDD	HxCDD	OCDD	PCB 52	PCB 138	PCB 180	HHCB	DEHP	BENZ	EDC	EDTA	LAS
Main uptake from		Air	Air	Soil (!)	Air	Air	Air	Air	Air	Air	Air	Soil	Soil
Degradation		X	X	X				X	X	X			X
Substance parameters	fPa	**-0.5**	**-44.7**	**-8924.4**		-0.4	**-1.6**		-0.1				
	kPlant	**-0.9**	**-1.0**	**-1.0**		-0.1	-0.2	**-1.0**	**-0.8**			-0.1	**-1.0**
	CAir	**1.0**	**0.9**		**1.0**	**1.0**	**1.0**	**0.9**	**1.0**	**1.0**	**1.0**		
	CPorewater		0.1	**1.0**				0.1				**1.0**	**1.0**
	Henry				**-0.9**	-0.5	-0.1		-0.1	**-1.0**	**-1.0**		
	K_{OW}				**0.9**	0.5	0.1		-0.1	0.6	0.3		
	Negligible impact: None												
Model parameters	b	0.2			**12.1**	**6.9**	**3.0**	**0.6**	**0.9**	**2.8**	**0.9**		
	RHOPlant	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**	**-1**
	Vleaf	**-1.0**	**-1.0**	**-1.0**	-0.1	**-0.5**	**-0.8**	**-1.0**	**-0.9**			**-1.0**	**-1.0**
	gPlant, AREAPlant	**1.0**	**0.9**		0.1	**0.5**	**0.8**	**0.9**	**0.9**				
	QTransp		0.1	**1.0**				0.1				**1.0**	**1.0**
	Temp				**0.9**	**0.5**	0.2		0.1	**1.0**	**1.0**		
	FlipidPlant				**0.9**	**0.5**	0.2		0.1	**0.6**	0.3		
	FwaterPlant									0.4	**0.7**		
	kgrowthPlant					-0.4	**-0.6**		-0.2				
	Negligible impact: FAirPlant												

Soil-plant as the main transfer pathway for OCDD leads to the extremely high impact of the pore water concentration, and the transpiration stream seems to be an artefact which can be explained

by the fact that the substance is only bounded to aerosols. Assigning to the particulate fraction a value of 0.99 (instead of 1) leads to a drastically reduced sensitivity of fPa and to a high sensitivity of CAir (according to the other dioxins).

Specifying the impact of parameters by means of the methodology applied here may cause problems for non-linear models, because varying the parameters by ±x% may lead to different sensitivity values for the same parameter. For the exposure module investigated here such an effect is possible for two parameters: K_{OW} and b. However, checking the possible outcomes by varying the input by ±20, ±10, ±5 and 0 (=derivative) per cent shows that possible deviations are negligible and do not alter qualitative statements. Fig. 7.3 illustrates such an investigation for input parameter b and PCB52, which reveals the largest deviations (ranging from 12.1 to 14.4 for b = 0.95).

The fraction of air in the plant (FAirPlant) is conspicuously unimportant for each of the conditions investigated here. It can even be hypothesised that it is without impact on each condition: The sensitivity of FAirPlant depends only on two physico-chemical properties, namely K_{OW} and the Henry coefficient. Analysing equation 4 (Section 4.3.4) shows that the lower K_{OW} and the higher the Henry coefficient is, the higher S(FAirPlant) becomes. Fig. 7.3 shows a plot of the Henry coefficient against sensitivity for a fictive substance with a low K_{OW} of 1. As can be seen in the figure, the sensitivity of FAirPlant will never reach an appreciable value unless the ratio FAirPlant/FWaterPlant and the Henry coefficient are given unrealistic values.

In order to interpret the results of this chapter it has to be taken into consideration that the sensitivity analysis depends on the input data used (Appendix A.5). Plausible, but also rounded data or data for which validity has not been evaluated were used for the determination of the sensitivity values. Refinement of these data or their adoption to scenarios can alter statements on the impact of one or the other parameter or exposure pathway. Hence, a re-evaluation is necessary later on.

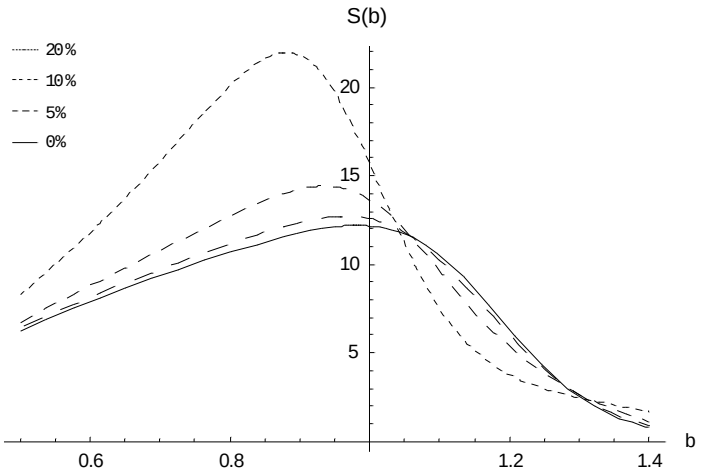


Fig. 7.2 Non-linearity in the plant model for substance PCB52: Sensitivity of parameter b is determined by varying it by ±20%, ±10%, ±5% and 0%.

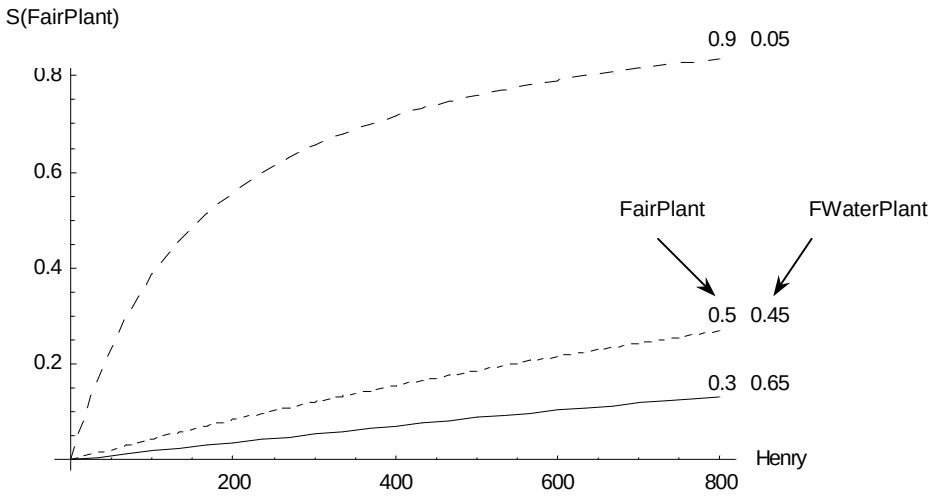


Fig. 7.3 Sensitivity of the fraction of air in plants for a fictive substance with K_{OW}=1 and three plant properties (FairPlant = 0.3 and FWaterPlant = 0.65 are the TGD default values).

7.4 Conclusions

The method used is applicable for the models analysed here, albeit low deviations in the sensitivity numbers arising from the amount of parameter variation may occur. But the conclusions drawn are not influenced by this. The parameters relevant for the calculation of the total daily intake may be classified into highly, moderately and negligibly sensitive ones. Except for the body weight, which is highly sensitive for all chemicals, each parameter is more or less sensitive in a certain situation. The highly sensitive parameters may be grouped and assigned to chemical classes (which are defined by their physico-chemical properties) as presented in Tab. 7.7. It has to be remarked that the classification scheme is only valid against the background of underlying general conclusions derived from the regional distribution model: the impact of emission rates relative to the degradation rates was found to be dependent on the ratio of these parameters. The higher the ratio, the greater the influence of the emission rates and physico-chemical properties and the lesser the influence of the degradation rates. Another example is the fact that the area of the EU shows a high sensitivity for EDTA, but not for LAS. The area of the region has a high impact for LAS. This, of course, is not a direct consequence of significant biodegradability. The reason is rather the different advective flow between the regional and continental scale of the regional distribution model. These general relationships are discussed in detail in BERDING (2000). However, this is not a contradiction and the presented chemical-dependent classification scheme works for all exposure module parameters and, if the emission rates are comparable, also for all other parameter groups. This is caused by the fact that the exposure module depends strongly on the lipophilicity of the substance and slight changes in other parameters normally do not alter the dominance of a certain exposure pathway. Thus, one should keep in mind that the ranking of important substance-specific parameters and those from the regional distribution model cannot be explained by substance-inherent properties alone.

Tab. 7.7 Sensitive parameters in relation to substance properties (impact on $DOSE_{total}$).

Class	Characteristics	Substances	Major exposure pathways*	Highly sensitive parameters
I	Highly lipophilic, less volatile	PCDDs (higher chlorinated)	Soil - Plant roots	Regional: AREAreg, RHOsolid, FsoildSoil, depthAgric, FOCSoil, fAgricReg), Exposure: b, BW, RHOplant, FlipidPlant, IHroot, Substance: EregirstWastewater, kdegsoil, ERegair
II	Lipophilic, less to semivolatile	PCDDs (higher chlorinated)	STP - Water - Fish Soil - Plant roots	Regional: heighAir, windspeed, ConJunge, SurfAer, Exposure: b, BW, RHOplant, kgrothplasnt, VLeaf, AREAplantgPlant, ICgrass, Substance: TempMelt, kdegsoil, ERegAir, VP
II a	Lipophilic, semivolatile	PCBs, HHCB	STP - Water - Fish Soil - Plant roots	Regional: FconnectSTP, AREAreg, fAgricReg, Exposure: b, BW, RHOplant, FlipidPlant, IHroot, Substance: ERegfirstWater, K_{OW}, VP, MOLW, SOL
II b	Lipophilic, semivolatile, high K_{OA}	DEHP	Soil - Plant roots	Regional: FconnectSTP, AREAreg, heighAir, FWaterSed, Exposure: b, BW, RHOplant, ICgrass, Substance: ERegAir
III	Less lipophilic, volatile	BENZ, EDC	Air	Regional: AREAreg, heighAir, windspeed, Exposure: BW, ICair, Substance: BIOinh, EContAir
IV	Hydrophilic, not volatile	EDTA	STP - Water - Drinking water	Regional: Rainrate, FrunoffSoil, AREAEU, FFlowOutReg, Exposure: BW, IHdrw, Substance: ERegfirstWater
IV a	Surface-active, not volatile, significantly biodegradable	LAS	STP - Water - Drinking water STP - Water - Fish Soil - Plant roots	STP: Fraction oc raw, Regional: FconnectSTP, AREAreg, RHOsolid, FsoildSoil, depthAgric, FOCSoil, fAgricReg, Exposure: BW, RHOplant, kgrowthPlant, Qtransp, IHleaf, Substance: ERegfirstWater, kdegsoil

* This column shows the main exposure pathway as calculated by the model (Tab. 7.4). An evaluation of these results is given later.

Besides this classification, the section provides the following results, which are important for further investigation.

- 40% of all parameters can be neglected with respect to calculating the total daily intake.
- The models contributing to the main exposure pathway show the most sensitive parameters (trivial!). Particularly for these models, accurate parameter values and an investigation of the underlying theory are necessary with respect to the following simulations. For the substances investigated here, plant roots, air and drinking water are the prevailing pathways.
- The plant in particular seems to be an unrealistically important exposure pathway for the lipophilic substances and requires further investigation.

- The parameters of the sewage treatment plant model are only relevant for waterborne substances with a significant biodegradation.
- With the intention of ameliorating an assessment, estimated parameters are (and should be) replaced by (often rounded) more plausible values (e.g. setting the estimated particulate fraction of OCDD to unity). But due to the effect of rounding, the value for the particulate fraction switches off the transfer pathway air-plant and yields unrealistic results.
- Because of its vanishing impact in each situation, the fraction of air in plants (FAirPlant) can definitely be ignored.

Finally, the findings of this section are relevant to the validity of the model: The investigation revealed many redundant parameters for certain substances, e.g. for EDTA the total dose is highly sensitive to only 7 out of 128 parameters from the overall system and to only 3 out of 34 parameters from the exposure module. This means that the overall system is of low relevance for assessing the total daily intake of EDTA. Against the task of the system to assess exposure for a variety of chemicals, more parameters show a key function, but there are still several parameters with negligible impact. In this context, due to their low complexity, all submodels of the exposure module are models with a high relevance. Also, the relative complex plant model, with the exception of the fraction of air in the plant, offers only key parameters.

7.5 Summary

Sensitivity analyses were performed using the differential approach to evaluate which input parameters are most important in assessing environmental concentrations and human exposure. It was shown that the total daily dose is sensitive to the majority of parameters if a variety of chemicals is investigated. However, there is a set of parameters with negligible impact. Few of the sensitive parameters show extremely sensitivity values and should be treated with caution. In order to assign sensitive parameters to substance classes, it is sufficient to distinguish between lipophilic, waterborne and airborne substances. Classification of the substances into six classes is suitable for a more refined view.

8 Scenario analyses and comparison with measured data

The comparison of measured concentrations with monitoring data forms a fundamental part of the model validation task. The calculated total daily dose should also be compared with observed doses. However, these doses do not exist in a strict sense since doses cannot be measured. But alternative assessments of the total daily dose are available in the scientific literature for most of the investigated chemicals. Data that can be compared with the calculated dose are in particular available for PCDD (TRAVIS ET AL. 1987, FÜRST ET AL. 1990, BRUNN 1993, FÜRST 1995, SCHREY ET AL. 1995), PCB (DFG 1988), DEHP (FÜRST 1995, KOCH 1995), HHCB (FORD 1996), EDC (BUA 1994, KOCH 1995, EC 1996C) and benzene (BUA 1993). These alternative estimations often consider further exposure pathways such as dermal uptake, or differentiate between, for instance, indoor and outdoor exposure. If available, they are nevertheless considered as more realistic doses. The first section of this chapter deals with the total daily intake, which is the final step of an exposure assessment according to the TGD. Individual models within the exposure module may become important for certain chemicals and, thus, were investigated in the following sections.

8.1 Bioconcentration model fish

8.1.1 Comparison with experimental data

A literature search was carried out and experimental data were compared with the model predictions in order to investigate the predictions of the K_{OW}/BCF model. The results are depicted in Fig. 8.1 and Fig. 8.2. Data availability is discriminative for the individual substances: Many experimental BCF are available for the lipophilic chemicals TCDD, OCDD and DEHP, but fewer exist for the remaining dioxins, for PCB, for HHCB and for the rather hydrophilic chemicals.

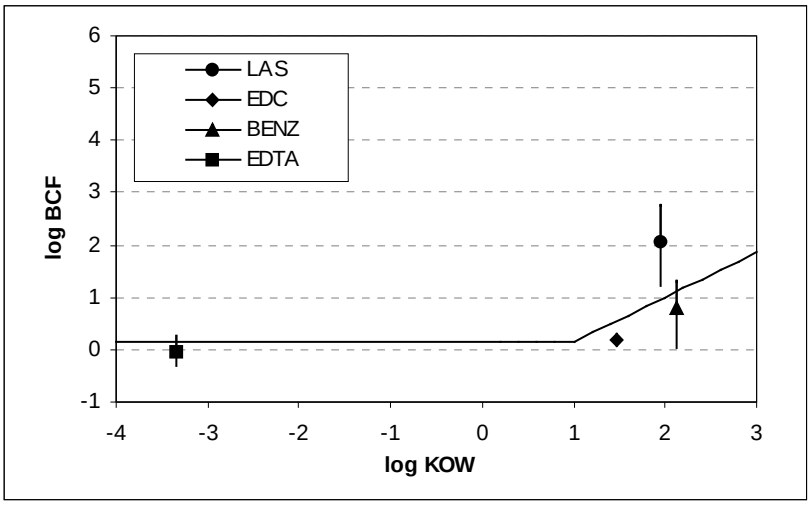


Fig. 8.1 Calculated and experimental bioconcentration factors for the rather hydrophilic compounds. Experimental values are shown by means of their range (minimum, maximum) and median.

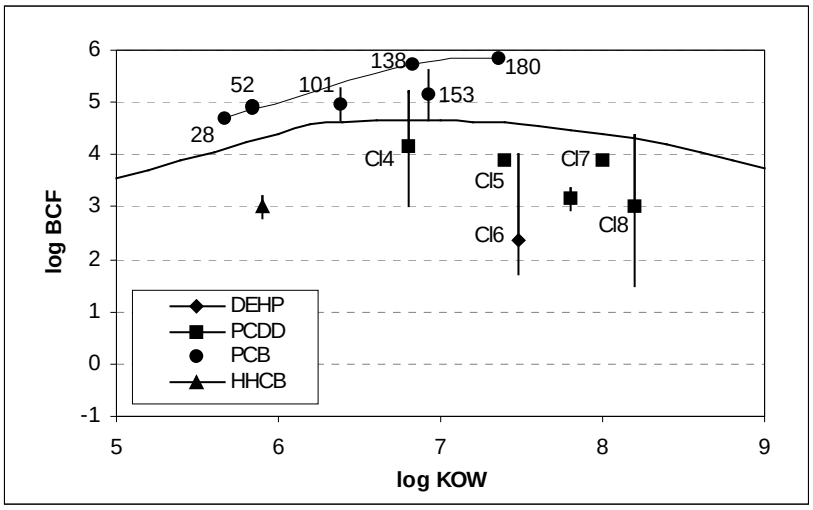


Fig. 8.2 Calculated and experimental bioconcentration factors for the lipophilic compounds. Experimental values are shown by means of their range (minimum, maximum) and median.

The measured bioconcentration factors of the rather hydrophilic chemicals EDTA, EDC and BENZ correspond to the predictions. Their range is small. The error is at maximum up to one log unit. All experimentally determined BCF exceed the prediction by a factor of 10 for LAS. The range of experimental values increases with increasing lipophilicity and results in higher uncertainty in model application for these substances. The bioconcentration potential of PCB congeners is underestimated, whereas the potential of PCDD and DEHP is overestimated. The deviations are up to three orders of magnitude (for DEHP and OCDD). The regression equation is therefore conservative for these chemicals.

The dioxins applied were the only lipophilic compounds in the training set of CONNELL AND HAWKER (1988). The polynomial curve results from fitting the relationship for these chemicals. Other lipophilic chemicals were not considered. The results of this study correspond to those of JAGER AND HAMERS (1997), who compared predictions with measurements for comprehensive datasets. Their datasets contained further lipophilic substances and also revealed the broad range of possible outcomes. They concluded that the majority of experimentally determined BCF are significantly underestimated for lipophilic chemicals (up to a factor of 1000). Furthermore, the model was judged to work well for the majority of substances with a log K_{OW} from 1 to 6. However, in individual cases deviations of up to two log units may occur.

8.1.2 Comparison to the monitoring data

The product of the dissolved fraction of a chemical in water and the estimated bioconcentration factor delivers the concentration in fish. Since total concentrations are mostly available in the literature, the dissolved fraction of the investigated chemical had to be determined first. According to Tab. 8.1, the bounded fraction is only relevant for PCDD, PCB and DEHP.

Tab. 8.1 Estimation of the molecular dissolved fraction by means of a TOC of 1.6 and 16.4 mg/l, respectively. The TOC corresponds to the lowest and highest measured value of the River Rhine (IKSR 1990). The second column shows the minimal dissolved fraction and the third column represents the maximal dissolved fraction. See Section 5.2.3 for the equation used.

	Dissolved fraction	
Substance	**Minimum [%]**	**Maximum [%]**
TCDD	2	19
PeCDD	1	6
HxCDD	0	2
HpCDD	0	1
OCDD	0	1
PCB 28	24	76
PCB 52	18	69
PCB 101	6	39
PCB 138	2	18
PCB 153	2	15
PCB 180	1	6
DEHP	0	5
HHCB	16	66
EDTA	100	100
LAS	100	100
EDC	100	100
BENZ	100	100

No realisable measured concentrations are available for dioxins. For polychlorinated biphenyls the material of the River Rhine segment between Village-Neuf and Lobith originating from 1990 (IKSR 1990, IKSR 1993, IKSR 1994) is appropriate and was used. Based on concentrations from six different sampling sites, the dissolved fraction was estimated by means of the content of suspended matter and organic carbon. The calculated concentrations in fish in comparison with measured concentrations are presented in Fig. 8.3. The figure shows the minimum and maximum for each congener and, thus, a survey of the whole river segment. The presented values are the median values of the individual sampling sites. Additionally, the material provides the possibility to assign the concentration for 7 species (roach, pike, zander, bream, perch, bass, eel) to individual sampling sites. The single values are depicted in Fig. 8.4.

The comparison of minimal and maximal measured concentrations shows different results (depending on the congener). It must be kept in mind that PCB 28 concentrations for both water and fish often fall below the detection limit

Tab. 8.2). While the lower chlorinated PCB congeners 28 and 52 match the monitoring data, the model significantly underestimates the measured data for PCB 101, 138, 153 and 180. Also the depiction of the single values indicates an underestimation of up to three orders of magnitude. However, the outcome strongly depends on the species under consideration. Deviations will decrease if eel is omitted. The reason for this is the eel's higher fat content, which results in higher concentrations for the whole fish. If concentrations were referred to the fat fraction, no deviations between the individual species would occur.

Tab. 8.2 Number of values below detection limit (324 samples for each congener).

Congener	**Not detectable**
PCB 28	118
PCB 52	21
PCB 101	12
PCB 138	11
PCB 152	8
PCB 180	18

Monitoring data from the River Rhine are also available for DEHP. A detailed survey is provided by NRW (1993). Concentrations in surface waters are reported as total water concentrations for DEHP as well. It is expected that approximately 95.2% to 99.5% are not dissolved (Tab. 8.1), and thus, also for DEHP the dissolved fraction first had to be determined. The calculated minimal and maximal concentration shows in contrast to PCB that the monitoring data seldom match the predictions and are often overestimated by more than two orders of magnitude. The average overestimation amounts to one log unit. Concentrations for HHCB originate from the River Ruhr (ESCHKE 1995) and are underestimated by 2 to 3 orders of magnitude. Concentrations for EDC and BENZ are usually below the detection limit for both water and fish. The minimum therefore represents the detection limits for both substances. The maximum corresponds to the maximum of literature values and represents an unrealistically high concentration. No fish concentrations for these chemicals were detected in the River Rhine. Thus, these substances are not useful in deriving statements on the model's validity. For LAS and EDTA, no measured concentrations are available for the real aquatic environment.

Based on the comparison of measured with predicted bioconcentration factors, an overestimation of the concentrations for higher chlorinated PCB and an underestimation for DEHP and HHCB had to be expected. This conclusion is confirmed by the measured concentrations from the Rivers Rhine and Ruhr.

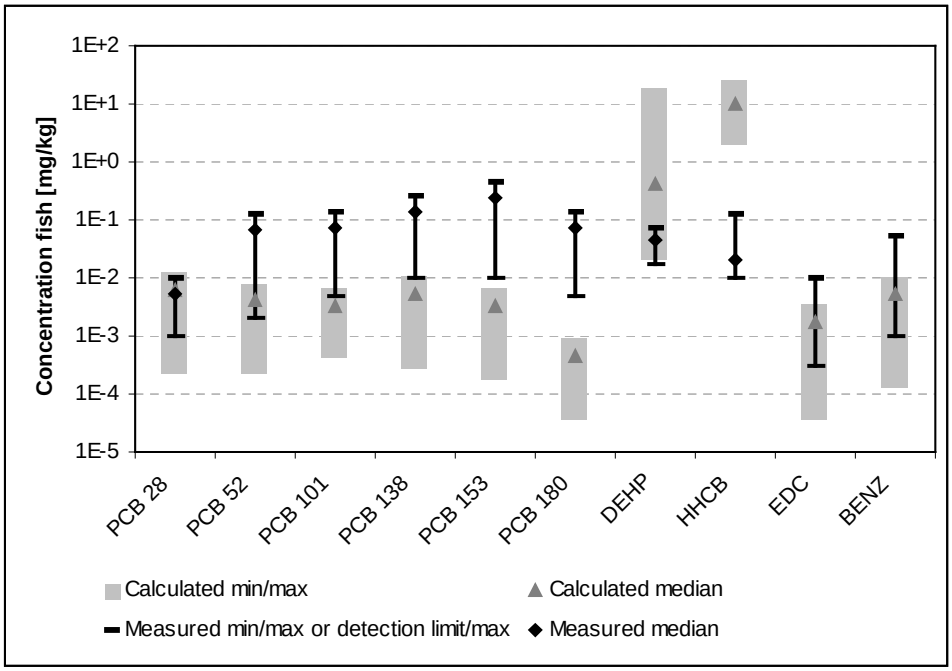


Fig. 8.3 Comparison of the median values of measured and calculated PCB concentrations in fish from the River Rhine (survey). For PCB 28, EDC and BENZ the minimum represents the detection limit.

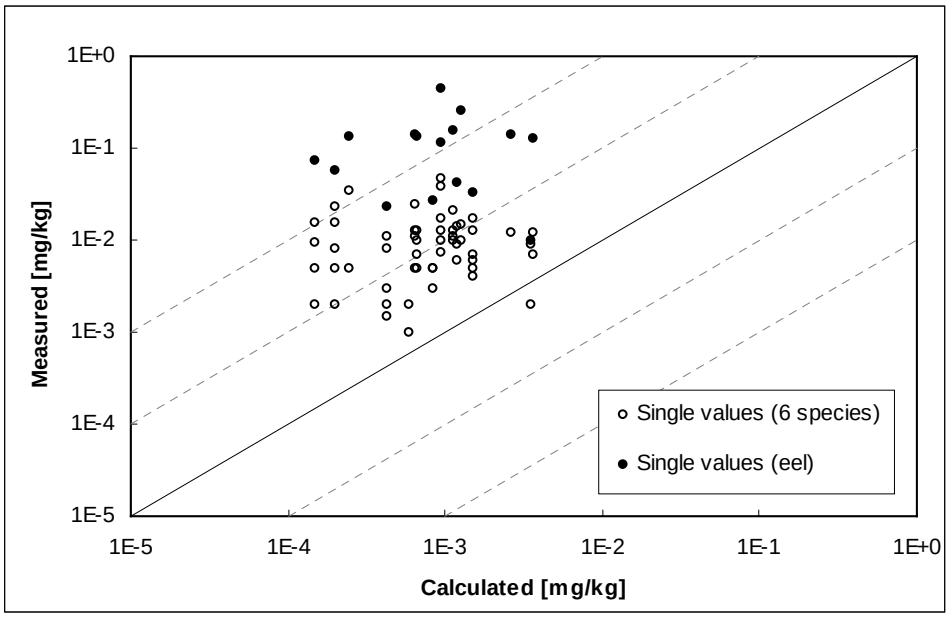


Fig. 8.4 Comparison of monitoring data (single values, seven species, River Rhine) for various PCB congeners with calculated concentrations.

Discussion: Particularly for lipophilic chemicals, significant deviations from measured data occur. The literature provides several explanations. The observed decrease of bioconcentration above K_{OW} > 6, which is also considered in the TGD relationship, is explained by a metabolism of substances. Another reason is the size of the molecule, which hampers the passing of biomembranes for molecules larger than approximately 9.5 Å (STEINBERG ET AL. 1992). GEYER ET AL. (1994) investigated the conditions of bioconcentration tests and pointed out the following aspects:

- Tests were often carried out with chemical concentrations that exceed the water solubility.
- A basic assumption of bioconcentration models the steady state between fish and water. This requires long-term periods for the superlipophilic compounds (usually several months). But the tests were carried out over shorter periods.
- In contrast to common belief, experiments show that large molecules are also able to absorb. However, this may depend on species and temperature.
- Superlipophilic chemicals are hardly metabolisable in aquatic organisms. Metabolism is consequently not suitable as an explanation of the observed reduced bioconcentration.

It can be concluded that the assumption of a decreasing bioconcentration potential with increasing log K_{OW} is not generally valid and, thus, the applicability of the TGD relationship is questionable for superlipophilic compounds.

A fast metabolism in aquatic organisms is arguable for DEHP, because of a less than 3d half-life in fish (NRW 1993), and is regarded as one reason for the low measured bioconcentration factors. The same effect is suitable for explaining the observations for HHCB. According to Fig. 8.1, the bioconcentration of LAS is underestimated. TOLLS ET AL. (1994) investigated the bioconcentration potential of surfactants and emphasised that experimentally determined BCF overestimate the real bioconcentration potential. The reason for this is the missing separation between parent substan-

ces and their degradation products in the context of the experiments. However, fast metabolism is commonly assumed. Furthermore, due to the partly lipophilic, and partly hydrophilic character of LAS and other surfactants, they are not expected to be able to pass the biomembranes. The measured concentration in an organism rather results from the accumulated degradation products. According to TOLLS ET AL. (1994), the degree of overestimation cannot yet be quantified. Against this background, the stated degree of overestimation (average one log unit) by the K_{OW}-based TGD model is disputable. The constant bioconcentration of ~1, as assumed in the TGD for extremely hydrophilic substances, is justified on the basis of the investigations for EDTA, since no bioaccumulation is expected for strongly polar and water-soluble chemicals.

The TGD neglects further processes with possible impacts, such as growth or biomagnification. Growth causes a dilution effect and leads to a reduction of the partitioning between fish and water. Biomagnification is important for lipophilic and persistent compounds (log K_{OW} > 5) and for fish of higher trophic levels (THOMANN 1989). Thus, it is expected that this effect is able to explain the increasing underestimation of PCB accumulation with an increasing degree of chlorination.

According to these findings, the neglect of underlying model assumptions is obvious. Although the model is applicable for the investigated chemicals with respect to regression range and molecular weight, bioconcentration alone is not the only relevant process. In particular, metabolism, growth and the organism's trophic rank must also be taken into account to avoid errors.

It can be concluded that the K_{OW} is not a perfect descriptor for the bioconcentration potential. Especially the validity of the TGD model is questionable for a log K_{OW} > 6 and eventually results in a false evaluation. However, if an error of approximately two orders of magnitude is acceptable then the results will be sufficient for lipophilic chemicals.

8.2 Biotransfer into milk and meat

The contamination of animal fat tissue by hydrophobic PCDD and PCB compounds is well known (FÜRST ET AL. 1990, BECK ET AL. 1989, WEIGERT ET AL. 1991). A compilation of concentration levels can be found in DFG (1988), BML (1993) or BALLSCHMITER (1996). Fewer or no datasets are available for all other chemicals. Thus, only PCDD and PCB are useful for evaluating the validity of the biotransfer model. Grass and, to a lower extent, soil are the relevant transfer pathways according to the TGD model (Tab. 8.3), which corresponds to the findings of MCLACHLAN (1992). Representative data are therefore indispensable for these environmental media. Intake occurs by more than 90% via the air for both EDC and benzene (even if unrealistically high concentrations in soil and grass are applied). Soil and grass are therefore negligible for these chemicals. For DEHP, grass is the dominating source of exposure (more than 80%). Polluted drinking water may be a significant pathway in extreme cases.

Tab. 8.3 Estimated averaged fractions of the environmental media for PCDD and PCB intake.

Substance	Grass [%]	Soil [%]
TCDD	72	28
PeCDD	91	9
HxCDD I	82	18
HxCDD II	86	14
HxCDD III	86	14
HpCDD	84	16
OCDD	84	16
PCB 28	99	1
PCB 52	99	1
PCB 101	98	2
PCB 153	95	5
PCB 180	92	8

Fig. 8.5 presents the datasets for concentrations in beef, which were taken from papers by FÜRST ET AL. (1990) and WEIGERT ET AL. (1991). The concentrations in the intake media originate from the *Chloraromaten* monitoring programme (NRW 1991A, NRW 1991B). Published values mostly refer to the fat content. Thus, they were first multiplied by a fat content of 25% (TRAVIS AND ARMS 1988A) to obtain values for meat.

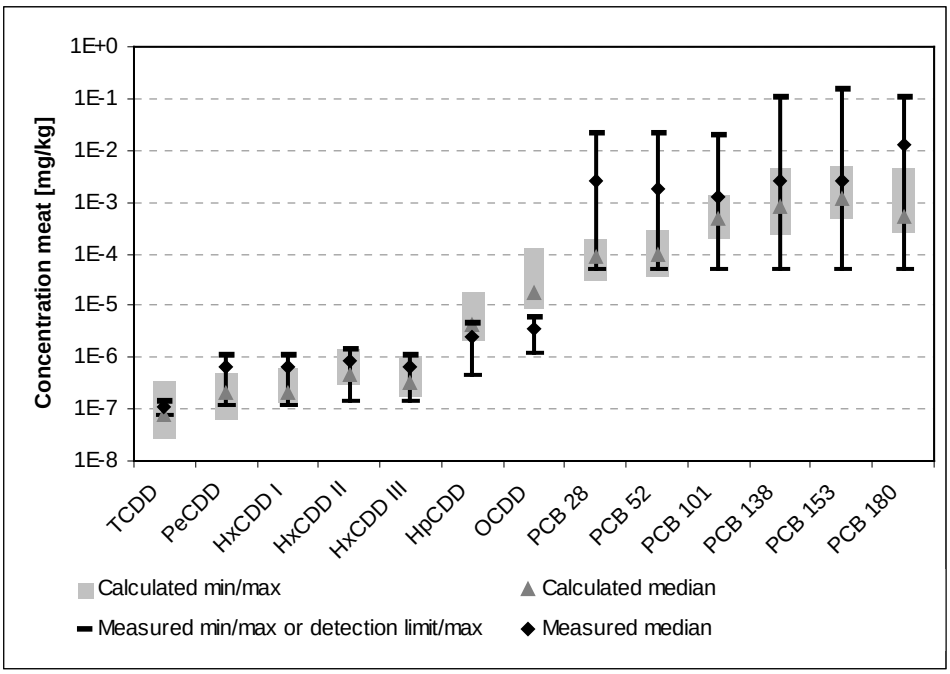


Fig. 8.5 Comparison of measured with predicted concentrations in beef for PCDD and PCB. The minimum for PCB and TCDD represents the detection limit.

The TGD model renders almost exactly the concentrations of the lower chlorinated dioxins. The deviation is less than one log unit. Concentrations of HpCDD and OCDD are overestimated. In contrast, PCB is underestimated by up to a factor of 100. Most of the values for PCB 28-101 admittedly fall below the detection limit (Tab. 8.4). Consequently, an overestimation for PCB congeners can usually be assumed. Information on percentiles in WEIGERT ET AL. (1991) points out that the majority of measured concentrations for PCB 138-180 correspond to the lower half of the range, as depicted in the figure. Regarding this complementary information, the model delivers good predictions of concentrations.

1,2-Dichloroethane and benzene may be underestimated by up to 4 orders of magnitude (no figure). However, these chemicals are mostly not detectable and measured values are often non-representatively high (BUA 1995, RIPPEN 1995). These data are not suitable for evaluating the model.

Tab. 8.4 Number of values below detection limit in beef.

Congener	Samples	Not detectable
PCB 28	1111	1017
PCB 52	1110	1009
PCB 101	1009	995
PCB 138	1111	306
PCB 152	1111	240
PCB 180	1111	430

The situation regarding concentrations in milk is similar: The transfer of PCDD and PCB from fodder into milk is well known (MCLACHLAN 1996, RUOFF 1995, NRW 1991A, NRW 1995, BML 1993). However, knowledge on the transfer of the other investigated compounds is scarce. The available datasets (originating from the *Chloraromaten* programme, NRW 1991A and 1991B) are very suitable for a validation study since the monitoring programme provides homogeneous single values for milk and all intake media. A comparison with the field data indicates an overestimation by the model. The concentrations of PCB 153 and 180 are well predicted on the one hand, but concentrations of the other chemicals are overestimated on the other. For PCB 101, deviation is up to a factor of 11, for PCB 28 and 52 a quantification of the deviations is not possible since nearly all concentrations fall below the detection limit. Deviation from measured values ranges between a factor of 3 (TCDD) and 116 (OCDD). The significantly higher overestimations for HpCDD and OCDD are striking.

DEHP concentrations of up to 3.1 mg/l in milk are reported. (EC 1996c). In any case, the fate of this chemical in the terrestrial food chain is poorly investigated. Only a few papers provide DEHP concentrations originating from food investigations, but measured concentrations are often influenced by plasticisers in the packing material (RIPPEN 1995). Representative data are unavailable. KLEIN ET AL. (1995) reported background concentrations in milk of 0.0074 mg/l for phthalates as a whole. The predicted value is 2.9 mg/l for DEHP and, hence, is significantly more than the background concentration.

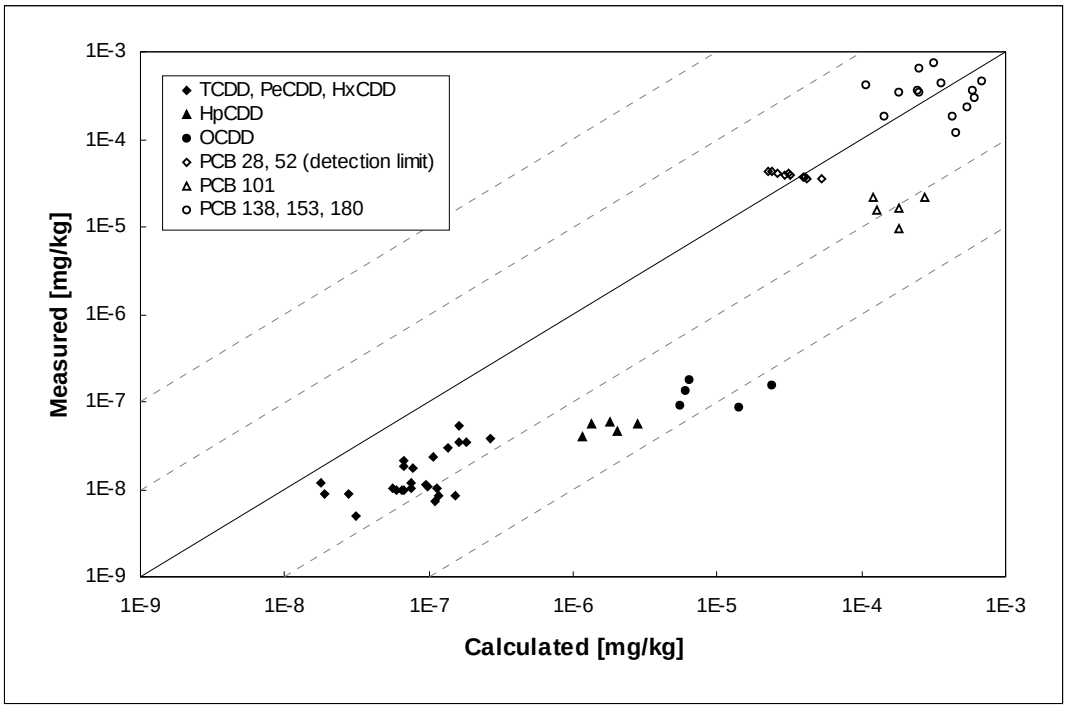


Fig. 8.6 Comparison of measured concentrations in milk (single values) with predictions for PCDD and PCB. Concentrations for PCB 28 and 52 represent the detection limit.

Uncertainties inhere the predictions. The assumed fat content of 25% for beef (TRAVIS AND ARMS 1988A) represents a considerably high value. Consumed beef usually shows a fat content ranging from 4.4% to 16.5% (ELMADFA ET AL. 1990). If one assumes a fat content of 10%, the concentration in the meat will diminish by 40% and the conclusions will correspond to those derived for predicting milk concentrations, i.e. a relatively good estimation for higher chlorinated PCB and an overestimation for lower chlorinated PCB. Fodder can be deemed as the only relevant transfer pathway for the investigated dioxins and PCB with a high degree of chlorination (RUOFF 1995). The TGD model assumes grass to be the only type of fodder. This is a conservative approach, because grass is more polluted than other sorts of fodder. Intake via grass averages approximately 2/3 of the total intake of pollutants, but is not the only sort of fodder (MCLACHLAN 1992). In particular, concentrations in non-leafy fodder are lower.

Discussion: Partition coefficients between octanol and water lie in between the regression range for PCB 28-101 in beef. Also the regression equation for milk is in a strict sense only applicable for these substances, for LAS and for benzene (Section 6.2.2). Against this background, good predictions are also obtained for the higher chlorinated PCB.

An explanation for the resulting pattern of congeners for dioxins and polychlorinated biphenyls is a possible metabolism and reduced bioavailability, respectively. According to MCLACHLAN (1992), the congeners PCB 128, 153 and 180 are classified as persistent and it can be expected that they are eliminated via milk. For PCB 28, 52 and 101, metabolism is hypothesised. These effects may explain the good predictions for higher chlorinated PCB 153 and 180 and overestimation for the lower chlorinated congeners. All of the 2,3,7,8-substitued PCDD congeners investigated here are regarded as persistent (BALLSCHMITER 1996, MCLACHLAN 1992).

Dioxin results are explained by a reduced bioavailability: The resorption of chemicals in the gastrointestinal tract depends on lipophilicity. For a log K_{OW} < 6.5, the resorption is constant and amounts to approximately 80%. Then resorption decreases with increasing lipophilicity and amounts to less than 20% for a log K_{OW} > 8 (MCLACHLAN 1992). This phenomenon may explain that the measured concentrations are low in relation to the model results for higher chlorinated PCDD. DEHP concentrations will be overestimated due to metabolism. The purpose of both regression equations is to produce average biotransfer factors. However, extreme concentrations in fat tissue may actually occur that exceed the calculated values by more than three log units. Examples are 1,2-dichloroethane or benzene in beef. But the models are not applicable in practice for estimating average concentrations for these chemicals. The steady-state assumption presumes a sufficiently long exposure time and a constant level of exposure. The former is no problem due to the high mobility of EDC and benzene. The latter, however, will seldom be fulfilled in practice, because air concentrations vary strongly on a regional scale (ranging from ng/m³ to µg/m³ in air, BUA 1993 and 1995). Furthermore, meat and milk are exposed in varying concentrations due to transporting. It has to be remarked that, against the background of human exposure, the model application is not relevant since uptake occurs exclusively via air (see Section 8.4.2). The model works with fresh weight-related concentrations. The fat content laid down in the regression equation for meat is unrealistically high.

It can be concluded that the model for the soil/plant/air/water-cattle transfer pathway is of restricted applicability for PCDD and PCB and similar lipophilic substances. It is not applicable for unstable compounds or in the case of a reduced resorption. Good results with deviations within a factor of 10 are obtained for chemicals such as PCB 138, 153 and 180, HxCDD and HpCDD. All in all, the model tends to overestimate the chemicals investigated in this study and therefore provides a conservative result. However, in extreme situations significant underestimations may occur.

8.3 Uptake by plants

The database caused similar results to those from the investigation of the previous models: Good datasets for PCDD and PCB are available, but only poor-quality or no data at all can be found for the other chemicals. Various calculations were carried out. Calculated concentrations were compared with those of the *Chloraromaten* programme. Air concentrations for PCB (which are not provided by the monitoring programme) were taken from HALSALL ET AL. (1995). These describe concentrations near the city of Ulm and are considered as typical background concentrations for rural areas.

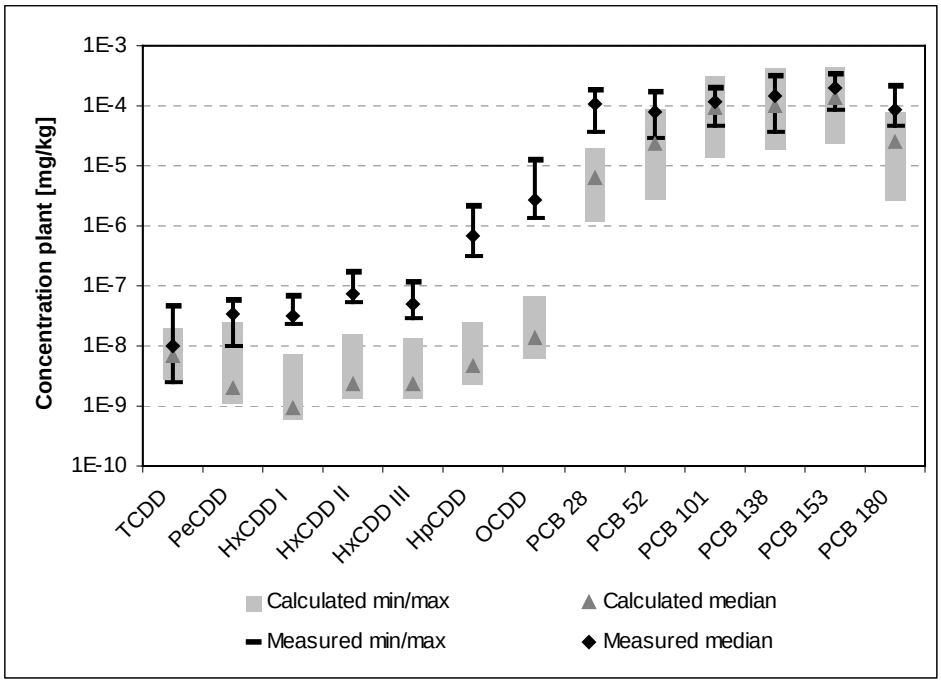


Fig. 8.7 Comparison of measured and predicted PCDD and PCB concentrations for grass. Data taken from NRW (1991B).

As can be seen in Fig. 8.7, the model produces deviations with an average error of less than a factor of 10 for concentrations in grass. Only the PCB 28 concentration is underestimated. The calculated concentrations for dioxins are also too low ; the higher the degree of chlorination, the lower the error is. The estimation for TCDD works well, whereas the estimation for PeCDD and HxCDD is fair with deviations of up to two log units. Poor results with deviations of more than two log units are archived for HpCDD and OCDD.

Further monitoring data for PCDD and PCB were additionally investigated. KAUPP (1996) analysed the atmospheric input of PCDD into maize under field and experimental conditions. The reported concentrations for PCDD in the air (near the city of Bayreuth) and in a maize field (leaves and flowers) were used in a further model calculation. The measured concentration is again underestimated by 1 to 2 log units. The measurement also revealed different contamination of the various plant parts: Flowers are more contaminated than leaves. KAUPP (1996) simultaneously carried out an analysis in a greenhouse in which maize leaves were exposed by filtrated and infiltrated air. Thus, the effect of an exclusively gaseous input in relation to a gaseous plus particulate input could be revealed. For lower chlorinated PCDD no difference was observed, whereas for higher chlorinated PCDD a difference of factor 2 to 3 was found (no figure). A significant input by particle deposition was concluded for slightly volatile compounds. With the intention of considering the effect of deposition on the results, the model was supplemented by a deposition part, as proposed by TRAPP AND MATTHIES (1998). A dataset for lettuce (*lactuca sativa*) from TRAPP ET AL. (1997) was applied for the calculation (soil and plant concentrations from Ulm, otherwise standard parameter). The effect of considering deposition is depicted in Fig. 8.8: Considering deposition ameliorates the result by a factor <5. The concentration of the lower chlorinated congeners is still strongly underestimated.

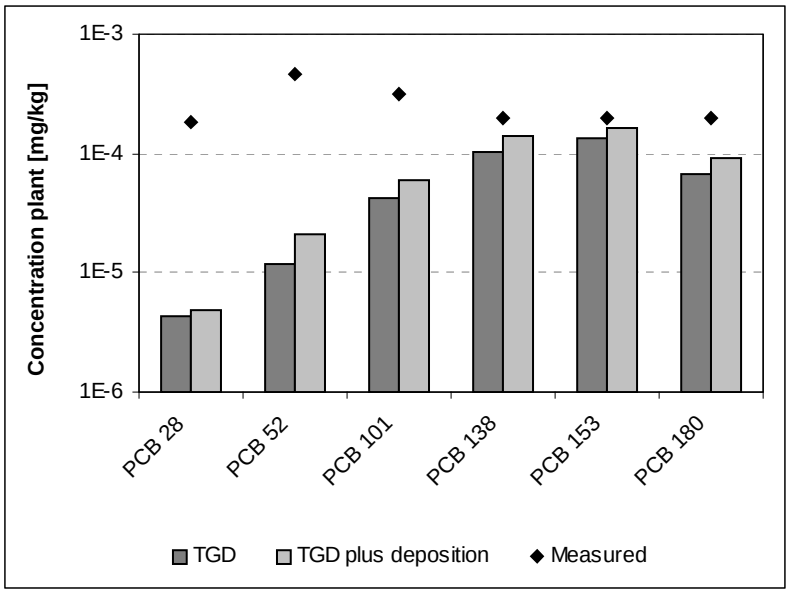


Fig. 8.8 PCB in lettuce with and without considering deposition. Data from TRAPP ET AL. (1997).

Only a few data are available for the remaining chemicals. Some investigations concerning LAS exist. These deal with the uptake of ^{14}C-marked LAS from soil into plants (FIGGE AND SCHÖBERL 1989, IPCS 1996). An uptake of detectable quantities of LAS or its degradation products was revealed for soils with sludge application by means of the radio-tracer technique. Fig. 8.9 presents these LAS concentrations in potatoes, grass, beans and radish in relation to the predictions. The model results seem to match the measurements, but it has to be noted that no distinction is made between LAS and its degradation products.

The DEHP concentrations used provide a survey on concentrations in grass on industrial sites. These are not representative for typical background concentrations in rural areas. Representative data are unavailable. Concentrations of plants like oats, maize, potatoes (leaves) on soils with sludge application also fall in the range of measured values. However, under extreme conditions concentrations of up to two orders of magnitude higher may occur (KÖRDEL AND MÜLLER 1995, RIPPEN 1995, UBA 1996). The applied air concentrations encompass both background concentrations and typical concentrations in industrial areas. But again, significantly higher values are possible (RIPPEN 1995). The calculated concentrations correspond rather to the measured concentrations in industrial areas.

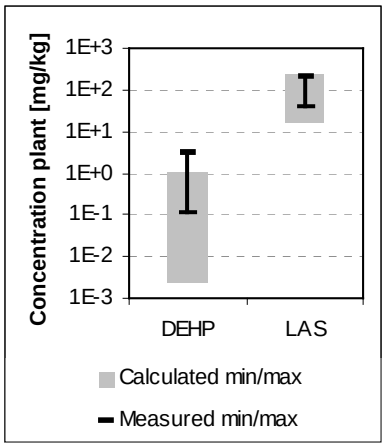


Fig. 8.9 Measured and predicted concentrations for DEHP and ^{14}C-marked LAS.

Uncertainties in estimating the particulate fraction: For the air compartment the fractions in the gaseous and particulate phase must be distinguished (Section 5.2.3). Measurements in less contaminated areas show that the lower chlorinated congeners occur mainly in gaseous form, while the higher chlorinated congeners are predominantly bounded to particles (KAUPP 1996, MCLACHLAN 1992).

Tab. 8.5 Measured, used and calculated particulate fractions (in %). A * represents the median of measurements. Data from KAUPP (1996) and UMLAUF (1994).

Substance	Measured	Used	Calculated[a]	Calculated[b]
TCDD	11-40	23*	32	11
PeCDD	32-58	43*	93	79
HxCDD	42-83	76*	98	91
HpCDD	61-94	88*	> 99	99
OCDD	66-96	96*	> 99	99
PCB 101	< 2	2	3	1
PCB 138	2-10	8	27	9
PCB 153	1-7	5	22	7
PCB 180	5-30	20	61	29

[a] TGD-default values for Junge-constant and surface area of particles and
[b] values proposed by COTHAM AND BIDLEMAN (1995)

Partitioning between gaseous and particulate phases significantly determines the uptake of plants (DOUBEN ET AL. 1997) and is, depending on the degree of chlorination, very sensitive for PCB and PCDD. The estimation of the particulate fraction (Junge-equation, Section 5.2.3) is influenced by the environmental temperature for PCDD and PCB (because the substances are solid under environmental conditions and the vapour pressure of the liquid chemical is used to estimate the particulate fraction). Due to its exponential influence, the temperature is the most sensitive parameter (besides the melting point). The vapour pressure is less sensitive in this case. The varying environmental temperature thus produces uncertainties in the estimation. Measured particulate fractions are available for PCDD (KAUPP 1996) and several PCB congeners (UMLAUF 1994). The measured fractions strongly deviate from the TGD estimations for some congeners (Tab. 8.5) and lead to a significant impact on the calculated concentrations in the plant: By using measured fractions the result is ameliorated by approximately one log unit for PeCDD to OCDD. Regarding the polychlorinated biphenyls, the best amelioration is obtained for PCB 180. A further dataset by JONES ET AL.

1997 was used to estimate PCDD concentrations. It represents typical background concentrations in a rural area of Great Britain. The results correspond to those obtained from the *Chloraromaten* monitoring programme (NRW 1991A-B) and thus confirm the findings.

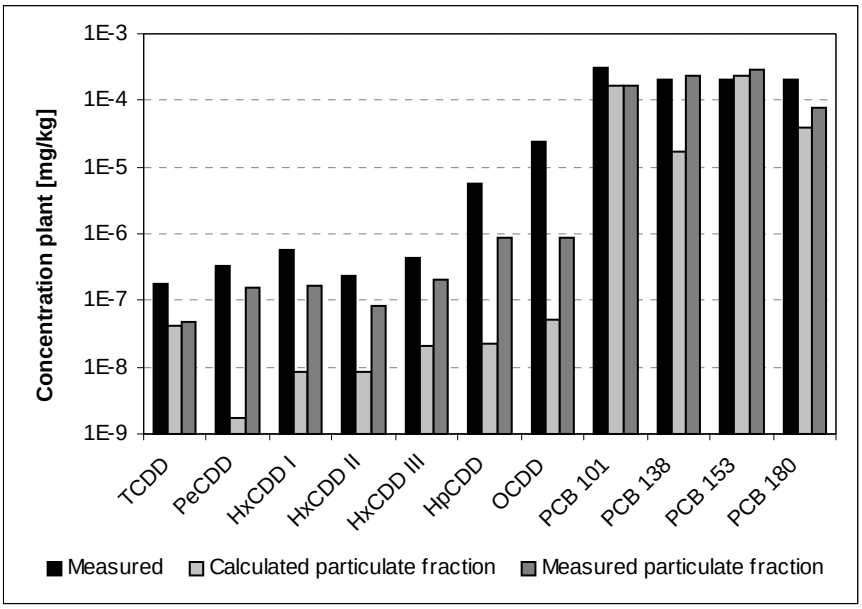


Fig. 8.10 Calculated concentrations in plants using measured and estimated particulate fractions. Concentrations in air are taken from JONES ET AL. (1997) for PCDD in grass and from TRAPP ET AL. (1997) for PCB in lettuce.

The estimation will significantly improve if alternative values for surface area s and Junge parameter c are used instead of the TGD default values. COTHAM AND BIDLEMAN (1995) proposed $s = 1.5 \cdot 10^{-6}$ [cm²/cm³] and c = 17.2 [Pa m] as reference values for a rural area and semi-volatile chemicals. The resulting fractions are better estimations regarding the measured values (Tab. 8.5). Analyses of the Junge equation by PANKOW (1997) and COTHAM AND BIDLEMAN (1995) show their general applicability within deviations of a factor of 2 (assuming representative values for c and s). Recently, K_{OA}-based approaches have been discussed (FINIZIO ET AL. 1997). LEE AND JONES (1999) compared the resulting concentrations for PCDD with measurements and found that the Junge equation produces overestimations with a factor ranging from 1.1 to 5. The K_{OA}-based equations result in lower deviations with a factor ranging from 1.2 to 1.5. It may be expected that experimental values for the K_{OA} will give even better results. Altogether, this approach produces the most realistic results.

Uncertainties arising from estimations of K_{OC} and TSCF: Partitioning between the water and soil matrix is estimated from the product of organic carbon content and the partition coefficient between octanol and organic carbon (K_{OC}). It is used by the plant model and by the bioconcentration model for aquatic systems for estimating concentrations in the liquid phase. The impact is the same as the concentration in pore water regarding the plant model. It is important for the upper plant part concentrations if translocation is a relevant process. Since several estimation functions are available for the K_{OC}, three often used K_{OW}-based regression equations were compared.

Especially for extremely hydrophilic and lipophilic chemicals, differences may be significant. For instance, K_{OC} for OCDD varies between 2.5E+6 and 6.5E+7. The TGD equation (proposed by SABLJIC ET AL. 1995) leads to mean values and shows the broadest regression range (Fig. 8.11). The range of applicability encompasses non-polar hydrophobic chemicals. The maximal deviation from measured values amounts to ± 0.5 for log K_{OW} < 4, otherwise ±1 order of magnitude.

Tab. 8.6 Overview of K_{OC} estimation functions for non-dissociating organic chemicals.

Equation	Range of applicability	Source
$0.411 \cdot K_{OW}$	n = 5, r^2 = 0.99, log K_{OW} from 1.0 to 6.72	KARICKHOFF (1981)
$1.26 \cdot K_{OW}^{0,1}$	n = 81 , r^2 = 0.89, log K_{OW} from 1.0 to 7.5	SABLJIC ET AL. (1995)
$10^{(0.72 \cdot \log KOW + 0.49)}$	n = 13 , r^2 = 0.95, log K_{OW} from 2.69 to 4.72	SCHWARZENBACH AND WESTALL (1981)

The TSCF is a sensitive parameter for chemicals that enter the plant via soil. Two estimation functions are available, of which that of the TGD is based on lesser lipophilic compounds and a larger training set. Estimations are higher and the most extreme value is for lower lipophilicity. For LAS (K_{OW} = 1.96), a high TSCF is calculated. By using the alternative estimation of HSU ET AL. (1990), the TSCF is only as half as high. However, impact is negligible regarding the model results depicted in Fig. 8.9.

Tab. 8.7 Overview of TSCF estimation functions for non-dissociating organic chemicals.

Equation	Range of applicability	Source
$0.784 \cdot \exp(-(\log K_{OW}-1.78)^2/2.44)$	n = 17 (Insecticides and herbicides in barley), log K_{OW} from -0.57 to 4.6	BRIGGS ET AL. (1982)
$0.700 \cdot \exp(-(\log K_{OW}-3.07)^2/2.78)$	n = 12 (Herbicides in soy beans), log K_{OW} from 0.96 to 5.3	HSU ET AL. (1990)

Discussion: For the chemicals investigated here the regression equations used are applicable in principle. Only higher chlorinated PCDD and PCB exceed the regression range, but their uptake via soil is negligible. The plant model rather shows underestimations in contrast to the previously investigated models, which tend to deliver rather high concentrations. Several reasons for this exist: Relevant transfer pathway may be neglected. Or the underlying database is not representative. For PCDD and PCB, homogenous concentrations are available and, hence, unconsidered processes are the most plausible reason for the observed underestimations. Further possible transfer pathways are (1) air-plant by dry and wet deposition, (2) soil-plant by volatilisation from soil into upper plant parts and (3) soil-plant by resuspension of particles from the soil and a subsequent adsorption.

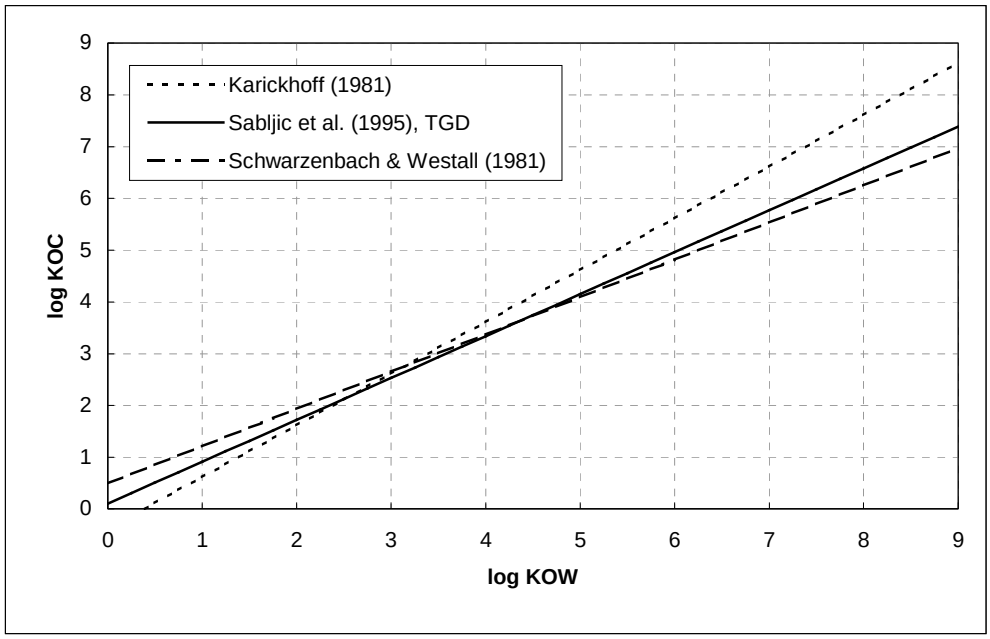


Fig. 8.11 Comparison of K_{OW}-based K_{OC} estimation functions.

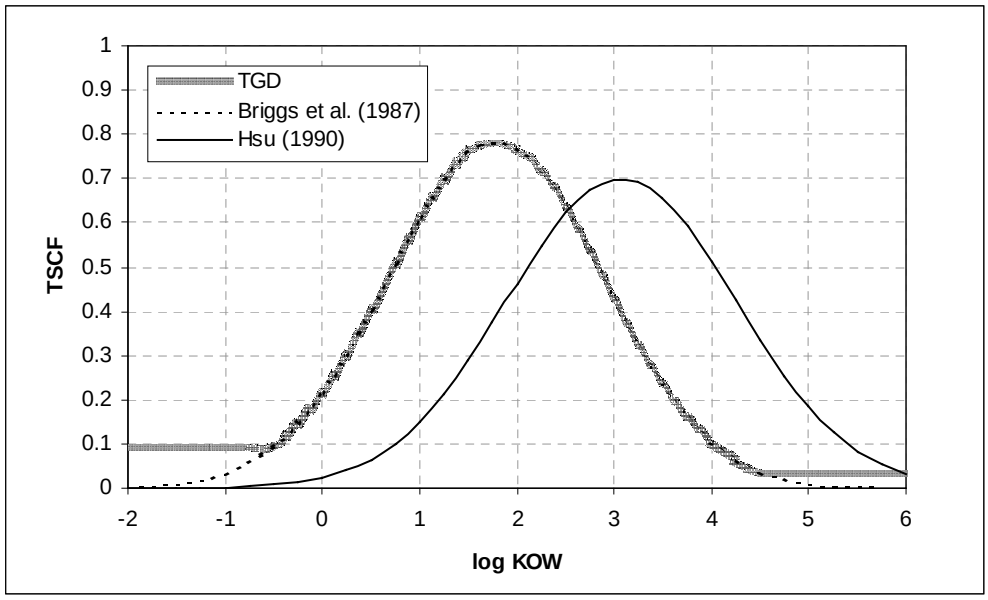


Fig. 8.12 Comparison of K_{OW}-based TSCF estimation functions.

Gaseous exchange with and deposition of particles are important pathways for PCDD uptake and uptake via soil is negligible (TRAPP ET AL. 1997, BALLSCHMITER 1996), which is confirmed by the application of soil-plant bioconcentration factors, leading to a high underestimation of measured concentrations (Section 10.3). The particulate fractions increase with an increasing degree of chlorination, which reveals a stronger impact of deposition for HpCDD and OCDD in relation to other congeners. Thus, a better result is expected when integrating deposition. The same phenomenon is assumed for the higher chlorinated PCB. Considering this process leads therefore to better estimations, but is not able to elucidate the deviations to measured values alone. TRAPP ET AL. (1997) found by means of an investigation of soil-plant transfer factors that volatilisation may play a role

for several substances and plants. However, this does not affect all kinds of plants, but only those located less than 5 cm above ground. Also adsorption of soil particles is deemed as relevant under certain conditions and was found to be the most important uptake pathway from soil for PCB and plants < 20 cm above ground. The relevancy of this pathway is particularly expected for lettuce (Fig. 8.8).

Investigations concerning the uptake of phthalates into plants from sludge-amended soils reveal that this transfer pathway is negligible (NRW 1993). This corresponds to the model results. Published DEHP concentrations are not suitable as background concentrations, since samples were taken in the proximity of emission sources. Thus, non-homogeneous concentrations are presumably the reason for the observed underestimation due to the low particulate fraction (calculated: 0.07) and the high sensitivity of the air concentration. A concluding statement on the validity of the model concerning this substance is not yet possible.

The presented LAS concentrations presumably also contain degradation products of LAS and not the parent substance alone. It can again be justified by the surface-active property and the degradation rate of this substance, and with the technique of measurement. Against this background and the apparently good match of measured and predicted concentrations, an overestimation of LAS concentrations by the model can be expected. The application of experimentally determined bioconcentration factors confirms this thought (Section 10.3).

The model assumes an environmental temperature of 12 °C. This values represents an upper limit of the yearly averaged temperature for NRW (1994). Monthly averaged values in NRW ranged from -2 °C to 23.5 °C for the year 1994. As previously shown, this parameter influences the estimation of the particulate fractions more than other parameters. Since plants grow in spring and summer, the temperature seems to be low. However, if a more realistic temperature for the summertime is entered, the particulate fractions will be even more overestimated. All in all, due to all the uncertainties when applying the Junge equation, it is important to consider representative values for particle surface area and the Junge constant for the regions, or to use measured fractions.

Field measurements regarding the uptake of benzene and 1,2-dichloroethane are unavailable. The findings of POLDER ET AL. (1996) are useful for evaluating the applicability of the model. They investigated the uptake of volatile organic compounds. It was found that, depending on the partition coefficient between leaves and air (K_{LA}), the model delivers good results for herbaceous plants. For a K_{LA} from 10^3 to 10^7 the error is within a factor of 5. The TGD model should be preferred to a simple bioconcentration model for substances with a larger K_{LA}, because the dilution effect caused by the plant growth becomes important. The error is larger for chemicals with high log K_{OW}, low water solubility and low vapour pressure. PCB (except PCB 180) show a K_{LA} between 10^3 and 10^7 for the chemicals investigated here. K_{LA} for benzene and EDC is less than 10^3. A larger error therefore has to be expected for all other substances.

It can be concluded that this simple generic TGD model yields an underestimation of measured concentrations for lipophilic chemicals. Deviations would be reduced significantly if the process of depositions of particles were integrated into the model. Furthermore, more realistic or, at best, measured particulate fractions are necessary.

8.4 Human exposure

8.4.1 Predicted doses

Four scenarios were elaborated in the context of the scenario analysis: Firstly, (if emission data were available) the total daily dose was calculated with the EUSES default values (scenario: s*tandard (default)*). Secondly, regional default values were replaced by more realistic and typical values for NRW (scenario: s*tandard* (*realistic*)). Thirdly, default intake rates were replaced by more realistic rates for an adult NRW inhabitant (man, 25-30 years old) due to the high sensitivity of the intake rates (scenario: *Adult* (*realistic*)). The intake rates of a child (boy, 7-8 years old) were finally used as a comparison (scenario: *Child* (*realistic*)). The results are presented together with available alternative estimations in Fig. 8.13. An appreciable reduction of the doses is obvious when avoiding EUSES default values. The three realistic scenarios deviate just slightly. For dioxins, the doses calculated using realistic parameters match the alternative estimations. But the calculations also correspond to the alternative estimations for volatile and other lipophilic compounds. The alternative dose for HHCB (FORD 1998) must be regarded as an upper limit, because uptake via the skin is considered. The dose, which is ingested via food, will be below this by orders of magnitudes.

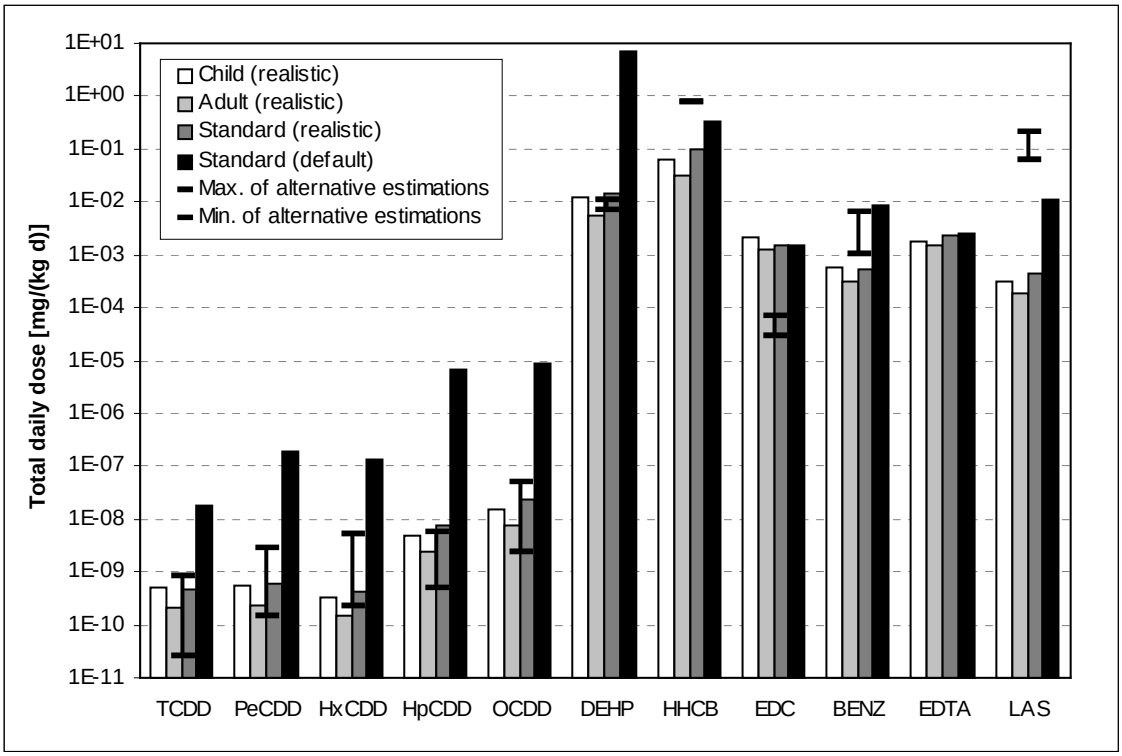


Fig. 8.13 Results of the scenario analyses of the total daily intake.

Altogether, deviations are within two log units for volatile and lipophilic substances. No alternative data are available for EDTA. LAS intake is underestimated by three orders of magnitude. Like HHCB, this value covers additional exposure pathways and it is also expected that the actual dose (ingested via the food chain) will be only orders of magnitude below the alternative estimations. More accurate data are lacking. The only slight deviations of the realistic scenarios do not exceed a

factor of five. However, the default intake rates rather correspond to those of a child and the use of realistic rates for adults results in the lowest calculated doses.

8.4.2 Contribution of the exposure pathways

The fraction of each individual exposure pathway can be estimated according to the formula derived in Section 7.1. As concluded from the sensitivity analysis, the fractions are equal to the sensitivities of intake rates and concentrations in the corresponding intake media. The fractions are listed in Tab. 2.2 for chemicals with available emission rates. If measured concentrations in the uptake media are used, the intake of dioxins is dominated by milk and meat pathways. Concentrations in plants also show a strong impact on the total dose for the higher chlorinated dioxins. HHCB is ingested via fish consumption and drinking water. Only air is relevant for 1,2-dichloroethane and benzene, whereas only drinking water is important for EDTA and LAS. PCB (not presented) show the same fractions as lower chlorinated dioxins. As already demonstrated in the sensitivity analysis, the exposure pathway root shows an unrealistic relevancy for the higher chlorinated dioxins. This outcome is misleading against the background of alternative (and more realistic) estimations (Appendix A.4, Tab. 36), because dioxins were found to be ingested equally via fish, meat and dairy products (each with one third, FÜRST (1995)). However, the calculated fractions for milk and meat are understandable. It will be discussed later in detail that an appreciable transfer of PCDD from soil into roots as well as the assumed high intake rates of roots is scientifically unjustifiable. For instance, FÜRST (1995) assumes a fruit and vegetable consumption of 0.38 kg (FW) per day. In contrast, the TGD default intake rate of plants amounts to 1.58 kg (FW). In addition, the alternative estimations show a more detailed differentiation of the various food products (diverse sorts of meat, eggs, bread, etc.). This fact causes further uncertainties.

Tab. 8.8 Contributions of the individual exposure pathways to the total dose (rounded). Both measured (1) and calculated (by EUSES) (2) concentrations in the intake media were used.

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	DEHP	HHCB	EDC	BENZ	EDTA	LAS
(1) Measured concentrations	Plant roots											
	Fish	0.9	0.8	0.1				0.7				
	Drinking water							0.3			1.0	1.0
	Air								1.0	1.0		
	Plant leaves			0.1	0.5	0.7	0.9					
	Meat		0.2	0.7	0.5	0.2						
	Milk											

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	DEHP	HHCB	EDC	BENZ	EDTA	LAS
(2) Calculated concentrations	Plant roots		0.3	0.8	0.9	0.9		0.9				
	Fish	0.1	0.1	0.1	0.1		0.3	0.1	0.1		0.1	0.2
	Drinking water								0.1		0.9	0.3
	Air								0.8	0.9		
	Plant leaves	0.3	0.2				0.2					0.5
	Meat	0.4	0.3	0.1			0.3					
	Milk	0.2	0.2				0.2					

It is generally assumed that intake of dioxins occurs by more than 90% via food (FÜRST 1995). On the other hand, direct intake via air, drinking water or soil is normally negligible with a contribution of less than 10%. Food originating from plants reveals only very low concentrations. Thus, the contribution of plants to the total daily PCDD intake is also low. In fact, the TGD model also estimates 90% as the contribution of indirectly ingested food, but similar fractions for fish, meat, and milk are not obtained. In particular, the contribution of the milk pathway is strongly underestimated. A separation according to “1/3 milk, 1/3 meat, and 1/3 fish” is unattainable (using realistic concentrations) without a more detailed consideration of intake rates. Altogether, the calculated fractions are rather suitable for finding out the sensitive intake media. According to the calculations, 1,2-dichloroethane and benzene are ingested by 99% from the air. In actual fact, the contribution of other exposure pathways may play an appreciable role: BUA (1993) estimated the intake of benzene via food to range between 10% and 40%. What is more, smoking 20 cigarettes per day will increase benzene intake by at least 50%.

8.5 Concluding evaluation

The investigations reveals that the TGD model will overestimate the actual exposure if the default intake rates are applied. The deviations amount typically to 1 to 3 orders of magnitude. This had to be expected against the background of the underlying assumptions. Although the calculated dose is conservative, several highly exposed groups (workers, babies, smokers, etc.) will actually show higher doses. By replacing the default intake rates by more realistic ones, the calculated doses fall exactly in the range of alternative estimations (with deviations within one order of magnitude of these results). However, these good results cannot be justified by the results from inspecting the model theory: The calculated contributions of the exposure pathways are only meaningful for *directly* ingested chemicals. The application of measured concentrations in the intake media confirms this conclusion. The calculated contributions are misleading for all other chemicals. This latter conclusion holds for diverse concentrations in the intake media and is independent of whether the concentrations are calculated or measured. The reason is not so much a non-representative concentration or a problem of the model, it is more the consequence of insufficiently detailed differentiation of the intake rates. Due to the high sensitivities of concentrations and intake rates, one obtains unrealistic results. A further problem arises from the interaction of overestimation and underestimation: The deviations in estimated concentrations may equalise and, thus, pretend to give an apparently good result for the total daily dose. After dwelling upon all problems and limitations mentioned before, the applicability of the human exposure model seems to be debatable. Nevertheless, the model produces good results for PCDD. But the apparently good result is based on overestimations for meat and milk concentrations on the one hand and on underestimations for the fish concentration on the other hand. These deviations balance each other out.

8.6 Summary

By using different scenarios, several model calculations were carried out and the results were compared with monitoring data and experimentally determined values. It was shown that the concentrations are overestimated by up to two orders of magnitude for the aquatic environment. For superlipophilic (log K_{OW} > 6) and persistent chemicals, higher uncertainties emerge and measured concentrations may also be underestimated. The deviations are caused by unrealistic bioconcentration factors or metabolism on the on hand and by neglecting biomagnification on the other hand. The degree of deviation also depends on the fat content of the fish species. The biotransfer model for meat and milk represents a conservative estimation. The overestimation is most significant for non-persistent or superlipophilic substances, with more than two orders of magnitude. A lack of steady state, metabolism and/or reduced resorption were presumed to be the reasons. The models deliver good results (with deviations within one order of magnitude) for persistent PCB. The model for describing uptake by plants was chiefly validated by grass concentrations and revealed deviations of less than one log unit for the higher chlorinated PCB. Stronger deviations must be expected for dioxins. This model often leads to an underestimation of the measured concentrations. Considerable uncertainties are caused by estimating the particulate fraction. The integrated estimation function results in quite high fractions. In addition, the model considers chemical uptake from air only via gas exchange. Integrating the process of particle deposition would ameliorate the results. The calculated total daily dose was compared with alternative estimations available from the literature. Accordingly, the calculated dose is usually more than that of alternative estimations. For several chemicals rates it corresponds with deviations within two orders of magnitude (for chemicals without a lack of data) when applying more realistic intake values. It was found that low deviations are sometimes caused by an equalising effect of overestimations and underestimations in the submodels.

9 Probabilistic uncertainty analyses

This chapter shows the results of the probabilistic assessment of the total daily dose. The probabilistic analyses reveal both cumulative distribution functions of the total daily dose and the contribution of the input parameters to the result's variance. First of all, the contribution of each parameter is discussed. This is helpful to understand the cumulative distribution functions discussed in turn below.

9.1 Uncertainty impact analyses of individual parameters

A ranked correlation was developed in order to determine the contribution of each parameter to the variance of the final result of the overall system (i.e. the total daily dose). Depending on the substance, between 5 and 17 out of 71 uncertain parameters are considered to be important due to correlation coefficients with an absolute value of more than 0.01. According to Tab. 9.1, the parameters of the exposure module are highly correlated for the lipophilic substances, particularly b (the correction coefficient between plant lipid and octanol). For the group of substance-specific parameters the emission rates play a role. For the lower chlorinated PCDD and DEHP the physico-chemical properties are also important. With the exception of the air compartment height, the parameters of the regional distribution model are not important. But they have a dominating relevancy for the rather hydrophilic compounds. In particular, the volumetric parameters (height and area of a compartment) are highly correlated. In contrast to the lipophilic substances, the rather hydrophilic ones show important parameters in each group. For example, the three most important parameters for LAS are the intake rate of drinking water (exposure module), the depth of the water compartment (distribution model) and emissions into waste water (substance-specific data). These results can be explained by the combination of sensitivities and uncertainties of individual parameters. A comparison with the sensitivity analyses (the methodology of which corresponds to an uncertainty analysis with equable uniform-distributed parameters) shows a similar ranking of many important parameters. However, exceptions do exist: The physico-chemical properties of HHCB show a high sensitivity, although their contribution to the dose is negligible. This results from high and relatively uncertain emissions into waste water. Based on both analyses, the physico-chemical properties show a low impact. They only play a role for the lower chlorinated dioxins and DEHP. This holds in general for all substances that partly occur in the gaseous phase and are partly bounded to particles. Changing physico-chemical properties yields deviating particulate fractions and, thus, a different mobility in air. The body weight, with its constantly high sensitivity, plays a minor role for lipophilic compounds, but is influential for all others. The reason for this is the always sensitive and, at the same time, uncertain parameters of the plant model. The parameters of the regional distribution model show the most marked difference between uncertainty and sensitivity analyses for the lipophilic substances: While sensitivity analyses reveal many important parameters of the distribution model, only some parameters of the soil compartment and the air compartment height are important against the background of uncertainty analyses. The vanishing impact of certain parameters is again explained by the combination of the high sensitivity and high uncertainty of other pa-

rameters. In other words: The coefficients of variation are low in relation to other important parameters, e.g. the emission rates. The parameters of the regional distribution model show a similar impact for the rather hydrophobic compounds in both analyses.

The aggregation of all parameters into three groups (parameters of the exposure module, of the regional distribution model and of the substance) and the presentation of the impact of these groups on the total dose is discussed in further detail in the following sections.

Tab. 9.1 Results of the ranked correlation between the total daily dose ($DOSE_{total}$) and each sensitive input parameter. Only coefficients with more than |0.01| are shown. The most influential parameter is presented in bold face for each substance.

		lipophilic							(rather) hydrophilic			
	Parameter	**TCDD**	**PeCDD**	**HxCDD**	**HpCDD**	**OCDD**	**DEHP**	**HHCB**	**EDC**	**BENZ**	**EDTA**	**LAS**
Exposure module parameters	A / V	0.11	0.16	0.18	0.22	0.17	0.25					-0.11
	b	**0.51**	**0.58**	**0.70**	**0.62**	**0.74**	0.36	**0.79**				
	BIOinh								0.33	0.11		
	BW						-0.11		-0.26	-0.22	-0.21	-0.15
	Flipid plant	0.22	0.18	0.21	0.17	0.20	0.14	0.41				
	g plant	0.11	0.28	0.20	0.27	0.20	0.32					
	IC grass											
	IH air								0.40	0.40		
	IH drwwater								0.14		0.51	0.27
	kgrowth plant	-0.11	-0.14	-0.17	-0.18	-0.15	-0.24					-0.14
	RHO plant	-0.11	-0.11	-0.11	-0.14		-0.15					
Regional distribution model parameters	DepthAgric			-0.11	-0.15	-0.14	-0.10					-0.20
	DepthWater Reg											**-0.54**
	FconnectSTP											-0.11
	FocSoil				-0.11	-0.11						-0.13
	FrunoffSoil							-0.11			**-0.57**	
	FsolidSoil					-0.11						-0.14
	fWater Reg								-0.12			-0.19
	HeightAir	-0.20	-0.17	-0.21	-0.17	-0.16	-0.20		**-0.56**	**-0.61**		
	Rainrate										-0.22	
	SurfAer						-0.14					
	Windspeed		-0.09		-0.11				-0.24	-0.21		
Substance-specific parameters	EcontAir								0.23	0.47		
	Econtfirstwastewater										0.13	
	EcontInd								0.01	0.01	-0.01	0.01
	EregAir	0.14	0.15	0.17	0.20	0.14	0.26		0.27	0.17		
	Eregfirstwastewater							0.26	0.21		0.50	0.47
	Eregfirstwater							-0.01	0.01	0.06	0.01	0.01
	EregInd								0.01	0.04	-0.02	0.05
	kdegair									0.11		0.01
	kdegsoil											0.14
	kdegwater											0.24
	KOW	0.49	0.20	0.15			-0.25					
	SOL	0.18	0.03				0.14					
	VP	-0.30	-0.21	-0.17			**0.38**					

Negligible: area Reg, BOD, CollEffAer, DepRateAer, depthInd, depthNat, depthSed, Econtfirstwater, Erosion, fAgric Reg, fInd Reg, FinfSoil, fNatural Reg, FocSed, FocSusp, Fwater plant, FwaterSed, FwaterSoil, IC air, IC drw, IC soil, IH fish, IH leaf, IH meat, IH milk, IH root, kasl air, kasl soilair, kasl soilwater, kdegsed, kwsSed, kwsWater, Qstp, Qtransp, RhoSolid, SETTLEvelocity, SuspEff, SuspWater Reg, Temperature

9.2 Cumulative distribution functions of the total daily dose

With the intention of quantifying uncertainties and determining which part of the overall system contributes most to the overall uncertainty, an iterative uncertainty analysis of the total daily dose was carried out for each chemical. Iterative means that the set of varied parameters is subsequently reduced (Tab. 9.2): The first simulation considers both model and substance parameters, whereas the second and third simulations only focus on the model parameters. On the one hand, the model parameters of the overall system, and on the other only the model parameters of the exposure module were considered. In simulation IV only intake rates were varied. In simulation V the estimated concentrations in the intake media were replaced by measured ones and, besides alternative estimations, are a means to evaluate the results. Only simulations IV and V represent variability. Due to the limited database, all other simulations represent a mixture of variability and true uncertainty.

Tab. 9.2 Survey of the five simulations carried out for each chemical.

No.	Varied parameters	Type of uncertainty
I	All parameters of the overall system (i.e. all model and substance parameters)	True uncertainty and variability
II	All model parameters	True uncertainty and variability
III	Model parameters of the exposure module	True uncertainty and variability
IV	Intake rates	Variability
V	Intake rates (but combined with measured concentrations in the intake media)	Variability

The shapes of the distribution functions visualise the preceding results. For example, the distributions for TCDD (Fig. 9.1) are similar for the variation of all model parameters or exposure module parameters. This points out the low impact of the regional distribution model, since ignoring uncertainties in its input parameters hardly influence the resulting distribution function. Deviating distributions are obtained when varying all parameters or just the intake rates. This demonstrates that the substance-specific parameters and exposure module parameters, respectively, cause most of the uncertainty. This finding corresponds to the values of Tab. 9.1 since the plant parameters b and FlipidPlant are the most important ones. These parameters predominantly cause the shape of the distribution. The largest range between two percentiles arise from varying all parameters (thick black line). Varying only the intake rates (thin grey line) spans the smallest range. The other distributions lie in between (thick grey lines). This is understandable because the more uncertain the parameters are, the more uncertain the results will be. For DEHP (Fig. 9.6), the distribution for variable intake rates is identical to the distribution for uncertain parameters of the exposure module. Consequently, the light grey line represents variability, which cannot be reduced by more accurate parameter values. The main source of uncertainty are the parameters of the substance and of the regional distribution model. The negligible impact of the exposure module in relation to other lipophilic chemicals is explained by the relatively low impact of b, which results from the high K_{OA} of this substance. Also for HHCB (Fig. 9.7), uncertainty predominately originates from the exposure module.

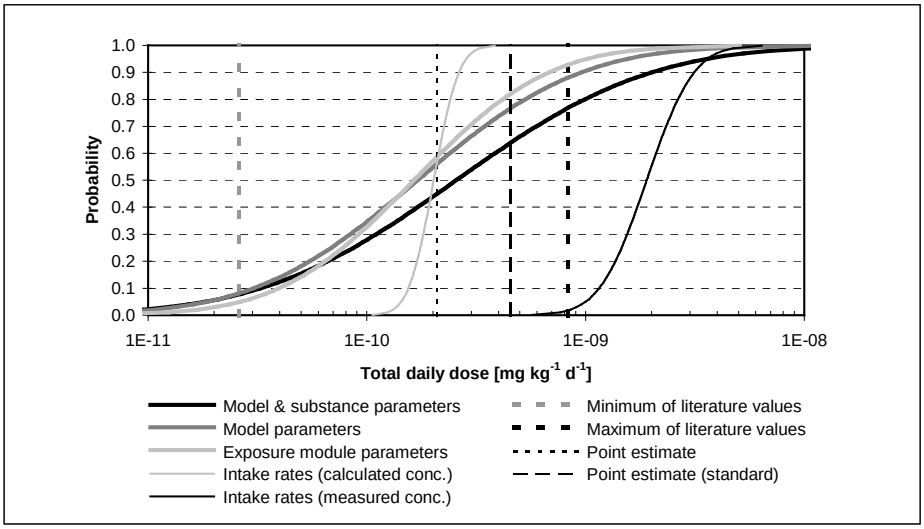


Fig. 9.1 Cumulative distribution of the total daily dose for TCDD.

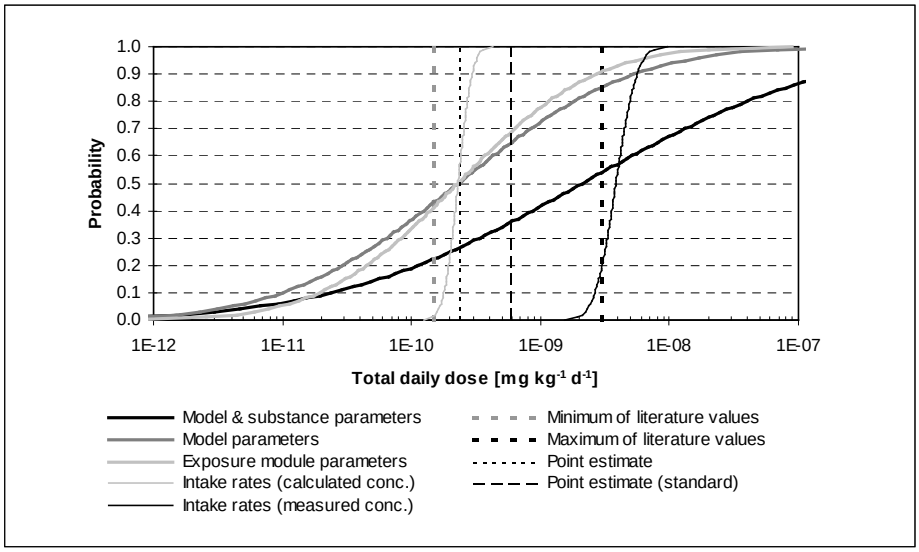


Fig. 9.2 Cumulative distribution of the total daily dose for PeCDD.

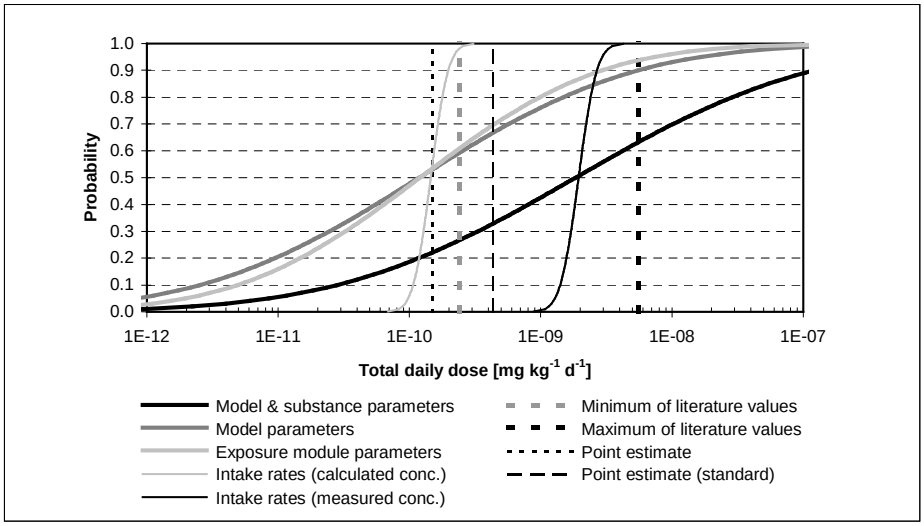


Fig. 9.3 Cumulative distribution of the total daily dose for HxCDD.

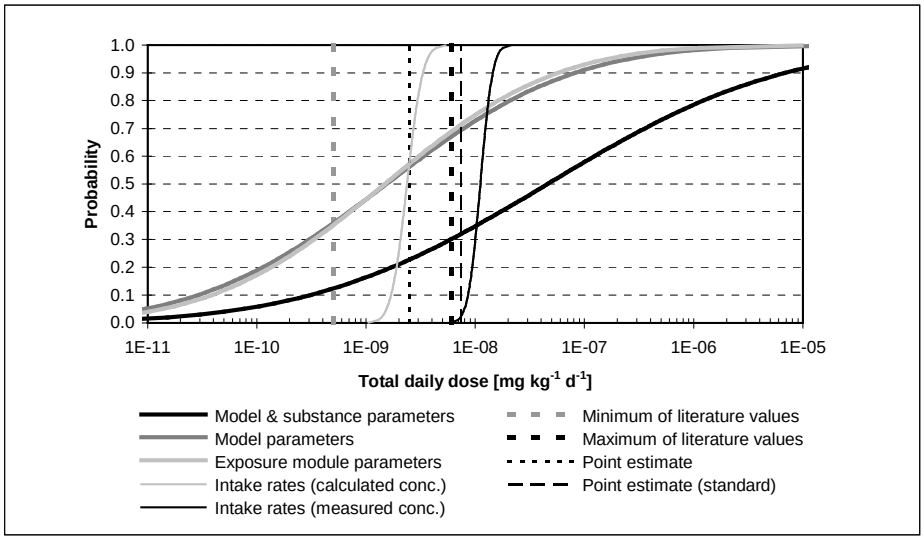


Fig. 9.4 Cumulative distribution of the total daily dose for HpCDD.

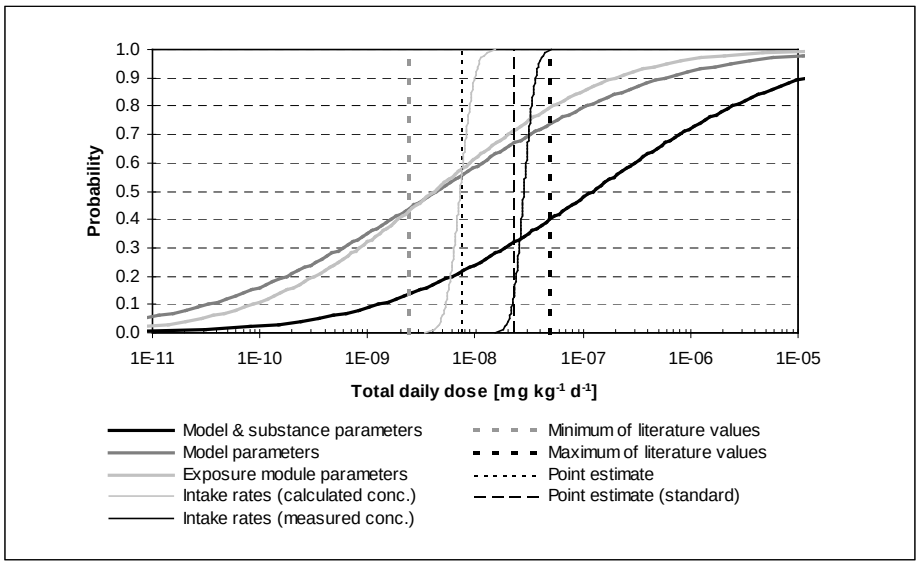


Fig. 9.5 Cumulative distribution of the total daily dose for OCDD.

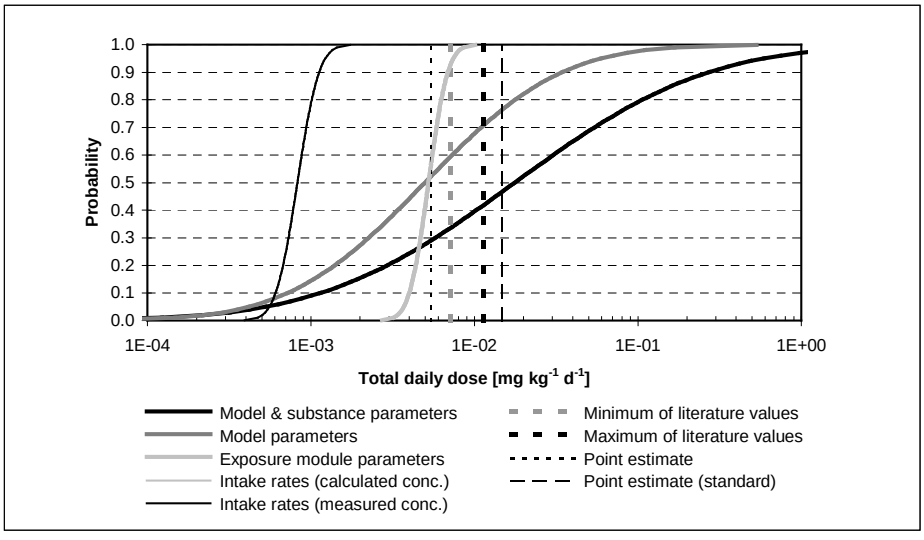


Fig. 9.6 Cumulative distribution of the total daily dose for DEHP.

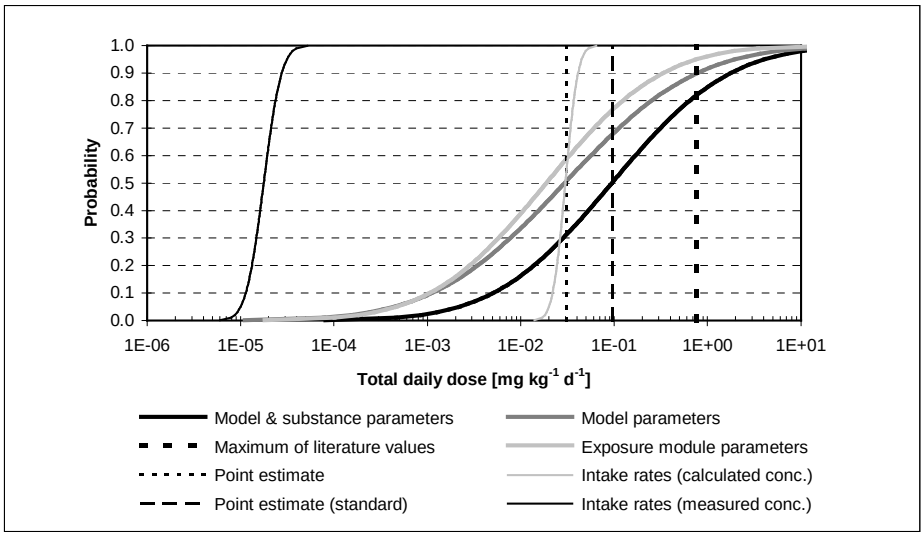


Fig. 9.7 Cumulative distribution of the total daily dose for HHCB.

1,2-Dichloroethane and benzene (Fig. 9.8, Fig. 9.9) are ingested exclusively via air. The thin black line draws an exact picture of the actual variability since the model covers this exposure pathway and the type of uncertainty is variability. The intake rates' variability is also the cause of the shapes for all other calculated curves. This implies that the uncertainties cannot be further reduced. Comparable statements are possible for EDTA and LAS (Fig. 9.10, Fig. 9.11), even if uncertainty is more in the individual parts of the system. Thus, the variability of the intake rates dominates uncertainty in the result for directly (i.e. via air or drinking water) ingested chemicals. A better database or more effort in the construction of probability distributions are not able to significantly reduce uncertainty in the result. On the contrary, both reducible uncertainty and variability cause the resulting uncertainty for indirectly (i.e. via multiple pathways) ingested chemicals. Because of the usually poor database, it may be assumed that the major source of uncertainty is caused by true (i.e. reducible) uncertainty. In this case, a more accurate quantification of distribution functions for the parameters will lead to a more certain result. Altogether, dioxins and other lipophilic compounds show the lowest kurtosis[3], i.e. the confidence intervals are the largest for these chemicals.

[3] Kurtosis refers to the peakedness of a distribution. A low kurtosis represents a "flat" distribution.

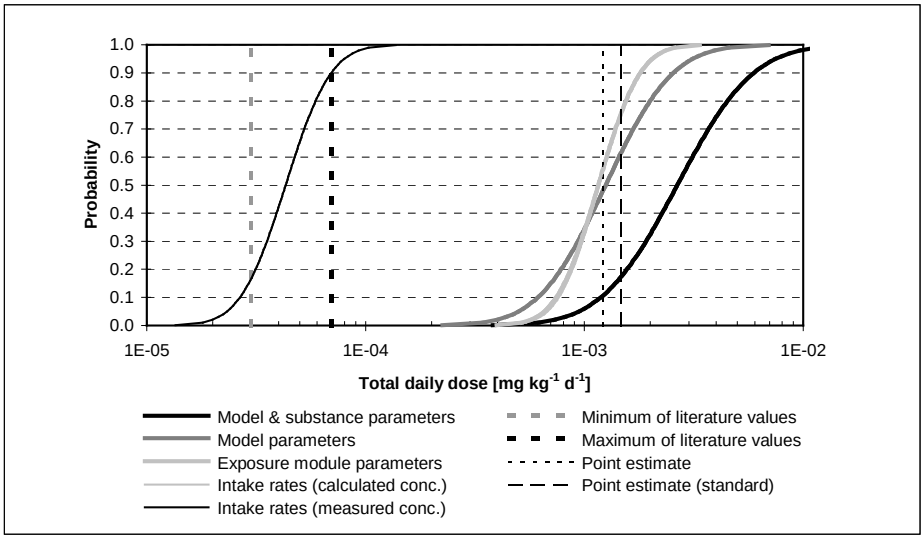


Fig. 9.8 Cumulative distribution of the total daily dose for 1,2-Dichloroethane (EDC).

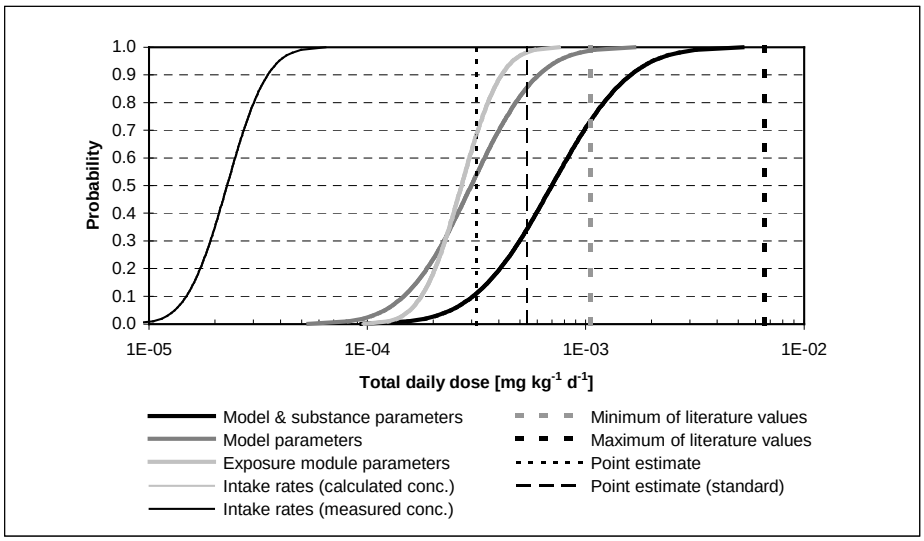


Fig. 9.9 Cumulative distribution of the total daily dose for benzene (BENZ).

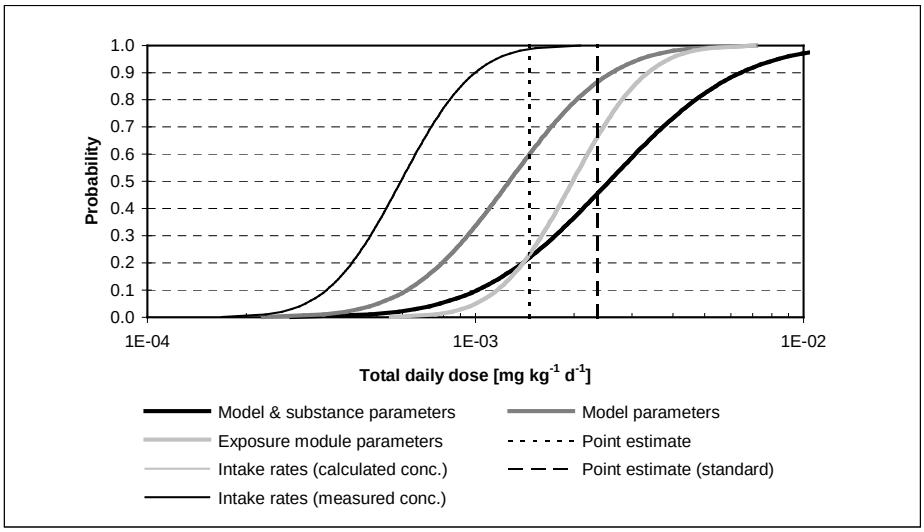


Fig. 9.10 Cumulative distribution of the total daily dose for EDTA.

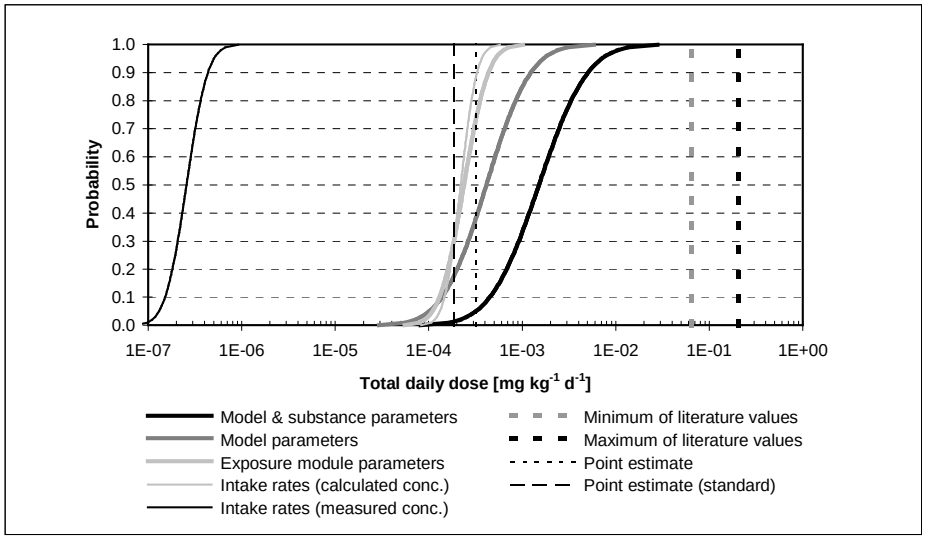


Fig. 9.11 Cumulative distribution of the total daily dose for LAS.

The higher resulting concentrations for all substances are obvious when assuming both substance and model parameters are uncertain. The reason is the triangular distributions used for the sensitive emissions. The central values of the triangular distribution are the constant values of the scenario analysis. Thus, the mean of the distribution is more than the central value, due to the negative skewness. This fact causes a shift of the whole resulting distribution towards higher concentrations. The mean of K_{OW}-literature values also exceeds the constant K_{OW} of the realistic scenarios and the mentioned effect is therefore excessive for the lipophilic compounds. Thus, the location of the probability function itself is uncertain due to the database. In order to interpret the results it must be regarded that, especially for emission and degradation rates, the location of the distribution is not absolutely realistic, but rather describes a possible and plausible range. Conclusions regarding the impact of various parameters or groups of parameters will therefore be more reliable than conclusions derived from a comparison of calculated percentiles with measured data.

The variation of preferably a large number of parameters (e.g. instead of substance-specific parameters alone) is sensible since only the variation of many parameters results in log-normal distributions (Chapter 3). The goodness of fit was checked for the resulting distributions according to the Kolmogorov-Smirnov test (Tab. 9.3). A test value less than 0.03 represents a good fit (DECISIONEERING 1999). The calculated distributions are similar to log-normal distributions for all chemicals with low uncertainties in the result. All chemicals with highly uncertain results and with distributions of low kurtosis do not correspond to log-normal distributions. The fact that the correspondence is low for simulations in which many parameters have been varied can be explained with several important input parameters that are not log-normally distributed and not multiplicatively connected. All in all, for chemicals with highly uncertain results also the type of resulting distribution is uncertain. In contrast to numerous published exposure assessments (Section 3.3.2), a log-normally distributed dose may not really be expected.

Tab. 9.3 Results of the goodness-of-fit test (according to Kolmogorov-Smirnov).

Substance	Simulation I	Simulation II	Simulation III	Simulation IV
TCDD	< 0.03	< 0.03	< 0.03	< 0.03
PeCDD	< 0.03	0.065	0.054	< 0.03
HxCDD	0.052	0.088	0.079	< 0.03
HpCDD	0.099	0.080	0.073	< 0.03
OCDD	0.113	0.075	0.068	< 0.03
DEHP	0.033	< 0.03	< 0.03	< 0.03
HHCB	0.054	0.060	0.053	< 0.03
EDC	< 0.03	< 0.03	< 0.03	< 0.03
BENZ	< 0.03	< 0.03	< 0.03	< 0.03
EDTA	< 0.03	< 0.03	< 0.03	< 0.03
LAS	< 0.03	< 0.03	< 0.03	< 0.03

The kurtosis of the result represents uncertainty and can clearly be arranged as the difference of 90%-ile and 10%-ile (Fig. 9.12). Accordingly, the difference amounts up to 4½ orders of magnitude for the lipophilic compounds. The variability in the intake rates is negligible in this case. Another situation arises for highly water-soluble or volatile substances, where the variability of the intake rates is important and the difference between the percentiles does not exceed one log unit. The lower uncertainties for OCDD in relation to HpCDD is elucidated with a better database and, hence, lower uncertainties in the input parameters for OCDD.

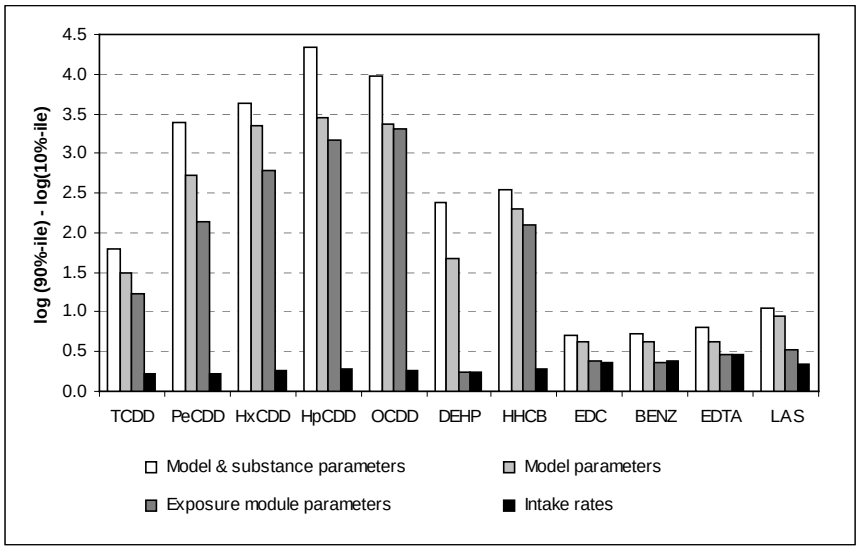


Fig. 9.12 Range of uncertainties of the total daily dose, expressed as log(90%-ile/10%-ile).

The results correspond to those of similar investigations. McKone (1994) emphasised the higher uncertainties in the estimated dose for indirectly ingested chemicals in contrast to directly ingested ones. McKone and Ryan (1989) stressed that much of the overall uncertainty in exposure is attributable to uncertainty in biotransfer factors and that uncertainties in the input data limit the precision of predictions to a 90% confidence range of roughly 2 orders of magnitude.

9.2.1 Comparison with point estimates

The cumulative distribution functions obtained from the variation of all parameters (thick black line) were compared to the point estimates. On the one hand, TGD standard intake rates were used and on the other hand, more realistic age-specific rates were employed. The latter deliver up to half a log unit lower concentrations (see scenario analysis). According to this investigation, the results of the point estimates correspond to the lower percentiles. The point estimate exceeds the median only for TCDD. Contrary to many published papers (Chapter 3), EUSES does not produce a cumulative worst case when using point estimates. The deterministic estimations tend rather to underestimate, with reference to the range of possible outcomes. This statement is caused by the distribution functions of the substance-specific data. In particular, the variation of the K_{OW} increases the negative skewness of the resulting probability distributions. Concurrently, the medians are shifted towards higher concentrations due to sensitive and uncertain emission rates. If only model parameters are assumed to be uncertain, all point estimates will be more than the median of the probabilistic estimates and, hence, show a conservative character (but also in this case never a cumulative worst case). All derived statements are valid against the background of the chosen probability distributions for the input parameters. The chosen triangular distributions for the input parameters are themselves uncertain and are only able to demonstrate a possible impact of uncertainties. It can be concluded that the EUSES calculations do not lead to a cumulative worst case, even if a *reasonable worst case* is used for the input parameters. The reason for this is the sensitive emission, which means that an accumulation of worst-case assumptions does not prevail.

9.2.2 Comparison with alternative assessments

For DEHP, EDTA, LAS or HHCB an evaluation of the calculated percentiles was not possible because none or no reliable exposure data are available in the scientific literature. Detailed information on human exposure exist for dioxins, EDC and benzene (Appendix A.4, Tab. 36). For dioxins, the calculated percentiles derived from varying all parameters cover the range of alternative doses reported in the literature (thick dashed lines). However, they rather correspond to the lower and central percentiles. The alternative estimations only correspond to the upper percentiles for benzene – which can be explained by the conservative character of the literature values, which consider additional sources of exposure such as smoking or traffic. Thus, benzene shows that EUSES does not necessarily deliver realistic doses, although realistic input data are used. Air as the solely relevant exposure pathway is in fact considered, but insufficiently detailed. The same applies to LAS, where all calculated percentiles are significantly lower than the alternative estimations. Higher percentiles than the values given in the literature were only obtained for EDC.

A generally valid and far-reaching conclusion can be drawn: The range of uncertainty in the model calculations (expressed by the difference of 90%-ile and 10%-ile) exceeds the range spanned by the minimum and maximum of alternative estimations. For chemicals with point estimations that correspond well to the alternative estimations this means that by an appropriate (and scientifically justifiable!) parameter selection, alternative estimations may be both overestimated and underestimated.

9.2.3 Impact of ignoring correlations

Various correlations regarding the physiological parameters are taken into account (Section 3.4.5). However, correlations between physico-chemical parameters were ignored due to a lack of data. If, e.g. a correlation between K_{OW} and water solubility were considered, the kurtosis of the resulting distribution would be lower (i.e. lower resulting uncertainties). Neglecting correlations in the context of this study is therefore arguable because it represents an alternative approach. Also for other possible correlations it was checked whether the deviations were small and whether their neglect does not lead to differing qualitative results.

9.2.4 Impact of unknown degradation rates

Input parameters required for the plant model are the degradation rates. However, appropriate data are usually unavailable. Thus, no degradation was assumed. The total elimination rate in plants is the sum of photodegradation and metabolism rates of the substance and the growth rate of the plant. It follows that equal degradation rates have equal sensitivities. According to the sensitivity analysis, the growth rate is sensitive for some lipophilic compounds. If substance-specific degradation were of the same value, these rates would also be sensitive. Only McCrady and Maggard (1993) and Schuler et al. (1998) reported PCDD-degradation rates of 0.84 [d^{-1}] for TCDD to 3.8 [d^{-1}] for OCDD. However, unpublished investigations suggest a significantly lower degradation in the real environment and indicate that quantification is not yet scientifically justifiable (Klasmeier 2000). Consequently, the published rates should at best be viewed as upper limits. A further uncertainty analysis for TCDD with a uniform distributed photodegradation rate ranging from 0 to 1 [d^{-1}] was carried out. This analysis showed no significantly different impact from this rate, since the total elimination rate is dominated by the distribution of the growth rate. Finally, it can be concluded that the substance-specific degradation rates in plants are negligible for all of the substances investigated here because the values are (1) neither sensitive nor sufficiently uncertain or (2) the exposure pathway plant is negligible.

9.2.5 Impact of other age-specific intake rates

The relevancy of realistic intake rates was already discussed in the context of the scenario analyses. The results of the probabilistic analyses provide some additional information: The deviations between the calculated dose for children and adults indicate that deviations between these age-specific intake rates are up to half a log unit for percentiles between 10 and 90 (Fig. 9.13). The negative value (i.e. a lower calculated dose) for EDTA is striking. This originates from the distribution for drinking water intake, the values of which are lower for children than for adults. Consequently, for children the resulting percentiles are in most cases considerably higher than for adults. But also lower doses for children may occur if only one exposure pathway is dominating. The deviations amount to between 0.3 and 0.5 log units for indirectly ingested chemicals and between 0 to 0.3 log units for all other chemicals.

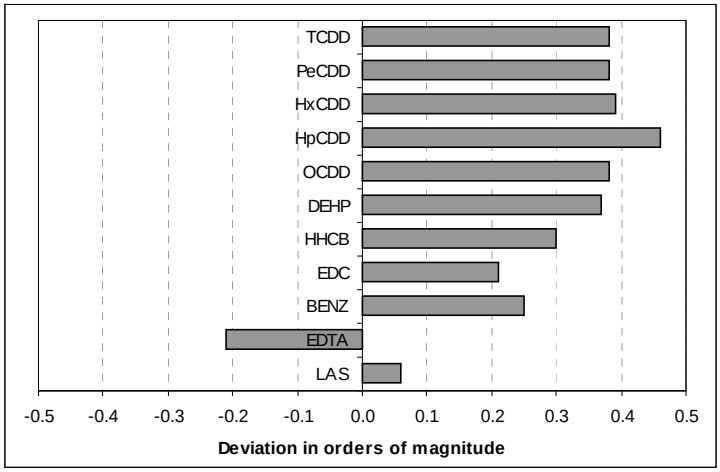


Fig. 9.13 Maximal deviation between two percentiles using intake rates for children (boy, 7-9 years old) instead of adults.

9.3 Uncertainty impact analyses of parameter groups

When summarising the results of the last two sections, the question arises as to which group of parameters mainly contributes to the uncertainty of the total daily dose. All of the uncertain parameters from Tab. 1.7 were grouped into substance-specific data, parameters of the regional distribution model and exposure parameters (e.g. breathing rates or plant properties). Their contribution to the total daily dose's variance is shown in Fig. 9.14. The diagram reveals that for lipophilic substances (TCDD – HHCB) the exposure module parameters show the highest contribution (often more than 50%). Changing the vapour pressure influences the particulate fraction, particularly for TCDD and DEHP, and the impact of substance-specific parameters is therefore also significant. For the lipophilic compounds the contribution of regional distribution model parameters is low. For chemicals that tend to distribute in one compartment (i.e. EDC and BENZ into air, EDTA and LAS into water), the contributions of the three groups of parameters are nearly balanced, but slightly dominated by those parameters of the regional distribution model.

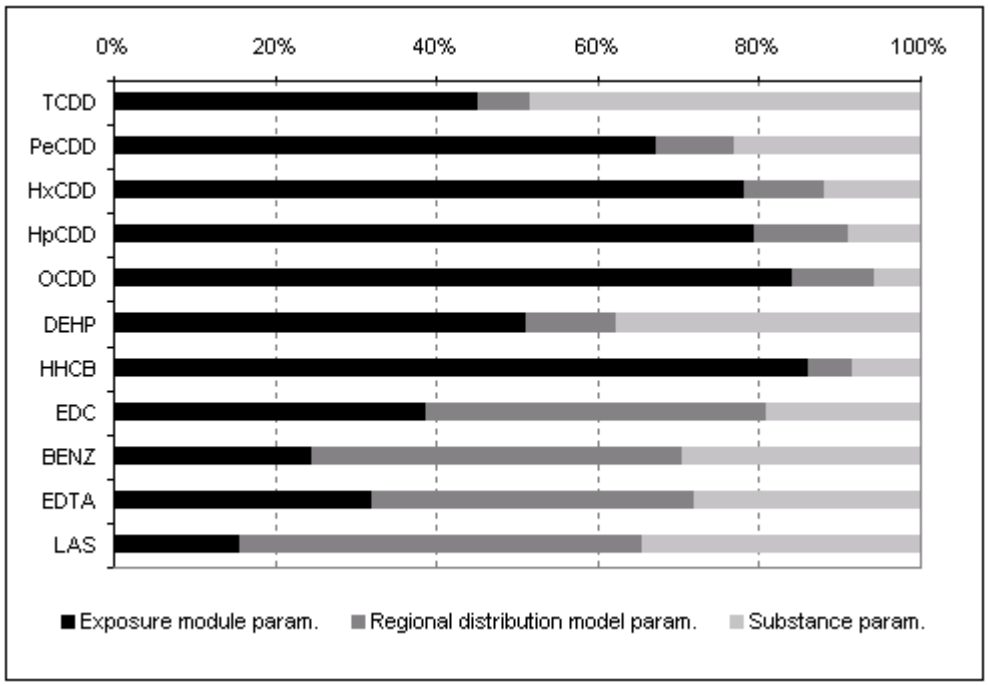
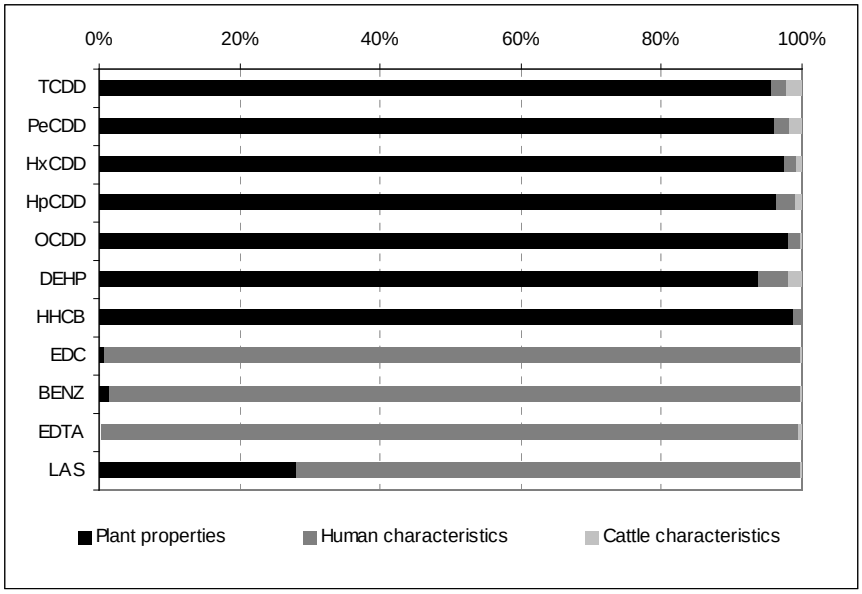


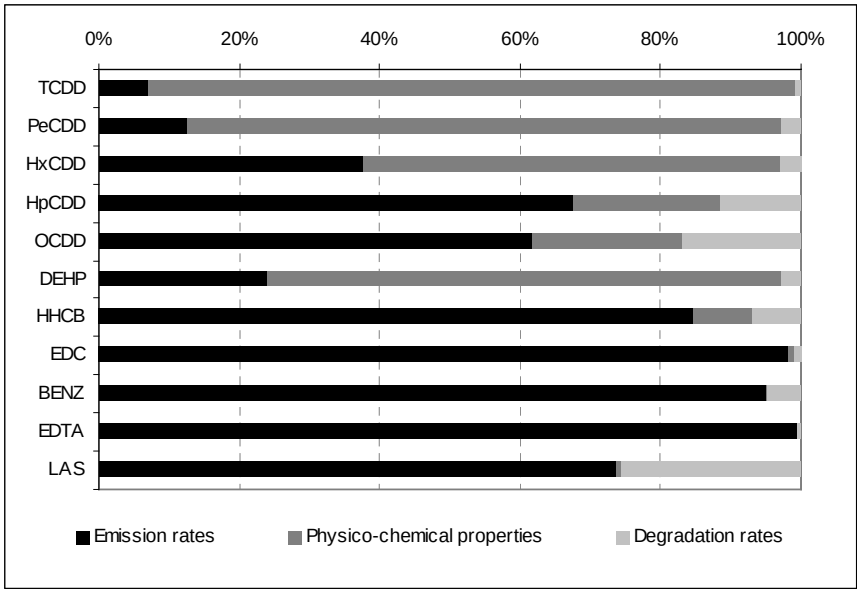
Fig. 9.14 Contribution of the three parameter groups to the variance of the total daily dose.

A refined view (Fig. 9.15) shows that the only relevant parameters of the exposure module are either the properties of the plant, in the case of lipophilic substances, or the human characteristics (i.e. body weight and intake rates) in the case of less lipophilic or lipophobic substances. The group of substance parameters is dominated by emission rates. Except for certain lipophilic chemicals (DEHP and lower chlorinated dioxins) physico-chemical properties will play a role if they show a significant impact on the mobility of the chemicals in the air. The contribution of degradation rates is low since the combination of uncertainty and sensitivity of other parameters prevails. The most important parameters of the group of regional distribution model parameters are the volumetric parameters of the compartments. Except for EDTA, only landscape characteristics (e.g. temperature, precipitation rate, etc.) are important. Regional human characteristics (i.e. the waste-water production rate and the number of inhabitants connected to sewage treatment plants) are insignificant due to their low uncertainty.

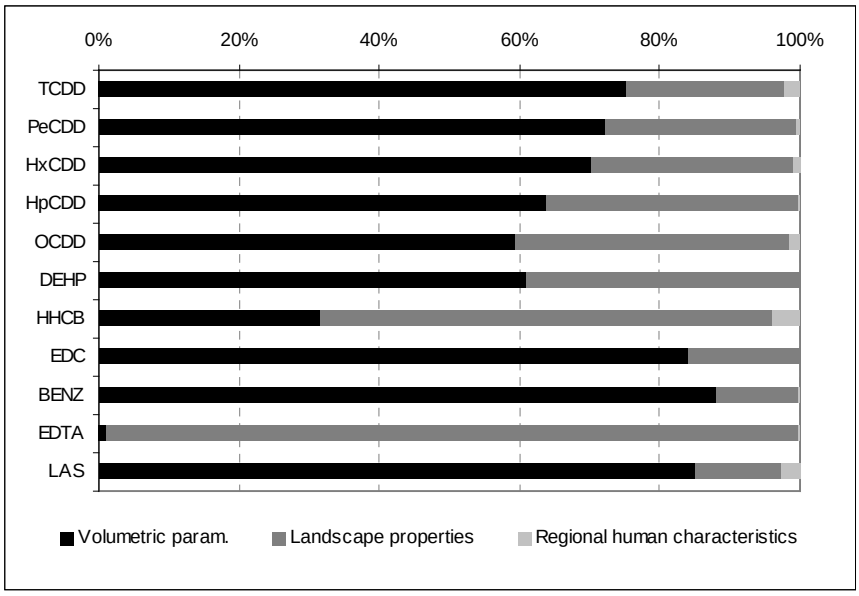
Two major aspects can be concluded from these findings: Firstly, the overall system strongly depends on the lipophilicity of the substance. Thus, in order to state the most important parameters, a first classification should be made for lipophilic and less lipophilic chemicals. In other words: chemicals ingested via the food chain on the one hand and chemicals predominantly ingested via the air or drinking water on the other. Secondly, for these two groups of chemicals only a few parameters contribute significantly to the total daily dose.



(a) Parameters from the exposure module.



(b) Substance-specific parameters.



(c) Parameters from the regional distribution model.

Fig. 9.15 Refined view of the contribution of the parameter groups to the variance of the total dose.

These results also correspond to similar investigations: HERTWICH ET AL. (1999) investigated diverse lipophilic substances by using CalTOX™ (DTSC 1993), which is also a combination of a Level III multimedia model and an exposure module. They pointed out that the variation in landscape properties, even in a state as diverse as California, is small compared to the effect of exposure factors. But most of the variance is due to chemical-specific input data, although certain exposure factors can be very important for chemicals for which dominant exposure is through the food chain.

9.4 Conclusions

Despite the poor database for some parameters, uncertainty analyses offer a means to attain a more detailed and realistic evaluation of the uncertainties inherent in the total daily dose. The range of possible outcomes (according to the selection of three realistic scenarios) of up to one order of magnitude lies in between the range of uncertainties spanned by the difference of the 90%-ile and 10%-ile (i.e. a maximum of 4½ orders of magnitude for lipophilic chemicals and 2 orders of magnitude for rather hydrophilic compounds). Hence, the scenario analysis is only able to reveal a subset of possible outcomes, and the probabilistic analysis leads to a significant wealth of information.

The strong dependence of the exposure module on the substance's lipophilicity is obvious. It can be concluded that the more lipophilic the chemical, the higher the expected uncertainties are and the lower the impact of variability of certain parameters is. The individual parts of the overall system are of different relevancy: Uncertainties of the exposure module are negligible for chemicals ingested directly via air. In this case, the variability of intake rates dominates the total uncertainty. But they play a role for chemicals ingested indirectly via the food chain and uncertainties of the exposure module then exceed the uncertainties of all other parts of the system. Due to the database, variability and true uncertainty can often not be separated in practice – only the variability of intake rates for various age-specific groups is quantifiable. Substance-specific properties are most important for partly particulate bounded and partly gaseous occurring chemicals, the mobility of which strongly fluctuates when changing properties.

Without respect to any substance, it can be concluded that only a small subset of all parameters (never more than 25%) is able to reproduce the uncertainties in the result. Several sensitive, but not sufficiently uncertain parameters exist that do not influence the resulting probability distributions.

After analysing the cumulative distribution functions, a propagation of uncertainties need not necessarily be expected for the input parameter distributions applied in this study. However, such an effect cannot be excluded if other distributions are chosen.

A further consequence affects the use of generic scenarios: The use of generic assessments is often criticised and the importance of differences in landscape characteristics is pointed out. This investigation indicates that the effect of uncertainty in landscape parameters will in many cases, especially for lipophilic substances, not be significant once the uncertainty in other parameters has been taken into account. The variance in the result does not increase significantly when uncertainty in landscape-specific parameters is included. For lipophilic substances, the contribution of landscape parameters to the overall uncertainty is usually less than 10%. For non-lipophilic substances,

the contribution is significantly higher, but always less than 50%. As a consequence, the use of a generic scenario is justified for chemicals ingested via the food chain (and not only air or drinking water). In this case, efforts to improve the exposure assessment should concentrate on a better characterisation of the substance or exposure model parameters.

9.5 Summary

The results of the probabilistic uncertainty analyses of the total daily intake were presented and discussed. Firstly, the contribution of individual parameters to the result was investigated. Secondly, the parameters were grouped in order to determine the contribution of each submodel on the daily intake. Additionally, the results were compared with deterministic values from the scenario analyses. It was found that the result only depends on a relatively small subset of parameters. Depending on the substance, up to a quarter of all parameters are important. The uncertainties are high for chemicals ingested via the food chain and lower for those that are ingested directly via air or drinking water. Only the parameters of the exposure module are important for the former, and parameters from all submodels are important for the latter. Against the background of the database used, a cumulative worst case could not be found.

10 Comparison with alternative models

This chapter provides a brief literature review of alternatives models that are suitable for comparison with the TGD models. On this occasion, both simple and complex models are presented.

10.1 Alternatives to the bioconcentration model for fish

K_{OW} as a descriptor for the bioaccumulation potential in fish is the simplest approach. More complex (i.e. mechanistic) models have undergone substantial research and development in the last decade (THOMAN 1989, BARBER ET AL. 1991, SIJM ET AL. 1995, CAMPFENS AND MACKAY 1997, GOBAS ET AL. 1993). ECETOC (1995A) provides a compilation of further models. In comparison to the TGD approach, a more complex model for describing steady state would integrate some or all of the following processes: bioconcentration, biomagnification, growth and elimination. JAGER AND HAMERS (1997) investigated a model suitable for considering bioconcentration, metabolism and growth. The model delivers up to a log K_{OW} of seven similar results like the TGD model. It yields a constant bioconcentration factor of six for superlipophilic substances. Against the background of calculated bioconcentration factors and theoretical considerations, the authors favour this model. The disadvantage of the model are its increased data requirements (weight, lipid and water content, growth and metabolism rates, etc.). They used a generic fish for dealing with this handicap. Using the model of THOMAN (1989), which considers biomagnification and the organism's rank in the food chain, and the example of PCB in the Great Lakes, CAMPFENS AND MACKAY (1997) showed that the increasing error with an increasing degree of lipophilicity can be explained by the model. But, due to its required database, this model is also only applicable with a great deal of effort and, hence, not practicable for the screening stage of chemical risk assessment. Using a generic fish seems to be problematical, due to highly variable properties, such as fat content (see concentrations in eels versus other species). Altogether, consideration of further processes may improve the results and is therefore desirable, but it is not suitable against the background of the model purpose (as a screening tool!). An alternative and equally simple method is the use of *quantitative structure-property relationships (QSPRs)*. LU (1999) demonstrated that these may lead to more realistic bioconcentration factors than the K_{OW}-based equations. QSPRs are investigated more closely in the next section.

10.2 Alternatives to the biotransfer model for meat and milk

A number of issues limit the reliability of K_{OW} as a BTF predictor. Because of the importance of the food chain exposure, and as a result of mainly unfulfilled assumptions (e.g. the assumption of a steady state, Section 6.2.2) by the TGD regression equations, there is a need to develop methods that can both determine biotransfer with a lower uncertainty and serve as a simple screening tool for risk assessment. It is possible to apply more detailed models on the one hand or models that do not depend on the K_{OW} on the other hand.

More detailed models, i.e. *physiologically-based pharmacokinetic models (PBPK)* have been developed by MCLACHLAN (1992), DERKS ET AL. (1994) AND VAN EIJKEREN ET AL. (1998). MCLACHLAN (1992) developed a mass balance under typical agricultural conditions and constructed a dynamic compartment model to describe the fate of hydrophobic organic chemicals in lactating cows. The model consists of three compartments: the gastrointestinal tract, blood and fat tissue. Diffusive flows are defined between them. Degradation takes place in the blood and the gastrointestinal tract. Three adjective flows exist: Fodder uptake, excretion and lactating. Thus, uptake via fodder is the only accumulation process. This model is able to explain the relatively low concentrations of higher chlorinated PCDD. Its disadvantage is the increased database, i.e. degradation rates in two compartments, fat content, lactating rate, etc. This and all other models of this type are based on several physiological compartments, which leads to an enormous enhancement of rarely measurable parameters. Furthermore, it was found that metabolism and absorption play an important role for many chemicals, but these are only slightly related to available physico-chemical properties. A generic PBPK model is therefore not suitable for the screening stage of risk assessment.

Another alternative approach is the use of *molecular connectivity indices (MCI).* As shown by DOWDY ET AL. (1996), the prediction of biotransfer using molecular connectivity indices as a quantitative structure-property relationship is, like the K_{OW}-based regression equation, not only a cost-effective and simple method, but also a more reliable one. The underlying idea of this method in its most simple form is to count the bonds of the hydrogen-suppressed molecular skeleton and to derive an index from them. The index correlates to experimentally determined biotransfer factors and, thus, can serve as a surrogate for a K_{OW}-based correlation. It is easy to quantify and it is able to reduce uncertainties. The range of applicability encompasses mainly non-polar chemicals, but its application to polar substances is possible when using correction factors. This approach was compared with the TGD approach. Particularly for lipophilic chemicals, the measured K_{OW} values for one compound may deviate by up to several orders of magnitude (MACKAY ET AL. 1991-1997). The uncertainty is propagated in the model calculation. Altogether, the TGD approach for estimating concentrations in milk and meat inheres three sources of uncertainty: (1) measurement of the K_{OW}, (2) goodness of the underlying regression and (3) uncertainties in other parameters used. The latter source holds for both the K_{OW}- and the MCI-based estimation.

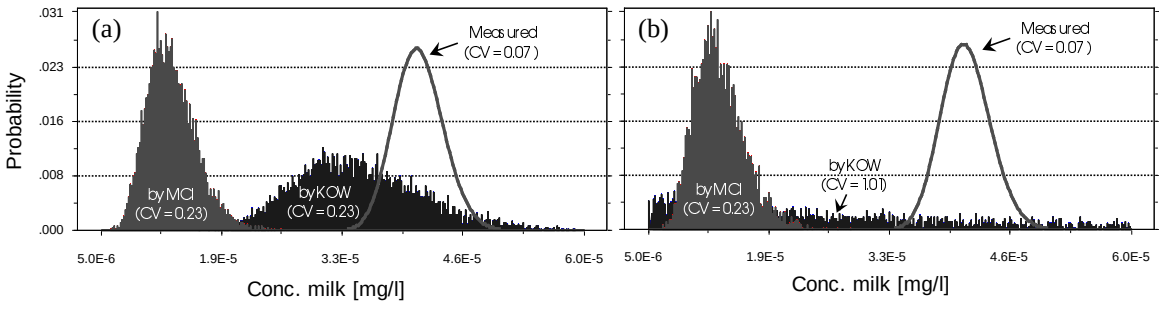


Fig. 10.1 Probability density functions for PCB 52 concentrations in milk using K_{OW} and MCI (a) constant K_{OW} (b) uncertain K_{OW}. Data from NRW (1991B) and (MACKAY ET AL. 1991-1997).

The second source was statistically evaluated by DOWDY ET AL. (1996). They emphasised the higher reliability of the MCI approach. The first source of uncertainty holds only for the K_{OW}-based approach. The impact of this source is illustrated in Fig. 10.1: (a) For a constant K_{OW} the TGD approach delivers better results than the MCI approach (with reference to the modus of the distribution). (b) For an uncertain K_{OW} the modus of the K_{OW}- based approach is less than that of the MCI-based method, and the coefficient of variations is four-times larger. Fig. 10.2 shows a comparison of measured with calculated concentrations for all investigated PCDD and PCB. The MCI methods result in better estimations for dioxins and in poorer estimations for PCB. However, the MCI methods always lead to lower concentrations.

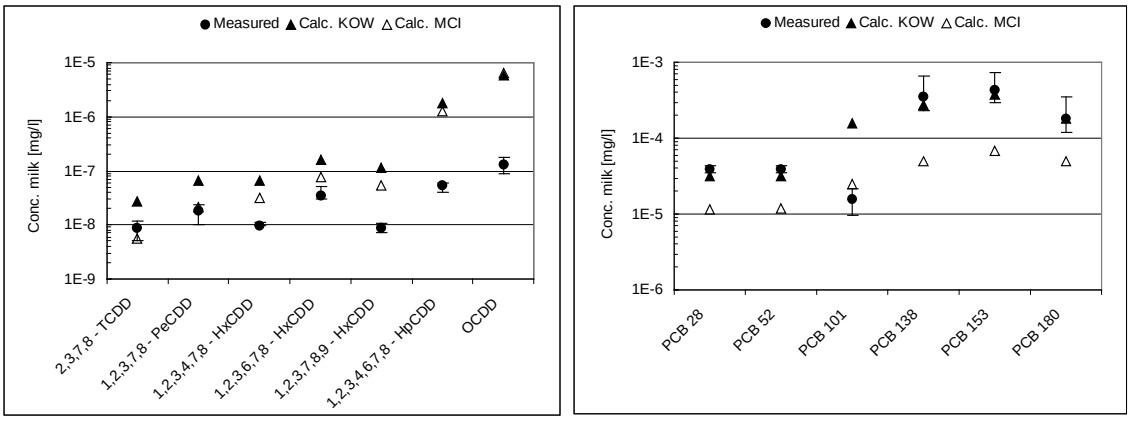


Fig. 10.2 Comparison of measured PCDD and PCB concentrations in milk with K_{OW} and MCI determined values. Data from NRW (1991B).

The calculations demonstrate that the MCI based method is necessarily closer to the monitoring data. But the method is able to reduce uncertainties that are concealed by the deterministic calculation of Fig. 10.2 and, thus, offers an advantage with regard to the K_{OW}- based method.

Experimentally determined *carryover rates (CR)* can be applied to estimate concentrations if these are available. Concentration in milk may be calculated by means of the intake rate of fodder Q_{Futter} [kg/d] and the lactating rate Q_{Milch} [l/d]:

$$C_{Milch} = \frac{C_{Futter} \cdot Q_{Futter} \cdot CR_{FM}}{Q_{Milch}}$$

Carryover rates of hydrophobic compounds are provided by several papers (Tab. 10.1). The available rates originate from different experiments. Due to the fact that a steady state is only reached after between 50 to 70 days, depending on the congener (MCLACHLAN 1992), only rates with an actually reached steady state were chosen.

According to the above-mentioned equation, the rates were applied to the concentrations in grass. The resulting concentrations in milk are depicted in Fig. 10.3. The results correspond well with the measured values and for PCB lead to similar results, while for PCDD appreciably better results than the TGD model are gained. Even the greatest deviations do not exceed one order of magnitude.

The good results achieved by applying carryover rates are caused by the fact that the rates are directly derived from the investigated congeners. Simultaneously, fodder is the only relevant pathway for the investigated chemicals. Carryover rates therefore lead to better results. However, their application is only allowed for hydrophobic and persistent compounds.

Tab. 10.1 Carryover rates for fodder/milk from six sources. Taken from RUOFF (1996) and MCLACHLAN (1992).

Substance	Olling	Stevens	Ruoff	McLachlan	Tuinstra	Heeschen
TCDD	0.30	0.40	0.35	0.36		
PeCDD	0.28		0.14	0.32		
HxCDD I			0.09			
HxCDD II	0.27		0.14			
HxCDD III			0.08			
HpCDD	0.02			0.03		
OCDD				0.04		
PCB 138				0.78	0.23	0.71
PCB 153				0.63	0.18	0.75
PCB 180				0.63	0.21	0.68

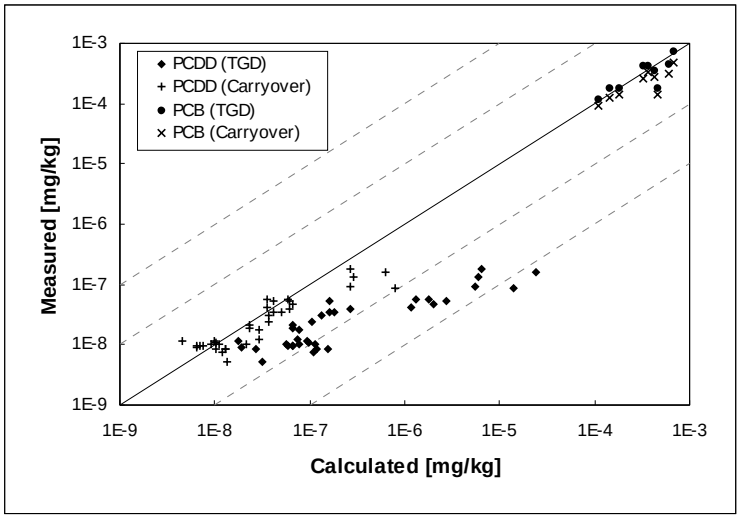


Fig. 10.3 Comparison of calculated milk concentrations (single values) with monitoring data. The calculation was carried out by means of the TGD approach and by means of carryover rates. Data from NRW (1991B).

10.3 Alternatives to the plant model

Applying bioconcentration factors (BRIGGS ET AL. 1983, TRAVIS AND ARMS 1988) is a more simple approach in relation to the model laid down in the TGD. By means of bioconcentration factors, calculated concentrations for PCDD and PCB were compared with the results of the TGD model (Fig. 10.4). The figure demonstrates that the use of bioconcentration factors leads to a significant underestimation of about two orders of magnitude. The TGD approach, which additionally considers the

transfer pathway air, achieves a good correspondence to measured values for chemicals in the gaseous phase (TCDD and PCB). Although the measured values are underestimated for stronger particulate-bound dioxins, the TGD model nevertheless results in better estimations with regard to using bioconcentration factors. The relatively strong deviations of PCB 28 and PCB 52 are obvious in contrast to the higher chlorinated congeners. The concentrations presented exceed the detection limits. Furthermore, concentrations in pore water do not contribute to the plant's concentration and the impact of the deposition of particles is negligible, because the particulate fraction is less than 1% (estimated according to the Junge equation). However, the plant concentrations originate from a more polluted region (*Northern Ruhrgebiet*), which might explain the high concentrations. More detailed monitoring data are necessary to answer this question. Concentrations were also estimated by means of a bioconcentration factor of 0.15 (FW plant/DW Soil) (MEM 1996). The resulting concentrations are considerably lower than those of the TGD model (no figure).

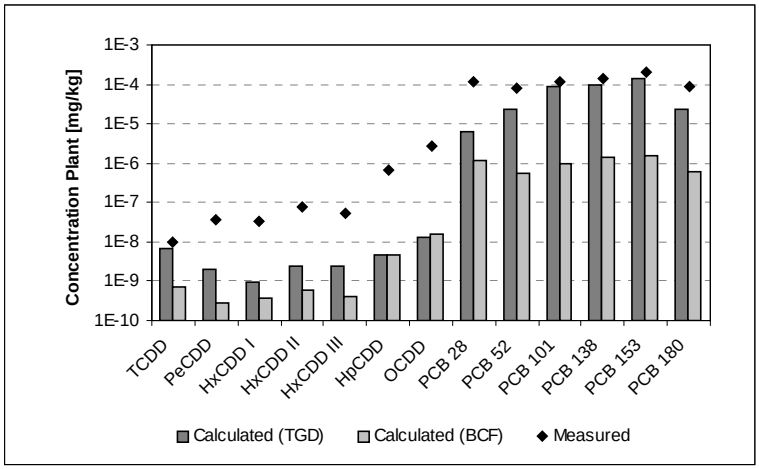


Fig. 10.4 Comparison of calculated (by means of bioconcentration factors soil/plant and TGD) and measured concentrations in plants. Data from NRW (1991B).

The results reveal that the use of bioconcentration factors is inadequate for a variety of substances, because they only consider one transfer pathway (e.g. soil/plant) and ignore the combined effect of K_{OW} and K_{AW}. On the other hand, more complex models (e.g. the numeric 4-compartment model of TRAPP (1995)) lead to non-acceptable data requirements for the screening phase of risk assessment. POLDER ET AL. (1998) evaluated the TGD model by comparing it with the 4-compartment model of RIEDERER (1990) and with experimental data on leaf/air partition coefficients found in the literature. For substances with $K_{LA} < 10^7$ they found that the air-to-plant pathway leads to deviations within a factor of 5 for most substances. For a higher partition coefficient growth dilution becomes a relevant process and the TGD model was assumed to be more adequate than the model of RIEDERER because it includes this process.

All in all, the TGD plant model represents a good compromise with regard to complexity. Furthermore, it can be improved by the integration of further processes (Section 8.3).

10.4 Alternative human exposure pathways

Estimating the total daily dose is realised by summing up diverse exposure pathways and, thus, no "alternative" models exist. However, viewing a system with a similar purpose is advisable. CalTOX™ (version 2.3, DTSC 1993) was chosen for this. It was introduced to improve the risk assessment process in the state of California. It is able to relate the concentration of a chemical in the soil to the daily intake by humans near the contaminated soil. Based on an eight-compartment multimedia distribution model and a comprehensive exposure model, it estimates the daily intake by humans. To reflect uncertainty, CalTOX™ has the capability to conduct Monte-Carlo simulations. A parameter value is described in terms of its mean and coefficient of variation. Model predictions are presented by confidence intervals. CalTOX™ covers considerably more exposure pathways for estimating the daily dose (Tab. 10.2). The pathways of EUSES represent a subset of those considered in CalTOX™. Even if the underlying models and the calculated relevancy of exposure pathways were not compared, a look at CalTOX™ demonstrates that EUSES describes only a minimum of possible pathways.

Tab. 10.2 Comparison of exposure pathways used by EUSES and CalTOX™.

Human exposure pathway		**EUSES**	**CalTOX™**
Inhalation	Outdoor air → Human	X	X
	Indoor air → Human		X
	Drinking and tap water → Human		X
	Soil (particle and volatilisation) → Human		X
Food	Freshwater fish → Human	X	X
	Seawater fish → Human		X
	Meat → Human	X	X
	Milk and dairy products → Human	X	X
	Eggs → Human		X
	Drinking and tap water → Human	X	X
	Plants → Human	X	X
	Roots → Human	X	X
	Surface water → Human (swimming)		X
	Soil → Human		X
	Human milk → Baby		X
Dermal	Drinking and tap water → Human (washing)		X
	Surface water → Human (swimming)		X
	Soil → Human (contact)		X

10.5 Conclusions

Alternatives for the submodels of the exposure module are basically two-fold: More simple or more complex models. Models for estimating concentrations in fish, milk and meat are quite simple. More complex models are in fact able to reduce occurring uncertainties, especially for the superlipophilic uncertainties. But they are not advisable for the screening stage of risk assessment, due to the extended database required for their application. Due to the interspecies variability, also a combination of these models with a generic organism is not useful. A future possibility are regression equations that are not based on the lipophilicity of the substance, but on the molecular structure. Although no better results can be expected from this methodology in general, its universal applica-

bility (besides BCF and BTF it is applicable for estimating several further partition coefficients) and the capability to reduce propagation of uncertainty reveals that this methodology might be an interesting alternative. Further investigations are necessary for a more detailed evaluation. Comparison with carryover rates shows that using substance-specific data leads to better results, but is not required for lipophilic chemicals since the TGD equations are particularly applicable for these chemicals. Replacing the plant model with regression equations is not advisable, because not all relevant transfer pathways would be considered. On the other hand, an extension of the plant model by further refinements or the inclusion of further compartments is not useful because, even in its current form, the model consists of several hardly quantifiable parameters. Using such a relatively complex plant model in contrast to the simple regression equations for fish, meat and milk is justified, since chemical uptake by plants occurs from the air, the soil or both pathways simultaneously. In contrast, the surrounding medium is always the most relevant pathway for uptake by fish, and fodder dominates chemical uptake by cattle. The submodel for estimating the total daily dose is based on many simplifying assumptions. The human exposure model only considers exposure pathways in a very aggregated form and does not differentiate between individual pathways, although it represents the final step of an exposure assessment according to the TGD and, thus, always contains sensitive parameters.

Altogether, it can be concluded that the submodels used are simple, but practical. Alternative models will not lead to better results if data are rare. However, the current approach seems to be extendable and correctable with respect to the number and refinement of integrated exposure pathways.

10.6 Summary

The models used were compared with alternative models. A comparison was carried out qualitatively for most of the models, and also quantitatively for the plant model. The models of the exposure module were classified as a good compromise between complexity and practicability. Only the number and refinement of integrated exposure pathways is regarded as insufficient.

11 Software evaluation

The following chapter yields the results of the software evaluation. A presentation of the results of this chapter may also be found in SCHWARTZ ET AL. (1998). Maintenance of EUSES 1.00 is in progress. The basis is the so-called *Blacklist* (ECB 2000), which is a compilation of errors and proposals. All errors described therein are not discussed here.

11.1 Product description

The description of EUSES (ECB 1997) presents the programme and theoretical background. A presentation of system requirements and interfaces to other programmes is lacking. Thus, the following questions remain:

1. Which operating system is exactly supported?
2. What are the minimal hardware requirements?
3. How can one import and export data?

Additionally, EUSES is presented by the ECB on a web page (ECB 2000), which also does not meet the requirements of a product description.

11.2 Documentation

The printed documentation consists of a description of the models used (EC 1996B) and a user manual (EC 1996D). Online documentation containing less information is also available. A tutorial does not exist.

11.2.1 Printed documentation

The model description presents the integrated models by means of considered processes, some assumptions and limitations, and provides an overview of the scientific background. Several equations are documented. The appendix provides tables with symbols and emission factors of the individual use categories. The manual contains a short reference, a description of the menu, an explanation of error messages and a technical reference. Results of validation studies and an index are not given.

The correspondence of manual to the TGD was checked. With the following exceptions, both papers are equivalent. According to an investigation of the documentation, EUSES contains only very few errors due to the conversion of equations from the literature. The Level III model (SimpleBox 1.6) is not documented. The same applies to the sewage treatment plant model (SimpleTreat 3.1). A comparison of TGD and the EUSES manual shows that formulae and parameters are usually equivalent. In two cases (Formulae 57 and 117 in the EUSES documentation EC (1996B)), the EUSES models were extended by one factor (Fconnect and Fresp). In Formula 93 in the EUSES

documentation EC (1996B), a factor is missing (correction factor a in the plant model), which is marked as an error in the original literature. Table III-22 of the manual does not correspond to the original literature. An exact description of the SimpleBox model is not available. Since no original literature exists for the model used in the TGD, it is not documented. Publications exist on SimpleBox versions 1.0 (VAN DE MEENT 1993) and 2.0 (BRANDES ET AL. 1996). The SimpleBox model in the TGD represents an interim version. The same applies to SimpleTreat: documentation for version 3.0 is available, but 3.1 is implemented.

Transparency of the equations implemented in the programme is lacking. The source code of the programme is not freely available or revealed, hence the implemented equations cannot be examined. Users can only follow the manual (EC 1997). Furthermore, the nomenclature between documentation/programme and TGD is not always equal: The term “bioaccumulation” (EUSES) is used instead of “biotransfer” (TGD) or “beef” (EUSES) in used instead of “meat” (TGD) for assessing concentrations in meat and milk (EC 1996B, III-70). In addition, the parameter of the concentration in plant roots is called C_{root} in the documentation on the on hand and $Croot_{plant}$ in the appendix on the other hand. Finally, several parameters and equations, respectively, are insufficiently explained. For instance:

- How is BCF_{biota} calculated (EC 1996B, III-45, (58))? Furthermore, these parameters can be altered within the software, but it is described as “closed” in the documentation.
- C_{Water} (EC 1996B, III-63) refers to the dissolved fraction. Hints are missing. The same holds for PECregwater (EC 1996B, III-47): Is this the dissolved or the total fraction?
- What is meant by *Respiratory fraction* and *Bioavailability oral* (EC 1996B, III-74, (117))?
- $TH_{der,worker}$ (EC 1996B, III-87) is listed in Tab. III-26, but no mention is made as to where these parameters are used.

11.2.2 Online documentation

An error was found: In the help window, “Main Category (MC)“ contradicts a presented combination of the allowed input in window “Emission input data“. According to the help guide?, only MC II and III are allowed for “Life cycle-step Formulation”. But EUSES allows the input of Ib and Ic (IC 0/UC 0).

11.3 Technical requirements

11.3.1 Installation and system requirements

EUSES 1.00 is executable with Microsoft® Windows operating system 3.1, 95, 98 and NT. Other systems (e.g. Macintosh, UNIX, etc.) are not supported. Test platforms ranging from INTEL 486 (32MB) to Pentium® Pro 200 (128MB) posed no problems. However, a time delay was observed on a Pentium® Pro 200 platform that could not be explained. The installation procedure was carried out without problems. A description on how to uninstall EUSES is lacking.

11.3.2 Stability and reliability

EUSES 1.00 runs stably. Programme crashes were not observed.

11.3.3 State-of-the-art

The file EUSES.INI is used for saving and loading the system's configuration. This is an obsolete concept that is necessary, however, to sustain a MS-Windows 3.1 compatibility. The state-of-the-art concept of Windows 98/NT makes arrangements to save the configuration settings in a system-immanent hierarchical database (registry).

11.3.4 Network-support

"EUSES is a stand-alone system. Network operations with more than one user can result in error-prone behaviour" (EC 1997, page 68). It was therefore investigated whether a single user is at least able to access the software on a network installation. It was found to be theoretically possible, but not practicable since the right to write configuration and data files is required by each individual user.

It must be concluded that the safe application of the programme on a network is not possible. The problems mentioned could be avoided if the configuration file EUSES.INI were surrendered.

11.3.5 Miscellaneous

A directory is shown below the menu item "System/Directories" that contains the path to all data files. In order to allow several users to work with EUSES or to enable an advance organisation of data, it would be desirable to choose between several databases located in different directories. EUSES allows access on only one database. Changing directory is impossible if the user is unable to write on the configuration file. Although a less meaningful error message occurs in this case, the directory entered is falsely presented as a new data directory. It would be better to allow the selection of a directory below the menu item "File/Study/Load-Save".

11.4 Correctness of calculations (verification)

Erroneous results in a formal sense were not observed. However, one exception exists. This involves the calculation of regional background concentrations. Even though a valid and plausible combination of input parameters is entered, negative concentrations may occur. This phenomenon indicates an erroneous implementation of certain equations. However, it cannot be explained in the absence of the source code. An exemplary parameter set for calculating a negative regional concentration in surface water is:

- FSolidSed = minimum, FWaterSed = minimum,
- FrunoffSoil = 0.6 (instead of 0.25),
- SuspWaterReg = 80 mg/l (instead of 15 mg/l).

All other model parameters receive default values. The substance-specific parameters should correspond to those of OCDD. It follows NetSedRate < 0 and PECregWater < 0.

11.5 User interface and operability

EUSES presents itself with a modern and uniform user interface. However, a multitude of input dialogues makes an overall view difficult. Over 100 dialogues (with up to 600 parameters) and 6 result windows (with more than 100 singe values) appear for a complete estimation, which leads users rapidly losing track. Use of the software only seems acceptable if the user completes the dialogues from beginning to end in the given order. If, however, individual values need to be changed after the estimation has been completed, it is extremely difficult to find the required input dialogues. EUSES is therefore not suitable for interactive use. A positive aspect is the possibility to enter values in different units. Comments on the input data would be desirable, but this is not possible. User hints and warnings are lacking. For example:

- An entered value that falls below the minimum possible value is set without warning as the minimum value.
- Decimals are separated using decimal points. If a value such as “0,9” is entered, “0” results without warning. Commas are used as separator in several languages, e.g. in German!
- Physically impossible values are made possible, e.g. K_{OW} = -1.

Contrary to the manual, the import of data from the up-to-date IUCLID version (EC 1996C) is not possible. The programme runs stably but slowly. It cannot be used on a network system. The following table compiles observed errors in detail.

Tab. 11.1 Examples of errors.

No.	Error	Example(s)
1.	Input of extreme values may cause problems.	Entering a log K_{OW}<-308 results in “1.234” (after pressing the “Accept value” button). Entering a log K_{OW}>308 results in “??”.
2.	Depending on the entered parameter value, the calculation stops abruptly.	It is impossible to finish the assessment if 0 is entered for the *Fraction connected to sewer systems* in window “Defaults/Release Estimation“. The programme stops at the window “Degradation and transformation input”.
3.	Erroneous parameter flags may occur.	The boxes *Oral NOAEL* and *NOEC via food* contain the flag “o” for output in the window “Mammalian effects input – (sub) chronic”, although they were entered by the user and not calculated by EUSES. This effect can be obtained in the following manner: A value is first entered in one of the two boxes. The flag is correctly set to “s”. In the next step, the user overwrites this value with “?”. The corresponding default or output value normally appears, but in this case the value entered beforehand sustains with the flag “o”. Thus, the entered value is presented as an output.
4.	Changing units may cause problems.	Errors will occur when changing from degradation rates to half-lives in the window “Distribution – Degradation and transformation rates – Characterization and STP”, if a degradation rate of 0 is entered. In this case, the half-life is set to a very small value instead of setting it to the maximum. When leaving the box (by pressing the tab-key) the value alters again to another small value.
5.	The status line is not al-	Information on “non-substance data“ in the status line is not set to “Stan-

No.	Error	Example(s)
	ways updated.	dard" after resetting the system, but the name of the previous dataset is shown. A correct entry appears only after selecting "Defaults – Edit – Defaults identification".
6.	Buttons do not always work.	Button "Print" in the window "Report Preview" does not work. Printing is only possible via "Assess – Print – Printer" (i.e. without preview).
7.	Formatting errors in the report may occur.	Line "NOEC for other terrestrial species" is printed out twice in the report.
8.	Fractions of more than 100% are possible.	Although the single fractions of natural soil, agricultural soil and industrial soil are checked as to whether they lie within the interval [0,1] in the windows "defaults/regional distribution" and "defaults/continental distribution", their sum is not checked. Consequently, a fraction of total soil of more than 1 is possible with respect to the area. Another example is that the emission may exceeds the tonnage (A-Tables): for LAS (IC 5/UC 9) 99% is emitted into waste water and 1% onto soil for "Private Use". These make 100% altogether. But further emissions due to "Production" and "Formulation" occur, which results in a total of over 100%. This phenomenon is even more significant for IC 5/UC 10. "Private Use" assumes an emission of 100% into waste water and 0.01% to 1% onto soils plus additional emissions for "Production" and "Processing".
9.	Check of parameter ranges may cause problems.	Putting in 0.5 days as the half-live for biodegradation in water (kdegWater) or 3 hours as the half-live in air (kdegair) produces the warning that the parameter is outside the valid range, although it falls within this range.

Furthermore, several annotations could be made to improve the operability of the software. These are listed in Tab. 11.2.

Tab. 11.2 Measures to improve the software's operability.

No.	Annotation/Proposal
1.	Some features should be implemented more consistently. For example, entering data on acute toxicity in the window "Aquatic effects input" allows the use of different units. But in the window "Mammalian effects input", this feature is lacking.
2.	Data input should be more flexible. EUSES currently only allows input of data in a fixed and linear form. A more model-oriented procedure would also be helpful (i.e. input of parameters used in a certain model). This would enable the user to calculate the models separately.
3.	Physically impossible values should not be made possible. For example, K_{OW}<0 or LC_{50} for fish<0 are possible. A warning does in fact appear, but the input is nevertheless possible.
4.	The ranges of estimated parameters should also be investigated. For example, K_{OW} = -1 results in BCF_{Worm} = -0.04 without an error message. Curiously, BCF_{Fish} is set to "1.41" (i.e. to the correct value) in the case of invalid *log* K_{OW} values, but to „??" in the case of invalid K_{OW} values.

No.	Annotation/Proposal
5.	No warnings appear if regression equations are used outside their range of applicability. If an entered value does not fall into the regression range, the result is set automatically, and without informing the user, to the minimum or maximum. Refer to, for example, the log K_{OW}/log BCF- regression equation (EC 1996B, III-60).
6.	Sometimes the result is calculated immediately, and sometimes the simulation has be started manually. For example, an immediate estimation for TSCF=f(K_{OW}) is possible, whereas non-immediate estimation is only available for $PEC_{oral,worm}$=f(APPLSludge) (EC 1996B, III-53, III-61, III-66).
7.	The user is not informed about inconsistency between parameters. For example, physico-chemical properties of a chemical are usually entered for an environmental temperature of 20-25°C. But the standard temperature of the system is 12°C.
8.	Several error messages should make more sense. The meaning of the field “Solution” in several error messages is questionable since the field is usually left empty or only contains a reference to the manual. For example, trying to load a substance in the case of a wrong data directory results in the message “Load substance failed” with the solution “See EUSES manual”.
9.	The width of columns in the table “Use patterns” should be variable. Otherwise, it is difficult to read results and a printout of the table is impossible.
10.	It is not clear where to find the “Main category” in the window “Emission input data” because this formulation is not used in that window. It would be better to use the term “Main category”.
11.	The button “Load Defaults” in the “Defaults” window is not placed ergonomically. It should be separated from the other buttons in the window and placed at the bottom of the window. It would be even better to remove the button from the window completely, since the respective function can also be carried out via the menu item “Defaults/Edit”.
12.	The status line is not legible on each computer screen (Fig. 11.1). This may be caused by the hardware’s configuration on certain computers.
13.	By pressing the right mouse button in the “Outline Mode”, a popup menu is available containing the entry “Information”. This entry is always selectable. However, it should only be selectable if it actually fulfils a function.
14.	The menu item for printing a report is found in menu “Assess”. However, this item is normally located in the “File” menu.
15.	Whether the column “Reference“ is printed into the report depends on the selection of the “interactive” or the “direct/outline” mode. The reason for this is not clear.
16.	In “Interactive mode assessment” and “Direct mode assessment”, when pressing the “Next” or “Prev” button the mouse pointer always jumps to the “Next” button of the following window, which makes jumping to prior windows uncomfortable. The pointer should return to the “Prev” button again after having pressed a “Prev” button.

Fig. 11.1 The status line.

11.6 Transparency

A complete reproducibility of the implemented equations in impossible. The source code of the programme is not freely available and a review of the implemented equations was not possible.

Thus, a check of the actual calculations can not be carried out. The documentation (EC 1996B) serves only as an orientation. The documentation reveals all models, but is incomplete (Section 11.2). A clear modularity of the modules is not given. It is hard to recognise which parameters are connected with which model. It is difficult to calculate models separately; the user must conduct complete risk assessments. The resulting complexity leads to users losing track and operates against the acceptance of models and software.

It must be concluded that EUSES does not fulfil the derived requirements of a transparent programme.

11.7 Features

Diverse mechanisms were integrated into EUSES to contribute to quality assurance: The range of entered parameter values is checked (plausibility check) or the user is able to enter values in different units. Unfortunately, these mechanisms were not always applied consistently. Tab. 11.3 compiles proposals for improving the software.

Tab. 11.3 Proposals for improvements concerning the programme's features.

No.	Proposal
1.	The plausibility check of input data can be further improved. For example, input of invalid CAS numbers is possible.
2.	A check of dependencies between parameters should be introduced. For example, melting point > boiling point is possible.
3.	As theoretically described in Section 6.3, a correction of partitioning coefficients should be integrated. This measure would broaden the applicability of the software to bases and acids.
4.	A warning should appear if a regression equation is used outside its range of applicability. Furthermore, information concerning underlying chemicals and goodness of fit could be shown.
5.	It should be possible to comment on each input parameter. This lack causes a loss of information regarding data source, data quality, time of measurement, etc. It is desirable to activate a window on demand for commenting on each single input field.
6.	The integration of alternative estimation functions is desirable. EUSES should at least contain the functions listed in the TGD.

11.8 Cooperation with other programmes

Data from other programmes can be imported by means of various formats (HEDSET/CIF, SNIF/SNF, EUSES/EXF). Export of entered data and results is possible by means of an own ASCII-based format (EUSES/EXF). However, none of the formats are documented and a real benefit for users is not visible. An inspection of an exported file reveals several parameters that do not exactly correspond to the entered values. For instance, entering "0.7" for the parameter FConnectSTP results in the following export file entry:

```
PA FConnectStp 2 0.699999999999999956 0 0 0
```

Hence, an error is hypothesised, which may be explained by different variable types in the implementation.

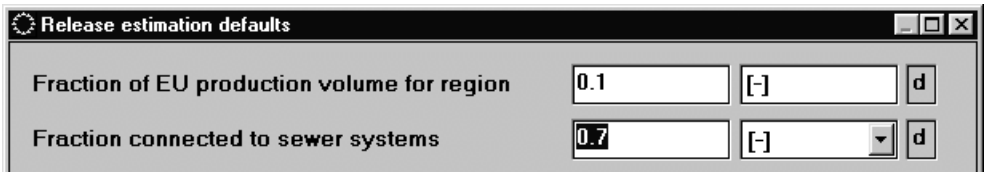


Fig. 11.2 Window Study/Defaults/Release Estimation

The possibility to load IUCLID datasets, as described in the manual (EC 1997, page 38), is not comprehensible, because the purchasable IUCLID-CD (EC 1996c) does not offer the possibility of exporting.

It has to be concluded that a data transfer with other programmes, as described in the documentation, is as yet impossible.

11.9 Uncertainty analyses capability

Uncertainty analyses cannot be carried out by means of the original EUSES version. Also coupling the software with add-ins (e.g. Crystal Ball®) is impossible. However, it is investigated as to how to integrate the possibility of uncertainty analyses in a EUSES update (JAGER 1997, page 8).

11.10 Support

Sources of information in the case of questions or problems are available. A compilation of all of the addresses listed in the programme and documentation shows that this information is inconsistent. Most comprehensive statements are found in the online help and product description. Facilities of support via the internet (e.g. in form of FAQ-listings[4] or current programme information) are not used sufficiently. However, an error list (the so-called “blacklist”) is available online (ECB 2000). Available sources of information are:

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European Chemicals Bureau
Joint Research Centre
I - 21020 Ispra (VA)
Tel. ++39 332 785 866
Fax ++39 332 785 862
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Only the product description (ECB 1997) provides a further address:

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National Institute of Public Health and the Environment
P.O. Box 1
NL – 3720 BA Bilthoven
Tel. ++31 30 274 30 04
Fax ++31 30 274 44 01
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[4] Frequently Asked Questions

The online help and web page (ECB 2000) point out a further e-mail address:

`euses.euses@jrc.it`

Only the web page provides the e-mail address of a contact person:

`christian.heidorn@jrc.it`

It is proposed to make available all known errors and all other useful information regarding EUSES on the EUSES web page. Furthermore, an internet news group could be introduced. These measures would further increase acceptability of the software.

11.11 Conclusions and proposals

EUSES 1.00 presents itself as a modern software product which basically meets the requirements of a DSS in the field of environmental risk assessment. Good knowledge of the theory and applicability of the implemented models is necessary to use the programme. Some assets and drawbacks should be emphasised: The programme basically meets common quality requirements such as product description, documentation, and the programme itself.

Positively remarkable is the fulfilment of the essential requirements concerning the correctness of calculations and stability. Implemented quality assurance measures, such as the opportunity to input parameters in various units or the plausibility check of parameters after input, are not self-evident for this type of programme.

One has, however, to find fault with the lack of transparency. This arises from the high complexity and low modularity of EUSES. It is difficult to recognise dependencies and links between parameters and whole models. A multitude of input dialogues makes an overall view difficult. Over 100 dialogues, including up to 600 parameters, appear for a complete assessment, which causes the user to become disorientated. Use of the software only seems acceptable if the user completes the dialogues from beginning to end in the given order. If, however, individual values need to be changed after the assessment has been completed, it is extremely troublesome to find the required input dialogues. EUSES is therefore not suitable for interactive use. A suitable insight into EUSES is aggravated by documentation lacking on the sewage treatment plant model SimpleTreat and the multimedia model SimpleBox. Publications exist on SimpleBox 1.0 and 2.0. The SimpleBox model in EUSES represents an interim version. The same applies to SimpleTreat: documentation for version 3.0 is available, but another version is implemented. Furthermore, a description of the import/export interface is not available to users of the programme. Contrary to the manual, the import of data from the up-to-date IUCLID version (EC 1996c) is not possible. Altogether, these facts lead to a black-box handling and might hamper the establishment of EUSES as a widely used programme for risk assessment.

Further points of criticism should be mentioned: comments on the input data are desirable, but this is not feasible. Some concepts, such as the plausibility check of data, are not consistently implemented (e.g. the user is given the opportunity to input physically impossible data or the range of

estimated data is not checked). With regard to technical requirements, it has to be stated that EUSES does not exercise the options offered by the Windows operating system (e.g. saving system configuration into an allocated systems database), nor does it provide a minimal network support.

Although EUSES fulfils several quality criteria, various alterations seem to be necessary and are possible. These can be divided into easily realisable and costly improvements.

Easily realisable improvements

- The documentation should be completed. This affects both models not yet documented and the import/export interface.
- Current information (e.g. regarding forthcoming updates, etc.) of EUSES should be made available (e.g. via the internet).
- Current errors should be rectified (see Tab. 6.1).
- Some minor changes regarding user interface and technical requirements would be advantageous (see Tab. 11.2 and Tab. 11.3).
- Users should be enabled to access various directories without having to change the configuration file. This may be easily realised by a possibility to select a directory in the menu File/Study-Substance-Block/Load-Save.

Costly improvements

- Although a function to check the plausibility of input data has already been implemented, it should be improved: as described above, a two-step plausibility check of all input parameters is proposed.
- Modularity should be increased by the isolation of single models.
- Alternative estimation functions (QPPRs) should be offered, at least those described in the TGD.
- More flexibility for data input and output, i.e. better interfaces. This of course requires exact documentation in the import/export format (EXF).

11.12 Summary

An evaluation of the software quality of *EUSES 1.00 (European Union System for the Evaluation of Substances*) was carried out. After testing the software and reviewing the documentation, EUSES presents itself as a modern software product that basically fulfils the postulated quality criteria. Particularly with regard to correctness and stability, (almost) no errors were found. EUSES contains some innovative features. However, numerous alterations are necessary. High complexity, low modularity, and incomplete documentation result in a lack of transparency and are emphasised as major points of criticism. Several proposals for improvements were made.

12 Conclusions

Until now, this paper investigated and evaluated the detailed aspects of the quality assurance task of models. The aggregation of the results and a concluding evaluation of all results together are both still missing. Thus, the goal of this chapter is to obtain a concluding evaluation in order to derive limitations of the models' applicability. A further section proposes ideas for both the improvement of the models and their integration into the entire procedure of risk assessment. The mathematical models and their integration into the software are considered.

It follows from the applied meaning of validation that the comparison of model results with monitoring data may be highly informative. But it alone is not a sufficient criteria for validation. Moreover, an inspection of the underlying theory, sensitivity, scenario and uncertainty analyses, a comparison with alternative models, and software evaluation are essential to fulfil the higher principle of quality assurance. Particularly, if the models are applied to new – and not yet released – chemicals for which environmental concentrations are unavailable, the properties of the models and the underlying assumptions play a central role. Of course, simulations have to be carried out for existing chemicals and, by means of inferences (substances with similar properties will lead to similar results), the models' behaviour regarding new chemicals can be characterised. This procedure necessitates a quantification of uncertainties. A combination of all of these methods renders evaluation of the entire system possible and leads to a derivation of a principle applicability and inherent problems, which are presented in the following sections. Proposals for improving existing models can be made regarding certain details of a model, but also conceptual changes of the overall system can be suggested. Initial proposals to improve the European Union exposure assessment scheme have already been made by TRAPP AND SCHWARTZ (2000).

12.1 Applicability of the models

If the underlying assumptions of the models are interpreted in a strict sense, the investigated models will only be applicable for persistent chemicals of low and medium lipophilicity (log K_{OW} from 0 to 6), the partitioning behaviour of which is not altered by dissociation or effects caused by the structure of the molecule. However, the numerous implicit assumptions are seldom accomplished simultaneously. Two effects are possible if one uses the models beyond their scope of applicability. On the one hand, the models may still estimate chemical concentrations or doses to a more or less accurate extent and, consequently, provide a valuable contribution to the risk assessment procedure. On the other hand, they may lead to qualitatively false statements, e.g. regarding the relevancy of a transfer pathway. In this case, they may mislead the user or even lull them into a sense of false security. An example of the first situation is the estimation of environmental concentrations by the regional distribution model. An example of the second situation is the false estimation of the relevancy of the root-human exposure pathway. However, it was shown that the latter problem is avoidable by an investigation of the models and a calibration of their parameters.

In general, for each model and the entire system hold: applicability comprises a variety of non-dissociating organic chemicals. All of the models investigated here are based on the lipophilicity of

the compound. These show highest uncertainties and strongest deviations from monitoring and experimental data for superlipophilic compounds. The best applicability for chemicals of low to medium K_{OW}-range was to be expected since lipophilicity forms the basis of the regression equations used. Mixtures like dioxins or polychlorinated biphenyls must be investigated congener-specifically because the different congeners occur in different environmental concentrations and act differently in food chains. All models ignore the transformation of a compound into its degradation products. When entering starting concentrations this also has to be taken into account. With respect to the submodels of the exposure module, the following remarks are essential.

Bioaccumulation fish: Estimation of the bioconcentration potential in fish poses two basic problems for lipophilic chemicals: on the on hand, no reliable data for the dissolved fraction in the water phase are available as a consequence of the high and variable fraction of suspended matter. An estimation of the dissolved fraction is possible, but involves high uncertainty. On the other hand, accuracy of the K_{OW}/BCF-relationship decreases with increasing lipophilicity. Normally, the model provides accurate results for non-superlipophilic compounds (log K_{OW} < 6) within an error of one order of magnitude. However, for some chemicals deviations of up to two orders of magnitude may occur. The model underestimates the bioconcentration of numerous superlipophilic substances with deviations amounting to between 2 and 3 orders of magnitude. For instance, the model reveals relatively good applicability for low-chlorinated PCB. For high-chlorinated PCB, the measured values are underestimated by 1 to 2 orders of magnitude. Concentrations for fatty species (e.g. eel) are not representative since the values are extremely high. Thus, the model should not be used for fatty species. Regarding the validation of regression equations for highly lipophilic compounds, a more thorough selection (by test method, test duration, etc.) of experimental bioconcentration factors is advisable in order to reduce the range of deviations. The question arises as to whether dioxins are the only compounds that justify the polynomial curve of the regression relationship. Further lipophilic chemicals should therefore be investigated. However, the need for further research does not seem to be urgent for this submodel.

In order to consider the variability of the fat content (i.e. the rudimental property of the fish regarding bioconcentration), the BCF should be referred to the fat content of the fish. This is justifiable for two reasons: (1) An estimation based on the whole fish may lead to high deviations due to variability. (2) Concentrations reported in the literature are mostly provided on a lipid basis. This validation study was carried out on the basis of fish from the River Rhine. But 90% of consumable fish originate from the sea (FÜRST 1995). Furthermore, concentrations in seafood are 2 orders of magnitude lower than those of fish from the River Rhine (BML 1993). In order to achieve a more realistic estimation of the chemical's intake caused by fish consumption, the differences between seawater vs. freshwater fish should be investigated.

Biotransfer into milk and meat: The biotransfer model is applicable for persistent organic chemicals of low lipophilicity, for which a steady state is justified. It then produces a conservative estimation of concentrations. Typical errors for substances with log K_{OW} < 7.5 can be up to a factor of 10. For more lipophilic chemicals, significant overestimations of 2 orders of magnitude are possible becau-

se the model does not consider a reduced resorption. Carry-over rates should be preferred if available. Airborne substances, such as benzene or 1,2-Dichloroethane, are not modelled appropriately by the TGD approach. The biotransfer model does not consider milk alone – it claims to comprise all dairy products. Depending on the fat content and the method of production, this fact leads to highly deviating concentrations. The model's scope of applicability seems to be small and consideration of the fat content seems to be advisable. Numerous chemicals lie beyond the scope of applicability; the application of a more complex model is obvious which, however, is not advisable for the screening stage of risk assessment. In addition, uncertainties arise from the fact that humans not only consume beef (which forms the basis of the regression equation), but also other sorts of meat or even no meat at all.

Uptake into plants: Various processes are important with regard to the uptake of chemicals into plants. By considering uptake from soil and gaseous diffusion with air, two rudimental processes are integrated. But consequently, this excludes the application for substances for which the deposition of particles is a major process (i.e. hardly water soluble and simultaneously highly adsorbing compounds, e.g. OCDD or PCB 180). Especially due to ignoring the deposition process, realistic or, at best, measured particulate fractions are important. With respect to the mobility and uptake into plants, the partition between gaseous and particulate phase was found to be a sensitive process. Hence, this submodel should be regularly updated to the present state of scientific knowledge. For the plant model, the impact of errors for estimating gas/particle partitioning will be reduced if (as proposed in TRAPP AND MATTHIES (1998), Pages 245-250) deposition of particles is considered. The particulate fraction according to the TGD seems to deliver high fractions. The type of plants consumed poses a further problem: if predominantly deep-growing plants are consumed, the resuspension of particles from the soil onto the plant (for strongly particle-bounded compounds) or the volatilisation off the soil into the plant (for chemicals with a high vapour pressure) will play a role. The model neglects both processes. It is surprising that the model contains as a parameter the fraction of air in a plant, which will never have a decisive impact on any result and, hence, can be neglected. Nevertheless, the ratio of key and redundant parameters is nearly unity for this model. It is therefore highly relevant against the background of its purpose (which of course only holds for compounds having deposition, resuspension and volatilisation as negligible processes). Serious problems occur when calculating root concentrations: A comparison of the model's outcome with measured concentrations is impossible since measured pore-water concentrations for the soil are missing. As a result, the partition coefficients (K_{SW} and K_{PW}) involved could not be confirmed experimentally. But even if the calculated concentrations correspond to real concentrations, they will only be valid for fine roots, those indeed that are not consumed by humans. Concurrently, the standard scenario assumes a relatively high intake of roots based on the consumption of potatoes (TGD, Chapter 2, Appendix VII, Tab. 5). But the estimations are never applicable for these tubers, leading to incorrect and deceptive interpretations. Consequently, each user of the model has to correct this value before carrying out a calculation. The intake rates for tubers (e.g. potatoes) and thick roots (e.g. carrots) should rather be assigned to the upper plant parts.

Exposure of humans: As a consequence of over- and underestimations in the submodels, individual errors may balance out and feign an apparently good result. In addition, the parameter set of the standard scenario can lead to the relevancy of an exposure pathway being falsely assessed. Of course, the proposed standard parameter values may be used to rank chemicals. But the models of the TGD are too complex for simple ranking procedures. The bioconcentration potential of a chemical could also be assessed without quantifying concentrations. However, if the goal of the models is to provide a realistic assessment of the total daily dose, improvements of the parameter values are necessary. This requires revision of the standard parameter values, including adoption of the values to the scientific theory (e.g. potatoes must not be considered as roots). Because of the highly aggregated intake rate, the system calculates possibly misleading fractions of the relevancy of certain exposure pathways. A more detailed consideration of intake rates is advisable. A separation (valid for a German region!) of the rates with respect to the fat content of the food product would be useful: e.g. dairy products should be divided into milk, cheese, and butter; meat should account for various sorts of meat; and fish should be separated into seawater and freshwater fish. Furthermore, for plants a separation with respect to fine and thick roots, cereals and vegetables is advisable. Individual values are well documented, e.g. in ADOLF ET AL. 1995 or EPA 1997C. This separation is also a necessity for realistically calculated contributions of certain exposure pathways. For instance, the contribution to the total PCDD exposure of 1/3 fish, 1/3 milk and 1/3 meat can only be obtained by using more detailed intake rates. A switcher, which enables users to turn a certain exposure pathway on or off, would be useful in this context with respect to the software.

Since the models' purpose is to characterise risk on a regional and local scale, respectively, it is inconsequent to choose a standard region and a standard person representing the European average. Taking average values for a specific region would be more consistent. Due to heterogeneity within the European Union, the implementation of various regional scenarios and exposure groups would then be required (e.g. scenarios for adults and children for a northern, central and southern European region).

The ratio of emission and degradation rates for all compartments is essential for estimating the exposure because it determines the background concentrations. These are important input parameters for the exposure module. Hence, all conclusions regarding the regional distribution model (BERDING 2000) hold also for estimating the total daily dose. Whereas a combination of several parameters determines the background concentration, which is provided by the complex regional distribution model, the exposure module with its different models depends strongly on the lipophilicity of the substance. Statements depending on the octanol/water partition coefficient are accordingly often possible. This is comprehensible since the plant model depends predominately and other models exclusively on the K_{OW}. This implies that the parameter uncertainty will make a minor impact on low partition coefficients, since the chemical is then ingested directly by air and water. The corresponding concentrations in air and water are calculated by the regional distribution model, which depends on the K_{OW} to a significantly lower degree. In this case, the most effective way to reduce uncertainties is a better definition of scenarios (e.g. turning away from the generic scena-

rio towards more realistic scenarios). Highly lipophilic compounds are ingested indirectly via the food chain and the high uncertainties intrinsic to high-partition coefficients influence the result.

A comparison of the findings from scenario and probabilistic analyses shows that the variation range of the total daily dose is higher for lipophilic compounds than for the rather hydrophilic compounds. In addition, changing the scenario influences the uncertainty in the total daily dose most significantly for hydrophilic compounds, whereas reducing the parameter uncertainty most significantly influences the resulting uncertainty for lipophilic compounds. This relationship is illustrated in Fig. 12.1 and is justified on the basis of the uncertainty analyses results, the database and scientific theory. A quantification was not possible due to the insufficient database. However, the resulting interception of both curves is surmised between log K_{OW} 3 and 5.

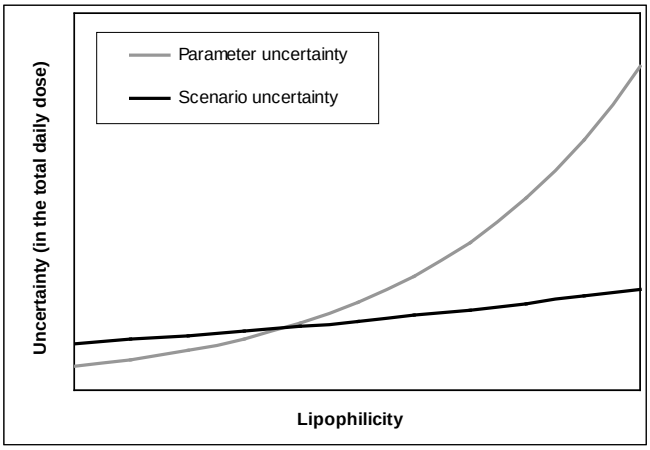

Fig. 12.1 Illustration of the impact of parameter and scenario uncertainty, respectively, on the total daily dose.

It must be kept in mind that this conclusion holds for the implemented models. Real accumulation processes are much more complicated. But models should be simplifications and the most relevant criteria for describing the accumulation of chemicals in food chains, namely lipophilicity, is considered. For certain chemicals, for which the dose is not dominated by lipophilicity, the models should be improved or the user should be warned of higher uncertainties in the calculations. The high relevancy of the exposure module in comparison to other parts of the TGD exposure assessment is comprehensible for lipophilic substances. But also for directly ingested chemicals, only the parameters of the substance and those of the regional distribution model are important. The parameters of the sewage treatment plant are only important for readily biodegradable and slowly absorbable chemicals, and if a large fraction of the regional population is connected to the sewer system (e.g. LAS in NRW). But again, an alteration of other sensitive parameters has a stronger impact on the result. The sewage treatment plant model may of course be important for estimating local concentrations, but it only plays a minor rule in the estimation of human exposure. Consequently, it would be consistent if the user, on demand, were able to switch off this model and use a more simple one. A more simple model, leading to equal results for most chemicals, is proposed in BERDING (2000).

12.2 Database

The database often limits conclusions: Despite a large amount of collected parameters, the claim of highly qualitative data was seldom fulfilled. Frequently investigated congeners of PCDD and PCB are suitable for the validation of a terrestrial food chain model. The several measurements of PCB provide a valid database for the aquatic environment. In particular, due to the homogeneous data for different environmental media of diverse monitoring programmes, accurate statements on the deviation of model results to measured data could be made. Also a good database is available for DEHP and HHCB for the aquatic environment. The database is poor for the remaining substances. Regarding the variability of data, a constant concentration in the environment often had to be assumed, concentration values are not always available for the considered time scale (at the beginning of the 1990s). This assumption is justifiable for benzene because the BUA (1993) points out constant emissions. In contrast, for DEHP a significant reduction of emission rates is assumed within the 1980s. In general, the material was inadequate to investigate temporal concentration profiles for all of the substances used. Spatial variability is also critical. Many measurements were carried out in the neighbourhood of emission sources and provide non-representative data. Furthermore, measurements for compounds such as DEHP, EDC or benzene indicate strongly decreasing concentrations from urban to rural areas. Although sewage sludge application represents a potential burden of plants and the overall food chain, the fate of LAS, HHCB and EDTA is hardly known in the terrestrial food chain. For exposure assessments one must distinguish between the substance itself and its degradation products, which is not possible for LAS. In this paper, the assumption is made that the LAS mixture is well represent by the properties of C_{12}-NaLAS. Against this background, further monitoring studies are required. As stated by the *Chloraromaten-programm* (NRW 1991A and 1991B), further monitoring programmes providing homogeneous concentrations in several compartments are needed. An additional field of applicability is therefore the combination of the models with measurements. *Before* doing costly and time-consuming experiments or measurements, the models are already able to reveal relevant processes and indicate which results can approximately be expected. This is valuable information: Confirmation of a hypothesis achieved from a model is easier to achieve than the construction of a hypothesis based on difficult to interpret and costly experiments. Hence, a combination of the model results and measurements is able to improve the efficiency of monitoring programmes.

12.3 Classes of chemicals posing problems

Exposure of the substances investigated here is chiefly determined by the intrinsic properties of the chemical. If a new notified substance shows the same properties, similar results may be expected. For chemicals and classes of chemicals, respectively, as listed in Tab. 12.1, limitations regarding applicability of the models occur.

Tab. 12.1 Evaluation of the applicability of problematic substances.

	Regional model	Exposure module	Overall system	Survey on conceivable problems
Inorganic compounds	±	-	-	The models may only be applied for inorganic chemicals if PC-data, partition coefficients, bioavailability, bioaccumulation and biotransfer factors are available. Quantifiable values are, however, already mostly missing for the physico-chemical properties. But also inorganic substances with quantifiable data (e.g. mercury with its relatively high vapour pressure) causes problems, since the regression equations are exclusively based on organic chemicals. Bioaccumulation of metals is moreover based on other mechanisms than for organic chemicals and, particularly, essential metals such as zinc do not show the typical dose-response relationship (CHAPMAN ET AL. 1998). For theoretical reasons, this fact forbids the application of a constant accumulation factor. Nevertheless, the TGD and JAGER (1998) provide some guidance on how to apply the models for metals. However, this approach cannot be applied in general and inherent uncertainties are not quantifiable.
Superlipophilic compounds	+	±	±	The estimation of bioaccumulation and other partitioning processes is principally highly uncertain for superlipophilic chemicals (i.e. log K_{OW} > 6). In addition, other processes become important, especially for these compounds. It follows that estimations are not just highly uncertain, they are even conceptually doubtful. Estimated values should be replaced by measured ones.
Persistent compounds	+	+	±	The models are very applicable for these compounds, since persistence is a presupposition for a ubiquitous and constant occurrence. Problems arise if a chemical risk is to be characterised on a continental or global scale.
Surfactants	±	±	±	Surfactants such as tensides or numerous pesticides contain a potential to accumulate in the food chain because adsorption on biological surfaces may also lead to accumulation. The estimated uptake of chemicals with a high adsorption capacity may result in incorrect bioconcentration factors. However, due to their high adsorption capacity these chemicals are not expected to be able to pass the regarding biomembranes. Integrated estimation functions must not be applied and estimated bioconcentration factors, respectively, have to be replaced by more realistic data.

	Regional model	Exposure module	Overall system	Survey on conceivable problems
Polar and dissociating compounds	+	±	±	Many of the produced and emitted chemicals show electrostatic interactions with the aqueous phase and are therefore termed as polar. Some of these dissociate under environmental conditions. Thus, it cannot be expected that the K_{OW} is a good descriptor for these chemicals. Ions do not have a measurable vapour pressure and the concept of partitioning (or the fugacity concept) is not applicable. (For ideal gases the fugacity in air is – per definition – equal to the vapour pressure BARROW 1973). Sorption and accumulation depend on the dissociation constant of the chemical and on the pH-value of the medium (Section 6.3). Both are ignored in the TGD. As a result, the estimated partition coefficients will either overestimate the real partitioning (if not only the neutral molecules are considered) or underestimate it (in the case of ion-taping).
Mixtures, metabolites and complexes	-	-	-	Risk assessment is carried out for an individual substance. No possibility to take the behaviour of metabolites into account exists. Moreover, neither synergistic nor antagonistic combinatory effects of mixtures are considered. But mixtures always occur in the environment. Since the use of average values is not advisable for exposure assessment, every substance and congener, respectively, has to be investigated separately. It is also a problem to apply the models to complexes since the complex will behave in a different manner to that of the individual substance. Consequently, the models are not applicable for EDTA (as acid) since it occurs only as salt or a complex in the environment.

Meaning of the symbols:

+ Application is possible in the scope of quantifiable uncertainties.

± Application is possible, but uncertainties are hardly to quantify and estimation functions should not be used.

- Application is impossible.

In order to solve some of the arising problems, integration of substance-specific regression equations would be advisable. Several regression equations are laid down in the TGD. But these are not integrated into EUSES. It would also be helpful to integrate regression equations that are not based on the K_{OW} alone. An alternative are QSPRs (e.g. molecular connectivity indices). Many of these regression equations were demonstrated as accurate methods to describe the behaviour of organic chemicals in the environment. QSPRs have already been successfully correlated with sorption coefficients of the soil, n-octanol/water partition coefficients, bioconcentration and biotransfer factors (KIER AND HALL 1985). For the latter correlation in particular, it was shown that a higher or at least an equal accuracy is achieved in relation to the TGD regression equation. Other limitations of the applicability may be avoided by slightly adopting the models. Estimation functions of partition coefficients for dissociating chemicals can be adopted in order to avoid conceptual overestimations. The according procedure is presented in Section 6.3. The effort required is very low.

12.4 General remarks regarding applicability

Some remarks concern all models. These are presented in this section. All TGD models assume a steady state, and partly also equilibrium in the thermodynamic sense. Consequently, the models are only useful for long-term periods, i.e. several months or even years, for which the environmental concentrations are constant over the exposure period. But since chemical uptake may occur very quickly, uptake and elimination rates must relate to each other. This is especially important for benzene, 1,2-dichloroethane and many waterborne chemicals. In general, it holds for all substances for which uptake occurs from the same media as emission: It can then be expected for compounds with short half-lives that elimination is faster than uptake. For this reason, the estimated bioaccumulation will be unrealistically high. On the contrary, a fast uptake in relation to elimination leads to a bioaccumulation effect, even if the substance is readily biodegradable. These phenomena are caused by ignoring dynamic processes and has to be investigated on a case-to-case basis under consideration of kinetic information for both processes. With the exception of the plant model and the BCF estimation for superlipophilic substances, the models are based on linear relationships. Due to several factors involved in the uptake process, linearity can hardly be expected for the real environment. For instance, continuation of the linear regression relationship for the BTF estimation of superlipophilic substances is unrealistic. However, these uncertainties are not yet quantifiable and must be evaluated on a case-to-case basis.

As previously shown, when interpreting the underlying assumptions in a strict sense potential applicability declines drastically. In order to solve this problem, a set of alternative models for various fields of application should be developed and made available. When using the software, warnings should appear if a model is applied outside its range of applicability. If no appropriate model is available, it is better not to use a model at all rather than use an inaccurate one , i.e. it is advisable to look for other sources of information. Measures for quality assurance are unfortunately underdeveloped within the existing system. This is not caused by a lack of concepts, but rather due to ignoring the importance of such measures. The low transparency and the black-box handling is the major point of concern not only for the software, but also for the entire system. Accordingly, providing more transparency by bringing all statements on model limitations into the documentation and user-interface of EUSES is advisable.

Finally, the question of how to characterise the model results remains. The quantification of exposure by means of averaged generic assumptions dominates. It does not consider – corresponding to the idea of generic modelling – spatial and temporal variability or true uncertainty. Moreover, a fictive averaged subpopulation prevails, rather than an actual exposure group. Averaged and worst-case values are used instead of parameter distributions, which could represent actual uncertainty. In this way, the exposure assessments according to the TGD lead to a mixture of averaged, reasonable worst-case and real worst-case results. A cumulative worst case, which in principle is possible for the models, was not observed within this study. This phenomenon is explained by the parameter values used. Particularly for the sensitive emission rates, average values were applied. But a cumulative worst case cannot be excluded if worst-case values are entered for various sensitive parameters. Due to the mixture of averaged and worst-case values and the uncertainties arising from the model theory, the results cannot be characterised in general. They tend

rather to overestimate, but have also been known to underestimate, too. This statement holds in fact for the standard parameter set. All in all, the claim of the models “to be on the safe side” is not always fulfilled. In order to avoid serious errors in risk characterisation (underestimations lead to a false sense of security) the models should only form the basis for decisions if their applicability is judged from case to case. If the models are used in the form of a black-box and without this judgement, they will miss their purpose. A further objective of the models is the possibility to refine the calculations and adapt the assessment to a more realistic situation. The findings in this paper show that refinement of the standard environment and standard exposure group, respectively, to a more realistic scenario leads to more realistic results. Thus, this aspect of the model objective is realised.

Tab. 12.2 Expected deviations (for both the standard and realistic scenario) in OoM. No values are given if monitoring data are missing or inadequate.

Substance	Water	Air	Soil	Fish	Milk	Meat	Plants	Humans	Maximum
TCDD	-	1	1-4	2	1	1	1	1-3	4
PeCDD	-	1	1-2	2	1	1	2	1-3	3
HxCDD	-	1	1-3	2	1	2	2	1-3	3
HpCDD	-	1	1-2	2	1	2	-	1-4	4
OCDD	-	2	1-3	2	2	-	-	1-4	4
PCB 28	-	-	-	1	1	2	2	-	2
PCB 52	-	-	-	1	1	2	2	-	2
PCB 101	-	-	-	2	1	2	1	-	2
PCB 138	-	-	-	2	1	2	1	-	2
PCB 153	-	-	-	2	1	1	1	-	2
PCB 180	-	-	-	2	1	1	1	-	2
DEHP	3	2	1-3	2	-	-	2	1-3	3
HHCB	1	-	-	2	-	-	-	-	2
EDC	2	2	-	-	-	-	-	1-2	2
BENZ	1	2-3	-	-	-	-	-	1-2	3
EDTA	1	-	-	1	-	-	-	-	1
LAS	1-2	-	3-4	-	-	-	-	2-3	4

The effect assessment part of the TGD is characterised as a conservative approach with possible deviations to experimental values up to three orders of magnitude for ecosystems and with an unknown degree of conservatism for humans (JAGER 1998). This characterisation suggests a comparison with the findings of this paper. It is interesting to contemplate whether the uncertainties in the entire risk assessment procedure arise from the effect assessment or from the exposure assessment part. Tab. 12.2 subsumes the deviations of the estimated dose to alternative estimations and estimated concentrations to monitoring data, respectively. The deviations are rounded up and represent an upper limit of possible errors in each submodel. The values depend strongly on the quality of the database and should rather be understood as an indicator of uncertainties. The table shows that one has to expect deviations of up to 2-4 orders of magnitude for humans. Deviations are often smaller, especially when realistic input parameters are used. Furthermore, the values will only be informative for highly lipophilic chemicals if the parameter uncertainty is kept low. Furthermore, the uncertainty analyses revealed that the 90%-ile and 10%-ile span a range of up to 5 or-

ders of magnitude for superlipophilic chemicals, a fact that raises uncertainty. However, for chemicals of low and medium lipophilicity the parameter uncertainty is negligible or lies between the stated deviations. Altogether, it can be concluded that the inherent uncertainties are often assessable and that the overall system provides the possibility to reduce the initially high uncertainties by means of appropriate scenarios and parameters to a magnitude that is smaller than or similar to that of the effect assessment.

12.5 Conceptual suggestions

Applicability of the models is limited by the underlying assumptions made during their construction. Unfortunately, the user is often not informed about the resulting limitations. The existence of a uniform methodology implemented as software for the evaluation of chemical substances tempts users to believe in a fast, automated risk assessment, where interpretation and evaluation of the procedure and database are lacking. For this reason, the tool used (here: EUSES) must guide users and reveal the limitations of the models. One should keep this aspect in mind with each improvement. A regular adaptation of TGD and EUSES to scientific knowledge is necessary and has already been proposed in the TGD: “The guidance is legally not binding, and the competent authorities may use other methods or approaches if they are more appropriate, provided that they are scientifically justified. The technical procedures ... may be subject to further refinement and development in the future.” (TGD, Chapter 1, page 6). A major conclusion in this paper is that detailed improvements of the existing models are useful, but no approach exists leading to acceptable results for all types of chemicals. There will never be a “supermodel”. The best procedure towards a commonly accepted and widely applicable exposure assessment is to provide a modular set of alternative approaches. This includes guidance on how to apply a certain approach. Users of the models should be able to carry out probabilistic assessments on demand, since they are (1) rudimental for the evaluation of models, and (2) helpful to obtain a more realistic evaluation of exposure on a local and regional scale.

Submodels should be interchangeable. For instance, one should replace the regional Level 3 regional distribution model with a Level 4 model if time-dependent concentration profiles are desired. Often, users may simply wish to interchange a regression equation with another one. The user should be able to choose between alternative models in such cases. For example, 19 different valid equations for estimating K_d for a variety of chemical classes are provided by the TGD. However, only one of these classes (one that is mainly only valid for very hydrophobic chemicals) is implemented in EUSES.

The selection of alternative models could be fixed during the implementation of the software; or users could be enabled to implement and integrate their own models. The best method of technical realisation (e.g. in the form of a component-based software engineering process) still has to be verified. In any case, modularisation would technically require a laborious definition of interfaces within the different modules. Experience from both model construction and software engineering reveal that such effort is necessary and worthwhile in order to facilitate a long-term use of the overall system.

The realisation of more modularity provides increased transparency of the overall system and facilitates the integration of new concepts for the evaluation of chemicals: Representatives of the risk assessment practice often complain about the current inert and time-consuming procedure of regulating chemicals and request acceleration of the process. One method is to bring more flexibility into the process of risk assessment and to consider the precautionary principle in the case of an insufficient database (AHLERS 1999). But even for excellent databases, certain risks posed by chemicals will never be quantifiable by the current PEC/PNEC approach because, for chemicals with a potential for long-range transport or for the marine environment, the concept fails. Such compounds are termed as POPs (*persistent organic pollutants*) and pose problems for the following reason: Measurements reveal relatively high concentrations of these chemicals in the marine environment and polar regions. This is caused by the interaction of bioaccumulation, persistence, partitioning and transport, and represents a major threat. Thus, POPs can pose a risk on a continental or a global scale. The current risk assessment methodology distinguishes between a local and a regional scale. It is true that the existing methodology calculates PECs on a continental scale, but due to the lack of scientific evidence, characterisation of the risk is not being carried out. Consequently, the EU methodology does not deal sufficiently with the risk on continental and global scales. Consideration of intrinsic chemical properties and the characterisation of a substance by, first of all, neglecting toxicological effects and emission rates is proposed as a solution (SCHERINGER AND BERG 1994, BERG AND SCHERINGER 1994). This proposal is termed as threat identification in the following paragraph.

The purpose of EUSES is to support the evaluation of new and existing chemicals. After realising the above-mentioned aspects, EUSES could be applied for a wide variety of recently discussed approaches for chemical evaluation (Tab. 12.3).

Tab. 12.3 Effort of conceptual changes.

Type of evaluation	**Spatial scale**	**Effort of the implementation**	**Necessary changes**
Threat identification by means of intrinsic properties	Continental and global	Low (as soon as evaluation criteria are available)	New models are not necessary
Generic risk assessment	Local and regional	None	No changes, the method is available
Scenario-based risk assessment	Local and regional	Low	Compilation of suitable scenarios, extension of EUSES by some technical features
Probabilistic risk assessment	Local and regional	High	Compilation and evaluation of suitable probability distributions as reference, extension of the entire system if necessary

The models, including their standard parameters, can be applied for a generic risk assessment. After adapting the numerous parameters, the generic assessment can be refined and applied suc-

cessfully for a real region. The implementation of concrete regions and exposure scenarios is imaginable as an improvement and, likewise, as standardisation. This would only require slight technical changes and the compilation of parameter sets for the scenarios. Exemplary parameter sets in the framework of a scenario-based risk assessment would be a northern, central and southern European region or regions with different population densities combined with different exposure groups.

An extension of the system is possible in two directions: In the direction of the precautionary principle in the case of insufficient data on the one hand. And towards more realistic and more scientific estimations in the case of sufficient data on the other hand. If one desires to account for the precautionary principle and consider threats on a continental and global scale, an appropriate methodology will be easy to integrate into the existing system. All information required for the evaluation of the persistence, bioaccumulation and long-range transport potential is already provided by the existing system or can easily be derived from existing models. (e.g. a characteristic travel distance to describe the long-range transport potential from the SimpleBox model). If a modularisation of the system is realised, after “turning off” unnecessary modules an evaluation based on the intrinsic chemical properties will become rapidly and easily possible. If, however, a more realistic and more scientific assessment is desired, one must also take into account which uncertainties may arise and which factors may remain unknown. A probabilistic analysis is a suitable method for the quantification of uncertainties.

12.6 Concluding remarks

After improving individual models and, particularly, after integrating new models or concepts new validation studies must be carried out. Often, one observes a discrepancy between the effort made towards the construction of models and the effort put into their subsequent quality assurance. On the one hand, high costs and endeavours are funded for the development and implementation of projects, but studies with the objective of analysing the applicability and limitations of such models are neglected. This insight is not new (MACKAY 1988):

> *And it is a long time-known insight that the existing review system does not give the models the scrutiny they need and deserve.*

Due to the fact that the models put forward by the TGD and EUSES, respectively, play a central role in the regulatory notification of chemical substances, the discrepancy poses problems. This study confirmed that the models fulfil their purpose within the framework of – in many cases – quantifiable uncertainties. Remaining uncertainties will be quantified more exactly, or even avoided if more effort is put into the analysis of the models. Changes to models require new evaluations and a final evaluation of the models can only be completed after their adaptation to all relevant fields of application.

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Appendix

A.1 Model parameters and probability distributions

The *Standard* column in the following tables shows the TGD default value of the standard scenario and the *Realistic* column shows the parameters of the Realistic scenario for both the point estimates and the probabilistic estimations. If available, the coefficient of variation (CV = SD / M) is listed in column *CV*.

Tab. 1 Volumetric parameters for the regional distribution model.

Name	Description	Unit	Standard	Realistic	CV	Source
area Reg	Area of region	m²	4E10	3.44E10 U(3.4056,3.434)		NRW (1998)
AreaEU	Area of EU	m²	3.56E12	3.56E12 (constant)		
depthAgric	Mixing depth of agricultural soil	m	0.2	0.2 L(0.2, 0.2)	1	CV: DTSC (1993)
depthInd	Mixing depth of industrial soil	m	0.05	0.05 L(0.05, 0.05)	1	CV: DTSC (1993)
depthNat	Mixing depth of natural soil	m	0.05	0.05 L(0.05, 0.05)	1	CV: DTSC (1993)
depthSed	Mixing depth of sediment	m	0.03	0.03 L(0.03, 0.03)	1	CV: DTSC (1993)
depthWater Cont *	Depth of water (continent)	m	3	3 (constant)		
depthWater Reg	Depth of water (region)	m	3	3 L(3, 4.74)	1.58	CV: DTSC (1993)
fAgric Cont	Area fraction agricultural soil (continent)	-	0.27	0.27 (constant)		
fAgric Reg	Area fraction agricultural soil (region)	-	0.27	0.52 L(0.520, 0.101)[#]	0.192	NRW (1998) for realistic value, STATISTISCHE ÄMTER (1998) for distributions
fInd Cont	Area fraction industrial soil (continent)	-	0.1	0.1 (constant)		
fInd Reg	Area fraction industrial soil (region)	-	0.1	0.202 L(0.1917, 0.047)[#]	0.240	NRW (1998) for realistic value, STATISTISCHE ÄMTER (1998) for distributions
fNatural Cont	Area fraction natural soil (continent)	-	0.6	0.6 (constant)		
fNatural Reg	Area fraction natural soil (region)	-	0.6	0.26 L(0.2707, 0.102)[#]	0.393	NRW (1998) for realistic value, STATISTISCHE ÄMTER (1998) for distributions
fWater Cont	Area fraction water (continent)	-	0.03	0.03 (constant)		
fWater Reg	Area fraction water region	-	0.03	0.018 L(0.0176, 0.007)[#]	0.395	NRW (1998) for realistic value, STATISTISCHE ÄMTER (1998) for distributions
heightAir	Atmospheric mixing height	m	1000	L(1000, 461)	0.461	CV: from ETIENNE (1997)

*: Not sensitive.

[#]: Values are based on the data for the five NRW *Regierungsbezirke*.

Tab. 2 Process parameters for the regional distribution model.

Name	Description	Unit	Standard	Realistic	CV	Source
CollEffAer	Aerosol collection efficiency	-	2E5	20000 L(20000, 10)	5E-4	ETIENNE (1997)
DepRateAer	Aerosol deposition velocity	m/d	86.4	86.4 L(86.4, 129.6)	1.5	CV: DTSC (1993)
Erosion	Soil erosion rate of regional system	m/d	8.22E-8	8.22E-8 L(8.22E-8, 1.644E-8)	0.2	CV: DTSC (1993)
FFlowOut Reg	Fraction of water flow from continental to regional scale	-	0.034	0.029		AreaReg / AreaEU * DepthWater
FrunoffSoil	Fraction of rain water running off soil	-	0.25	L(0.4, 0.22)	0.55	ECETOC (1994) for realistic value, CV: DTSC (1993)
kasl air	Air-film partial mass-transfer coefficient (air-soil interface)	m/d	120	120 L(120, 8.64E6)	72000	ETIENNE (1997)
kasl soilair	Soil-air partial mass-transfer coefficient (air-soil interface)	m/d	0.48	0.48 L(0.48, 8.64E6)	1.8E7	ETIENNE (1997)
kasl soilwater *	Soilwater-water film partial mass-transfer coefficient (air-soil interface)	m/d	4.8E-5	4.8E-5 L(4.8E-5, 8.64E6)	1.8E11	ETIENNE (1997)
kawAir	Air-film partial mass-transfer coefficient (air-water interface)	m/d	120	0.01 * (0.004 + 0.00004 * windspeed) * (0.032 / mol-weight)$^{0.4047}$		BRANDES ET AL. (1996)
kawWater	Water-film partial mass-transfer coefficient (air-water interface)	m/d	1.2	0.01 * (0.3 + 0.2*windspeed) * (0.018 / mol weight)$^{0.4355}$		BRANDES ET AL. (1996)
kwsSed	Pore water film partial mass-transfer coefficient (sediment-water interface)	m/d	0.0024	0.0024 L(0.0024, 8.634E6)	3.6E9	ETIENNE (1997)
kwsWater *	Water-film partial mass-transfer coefficient (sediment-water interface)	m/d	0.24	0.24 L(0.24, 8.64E6)	3.6E7	ETIENNE (1997)
Rainrate	Average daily precipitation	m/d	1.92E-3	1.86E-3 L(2.32E-3, 4.19E-4)	0.181	NRW (1995A, 1995B, 1995C)
SETTLEveloc-ity	Settling velocity of suspended solids	m/d	2.5	2.5 L(2.5, 0.75)	0.3	CV: DTSC (1993); value for deposition in kg/m^2/d
windspeed	Wind speed in the system	m/d	2.59E5	2.69E5 L(2.69E5, 4.82E4)	0.179	NRW (1995A, 1995B, 1995C)

*: Not sensitive.

Tab. 3 Other model parameters for the regional distribution model.

Name	Description	Unit	Standard	Realistic	CV	Source
BIOwater *	Concentration of biota	kg/m³	0.001			
BOD	Mass of O_2 binding material	kg/eq/d	0.054	0.06 U(0.054, 0.06)		German standard for BSB
ConJunge	Constant of Junge equation	Pa m	0.01	0.172 (constant)		FALCONER AND BIDLEMAN (1997)
FAirSoil *	Volume fraction air in soil	m³/m³	0.2			
FconnectSTP	Fraction connected to sewer systems	-	0.7	0.92 L(0.904, 0.045)[#]	0.05	NRW (1998) for realistic NRW-values, STATISTISCHE ÄMTER (1998) for distributions
FInfSoil	Fraction of rain water infiltrating soil	-	0.25	0.25 L(0.25, 0.1375)	0.55	CV: DTSC (1993)
FocSed	Weight fraction of organic carbon in sediment	kg/kg	0.05	0.05 U(0.04, 0.05)		EUSES standard value and MACKAY ET AL. (1991-1997)
FocSoil	Weight fraction of organic carbon in soil	kg/kg	0.02	0.02 L(0.0577, 0.03627)	0.629	SCHEFFER & SCHACHTSCHABEL (1994)
FocSusp	Weight fraction of organic carbon in suspended matter	kg/kg	0.1	0.1 U(0.1, 0.2)		EUSES standard value and MACKAY ET AL. (1991-1997)
FSolidSed	Volume fraction of solids in sediment	m³/m³	0.2	0.2 L(0.2, 0.04)	0.2	CV: DTSC (1993)
FSolidSoil	Volume fraction of solids in soil	m³/m³	0.6	0.6 T(0, 0.4, 1)		ETIENNE (1997)
FWaterSed	Volume fraction of water in sediment	m³/m³	0.8	0.8 T(0.5, 0.8, 0.999)		ETIENNE (1997)
FWaterSoil	Volume fraction of water in soil	m³/m³	0.2	0.2 U(0.2, 0.3)		EUSES standard value and MACKAY ET AL. (1991-1997)
N Reg *	Number of inhabitants in region	-	2E7	17,816,100 (constant)		NRW (1998)
NEU *	Number of EU inhabitants	-	3.7E8	3.7E8 (constant)		
OHconcair *	Concentration of OH-radicals in the atmosphere	molec/m³	5E11			
Qstp	Sewage flow	m³/eq/d	0.2	0.202 L(195.67, 22.94)[#]	0.12	NRW (1998) for realistic NRW-values, STATISTISCHE ÄMTER (1998) for distributions
RHOair *	Density of air phase	kg/m³	1.3			
RhoSolid	Density of solid phase	kg/m³	2,500	2500 L(2500, 125)	0.05	CV: DTSC (1993)
RhoWater	Density of water phase	kg/m³	1,000	1000 (constant)		
SurfAer	Surface area of aerosols	m²/m³	0.01	4.2E-5 T(4.2E-5, 1.5E-4, 1.1E-3)		FALCONER AND BIDLEMAN (1997)
SuspEff	Concentration of solids in effluent	kg/m³	0.03	0.03 L(0.03, 0.0234)	0.78	Like river water (IKSR 1996); parameter is only less sensitive
SuspWater Reg	Suspended solids concentration of region	kg/m³	0.015	0.015 L(0.0297, 0.02315)	0.78	Derived from IKSR (1996)
SuspWater-Cont	Suspended solids concentration of continent	kg/m³	0.025	0.025 (constant)		
Temperature	Environmental temperature	°C	11	10 L(10.3, 1.6)	0.15	NRW(1995A-C)

*: Not sensitive.

[#]: Values are based on the data for the five NRW *Regierungsbezirke*.

Tab. 4 Parameters for the plant model.

Name	Description	Unit	Standard	Realistic	CV	Source
k_G (kGrowth-Plant)	Growth rate	1/d	0.035	0.035 L(0.043, 0.022)[$]	0.51	TRAPP ET AL (1997)
Q (QTransp)	Transpiration stream	m³/d	0.001	6.5E-4 L(6.5E-4, 1.2E-4)[$]	0.18	TRAPP ET AL (1997)
A_L / V_L (VLeaf / AREAPlant)	Ratio area / volume of upper plant parts	m²/m³	2500 (=5/2E-3)	1804 L(1804, 999)[#]	0.55	TRAPP ET AL (1997), BÖHME ET AL. (1999)
f_A (FAirPlant)	Air content of upper plant parts	m³/m³	0.3	0.3 (constant)		
f_W (FWaterPlant)	Water content of upper plant parts	m³/m³	0.65	0.65 T(0.47, 0.65, 0.93)[$]		TRAPP ET AL (1997)
f_{Li} (FLipidPlant)	Lipid content of upper plant parts	m³/m³	0.01	7.8E-3 L(7.8E-3, 9.0E-3)[#]	1.15	ELMADFA ET AL. (1990), TRAPP ET AL. (1997), BÖHME ET AL. (1999)
g_L (gPlant)	Conductance (of diffusive gas exchange)	m/d	86.4	86.4 T(8.64, 86.4, 432)		TRAPP AND MATTHIES (1998)
ρ_L (RHOPlant)	Plant density	kg/m³ (FW)	700	750 L(750, 166)	0.22	HUNG AND MACKAY (1997), TRAPP ET AL. (1997), RIEDERER (1990)
b	Correction factor for lipid/octanol difference	-	0.95	0.95 L(0.95, 0.128)[§]	0.13	CV: TRAPP AND MATTHIES (1995)
DW	Dry weight fraction of plant	% (FW)	25	23.5 L(23.5, 5.08)[#]	0.22	BÖHME ET AL. (1999)

[$] Derived from values for lettuce, kale, spinach and wheat.
[#] Derived from values for rye, thistle, dandelion, ribwort, yarrow, lady's mantle, sunflower leaves, trefoil, cereals and maize leaves.
[§] see chapter five.

Tab. 5 Parameters for the biotransfer model meat/milk.

Name	Description	Unit	Standard	Realistic	CV	Source
IC_{Gras}	Intake rate grass	kg/d (FW)	67.7	67.6 L(85,17); co-varied with IC_{Soil} (+0.5)	0.2	MCKONE AND RYAN (1989), MCKONE (1994)
IC_{Soil}	Intake rate soil	kg/d (FW)	0.46	0.46 L(0.4, 0.28); co-varied with IC_{Gras} (+0.5)	0.7	MCKONE AND RYAN (1989), MCKONE (1994)
IC_{Air}	Intake rate air	m³/d	122	122 L(122, 36.6)	0.3	CV: DTSC (1993)
IC_{Water}	Intake rate drinking water	l/d	55	55 L(55, 11)	0.2	CV: DTSC (1993)

Tab. 6 Physiological parameters for humans.

Name	Description	Unit	Standard	Realistic	CV	Source
IH_{Fish}	Daily intake of fish (Adults)	kg/d (FW)	0.115	0.018 L(0.018, 7.2E-3)	0.4	ADOLF ET AL. (1995), CV: DTSC (1993)
	(Children)			7.8E-3, L(7.8E-3, 3.2E-3)	0.4	
IH_{Meat}	Daily intake of meat (Adults)	kg/d (FW)	0.301	0.199 L(0.199, 0.040)	0.2	ADOLF ET AL. (1995), CV: DTSC (1993)
	(Children)			0.110 L(0.110, 0.022)	0.2	
IH_{Dairy}	Daily intake of dairy products (Adults)	kg/d (FW)	0.561	0.214 L(0.214, 0.043)	0.2	ADOLF ET AL. (1995), CV: DTSC (1993)
	(Children)			0.372 L(0.372, 0.074)	0.2	
IH_{Water}	Daily intake of drinking water (Adults)	m³/d	0.002	1.4E-3, L(2.1E-3, 8.4E-4)	0.4	STUBENRAUCH ET AL. (1999), MCKONE AND BOGEN (1991)
	(Children)			6E-4, L(4.5E-4, 1.8E-4)	0.4	
IH_{Plant}	Daily intake of upper plant parts (Adults)	kg/d (FW)	1.2	0.514 L(0.514, 0.103)	0.2	ADOLF ET AL. (1995), CV: DTSC (1993)
	(Children)			0.411 L(0.411, 0.082)	0.2	
IH_{Root}	Daily intake of roots (Adults)	kg/d (FW)	0.384	0.139 L(0.139, 0.028)[$]	0.2	ADOLF ET AL. (1995), CV: DTSC (1993)
	(Children)			0.099 L(0.099, 0.020)	0.2	
IH_{Air}	Inhalationsrate (Adults, active person)	m³/d	20	19.2 L(19, 5.7)	0.3	STUBENRAUCH ET AL. (1999), CV: DTSC (1993)
	(Children)			12 L(12, 3.6)	0.3	
IH_{Soil}	Soil ingestion rate (only 2-5 years old children)	kg/d (FW)	-	2.1E-5, L(2.98E-2, 4.71E-2)	1.58	FINLEY ET AL. (1994)
BIO_{Inh}	Bioavailability for inhalation	-	0.75	0.75 U(0.46, 1)[§]		COPELAND ET AL. (1994)
BIO_{Oral}	Bioavailability for oral uptake	-	1	1 (constant)		
BW[#]	Body weight (Adults)	kg	70	74.5 L(76.91, 12.00)	0.16	BAGS (1995)
	Body weight (Children)	kg		26.1 L(26.69, 4,06)	0.15	

[#] Correlation between body weight and intake rates are considered by means of age-specific consumption data, i.e. adults (men, 25-50 a) and children (7-9 a).

[$] Derived from intake rates of potatoes, onions, carrots.

[§] Only assumed, if not more appropriate substance-specific data are available.

A.2 Substance data survey

Tab. 7 Substance identification.

Substance	Chemical name	Sum formula	CAS
PCDD	**Polychlorinated dibenzo-*p*-dioxins**		**262 - 12 - 4**
TCDD	2,3,7,8 – Tetrachloro-dibenzo-*p*-dioxin	$C_{12}H_4O_2Cl_4$	1746 - 01 - 6
PeCDD	1,2,3,7,8 – Pentachloro-dibenzo-*p*-dioxin	$C_{12}H_3O_2Cl_5$	40321 - 76 - 4
HxCDD-I	1,2,3,4,7,8 – Hexachloro-dibenzo-*p*-dioxin	$C_{12}H_2O_2Cl_6$	39227 - 28 - 6
HxCDD-II	1,2,3,6,7,8 – Hexachloro-dibenzo-*p*-dioxin	$C_{12}H_2O_2Cl_6$	57653 - 85 - 7
HxCDD-III	1,2,3,7,8,9 – Hexachloro-dibenzo-*p*-dioxin	$C_{12}H_2O_2Cl_6$	19408 - 74 - 3
HpCDD	1,2,3,4,6,7,8 – Heptachloro-dibenzo-*p*-dioxin	$C_{12}HO_2Cl_7$	5822 - 46 - 9
OCDD	Octachloro-dibenzo-*p*-dioxin	$C_{12}O_2Cl_8$	3268 - 87 - 9
PCB	**Polychlorinated biphenyls**		**1336 - 36 - 3**
PCB 28	2,4,4‘ – Trichlorobiphenyl	$C_{12}H_7Cl_3$	7012 - 37 - 5
PCB 52	2,2‘,5,5‘ – Tetrachlorobiphenyl	$C_{12}H_6Cl_4$	35693 - 99 - 3
PCB 101	2,2‘,4,5,5‘ – Pentachlorobiphenyl	$C_{12}H_5Cl_5$	37680 - 73 - 2
PCB 138	2,2‘,3,4,4‘,5‘ – Hexachlorobiphenyl	$C_{12}H_4Cl_6$	35065 - 28 - 2
PCB 153	2,2‘,4,4‘,5,5‘ – Hexachlorobiphenyl	$C_{12}H_4Cl_6$	35065 - 27 - 1
PCB 180	2,2‘,3,4,4‘,5,5‘ – Heptachlorobiphenyl	$C_{12}H_3Cl_7$	35065 - 29 - 3
DEHP	Di-(2-ethylhexyl)phthalate	$C_{24}H_{38}O_4$	117 - 81 - 7
HHCB	1,3,4,6,7,8-Hexahydro-4,6,6,7,8,8-hexamethyl-cyclopenta-[g]-2-benzopyrane	$C_{18}H_{26}O$	1222 - 05 - 5
EDTA	Ethylendiaminetetra acetic acid	$C_{10}H_{16}N_2O_8$ (*salt:* $C_{10}H_{12}N_2O_8Na_4$)	60 - 00 - 4 (*salt:* 64 - 02 - 8)
LAS	Linear alkyl benzene sulfonates	$C_{18}H_{30}O_3S$ (*salt:* $C_{18}H_{29}NaO_3S$)	85536 - 14 - 7 (*salt:* 25155 - 30 - 0)
EDC	1,2-Dichlorethane or ethylene dichloride	$C_2H_4Cl_2$	107 - 06 - 2
BENZ	Benzene	C_6H_6	71 - 43 - 2

Tab. 8 Physico-chemical properties.

Substance	M [g/mol]	BP [K]	MP [K]	VP [Pa at 25°C]	WS (25°C) [mg/l]	H [Pa·m³/mol]
TCDD	322 (1)	720 (2)	578 (1)	2.0E-07 (1)	1.9E-5 (1)	3.34 (1)
PeCDD	356.4 (1)	738 (2)	469 (1)	8.8E-08 (1)	1.2E-4 (1)	0.26 (1)
HxCDD	391 (1)	761 (2)	546 (1)	5.1E-09 (1)	4.4E-6 (1)	0.45 (1)
HpCDD	425.2 (1)	780 (2)	538 (1)	7.5E-10 (1)	2.4E-6 (1)	0.13 (1)
OCDD	460 (1)	783 (2)	608 (1)	1.1E-10 (1)	7.4E-8 (1)	0.68 (1)
PCB 28	257.55 (2)	479 (2)	330 (3)	1.0E-02 (2)	8.9E-2 (4)	28.86 (4)
PCB 52	291.99 (2)	633 (2)	360 (3)	3.6E-03 (2)	3.3E-2 (4)	32.23 (4)
PCB 101	326.44 (2)	654 (2)	350 (3)	7.2E-04 (2)	9.4E-3 (4)	24.79 (4)
PCB 138	360.88 (2)	673 (2)	352 (3)	5.4E-05 (2)	1.5E-3 (4)	13.13 (4)
PCB 153	360.88 (2)	673 (2)	376 (3)	4.1E-05 (2)	8.8E-4 (4)	16.65 (4)
PCB 180	395.33 (2)		382 (3)	6.4E-06 (2)	2.3E-4 (4)	10.85 (4)
DEHP	390.56 (5)	658 (2)	233 (5)	1.9E-03 (5)	2.9E-2 (5)	17.51 (2)
HHCB	258.4 (6)	605 (6)		7.3E-02 (6)	1.75 (6)	11.3 (6)
EDTA	292.25 (7)		493 (7)	-	500 (20°C) (7)	-
LAS	348.48 (5)		263 (7)	-	1,100 (5)	-
EDC	98.96 (5)	356 (1)	238 (1)	11,300 (5)	8,600 (5)	96.66 (E)
BENZ	78.12 (5)	353 (5)	279 (5)	12,700 (5)	1,760 (5)	448.30 (E)

	log K_{OW} [-]	log K_{OC} [l/kg]	log K_{OA} [-]	fPa [-]	VP' (12°C) [Pa]	^1X
TCDD	6.8 (1)	5.61 (E)	9.67 (E)	0.32 (E)	2.2E-4 (E)	8.54 (E)
PeCDD	7.4 (1)	6.09 (E)	11.38 (E)	0.93 (E)	7.1E-6 (E)	8.97 (E)
HxCDD	7.8 (1)	6.42 (E)	10.54 (E)	0.98 (E)	2.6E-6 (E)	9.40 (E)
HpCDD	8.0 (1)	6.58 (E)	12.28 (E)	1.00 (E)	3.1E-7 (E)	9.82 (E)
OCDD	8.2 (1)	6.74 (E)	11.76 (E)	1.00 (E)	2.3E-7 (E)	10.25 (E)
PCB 28	5.67 (2)	4.69 (E)	7.92 (10)	0.00 (E)	3.3E-2 (E)	7.15 (E)
PCB 52	5.84 (2)	4.83 (E)	8.22 (10)	0.00 (E)	9.0E-3 (E)	7.58 (E)
PCB 101	6.38 (2)	5.27 (E)	8.8 (10)	0.03 (E)	3.4E-3 (E)	7.99 (E)
PCB 138	6.83 (2)	5.63 (E)	9.51 (10)	0.27 (E)	2.7E-4 (E)	8.40 (E)
PCB 153	6.92 (2)	5.71 (E)	9.37 (10)	0.22 (E)	3.6E-4 (E)	8.40 (E)
PCB 180	7.36 (2)	6.06 (E)	9.88 (10)	0.61 (E)	6.5E-5 (E)	8.82 (E)
DEHP	7.48 (5)	4.94 (9)	10.85 (E)	0.05 (E)	VP (E)	
HHCB	5.9 (6)	4.86 (6)	8.26 (E)	0.00 (E)	VP (E)	8.76 (E)
EDTA	-3.34 (7)	-2.61(E)	-	1.00 (E)	VP (E)	
LAS	1.96 (5)	1.69 (E)	-	1.00 (E)	VP (E)	
EDC	1.46 (5)	2.3 (5)	2.73 (E)	0.00 (E)	VP (E)	3.25 (E)
BENZ	2.12 (5)	1.96 (5)	2.78 (E)	0.00 (E)	VP (E)	3.00 (E)

Sources: (1) SHIU ET AL. (1988), (2) MACKAY ET AL. (1999), (3) BRODSKY (1986), (4) DUNNIVANT ET AL. (1992), (5) RIPPEN (1995), (6) PLASSCHE AND BALK (1997), (7) EC (1996C), (8) BUA (1995), (9) KÖRDEL UND MÜLLER (1995), (10) KÖMP AND MCLACHLAN (1997), (E) Estimated from available data.

M = Molecular weight, BP = Boiling point, MP = Melting point, VP = Vapour pressure, WS = Water solubility, H = Henry-constant, fPa = Particulate fraction, VP' = Vapour pressure of the sub-cooled liquid form, ^1X = MCI 1. order (Randic-number).

Tab. 9 Degradation rates and half-lives.

Sub-stance	BC	Air [h]	[1/h]	Water [h]	[1/h]	Soil [h]	[1/h]	Sediment [h]	[1/h]	Plants [h]	[1/h]
TCDD	0	170 (1)	4.1E-3	550 (1)	1.3E-3	17000 (1)	4.1E-5	55000 (1)	1.3E-5	20	0.035 (4)
PeCDD	0	550 (1)	1.3E-3	550 (1)	1.3E-3	17000 (1)	4.1E-5	55000 (1)	1.3E-5	6.4	0.109 (4)
HxCDD	0	550 (1)	1.3E-3	1700 (1)	4.1E-4	55000 (1)	1.3E-5	55000 (1)	1.3E-5	6.9	0.101 (4)
HpCDD	0	550 (1)	1.3E-3	1700 (1)	4.1E-4	55000 (1)	1.3E-5	55000 (1)	1.3E-5	5.1	0.137 (4)
OCDD	0	550 (1)	1.3E-3	5500 (1)	1.3E-4	55000 (1)	1.3E-5	55000 (1)	1.3E-5	4.3	0.161 (4)
PCB 28	0	550 (1)	1.3E-3	17000 (1)	4.1E-5	55000 (1)	1.3E-5	55000 (1)	1.3E-5		
PCB 52	0	1700 (1)	4.1E-4	55000 (1)	1.3E-5	55000 (1)	1.3E-5	55000 (1)	1.3E-5		
PCB 101	0	1700 (1)	4.1E-4	55000 (1)	1.3E-5	55000 (1)	1.3E-5	55000 (1)	1.3E-5		
PCB 138	0	5500 (1)	1.3E-4	55000 (1)	1.3E-5	55000 (1)	1.3E-5	55000 (1)	1.3E-5		
PCB 153	0	5500 (1)	1.3E-4	55000 (1)	1.3E-5	55000 (1)	1.3E-5	55000 (1)	1.3E-5		
PCB 180	0	17000 (1)	4.1E-5	55000 (1)	1.3E-5	55000 (1)	1.3E-5	55000 (1)	1.3E-5		
DEHP	1(7)	17.5 (7)	0.04	360 (7)	1.9E-3	1663 (7)	4.2E-4	7200 (7)	9.6E-5	96 (13)	7.2E-3
HHCB	0(3)	3.22 (3)	0.22	11880 (S)	5.8E-5	2.4E+7 (S)	2.9E-8	2.4E+8 (S)	2.9E-9		
EDTA	0(16)			11880 (9)	5.8E-5	2.4E+7 (S)	2.9E-8	2.4E+8 (S)	2.9E-9		
LAS	4(15)	24 (10)	2.9E-2	24 (12)	5.8E-2	504 (11)	1.4E-3	480 (12)	1.4E-3		
EDC	0(8)	2772 (8)	2.5E-4	2.4E+8(S)	2.9E-9	2160 (8)	3.2E-4	2.4E+8 (S)	2.9E-9		
BENZ	4(7)	240 (7)	2.9E-3	191(7)	3.6E-3	1680(S)	9.6E-4	7200 (S)	9.6E-5		

Tab. 10 Substance data for the standard assessment.

Substance	EU production/ import [t/a]	IC/UC#	LC/MC#
TCDD	-	-	-
PeCDD	-	-	-
HxCDD	-	-	-
HpCDD	-	-	-
OCDD	-	-	-
PCB$	-	-	-
DEHP	1E6 / 0 (14)	3/47 (8)	1-5/III, III, Ic
HHCB	0 / 2400 (3)	5/9 (3)	4/-
EDTA	2.65E4 / 0 (9)	5/9 (8%), 2/2 (26%), 10/42 (29%), 13/11 (4%), 15/55 (17.2%), 12/11 (3%), 8/17 (7%), 1/19 (3%), 5/15 (3%) (9)	4; 2, 3; 2, 3, 4; 3; 3; 3; 4; 1, 2, 3, 4
LAS	5E5 / 0 (10)	5/9	1/III, 2/III, 3/IV
EDC	8.5E6 / 0 (7)	3/33 (8)	1-5/Ic, III, Ic
BENZ	6.1E6 / 0 (7)	9/28 (7)	1/III, 2/III, 3/IV, 4

#: The meaning of the categories is explained in EC (1996B).

$: PCB are only used for validating submodels.

BC = Biodegradation class, assigned biodegradation according to TGD in [h] and ([1/h]). **4 (ready):** STP 0.7 (1), water 360 (1.9E-3), soil >720 (<9.6E-4), **3 (ready and falling in 10d window):** STP 2.3 (0.3), water 1200 (5.8E-4), soil >2160 (<3.2E-4), **2 (inherently):** STP 6.9 (0.1), water 3600 (1.9E-4), soil >7200 (<9.6E-5), biodegradation in sediment is assumed to be a factor of 10 higher than in soil, **1 (inherently, but not fulfilling specific criteria)**, **0 (not biodegradable).**

Sources: (1) MACKAY ET AL. (1999), (2) SETAC-POSTER (?), (3) PLASSCHE AND BALK (1997), calc., (4) SCHULER ET AL. (1998), sunlight photodegradation in cuticular wax, (7) EC (1999), (8) BUA (1995), (9) BUA (1996), (10) EC (1996C), (11) JENSEN (1999), (12) WHO (1996), (13) EUSES-LCA, (14) LWA (1993), (15) WOLTERING ET AL. (1988), (16) EUROPEAN AMINI-CARBOXYLATES PRODUCERS COMMITTEE (1990) (S) TGD standard value assumed for the given biodegradability.

A.3 Substance-specific parameters and probability distributions

In the following tables the *Realistic* column lists the data for the scenario analyses *and* the distributions for the probabilistic analyses. If available, the coefficient of variation (CV = SD / M) is shown in column *CV*. The remaining substance-specific data and the data used for the standard scenario can be found in Tabs 9 to 11. The triangular distributions for emission rates and half-lives is abbreviated by T*(central value). The procedure for selecting these probability distributions is explained in Chapter 6. Emission rates are calculated (according to EUSES) on the basis of tonnage and use category (*Industry Category: IC, Use Category: UC*). Degradation rates are derived (by EUSES) from the class of biodegradability. Only regional emissions are available for PCDD; continental estimations were estimated according to the 10%-rule (Section 5.2.1).

Tab. 11 TCDD.

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	322		SHIU ET AL. (1988)
K_{OW}	Octanol/water partition coefficient	-	6.31E6 L(3.08E7, 1.26E8)	4.09	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Sol	Water solubility	mg/L	1.9E-5 L(1.65E-4, 1.25E-4)	0.76	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
TempMelt	Melting point	K	578		SHIU ET AL. (1988)
Vp	Vapour pressure	Pa	2E-7 L(1.239E-5, 3.065E-5)	2.47	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Kdegair	Half-life in air	d	T*(7.08)		MACKAY ET AL. (1991-1997)
Kdegwater	Half-life in water	d	T*(22.92)		MACKAY ET AL. (1991-1997)
kdegsed	Half-life in sediment	d	T*(2292)		MACKAY ET AL. (1991-1997)
kdegsoil	Half-life in soil	d	T*(708.3)		MACKAY ET AL. (1991-1997)
ERegAir	Total regional emissions into air	kg/d	T*(7.4E-5)		NRW (1996)
ERegfirstwater	Total direct regional emission into surface water	kg/d	0		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
ERegfirstwastewater	Total direct regional emission into waste water	kg/d	T*(2.04E-7)		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
ERegInd	Total direct regional emission on industrial and urban soil	kg/d	0		
EContAir	Total continental emissions into air	kg/d	T*(6.81E-4)		
EContfirstwater	Total direct continental emission into surface water	kg/d	T*(4.35E-11)		
EContfirstwastewater	Total direct continental emission into waste water	kg/d	T*(1.84E-6)		
EContInd	Total direct continental emission on industrial and urban soil	kg/d	0		

Tab. 12 PeCDD.[#]

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	356.4		SHIU ET AL. (1988)
K_{OW}	Octanol/water partition coefficient	-	2.5E7 L(2.84E9; 3.37E9)	1.19	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Sol	Water solubility	mg/L	1.2E-4 L(1.18E-3; 2.62E-3)	2.22	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
TempMelt	Melting point	K	469		SHIU ET AL. (1988)
Vp	Vapour pressure	Pa	8.8E-8 L(6.96E-7; 5.26E-7)	0.76	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
kdegair	Half-life in air	d	T*(22.92)		
kdegwater	Half-life in water	d	T*(22.92)		
kdegsed	Half-life in sediment	d	T*(2292)		
kdegsoil	Half-life in soil	d	T*(708.3)		
EregAir	Total regional emissions into air	kg/d	T*(2.65E-4)		NRW (1996)
Eregfirstwater	Total direct regional emission into surface water	kg/d	T*(3.44E-7)		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
Eregfirstwastewater	Total direct regional emission into waste water	kg/d	T*(3.44E-7)		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
EregInd	Total direct regional emission on industrial and urban soil	kg/d	0		
EcontAir	Total continental emissions into air	kg/d	T*(2.38E-3)		
Econtfirstwater	Total direct continental emission into surface water	kg/d	T*(1.74E-10)		
Econtfirstwastewater	Total direct continental emission into waste water	kg/d	T*(3.10E-6)		
EcontInd	Total direct continental emission on industrial and urban soil	kg/d	0		

[#]: The probability distributions are derived from data for 1,2,3,4,7-PeCDD.

Tab. 13 HxCDD.

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	391		SHIU ET AL. (1988)
K_{OW}	Octanol/water partition coefficient	-	6.31E7 L(1.402E10, 2.107E10)	1.50	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Sol	Water solubility	mg/L	4.4E-6 L(8.091E-6, 7.685E-6)	0.95	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
TempMelt	Melting point	K	546		SHIU ET AL. (1988)
Vp	Vapour pressure	Pa	5.10E-9 L(1.714E-5, 4.367E-5)	2.55	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Kdegair	Half-life in air	d	T*(22.92)		MACKAY ET AL. (1991-1997)
Kdegwater	Half-life in water	d	T*(70.93)		MACKAY ET AL. (1991-1997)
Kdegsed	Half-life in sediment	d	T*(2292)		MACKAY ET AL. (1991-1997)
Kdegsoil	Half-life in soil	d	T*(2292)		MACKAY ET AL. (1991-1997)
ERegAir	Total regional emissions into air	kg/d	T*(1.18E-4)		NRW (1996)
ERegfirstwater	Total direct regional emission into surface water	kg/d	0		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
ERegfirstwastewater	Total direct regional emission into waste water	kg/d	T*(4.57E-6)		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
ERegInd	Total direct regional emission on industrial and urban soil	kg/d	0		

Name	Description	Unit	Realistic	CV	Source
EContAir	Total continental emissions into air	kg/d	T*(1.06E-3)		
EContfirstwater	Total direct continental emission into surface water	kg/d	T*(3.04E-9)		
EContfirstwastewater	Total direct continental emission into waste water	kg/d	T*(4.12E-5)		
EContInd	Total direct continental emission on industrial and urban soil	kg/d	0		

Tab. 14 HpCDD.

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	425.2		SHIU ET AL. (1988)
K_{OW}	Octanol/water partition coefficient	-	1E8 L(1.176E11, 2.121E11)	1.81	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Sol	Water solubility	mg/L	2.40E-6 L(1.124E-4, 2.596E-4)	2.31	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
TempMelt	Melting point	K	265		SHIU ET AL. (1988)
Vp	Vapour pressure	Pa	7.5E-10 L(1.709E-6, 3.286E-6)	1.92	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
kdegair	Half-life in air	d	T*(22.92)		MACKAY ET AL. (1991-1997)
kdegwater	Half-life in water	d	T*(70.83)		MACKAY ET AL. (1991-1997)
kdegsed	Half-life in sediment	d	T*(2292)		MACKAY ET AL. (1991-1997)
kdegsoil	Half-life in soil	d	T*(2292)		MACKAY ET AL. (1991-1997)
ERegAir	Total regional emissions into air	kg/d	T*(2.04E-3)		NRW (1996)
ERegfirstwater	Total direct regional emission into surface water	kg/d	0		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
Eregfirstwastewater	Total direct regional emission into waste water	kg/d	T*(1.04E-4)		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
EregInd	Total direct regional emission on industrial and urban soil	kg/d	0		
EcontAir	Total continental emissions into air	kg/d	T*(1.83E-2)		
Econtfirstwater	Total direct continental emission into surface water	kg/d	T*(3.04E-8)		
Econtfirstwastewater	Total direct continental emission into waste water	kg/d	T*(9.36E-4)		
EcontInd	Total direct continental emission on industrial and urban soil	kg/d	0		

Tab. 15 OCDD.

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	460		SHIU ET AL. (1988)
K_{OW}	Octanol/water partition coefficient	-	1.585E8 L(8.075E11, 2.239E12)	2.77	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Sol	Water solubility	mg/L	7.4E-8 L(1.564E-5, 4.665E-5)	2.98	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
TempMelt	Melting point	K	335		SHIU ET AL. (1988)
Vp	Vapour pressure	Pa	1.1E-10 L(6.201E-6, 9.185E-6)	1.48	SHIU ET AL. (1988) for realistic value, MACKAY ET AL. (1991-1997) for distribution
kdegair	Half-life in air	d	T*(22.92)		MACKAY ET AL. (1991-1997)
kdegwater	Half-life in water	d	T*(229.2)		MACKAY ET AL. (1991-1997)
kdegsed	Half-life in sediment	d	T*(2292)		MACKAY ET AL. (1991-1997)
kdegsoil	Half-life in soil	d	T*(2292)		MACKAY ET AL. (1991-1997)
ERegAir	Total regional emissions into air	kg/d	T*(2.96E-3)		NRW (1996)

Name	Description	Unit	Realistic	CV	Source
ERegfirstwater	Total direct regional emission into surface water	kg/d	0		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
ERegfirstwastewater	Total direct regional emission into waste water	kg/d	T*(7.86E-4)		HORSTMANN AND MCLACHLAN (1994), JONES AND STEWARD (1997)
ERegInd	Total direct regional emission on industrial and urban soil	kg/d	0		
EContAir	Total continental emissions into air	kg/d	T*(2.67E-2)		
EContfirstwater	Total direct continental emission into surface water	kg/d	0		
EContfirstwastewater	Total direct continental emission into waste water	kg/d	T*(7.08E-3)		
EContInd	Total direct continental emission on industrial and urban soil	kg/d	0		

Tab. 16 DEHP.

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	390.56		RIPPEN (1996)
K_{OW}	Octanol/water partition coefficient	-	3.02E7 L(3.505E8, 1.105E9)	3.151	RIPPEN (1996) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Sol	Water solubility	mg/L	0.029 L(8.651, 25.953)	3.00	RIPPEN (1996) for realistic value, MACKAY ET AL. (1991-1997) for distribution
TempMelt	Melting point	K	233		
Vp	Vapour pressure	Pa	1.9E-3 L(2.549E-4, 6.491E-4)	2.547	RIPPEN (1996) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Kdegair	Half-life in air	d	T*(7.29E-1)		Estimated from molecular structure according to SRC-AOP (1998)
kdegwater	Half-life in water	d	T*(15.2)		EC (1997)
kdegsed	Half-life in sediment	d	T*(300)		EC (1997)
kdegsoil	Half-life in soil	d	T*(68)		EC (1997)
ERegAir	Total regional emissions into air	kg/d	T*(3330)		UBA (1996)
ERegfirstwater	Total direct regional emission into surface water	kg/d	0		UBA (1996)
ERegfirstwastewater	Total direct regional emission into waste water	kg/d	T*(1576)		UBA (1996)
ERegInd	Total direct regional emission on industrial and urban soil	kg/d	0		UBA (1996)
EContAir	Total continental emissions into air	kg/d	T*(30000)		Standard value
EContfirstwater	Total direct continental emission into surface water	kg/d	T*(8.70E-1)		Standard value
EContfirstwastewater	Total direct continental emission into waste water	kg/d	T*(1.42E4)		Standard value
EContInd	Total direct continental emission on industrial and urban soil	kg/d	0		Standard value

Tab. 17 HHCB.

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	258.4		PLASSCHE AND BALK (1997)
Kow	Octanol/water partition coefficient	-	log: 5.9 U(7.9E5; 1.8E6)		PLASSCHE AND BALK (1997)
Sol	Water solubility	mg/L	1.75 U(1.5; 2.0)		PLASSCHE AND BALK (1997)
TempMelt	Melting point	K			
Vp	Vapour pressure	Pa	0.0727		PLASSCHE AND BALK (1997)
kdegair	Half-life in air	d	T*(1.34E-1)		PLASSCHE AND BALK (1997)
kdegwater	Half-life in water	d	T*(5E5)		Standard value
kdegsed	Half-life in sediment	d	T*(1E7)		Standard value
kdegsoil	Half-life in soil	d	T*(1E6)		Standard value
ERegAir	Total regional emissions into air	kg/d	0		PLASSCHE AND BALK (1997)
ERegfirstwater	Total direct regional emission into surface water	kg/d	T*(4.78E-2)		PLASSCHE AND BALK (1997)
ERegfirstwastewater	Total direct regional emission into waste water	kg/d	T*(3.21E2)		PLASSCHE AND BALK (1997)
ERegInd	Total direct regional emission on industrial and urban soil	kg/d	0		PLASSCHE AND BALK (1997)
EContAir	Total continental emissions into air	kg/d	0		PLASSCHE AND BALK (1997)
EContfirstwater	Total direct continental emission into surface water	kg/d	0		PLASSCHE AND BALK (1997)
EContfirstwastewater	Total direct continental emission into waste water	kg/d	T*(6.25E3)		PLASSCHE AND BALK (1997)
EContInd	Total direct continental emission on industrial and urban soil	kg/d	0		PLASSCHE AND BALK (1997)

Tab. 18 BENZ (benzene).

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	0.07812		RIPPEN (1995)
Kow	Octanol/water partition coefficient	-	131.83 L(151.84, 60.71)	0.400	RIPPEN (1995); MACKAY ET AL. (1991-1997) for distribution
Sol	Water solubility	mg/L	1760 L(1767.725, 511.620)	0.289	RIPPEN (1995); MACKAY ET AL. (1991-1997) for distribution
TempMelt	Melting point	K	279		RIPPEN (1995)
Vp	Vapour pressure	Pa	1.27E4 L(1.259E4, 824.9)	0.066	RIPPEN (1995)
kdegair	Half-life in air	d	T*(10)		RIPPEN (1995)
kdegwater	Half-life in water	d	T*(7.95)		RIPPEN (1995)
kdegsed	Half-life in sediment	d	T*(300)		Standard value
kdegsoil	Half-life in soil	d	T*(30)		Standard value
ERegAir	Total regional emissions into air	kg/d	T*(3.43E4)		UBA (1996a)
ERegfirstwater	Total direct regional emission into surface water	kg/d	T*(792)		Estimated
ERegfirstwastewater	Total direct regional emission into waste water	kg/d	T*(3722)		Estimated
ERegInd	Total direct regional emission on industrial and urban soil	kg/d	T*(2120)		Estimated
EContAir	Total continental emissions into air	kg/d	T*(1.37E6)		Standard value
Econtfirstwater	Total direct continental emission into surface water	kg/d	T*(1.46E3)		Standard value
Econtfirstwastewater	Total direct continental emission into waste water	kg/d	T*(1.05E5)		Standard value
EcontInd	Total direct continental emission on industrial and urban soil	kg/d	T*(1.91E4)		Standard value

Tab. 19 EDC (1,2-dichloroethane).

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	98.96		BUA (1995)
K_{OW}	Octanol/water partition coefficient	-	28.84 L(30.2, 8.73)	0.27	BUA (1995) for realistic value, MACKAY ET AL. (1991-1997) for distribution
Sol	Water solubility	mg/L	8.6E3 L(8.611E3, 2.26E2)	0.03	BUA (1995) for realistic value, MACKAY ET AL. (1991-1997) for distribution
TempMelt *	Melting point	K	238		BUA (1995)
Vp	Vapour pressure	Pa	1.13E4 L(1.040E4, 9.33E2)	0.09	BUA (1995) for realistic value, MACKAY ET AL. (1991-1997) for distribution
kdegair	Half-life in air	d	T*(115)		BUA (1995)
kdegwater	Half-life in water	d	T*(9.92E4)		BUA (1995)
kdegsed	Half-life in sediment	d	T*(1E7)		Standard value
kdegsoil	Half-life in soil	d	T*(90)		BUA (1995)
ERegAir	Total regional emissions into air	kg/d	T*(1.28E5)		Standard value
ERegfirstwater	Total direct regional emission into surface water	kg/d	T*(73.9)		Standard value
Eregfirstwastewater	Total direct regional emission into waste water	kg/d	T*(2.98E4)		Standard value
ERegInd	Total direct regional emission on industrial and urban soil	kg/d	T*(69.2)		Standard value
EContAir	Total continental emissions into air	kg/d	T*(1.15E6)		Standard value
EContfirstwater	Total direct continental emission into surface water	kg/d	T*(21.7)		Standard value
Econtfirstwastewater	Total direct continental emission into waste water	kg/d	T*(2.69E5)		Standard value
EContInd	Total direct continental emission on industrial and urban soil	kg/d	T*(6.23E3)		Standard value

*: Not sensitive.

Tab. 20 EDTA (acid).

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	292.25		BUA (1996)
K_{OW}	Octanol/water partition coefficient	-	log: -3.34 log: U(-5.01; -3.34)		BUA (1996)
Sol	Water solubility	mg/L	T*(500)		EC (1996D)
TempMelt	Melting point	K	493		EC (1996D)
Vp	Vapour pressure	Pa	T*(1E-6)		Minimum of EUSES parameter range
kdegair	Half-life in air	1/d	0		Standard value
kdegwater	Half-life in water	d	T*(4.81E2)		Standard value
kdegsed	Half-life in sediment	d	T*(1E7)		Standard value
kdegsoil	Half-life in soil	d	T*(1E6)		Standard value
ERegAir	Total regional emissions into air	kg/d	0		Estimated
ERegfirstwater	Total direct regional emission into surface water	kg/d	T*(10.4)		Standard value
ERegfirstwastewater	Total direct regional emission into waste water	kg/d	T*(4.78E3)		Standard value
ERegInd	Total direct regional emission on industrial and urban soil	kg/d	T*(49.2)		Standard value
EContAir	Total continental emissions into air	kg/d	0		Estimated
EContfirstwater	Total direct continental emission into surface water	kg/d	T*(1.04E2)		Standard value
EContfirstwastewater	Total direct continental emission into waste water	kg/d	T*(4.45E4)		Standard value
EcontInd	Total direct continental emission on industrial and urban soil	kg/d	T*(4.5E2)		Standard value

Tab. 21 LAS.

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	348.48		RIPPEN (1995); value for C_{12}-LAS
K_{OW}	Octanol/water partition coefficient	-	T*(91.2)		LWA (1993); value for C_{12}-LAS
Sol	Water solubility	mg/L	T*(1.1E3)		RIPPEN (1995)
TempMelt *	Melting point	K	263		EC (1996)
Vp	Vapour pressure	Pa	T*(1E-6)		Minimum of EUSES parameter range
kdegair *	Half-life in air	d	T*(1.1)		EC (1996)
kdegwater	Half-life in water	d	T*(0.5)		GROB (1996)
kdegsed	Half-life in sediment	d	T*(1)		WOLTERING ET AL. (1988)
kdegsoil	Half-life in soil	d	T*(14)		JENSEN (1999)
ERegAir	Total regional emissions into air	kg/d	0		Estimated
ERegfirstwater	Total direct regional emission into surface water	kg/d	T*(3.91)[a]		
ERegfirstwastewater	Total direct regional emission into waste water	kg/d	T*(6.78E4)[a]		
ERegInd	Total direct regional emission on industrial and urban soil	kg/d	T*(6.95E2)[a]		
EContAir	Total continental emissions into air	kg/d	0		
EContfirstwater	Total direct continental emission into surface water	kg/d	0		Estimated
Econtfirstwastewater	Total direct continental emission into waste water	kg/d	T*(1.29E6)		Standard value
EContInd	Total direct continental emission on industrial and urban soil	kg/d	T*(1.32E4)		Standard value

[a]: The estimated emissions are too high. Based on a per-capita consumption of 3.8 g/d (HUBER 1989), a realistic regional production volume amounts half of standard volume. Thus, all emissions are by 50% lower than the standard emissions.

Tab. 22 PCB28.[$]

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	257.55		MACKAY ET AL. (1991-1997)
Kow	Octanol/water partition coefficient	-	log: 5.57 L(4.4E5; 1.8E5)	0.41	HAWKER (1988), MACKAY ET AL. (1991-1997)
Sol	Water solubility	mg/L	8.9E-2		DUNNIVANT ET AL. (1992)
TempMelt	Melting point	K	330		BRODSKY (1986)
Vp	Vapour pressure	Pa	1.0E-2		DUNNIVANT ET AL. (1992)

Tab. 23 PCB52. [$]

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	291.99		MACKAY ET AL. (1991-1997)
Kow	Octanol/water partition coefficient	-	log: 5.84 L(1.3E6; 1.6E6)	1.25	HAWKER (1988), MACKAY ET AL. (1991-1997)
Sol	Water solubility	mg/L	3.3E-2		DUNNIVANT ET AL. (1992)
TempMelt	Melting point	K	360		BRODSKY (1986)
Vp	Vapour pressure	Pa	3.6E-3		DUNNIVANT ET AL. (1992)

Tab. 24 PCB101. [$]

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	326.44		MACKAY ET AL. (1991-1997)
Kow	Octanol/water partition coefficient	-	log: 6.38 L(1.0E7; 1.4E7)	1.42	HAWKER (1988), MACKAY ET AL. (1991-1997)
Sol	Water solubility	mg/L	9.4E-3		DUNNIVANT ET AL. (1992)
TempMelt	Melting point	K	350		BRODSKY (1986)
Vp	Vapour pressure	Pa	7.2E-4		DUNNIVANT ET AL. (1992)

Tab. 25 PCB138. [$]

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	360.88		MACKAY ET AL. (1991-1997)
Kow	Octanol/water partition coefficient	-	log: 6.83 L(9.4E6; 9.6E6)	1.02	HAWKER (1988), MACKAY ET AL. (1991-1997)
Sol	Water solubility	mg/L	1.5E-3		DUNNIVANT ET AL. (1992)
TempMelt	Melting point	K	352		BRODSKY (1986)
Vp	Vapour pressure	Pa	5.4E-5		DUNNIVANT ET AL. (1992)

Tab. 26 PCB153. [$]

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	360.88		MACKAY ET AL. (1991-1997)
Kow	Octanol/water partition coefficient	-	log: 6.92 L(4.9E7; 7.5E7)	1.53	HAWKER (1988), MACKAY ET AL. (1991-1997)
Sol	Water solubility	mg/L	8.8E-4		DUNNIVANT ET AL. (1992)
TempMelt	Melting point	K	376		BRODSKY (1986)
Vp	Vapour pressure	Pa	4.1E-5		DUNNIVANT ET AL. (1992)

Tab. 27 PCB180. [$]

Name	Description	Unit	Realistic	CV	Source
Molw	Molecular weight	g/mol	395.33		MACKAY ET AL. (1991-1997)
Kow	Octanol/water partition coefficient	-	log: 7.36 L(1.4E7; 5.6E6)	0.39	HAWKER (1988), MACKAY ET AL. (1991-1997)
Sol	Water solubility	mg/L	2.3E-4		DUNNIVANT ET AL. (1992)
TempMelt	Melting point	K	382		BRODSKY (1986)
Vp	Vapour pressure	Pa	6.4E-6		DUNNIVANT ET AL. (1992)

[$]: PCB are only used for validating submodels. Thus, a reduced parameter set is presented.

A.4 Chemical concentrations and intake rates

This section presents measured concentrations. The following tables show the concentrations used and present further concentrations as a comparison. Units are usually refer to the fresh weight and correspond to the units used in EUSES. Column *M* represents the mean, unless otherwise stated.

Tab. 28 Concentrations (dissolved) in surface water [mg/l].

Substance	Min	Max	M	Comment	Source
TCDD			2.2E-11	Representative total concentrations rivers in GB	DUARTE-DAVIDSON ET AL. (1997B)
PeCDD			3.0E-11	s.a.	s.a.
HxCDD I			2.7E-11	s.a.	s.a.
HxCDD II			6.0E-11	s.a.	s.a.
HxCDD III			3.8E-11	s.a.	s.a.
HpCDD			2.3E-10	s.a.	s.a.
OCDD			1.4E-09	s.a.	s.a.
PCB 28	1.7E-08	9.3E-07		River Rhine 1990, estimated from total concentration, min = NG	IKSR (1990)
PCB 52	1.2E-08	4.3E-07		s.a., Min = NG	s.a.
PCB 101	1.0E-08	1.5E-07		s.a.	s.a.
PCB 138	6.1E-09	2.3E-07		s.a.	s.a.
PCB 153	3.7E-09	1.4E-07		s.a.	s.a.
PCB 180	9.0E-10	2.2E-08		s.a.	s.a.
DEHP	5.4E-07	4.9E-04	1.1E-05	River Rhine between Honnef and Bimmen 1991-92, M = median, estimated from total concentration	NRW (1993)
	1.1E-04	1.0E-02	5.2E-04	Total concentration of the previous values	NRW (1993)
	1.0E-07	3.0E-01		Range of concentrations in surface waters	NRW (1993)
HHCB	1.0E-4	1.2E-3	5.0E-4	30 samples River Ruhr, 1994/95	ESCHKE ET AL. (1994, 1995)
EDTA	1.0E-03	5.4E-02		River Rhine and tributaries 1980-81, River Rhine and River Main are highly contaminated, Min = NG	BUA (1996)
	1.3E-03	4.4E-03		Bodensee 1989-90	BUA (1996)
		4.2E+00		Maximum value, River Thames (GB)	WOLF ET AL. (1992)
LAS	1.0E-03	6.0E-01	1.0E-02	River Rhine	RIPPEN (1995)
	1.0E-03	2.7E+00		Range of concentrations in surface waters	RIPPEN (1995)
EDC	1.0E-05	1.0E-03		River Rhine near Lobith 1987-1992, Min = NG	BUA (1995)
		3.6E-01		Maximum value, River Rhine (1994)	BUA (1995)
BENZ	1.0E-05	8.0E-04		River Rhine, Values usually below NG, Min = NG	RIPPEN (1995)
		2.0E-01		Maximum value for rivers	BUA (1988)

Tab. 29 Concentrations in air [mg/m³].

Substance	Min	Max	M	Comment	Source
TCDD	6.0E-13	3.6E-12	1.4E-12	Chloraromaten monitoring programme 1990, M = median	NRW (1991a)
PeCDD	1.8E-12	4.0E-11	3.4E-12	s.a.	s.a.
HxCDD I	2.7E-12	3.3E-11	4.4E-12	s.a.	s.a.
HxCDD II	6.3E-12	6.7E-11	1.1E-11	s.a.	s.a.
HxCDD III	6.5E-12	5.9E-11	1.1E-11	s.a.	s.a.
HpCDD	8.0E-11	8.1E-10	1.7E-10	s.a.	s.a.
OCDD	3.1E-10	2.8E-09	6.7E-10	s.a.	s.a.
TCDD	1.0E-11	5.7E-10		Cities in GB, Min = NG	DUARTE-DAVIDSON ET AL. (1997A)
PeCDD	1.0E-11	4.0E-10		s.a.	s.a.
HxCDD I	1.0E-11	6.7E-10		s.a.	s.a.

Substance	Min	Max	M	Comment	Source
HxCDD II	1.0E-11	6.7E-10		s.a.	s.a.
HxCDD III	1.0E-11	6.7E-10		s.a.	s.a.
HpCDD	1.0E-11	1.1E-08		s.a.	s.a.
OCDD	1.0E-11	6.2E-08		s.a.	s.a.
PCB 28	4.0E-09	6.8E-08	2.3E-08	Vicinity of Ulm, Rural and industrial area, M = mean, samples of WITTLINGER AND BALLSCHMITER	HALSALL ET AL. (1995)
PCB 52	7.0E-09	2.3E-07	6.1E-08	s.a.	s.a.
PCB 101	1.0E-08	2.3E-07	6.6E-08	s.a.	s.a.
PCB 138	6.0E-09	1.1E-07	3.4E-08	s.a.	s.a.
PCB 153	7.0E-09	1.3E-07	4.3E-08	s.a.	s.a.
PCB 180	1.0E-09	2.7E-08	9.0E-09	s.a.	s.a.
DEHP	3.0E-07	1.9E-06	1.1E-06	Rural area 1985	RIPPEN (1995)
	3.0E-05	1.3E-04		Antwerpen (B), 1985	RIPPEN (1995)
	1.5E-01	2.6E-01		Indoor, highly contaminated	EC (1996C)
		6.6E+01		Conurbation area, maximum value	EC (1996C)
HHCB	1.1E-7	2.2E-7	1.2E-7	Norway, 5 samples	KALLENBORN ET AL. (1999)
EDC	2.0E-04	1.2E-01		Hamburg	RIPPEN (1995)
	2.1E-02	3.7E-02		Typical concentrations in industrial areas	BUA (1995)
	4.0E-05	1.8E-04		Rural areas, Schwäbische Alb, Germany	BUA (1995)
		1.6E+00		Maximum value, congested cities	BUA (1995)
			2.5E-04	M = median for 455 cities	RIPPEN (1995)
			1.0E-04	Background concentration	BUA (1995)
BENZ	2.0E-04	8.4E-02	2.0E-04	Rural areas in Germany, Min = NG	BUA (1988)
	5.0E-03	1.0E-02		Conurbation areas	BUA (1993)
	5.0E-04	1.6E+00		Industrial areas and petrol stations	BUA (1988)

Tab. 30 Concentrations in soil [mg/kg].[#]

Substance	Min	Max	M	Comment	Source
TCDD	3.5E-07	2.1E-06	5.8E-07	Chloraromaten monitoring programme 1990, estimated from DW, M = median	NRW (1991B)
PeCDD	2.7E-07	2.7E-06	4.9E-07	s.a.	s.a.
HxCDD I	1.8E-07	2.6E-06	1.1E-06	s.a.	s.a.
HxCDD II	5.3E-07	6.7E-06	1.8E-06	s.a.	s.a.
HxCDD III	3.5E-07	4.9E-06	1.2E-06	s.a.	s.a.
HpCDD	1.1E-05	1.2E-04	1.8E-05	s.a.	s.a.
OCDD	4.0E-05	1.5E-03	7.6E-05	s.a.	s.a.
PCB 28	9.7E-05	3.7E-03	1.9E-04	Chloraromaten monitoring programme 1990, estimated from DW, M = median	NRW (1991B)
PCB 52	4.4E-05	4.4E-03	1.2E-04	s.a.	s.a.
PCB 101	1.5E-04	1.2E-02	4.2E-04	s.a.	s.a.
PCB 153	6.5E-04	7.2E-02	1.4E-03	s.a.	s.a.
PCB 180	9.7E-05	8.1E-02	1.0E-03	s.a.	s.a.
DEHP	3.5E-02	2.7E-01	2.1E-01	Soils in Germany, estimated from DW M = mean for grassland soil	MÜLLER AND KÖRDEL (1995)
		4.5E+00		After sludge application, estimated from DW	s.a.
EDTA	0.02 - 0.1 mg/kg after application of nutrient fertiliser (estimated)				BUA (1996)
LAS		2.2E+03		Maximum value after sludge application	RIPPEN (1995)
		1.4E+00		Maximum value without sludge application	RIPPEN (1995)
BENZ	2.0E-03	2.0E-01		Contaminated soil, values are usually below NG	HOWARD (1990)

[#]: Conversion factor DW/FW = 1.13.

Tab. 31 Concentrations in grass [mg/kg].[#]

Substance	Min	Max	M	Comment	Source
TCDD	2.5E-09	4.5E-08	1.0E-08	Chloraromaten monitoring programme 1990, estimated from DW, M = median	NRW (1991B)
PeCDD	1.0E-08	6.0E-08	3.5E-08	s.a.	s.a.
HxCDD I	2.3E-08	7.0E-08	3.3E-08	s.a.	s.a.
HxCDD II	5.3E-08	1.7E-07	7.6E-08	s.a.	s.a.
HxCDD III	3.0E-08	1.2E-07	5.1E-08	s.a.	s.a.
HpCDD	3.3E-07	2.2E-06	6.6E-07	s.a.	s.a.
OCDD	1.4E-06	1.2E-05	2.7E-06	s.a.	s.a.
PCB 28	3.8E-05	1.8E-04	1.1E-04	Chloraromaten monitoring programme 1990, estimated from DW, M = median	NRW (1991B)
PCB 52	3.0E-05	1.7E-04	8.1E-05	s.a.	s.a.
PCB 101	4.5E-05	2.1E-04	1.2E-04	s.a.	s.a.
PCB 153	8.5E-05	3.4E-04	2.0E-04	s.a.	s.a.
PCB 180	4.5E-05	2.2E-04	8.8E-05	s.a.	s.a.
DEHP	1.1E-01	3.3E+00		Vicinity of emitters	RIPPEN (1995)
EDC		1.0E-03		Maximum value for biota	EC (1996C)
BENZ		1.0E-03		Maximum value for biota	EC (1996C)

[#]: Conversion factor DW/FW = 4.

Tab. 32 Concentrations in drinking water [mg/l].

Substance	Min	Max	M	Comment	Source
PCDD			3.0E-12	Estimated average drinking water concentration in I-TE, no positive samples in Germany (NG ~ fg/l)	BALLSCHMITER AND BACHER (1996)
OCDD	9.0E-09	1.8E-07		Canada, 36 out of 37 samples positive	JOBB (1990)
DEHP	6.0E-04	1.7E-01	1.0E-03	Survey of concentrations, M = median	RIPPEN (1995)
EDTA	5.0E-04	1.9E-02	2.3E-02	Cities in NRW, concentrations, Min = NG	BUA (1996)
		3.1E-02		Maximum value	BUA (1996)
LAS	3.0E-04	2.5E-03	1.4E-03	30% out of all LAS, diverse countries	IPCS (1996)
EDC	3.5E-04	1.3E-03		River Rhine 1975/76, current data unavailable	BUA (1995)
	1.0E-04	5.8E-02	5.0E-04	Spain 1987, M = mean	EC (1996C)
BENZ	1.8E-05	4.5E-05		Western Germany 1980	BUA (1988)
		1.0E-02		Maximum value	BUA (1988)

Tab. 33 Concentrations in fishes [mg/kg].

Substance	Min	Max	M	Comment	Source
TCDD	6.9E-06	6.9E-06	6.9E-06	Trouts, background concentration Sweden, estimated from lipid concentration (fat content 16%)	BALLSCHMITER AND BACHER (1996)
PeCDD	8.5E-06	8.5E-06	8.5E-06	s.a.	s.a.
HxCDD I	2.4E-07	2.4E-07	2.4E-07	s.a.	s.a.
HxCDD II	2.7E-06	2.7E-06	2.7E-06	s.a.	s.a.
HxCDD III	2.1E-07	2.1E-07	2.1E-07	s.a.	s.a.
HpCDD	2.7E-07	2.7E-07	2.7E-07	s.a.	s.a.
OCDD	6.1E-07	6.1E-07	6.1E-07	s.a.	s.a.
TCDD	2.0E-09	1.8E-06		Diverse sea fishes, estimated from lipid concentration	FÜRST ET AL. (1990)
PeCDD	2.0E-09	5.4E-06		s.a.	s.a.
HxCDD I	2.0E-09	6.1E-07		s.a.	s.a.
HxCDD II	2.0E-09	4.2E-06		s.a.	s.a.
HxCDD III	2.0E-09	1.8E-06		s.a.	s.a.
HpCDD	5.6E-09	5.4E-06		s.a.	s.a.
OCDD	1.3E-08	2.7E-05		s.a.	s.a.
PCB 28	1.0E-03	1.0E-02	5.5E-03	Diverse fishes in River Rhine 1990	IKSR (1993)

PCB 52	2.0E-03	1.3E-01	6.6E-02	s.a.	s.a.
PCB 101	5.0E-03	1.4E-01	7.3E-02	s.a.	s.a.
PCB 138	1.0E-02	2.6E-01	1.4E-01	s.a.	s.a.
PCB 153	1.0E-02	4.6E-01	2.4E-01	s.a.	s.a.
PCB 180	5.0E-03	1.4E-01	7.2E-02	s.a.	s.a.
DEHP	1.7E-02	7.0E-02	4.4E-02	Diverse fishes in River Rhine, near Rees 1981	RIPPEN (1995)
	1.0E-05	3.2E+00		Survey	EC (1997)
HHCB	1.0E-2	1.3E-1	2.0E-2	River Ruhr 1994/95	ESCHKE ET AL. (1994, 1995)
EDC	Usually undetectable (NG ~ 1E-4 to 1E-2)				RIPPEN (1995), HOWARD (1990)
BENZ	Usually undetectable (NG ~ 1E-3). However, in diverse sea fishes (urban areas, USA 1980/81) concentrations up to 5.2E-02 were found.				HOWARD (1990)

Tab. 34 Concentrations in beef [mg/kg][#].

Substance	Min	Max	M	Comment	Source
TCDD	7.5E-08	1.5E-07	1.1E-07	Samples in NRW, estimated from lipid concentration, Min = NG	FÜRST ET AL. (1990), BECK ET AL. (1989)
PeCDD	1.3E-07	1.2E-06	6.4E-07	s.a.	s.a.
HxCDD I	1.3E-07	1.2E-06	6.4E-07	s.a.	s.a.
HxCDD II	1.5E-07	1.5E-06	8.3E-07	s.a.	s.a.
HxCDD III	1.5E-07	1.1E-06	6.4E-07	s.a.	s.a.
HpCDD	4.5E-07	4.5E-06	2.5E-06	s.a.	s.a.
OCDD	1.2E-06	6.3E-06	3.7E-06	s.a.	s.a.
PCB 28	5.0E-05	2.3E-02	2.5E-03	Estimated from lipid concentration, Min = NG	WEIGERT ET AL. (1991)
PCB 52	5.0E-05	2.3E-02	1.9E-03	s.a.	s.a.
PCB 101	5.0E-05	1.9E-02	1.3E-03	s.a.	s.a.
PCB 138	5.0E-05	1.1E-01	2.5E-03	s.a.	s.a.
PCB 153	5.0E-05	1.6E-01	2.5E-03	s.a.	s.a.
PCB 180	5.0E-05	1.1E-01	1.3E-02	s.a.	s.a.

[#]: Assumed fat content: 25%.

Tab. 35 Concentrations in milk [mg/l][#].

Substance	Min	Max	M	Comment	Source
TCDD	2.9E-09	2.0E-08	7.4E-09	Chloraromaten monitoring programme 1990, estimated from lipid concentration, M = median	NRW (1991B)
PeCDD	6.3E-09	2.9E-08	1.5E-08	s.a.	s.a.
HxCDD I	2.2E-09	1.4E-08	9.2E-09	s.a.	s.a.
HxCDD II	1.8E-08	5.9E-08	3.0E-08	s.a.	s.a.
HxCDD III	2.2E-09	1.5E-08	7.4E-09	s.a.	s.a.
HpCDD	1.1E-08	1.3E-07	4.5E-08	s.a.	s.a.
OCDD	4.7E-08	3.3E-07	1.0E-07	s.a.	s.a.
PCB 28	3.7E-05	3.7E-05	3.7E-05	Chloraromaten monitoring programme 1990, estimated from lipid concentration, M = median, Concentrations of PCB 28 and 52 were below NG	NRW (1991B)
PCB 52	3.7E-05	3.7E-05	3.7E-05	s.a.	s.a.
PCB 101	3.7E-06	2.5E-05	1.7E-05	s.a.	s.a.
PCB 153	1.6E-04	8.7E-04	3.6E-04	s.a.	s.a.
PCB 180	5.9E-05	4.0E-04	1.4E-04	s.a.	s.a.
DEHP			7.4E-03	Various phthalates together	FÜRST (1995)
EDC			2.9E-05	Diverse dairy products, M = mean	RIPPEN (1995)

[#]: Assumed fat content: 3.68%.

Tab. 36 Total daily doses reported in the literature [mg/(kg*d)].

Substance	Min	Max	Comment	Source
TCDD	2.60E-11	8.30E-10	More than 90% via food	SCHREY ET AL. (1996)
PeCDD	1.50E-10	3.00E-09	s.a.	s.a.
HxCDD	2.40E-10	5.50E-09	s.a.	s.a.
HpCDD	5.00E-10	6.00E-09	s.a.	s.a.
OCDD	2.40E-09	5.00E-08	s.a.	s.a.
PCB 28			No data available	
PCB 52			No data available	
PCB 101	4.00E-08	9.60E-06	More than 90% via milk, meat and fish	PETZOLD ET AL. (1999)
PCB 138	2.70E-07	1.38E-05	s.a.	s.a.
PCB 153	6.00E-07	1.51E-05	s.a.	s.a.
PCB 180	3.20E-07	4.14E-06	s.a.	s.a.
DEHP	7.14E-03	1.14E-02	More than 90% via food	EIKMANN (1995)
HHCB		7.64E-01	Via detergents, soaps, etc.	FORD (1998)
EDC	3.00E-05	7.00E-05	More than 75% via inhalation	HUGHES ET AL. (1994)
BENZ	1.06E-03	6.54E-03	More than 95% via inhalation	GENNART ET AL. (1994)
EDTA			No data available	
LAS	6.43E-02	2.07E-01	Via drinking water and washing	IPCS (1996)

A.5 Input data for the sensitivity analyses

The sensitivity analyses are based on a parameter set which corresponds to the *Realistic* scenario (i.e. it is based on regional parameters for NRW, and measured substance-specific data were used if available). However, estimable parameters were not replaced. This scenario corresponds to that used in the uncertainty analyses.

For the sensitivity analyses of the exposure module alone (i.e. without emission tables and regional distribution model) the values of the following table are used. EUSES default values were used as model parameters.

Tab. 37 Input parameters for the sensitivity analyses of the exposure module.

Parameter	Unit	TCDD	HxCDD	OCDD	PCB52	PCB138	PCB180	HHCB	DEHP	BENZ	EDC	EDTA	LAS
Substance properties													
Henry law constant (not less than 0.01)	Pa m³/mol	3.3	1.1	0.7	32.2	13.1	10.9	11.3	17.5	448.3	96.7	0.01	0.01
Octanol-water partition coefficient (log K_{OW})	-	6.8	7.8	8.2	5.84	6.83	7.36	5.9	7.48	2.12	1.46	-3.34	1.96
Particulate fraction	-	0.32	0.98	1	0	0.27	0.61	0	0.05	0	0	1	1
Degradation rate of chemical in plants (not less than 0.01)	1/d	0.84	2.42	3.86	0.01	0.01	0.01	5.28*	0.17	0.07*	0.01	0.01	0.69*
Half-live for biodegradation in water	d	>10	>10	> 10	>10	>10	>10	>10	>10	< 10	>10	>10	< 10
* Degradation rate in air is assumed as estimation													
Input concentration for the exposure module													
PEC in water (dissolved)	kg/m³	1.0E-15	1.0E-14	2.4E-12	2.2E-10	1.2E-10	1.1E-11	5.0E-07	1.1E-08	4.1E-07	5.1E-07	2.8E-05	1.5E-05
PEC in air (total)	kg/m³	1.4E-18	4.4E-18	6.7E-16	6.1E-14	3.4E-14	9.0E-15	1.8E-11	1.1E-12	2.0E-10	2.5E-10	0	0
PEC in agricultural soil	kg/kg (FW)	5.8E-13	1.1E-12	7.6E-11	1.2E-10	1.2E-09	1.0E-09	6.4E-06	2.1E-07	0	0	1.0E-07	1.4E-06
PEC in pore water of agricultural soil	kg/m³	8.1E-14	1.0E-13	7.8E-13	1.2E-10	1.0E-10	5.0E-11	1.0E-05	1.6E-07	7.7E-08	2.2E-07	8.5E-04	1.4E-03
Input concentrations for the direct calculation of the total daily dose													
PEC in drinking water	kg/m³	1.0E-18	1.0E-18	1.0E-18	1.0E-15	1.0E-15	1.0E-15	5.0E-07	1.1E-08	3.0E-08	5.0E-07	2.3E-05	1.4E-06
PEC in air	kg/m³	1.4E-18	4.4E-18	6.7E-16	6.1E-14	3.4E-14	9.0E-15	1.8E-11	1.1E-12	2.0E-10	2.5E-10	0	0
PEC in fish	kg/kg	6.9E-12	2.4E-13	6.1E-13	6.6E-08	1.4E-07	7.2E-08	1.3E-07	4.4E-08	0	0	0	0
PEC in meat	kg/kg	1.1E-13	6.4E-13	3.7E-12	1.9E-09	2.5E-09	1.3E-08	0	0	0	0	0	0
PEC in milk	kg/kg	8.8E-15	9.9E-15	1.3E-13	3.9E-11	3.6E-10	1.8E-10	0	7.4E-09	0	2.9E-11	0	0
PEC in plant leaves and grass	kg/kg	1.1E-13	3.3E-13	2.8E-12	8.3E-11	1.4E-10	8.7E-11	0	1.0E-09	1.0E-09	0	0	0
PEC in plant roots*	kg/kg	3.3E-12	3.7E-11	6.9E-10	5.8E-10	4.4E-09	7.0E-09	5.8E-05	2.9E-05	1.9E-10	2.8E-10	7.9E-07	2.8E-06
*Estimated from soil water concentration according to TGD													

A.6 Results of the sensitivity analyses

This section presents the detailed results of the sensitivity analyses regarding the total daily dose and the doses in air, drinking water, fish, plants, roots, meat and milk.

Tab. 38 Sensitivities of $DOSE_{total}$. Negligible parameters are not shown.

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
STP model parameters	Fraction oc raw sewage											**0.5**
	Input solids in raw sewage											**0.5**
	Sludge loading rate											0.2
	C activated sludge											0.2
	BOD						0.1	-0.1				-0.2
	Density solids PS											0.2
	Density solids raw sewage											-0.2
	K waterM									-0.1		
	Depth aerator									0.1		
Regional distribution module parameters	FconnectSTP				0.1	0.4	**0.8**	**-1.5**	-0.2	-0.2		**-1.2**
	AREA reg	-0.4	-0.5	**-0.6**	**-0.6**	**-0.8**	**-1.0**	**-0.7**	-0.2	-0.5	-0.2	**-1.0**
	RhoSolid		-0.3	**-0.8**	**-1.0**	**-1.0**	-0.1	-0.2				-0.5
	FSolidSoil		-0.3	**-0.8**	**-1.0**	**-1.0**	-0.1	-0.1				-0.5
	depthAgric		-0.3	**-0.8**	**-0.9**	**-1.0**	-0.1					**-0.5**
	FocSoil		-0.3	**-0.8**	**-0.9**	**-1.0**	-0.1					-0.5
	fAgric Reg		-0.1	-0.3	-0.3	**-0.6**	**-0.9**	-0.1			-0.1	**-0.6**
	heightAir	**-0.9**	**-0.7**	**-0.5**	-0.5	-0.2		**-0.7**	**-0.9**	**-0.7**		
	Rainrate			0.3	0.4	0.2	-0.2				**-0.9**	
	FrunoffSoil						-0.1				**-0.8**	
	windspeed	**-0.7**	**-0.6**	**-0.5**	-0.5	-0.2		-0.3	**-0.7**	**-0.7**		
	AREA EU	-0.3	-0.2	-0.2	-0.1	-0.1		-0.1	-0.4	-0.2	**-0.7**	
	kasl soilair						**-0.6**					
	FFlowOut Reg										**-0.5**	
	ConJunge	-0.1	**-0.5**	-0.1								
	SurfAer	-0.1	**-0.5**	-0.1								
	FWaterSed	0.1	0.3	0.2	0.1	0.1		0.5				
	fWater Reg						-0.1	-0.2	-0.1	-0.3		-0.4
	depthWater Reg											-0.4
	CollEffAer			0.3	0.4	0.2						
	fNatural Cont		-0.1	-0.1	-0.1						-0.4	
	Qstp						-0.1	0.1		0.1	-0.1	-0.3
	FocSusp		-0.1	-0.1				-0.2				
	kawWater								-0.1	-0.2		
	Temperatur	-0.1	-0.2	-0.1								
	fAgric Cont										-0.2	
	FInfSoil						-0.1					
	SuspEff						-0.1	0.1				
	fWater Cont										-0.1	
	SETTLEvelocity							-0.1				
	DepRateAer			0.1	0.1							
	depthSed							-0.1				
	SuspWater Reg							0.1				
	FWaterSoil											-0.1
	fInd Cont										-0.1	
	fNatural Reg		-0.1								-0.1	
Exposure module parameters	b	**3.4**	**6.3**	**20.8**	**26.1**	**27.8**	**15.3**	**4.3**				
	RHO plant	**-1.0**	**-0.9**	**-0.9**	**-1.0**	**-1.0**	**-0.9**	**-0.7**				**-0.6**
	Flipid plant	0.2	0.3	**0.8**	**0.9**	**1.0**	**0.9**	0.3				
	IH root		0.3	**0.8**	**0.9**	**1.0**	**0.9**					
	IH drw									0.2	**0.9**	0.3
	IH air								**0.9**	**0.7**		
	kgrowth plant	**-0.7**	**-0.6**	-0.2				-0.4				**-0.5**
	V leaf	**-0.7**	**-0.6**	-0.2				-0.4				**-0.5**
	AREA plant	**0.7**	**0.6**	0.2				0.4				
	g plant	**0.7**	**0.6**	0.2				0.4				
	IC grass	**0.6**	0.4	0.1				0.5				
	Qtransp											**0.5**
	IH leaf	0.3	0.2	0.1				0.2				**0.5**
	IH meat	0.4	0.3	0.1				0.3				
	IH fish		0.1	0.1			0.1	0.3	0.1	0.1	0.1	0.2
	IH milk	0.2	0.2					0.2				

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
Substance parameters	TempMelt	**1.1**	**2.4**	**0.7**	0.1							
	ERegfirstwastewater			0.2	0.2	**0.6**	**1.0**	0.3		0.4	**0.8**	**1.0**
	kdegsoil		-0.3	**-0.8**	**-1.0**	**-1.0**	-0.1					**-0.5**
	BIO inh								**0.9**	**0.7**		
	Kow	0.2	-0.1		0.1	0.1	**0.9**	-0.1		0.1		0.2
	ERegAir	**0.7**	**0.8**	**0.7**	**0.6**	0.3		**0.6**	0.2	0.3		
	EContAir	0.3	0.2	0.2	0.1	0.1		0.1	**0.6**	0.2		
	Vp	-0.1	0.5	0.1			**-0.6**	-0.2		-0.1		
	Molw	-0.2					**-0.6**	-0.2		-0.1		
	Sol	0.2					**0.6**	0.2		0.1		
	kdegwater										-0.1	-0.4
	kdegair	-0.2						-0.3	-0.2			
	EContfirstwastewater									0.1	0.2	
	kdegsed							-0.1				
	ERegfirstwater								0.1			

Tab. 39 Sensitivities of $DOSE_{Air}$. Negligible parameters are not shown.

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
Si	Input solids in raw sewage	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1
	Fraction oc raw sewage											0.1
Regional distribution module parameters	Rainrate	-0.1	-0.3	-0.3	-0.3	-0.3	-0.2				**-2.0**	**-1.2**
	heightAir	**-0.9**	**-0.7**	**-0.7**	**-0.7**	**-0.6**	**-1.0**	**-1.0**	**-1.0**	**-1.0**		
	AREA reg	-0.4	-0.5	**-0.5**	**-0.5**	**-0.5**	**-0.8**	**-0.6**	-0.2	-0.3	**-0.7**	**-1.0**
	windspeed	**-0.8**	**-0.7**	**-0.6**	**-0.6**	**-0.6**	-0.2	**-0.5**	**-0.7**	**-1.0**		
	ConJunge	-0.1	-0.1								0.1	**1.0**
	SurfAer	-0.1	-0.1								0.1	**1.0**
	kasl air										**0.5**	**1.0**
	depthInd											**-0.7**
	RhoSolid						-0.1					**-0.7**
	FSolidSoil						-0.1					**-0.7**
	FocSoil						-0.1					**-0.7**
	FrunoffSoil						-0.1				**-0.6**	-0.1
	fAgric Reg		-0.1	-0.1	-0.1	-0.1					**-0.6**	**-0.5**
	AREA EU	-0.3	-0.2	-0.2	-0.2	-0.2	-0.1	-0.1	-0.5	-0.3	-0.3	
	kawAir						0.1				0.5	
	fWater Reg						0.1				0.4	
	CollEffAer	-0.1	-0.3	-0.3	-0.3	-0.3						
	fNatural Reg			-0.1	-0.1	-0.1					-0.3	-0.3
	FInfSoil						-0.1				-0.3	-0.1
	FFlowOut Reg						-0.1				-0.2	
	fInd Reg										-0.2	-0.2
	kasl soilair						0.2					
	fNatural Cont		-0.1	-0.1	-0.1	-0.1					-0.2	
	Temperatur											0.2
	FWaterSed						0.1					
	depthAgric						-0.1					-0.1
	FWaterSoil											-0.1
	Qstp											-0.1
	fAgric Cont										-0.1	
	DepRateAer		-0.1	-0.1	-0.1	-0.1						
	FconnectSTP							-0.1				0.1
Ei	IH air	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**
Substance parameters	Sol						-0.2				**-2.0**	**-2.0**
	Molw						0.2				**2.0**	**2.0**
	Vp	0.1	0.1				0.2				**1.9**	**1.0**
	BIO inh	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**
	ERegfirstwastewater						**0.9**			0.1	0.4	0.1
	ERegAir	**0.7**	**0.8**	**0.8**	**0.8**	**0.8**		**0.9**	0.3	0.5		
	ERegInd										**0.5**	**0.9**
	kdegair	-0.2					**-0.8**	-0.5	-0.3			
	TempMelt	**0.5**	0.3	0.2							**-0.8**	0.2
	kdegsoil						-0.1					**-0.8**
	EContAir	0.3	0.2	0.2	0.2	0.2		0.1	**0.7**	0.4		
	Kow						-0.1					-0.5
	EContfirstwastewater						0.1			0.1	0.1	

Tab. 40 Sensitivities of $DOSE_{DrinkingWater}$. Negligible parameters are not shown.

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
STP model parameters	BOD	-0.1	0.0	0.0	0.0	-0.4	0.1	-0.4	-0.1	-0.1	0.0	-0.5
	C activated sludge								0.1	0.1		0.5
	Sludge loading rate								0.1	0.1		0.5
	K waterM									-0.2		
	Depth aerator									0.2		
	Depth SLS									0.1		
	HRTSLS									-0.1		
	Aeration rate									-0.1		
	Factor Hsieh et al. (1993b)									-0.1		
	Height air column									-0.1		
	Fraction oc activated sludge						0.1					
Regional distribution module parameters.	FconnectSTP	**-0.8**	-0.2	0.2	**-1.3**	**-4.3**	**1.0**	**-5.2**	**-2.2**	**-0.5**		**-4.1**
	FWaterSed	**1.9**	**2.6**	0.2	**0.7**	**3.4**		**1.6**				
	AREA reg	-0.3	-0.3	**-0.6**	**-0.6**	**-0.8**	**-1.0**	**-0.9**	**-0.9**	**-0.9**	-0.2	**-1.0**
	fAgric Reg	-0.2	-0.3	-0.3	-0.3	-0.3	**-1.0**	-0.1			-0.1	
	RhoSolid	-0.4	**-0.6**	**-1.0**	**-1.0**	**-0.7**	-0.1	-0.4				
	FSolidSoil	-0.3	-0.4	**-1.0**	**-1.0**	-0.5	-0.1	-0.1				
	FocSoil			**-1.0**	**-1.0**		-0.1					
	depthAgric			**-1.0**	**-1.0**		-0.1					
	FocSusp	**-0.6**	**-0.9**			**-1.0**		**-0.8**				
	fWater Reg	0.2	0.4			-0.2		**-0.8**	**-0.9**	**-0.9**		**-1.0**
	depthWater Reg	-0.2	-0.1					-0.1	-0.2			**-1.0**
	Rainrate		0.3	0.4	0.4	-0.1	-0.3	-0.1	-0.1	-0.1	**-0.9**	
	SuspWater Reg	0.2	0.5			**0.8**		0.3				
	FrunoffSoil	-0.1	-0.1			-0.1	-0.1	-0.1	-0.1	-0.1	**-0.8**	
	heightAir	**-0.8**	**-0.6**	**-0.5**	-0.5	-0.1						
	kawWater							-0.1	**-0.7**	**-0.8**		
	AREA EU	-0.4	-0.3	-0.1	-0.1	-0.2		-0.1	-0.1	-0.1	**-0.7**	
	windspeed	**-0.6**	**-0.6**	**-0.5**	-0.5	-0.1						
	kasl soilair						**-0.6**					
	FFlowOut Reg	-0.1	-0.1			-0.2		-0.1	-0.1	-0.1	**-0.5**	
	FSolidSed	0.3	0.4			**0.5**		0.1				
	Qstp	0.1				0.3	-0.2	0.4	0.2	0.2	-0.1	0.5
	Erosion	-0.3	-0.4			-0.5	-0.1	-0.1				
	CollEffAer	0.2	0.5	0.4	0.4	0.1						
	SuspEff	0.1				0.4	-0.1	0.4				
	fNatural Cont	-0.1	-0.2	-0.1	-0.1	-0.1					-0.4	
	SETTLEvelocity	-0.1	-0.1			-0.1		-0.3				
	kawAir	0.3	0.1					-0.1		-0.1		
	depthSed	-0.1	-0.2			-0.2		-0.2				
	fAgric Cont		-0.1								-0.2	
	SurfAer	0.1	0.1									
	ConJunge	0.1	0.1									
	SuspWaterCont	-0.1	-0.1			-0.1						
	fNatural Reg	-0.1	-0.1			-0.1					-0.1	
	FInfSoil						-0.1					
	fWater Cont										-0.1	
	DepRateAer		0.1	0.1	0.1							
	fInd Reg	-0.1	-0.1			-0.1						
	Temperatur	0.1										
	fInd Cont										-0.1	
E.	IH drw	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**
Substance parameters	TempMelt	**-1.0**	-0.3	-0.3								
	kdegsoil		-0.1	**-1.0**	**-1.0**		-0.1					
	ERegfirstwastewater	0.2		0.2	0.2	**0.9**	**1.0**	**1.0**	0.4	**1.0**	**0.8**	**1.0**
	kdegwater	-0.2	-0.2					-0.1	-0.2		-0.1	**-1.0**
	Kow	**-0.5**	**-0.7**	**-0.8**	**-0.8**	**-0.8**		**-0.7**				
	ERegAir	**0.5**	**0.7**	**0.7**	**0.6**	0.1						
	Vp	-0.4	-0.1				**-0.6**	-0.1	-0.1	-0.3		
	Molw	-0.2					**-0.6**	-0.1	-0.1	-0.3		
	Sol	0.2					**0.6**	0.1	0.1	0.3		
	ERegfirstwater								**0.6**			
	EContAir	0.3	0.2	0.1	0.1							
	kdegsed	-0.1	-0.2			-0.2		-0.2				
	EContfirstwastewater										0.2	
	kdegair	-0.2										

Tab. 41 Sensitivities of $DOSE_{Fish}$. Negligible parameters are not shown.

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
STP model parameters	BOD	-0.1	0.0	-0.2	-0.2	-0.4	-0.2	-0.4	-0.1	-0.1	0.0	-0.5
	C activated sludge								0.1	0.1		0.5
	Sludge loading rate								0.1	0.1		0.5
	K waterM									-0.2		
	Depth aerator									0.2		
	Depth SLS									0.1		
	HRTSLS									-0.1		
	Fraction oc activated sludge						-0.1					
	Aeration rate									-0.1		
	Factor Hsieh et al. (1993b)									-0.1		
	Height air column									-0.1		
Regional distribution module parameters	FconnectSTP	**-0.8**	-0.2	**-2.7**	**-3.1**	**-4.7**	**-1.8**	**-5.2**	**-2.2**	**-0.5**		**-4.1**
	FWaterSed	**1.9**	**2.8**	**3.3**	**3.3**	**3.4**	0.4	**1.6**				
	AREA reg	-0.3	-0.3	**-0.6**	**-0.6**	**-0.8**	**-0.8**	**-0.9**	**-0.9**	**-0.9**	-0.2	**-1.0**
	FocSusp	**-0.6**	**-0.9**	**-1.0**	**-1.0**	**-1.0**	-0.1	**-0.8**				
	fWater Reg	0.2	0.4			-0.2	**-0.7**	**-0.8**	**-0.9**	**-0.9**		**-1.0**
	depthWater Reg	-0.2	-0.1					-0.1	-0.2			**-1.0**
	Rainrate		0.3	0.1	0.1	-0.1		-0.1	-0.1	-0.1	**-0.9**	
	SuspWater Reg	0.2	0.5	**0.7**	**0.7**	**0.8**	0.1	0.3				
	FrunoffSoil	-0.1	-0.1	-0.1	-0.1	-0.1		-0.1	-0.1	-0.1	**-0.8**	
	heightAir	**-0.8**	**-0.6**	-0.3	-0.3	-0.1						
	kawWater						-0.2	-0.1	**-0.7**	**-0.8**		
	RhoSolid	-0.4	**-0.6**	**-0.7**	**-0.7**	**-0.7**		-0.4				
	AREA EU	-0.4	-0.3	-0.3	-0.2	-0.2	-0.2	-0.1	-0.1	-0.1	**-0.7**	
	windspeed	**-0.6**	**-0.6**	-0.3	-0.3	-0.1						
	FFlowOut Reg	-0.1	-0.1	-0.1	-0.1	-0.2	-0.2	-0.1	-0.1	-0.1	**-0.5**	
	FSolidSed	0.3	0.4	0.5	**0.5**	**0.5**	0.1	0.1				
	FSolidSoil	-0.3	-0.4	-0.5	-0.5	-0.5		-0.1				
	kawAir	0.3	0.1				-0.5	-0.1		-0.1		
	Qstp	0.1		0.2	0.2	0.3	0.3	0.4	0.2	0.2	-0.1	0.5
	Erosion	-0.3	-0.4	-0.4	-0.4	-0.5		-0.1				
	CollEffAer	0.2	0.5	0.2	0.2	0.1						
	SuspEff	0.1		0.2	0.2	0.4	0.1	0.4				
	fNatural Cont	-0.1	-0.2	-0.1	-0.1	-0.1	-0.1				-0.4	
	SETTLEvelocity	-0.1	-0.1	-0.1	-0.1	-0.1		-0.3				
	fAgric Reg	-0.2	-0.3	-0.3	-0.3	-0.3	-0.1	-0.1			-0.1	
	depthSed	-0.1	-0.2	-0.2	-0.2	-0.2		-0.2				
	kasl soilair						-0.2					
	fAgric Cont		-0.1	-0.1	-0.1						-0.2	
	SurfAer	0.1	0.1									
	ConJunge	0.1	0.1									
	SuspWaterCont	-0.1	-0.1	-0.1	-0.1	-0.1						
	fNatural Reg	-0.1	-0.1	-0.1	-0.1	-0.1					-0.1	
	fWater Cont										-0.1	
	DepRateAer		0.1	0.1								
	fInd Reg	-0.1	-0.1	-0.1	-0.1	-0.1						
	Temperatur	0.1										
	fInd Cont										-0.1	
	FocSoil						0.1					
E_i	IH fish	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**
Substance parameters	Kow	-0.5	**-1.0**	**-1.2**	**-1.3**	**-1.4**	**0.7**	**-0.9**	**0.8**	**0.8**		**0.8**
	TempMelt	**-1.0**	-0.3	-0.1								
	ERegfirstwastewater	0.2		**0.5**	**0.6**	**0.9**	**1.0**	**1.0**	0.4	**1.0**	**0.8**	**1.0**
	kdegwater	-0.2	-0.2					-0.1	-0.2		-0.1	**-1.0**
	ERegAir	**0.5**	**0.7**	0.4	0.3	0.1						
	Vp	-0.4	-0.1				**-0.7**	-0.1	-0.1	-0.3		
	Molw	-0.2					**-0.7**	-0.1	-0.1	-0.3		
	Sol	0.2					**0.7**	0.1	0.1	0.3		
	ERegfirstwater								**0.6**			
	EContAir	0.3	0.2	0.1	0.1							
	kdegsed	-0.1	-0.2	-0.2	-0.2	-0.2		-0.2				
	EContfirstwastewater										0.2	
	kdegair	-0.2										
	kdegsoil		-0.1	-0.1	-0.1							

Tab. 42 Sensitivities of $DOSE_{Leaf}$. Negligible parameters are not shown.

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
STP model parameters	Fraction oc raw sewage	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.3	0.9
	Input solids in raw sewage										0.3	**0.9**
	Density solids PS										0.1	0.3
	Density solids raw sewage										-0.1	-0.3
	BOD										0.2	
	Fraction oc activated sludge										0.1	
	Density solids SLS										-0.1	
	Density solids activated sludge										0.1	
Regional distribution module parameters	Rainrate	-0.1	-0.3	-0.3	-0.3	-0.3	-0.2				**-1.5**	-0.1
	AREA reg	-0.4	-0.5	**-0.5**	**-0.5**	**-0.5**	**-0.8**	**-0.6**	-0.2	-0.3	**-0.8**	**-1.0**
	fAgric Reg		-0.1	-0.1	-0.1	-0.1	-0.3				**-0.8**	**-1.0**
	heightAir	**-0.9**	**-0.7**	**-0.7**	**-0.7**	**-0.6**	**-0.8**	**-1.0**	**-1.0**	**-1.0**		
	ConJunge	-0.2	**-0.9**	**-0.9**	**-1.0**	**-1.0**		-0.1				
	SurfAer	-0.2	**-0.9**	**-0.9**	**-1.0**	**-1.0**		-0.1				
	FconnectSTP						0.3	-0.1			0.5	**1.0**
	windspeed	**-0.8**	**-0.7**	**-0.6**	**-0.6**	**-0.6**	-0.1	**-0.5**	**-0.7**	**-1.0**		
	Qstp										**-0.5**	**-1.0**
	depthAgric						-0.1					**-1.0**
	RhoSolid						-0.1					**-0.8**
	FSolidSoil						-0.1					**-0.8**
	FocSoil						-0.1					**-0.8**
	FrunoffSoil						-0.1				**-0.8**	
	FInfSoil						-0.1				**-0.6**	
	Temperatur	-0.1	-0.4	-0.5	-0.5	**-0.6**						
	AREA EU	-0.3	-0.2	-0.2	-0.2	-0.2	-0.1	-0.1	-0.5	-0.3	-0.2	
	CollEffAer	-0.1	-0.3	-0.3	-0.3	-0.3						
	kasl air										0.3	
	kawAir						0.1				0.2	
	fWater Reg						0.1				0.2	
	fNatural Reg			-0.1	-0.1	-0.1					-0.2	
	FFlowOut Reg										-0.1	
	fInd Reg										-0.1	
	FWaterSoil											-0.1
	FWaterSed						0.1					
	fNatural Cont		-0.1	-0.1	-0.1	-0.1					-0.1	
	DepRateAer		-0.1	-0.1	-0.1	-0.1						
Exposure module parameters	b	**3.3**	0.2	0.1		0.1	**12.1**	**4.8**	**2.8**	**0.9**		
	RHO plant	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**
	V leaf	**-0.8**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	-0.1	**-0.7**			**-1.0**	**-1.0**
	kgrowth plant	**-0.8**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	-0.1	**-0.7**			**-1.0**	**-1.0**
	IH leaf	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**
	Qtransp						0.3				**1.0**	**1.0**
	AREA plant	**0.8**	**1.0**	**1.0**	**1.0**	**1.0**	-0.1	**0.7**				
	g plant	**0.8**	**1.0**	**1.0**	**1.0**	**1.0**	-0.1	**0.7**				
	Flipid plant	0.2					**0.9**	0.3	**0.6**	0.3		
	Fwater plant								0.4	**0.7**		
Substance parameters	TempMelt	**1.3**	**4.0**	**6.4**	**6.7**	**8.8**		-0.1				
	ERegfirstwastewater						**0.9**			0.1	**0.7**	**1.0**
	Vp	-0.1	**0.8**	**0.9**	**1.0**	**1.0**	**-0.9**	-0.3	**-1.0**	**-1.0**	**0.5**	
	Molw	-0.2					**-0.9**	-0.3	**-1.0**	**-1.0**	**0.5**	
	Sol	0.2					**0.8**	0.3	**0.9**	**1.0**	**-0.5**	
	kdegsoil						-0.1					**-1.0**
	ERegAir	**0.7**	**0.8**	**0.8**	**0.8**	**0.8**		**0.9**	0.3	0.5		
	Kow	0.2					**0.8**	0.3	**0.6**	0.3	0.4	0.1
	EContAir	0.3	0.2	0.2	0.2	0.2		0.1	**0.7**	0.4		
	kdegair	-0.2					**-0.6**	-0.5	-0.3			
	ERegInd										0.3	
	EContfirstwastewater						0.1			0.1		

Tab. 43 Sensitivities of $DOSE_{Root}$. Negligible parameters are not shown.

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
STP model parameters	Fraction oc raw sewage	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.5	0.4	0.3	0.9
	Input solids in raw sewage								0.5	0.4	0.3	**0.9**
	Density solids PS								0.2	0.1	0.1	0.3
	Density solids raw sewage								-0.1	-0.1	-0.1	-0.3
	BOD						0.1	0.1		0.2	0.2	
	Fraction oc activated sludge						0.1			0.1	0.1	
	Density solids SLS									-0.1	-0.1	
	Density solids activated sludge									0.1	0.1	
Regional distribution module parameters	Rainrate	**0.7**	**0.5**	0.4	0.4	0.2	-0.3				**-1.5**	-0.1
	AREA reg	-0.4	-0.5	**-0.6**	**-0.6**	**-0.8**	**-1.0**	**-1.0**	**-0.6**	**-0.7**	**-0.8**	**-1.0**
	fAgric Reg	-0.1	-0.1	-0.3	-0.3	**-0.7**	**-1.0**	**-1.0**	**-0.5**	**-0.6**	**-0.8**	**-1.0**
	RhoSolid	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	-0.1	**-1.0**	-0.1			**-0.8**
	FSolidSoil	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	-0.1	**-1.0**	-0.1			**-0.8**
	FocSoil	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	-0.1	**-1.0**	-0.1			**-0.8**
	depthAgric	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	-0.1	**-1.0**	-0.1			**-1.0**
	FconnectSTP	0.1		0.2	0.2	**0.6**	**1.0**	**1.0**	0.5	**0.6**	0.5	**1.0**
	Qstp						-0.2	-0.1	-0.5	**-0.6**	**-0.5**	**-1.0**
	heightAir	**-0.8**	**-0.7**	**-0.5**	-0.5	-0.2			**-0.5**	-0.5		
	FrunoffSoil						-0.1				**-0.8**	
	ConJunge	**0.7**	0.1									
	SurfAer	**0.7**	0.1									
	windspeed	**-0.7**	**-0.6**	**-0.5**	-0.5	-0.2			-0.4	-0.4		
	CollEffAer	**0.7**	**0.5**	0.4	0.4	0.2						
	FInfSoil						-0.1				**-0.6**	
	kasl soilair						**-0.6**		-0.4	**-0.5**		
	Temperatur	0.4	0.1									
	kasl air										0.3	
	kawAir										0.2	
	AREA EU	-0.2	-0.2	-0.1	-0.1	-0.1			-0.2	-0.1	-0.2	
	fWater Reg										0.2	
	DepRateAer	0.2	0.1	0.1	0.1							
	fNatural Reg										-0.2	
	FFlowOut Reg										-0.1	
	fInd Reg										-0.1	
	FWaterSoil											-0.1
	fNatural Cont		-0.1	-0.1	-0.1						-0.1	
	SuspEff						-0.1	-0.1				
	Erosion						-0.1					
E_i	b	**21.0**	**24.2**	**26.6**	**27.9**	**29.2**	**16.8**	**24.7**	**2.9**	**0.9**		**2.3**
	RHO plant	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**
	IH root	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**
	Flipid plant	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**0.6**	0.3		**0.5**
	Fwater plant								0.4	**0.7**	**1.0**	0.5
Substance parameters	TempMelt	**-5.3**	**-0.7**	-0.3								
	kdegsoil	**-1.0**	**-1.0**	**-1.0**	**-1.0**	**-1.0**	-0.1	**-1.0**	-0.1			**-1.0**
	ERegfirstwastewater	0.1		0.2	0.2	**0.6**	**1.0**	**1.0**	**0.5**	**0.6**	**0.7**	**1.0**
	Vp	**-0.7**	-0.1				**-0.6**		**-0.9**	**-1.0**	**0.5**	
	Molw						**-0.6**		**-0.9**	**-1.0**	**0.5**	
	Sol						**0.6**		**0.9**	**1.0**	**-0.5**	
	Kow	0.1	0.1	0.1	0.1	0.1	**0.9**	0.1	**0.9**	**0.7**	0.4	**0.6**
	ERegAir	**0.6**	**0.8**	**0.7**	**0.6**	0.3			0.1	0.2		
	EContAir	0.3	0.2	0.1	0.1	0.1			0.3	0.2		
	ERegInd										0.3	
	kdegair	-0.1							-0.1			

Tab. 44 Sensitivities of $DOSE_{Meat}$. Negligible parameters are not shown.

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
STP model parameters	Fraction oc raw sewage	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.7
	Input solids in raw sewage											**0.7**
	Density solids PS											0.2
	Density solids raw sewage											-0.2
	Sludge loading rate									0.1		0.1
	BOD						0.1			-0.1		-0.1
	C activated sludge									0.1		0.1
	K waterM									-0.1		
	Depth aerator									0.1		
	Depth SLS									0.1		
	HRTSLS									-0.1		
	Fraction oc activated sludge						0.1					
Regional distribution module parameters	AREA reg	-0.4	-0.5	**-0.5**	**-0.6**	**-0.7**	**-1.0**	**-0.6**	-0.3	**-0.6**	-0.2	**-1.0**
	heightAir	**-0.9**	**-0.7**	**-0.7**	**-0.6**	-0.5		**-1.0**	**-0.9**	-0.5		
	fAgric Reg		-0.1	-0.1	-0.2	-0.4	**-1.0**				-0.1	**-0.8**
	FconnectSTP				0.1	0.3	**1.0**	-0.1	-0.3	-0.3		-0.1
	ConJunge	-0.2	**-0.8**	**-0.9**	**-0.7**	**-0.5**		-0.1				
	SurfAer	-0.2	**-0.8**	**-0.9**	**-0.7**	**-0.5**		-0.1				
	Rainrate	-0.1	-0.3	-0.3	-0.1	-0.1	-0.3			-0.1	**-0.9**	
	FrunoffSoil						-0.1				**-0.8**	
	depthAgric				-0.3	-0.5	-0.1					**-0.8**
	windspeed	**-0.8**	**-0.7**	**-0.6**	**-0.6**	-0.4		**-0.5**	**-0.7**	-0.5		
	FocSoil						**0.7**					**-0.7**
	AREA EU	-0.3	-0.2	-0.2	-0.2	-0.1		-0.1	-0.4	-0.2	**-0.7**	
	Qstp						-0.2			0.1	-0.1	**-0.7**
	RhoSolid				-0.2	-0.4						**-0.7**
	FSolidSoil				-0.2	-0.4						**-0.7**
	kasl soilair						**-0.6**					
	FFlowOut Reg										**-0.5**	
	fWater Reg								-0.1	-0.5		-0.2
	Temperatur	-0.1	-0.4	-0.5	-0.4	-0.3						
	kawWater								-0.1	-0.4		
	fNatural Cont		-0.1	-0.1	-0.1	-0.1					-0.4	
	CollEffAer	-0.1	-0.3	-0.2	-0.1	-0.1						
	depthWater Reg											-0.2
	fAgric Cont										-0.2	
	FInfSoil						-0.1					
	fWater Cont										-0.1	
	RhoWater					-0.1	-0.1					
	FWaterSoil					-0.1	-0.1					-0.1
	SuspEff						-0.1					
	kawAir									-0.1		
	DepRateAer		-0.1	-0.1								
	fInd Cont										-0.1	
	fNatural Reg			-0.1	-0.1						-0.1	
	Erosion						-0.1					
Exposure module parameters	b	**3.3**	0.1	0.1			0.5	**4.8**				
	RHO plant	**-1.0**	**-1.0**	**-1.0**	**-0.7**	**-0.5**		**-1.0**				**-0.8**
	IH meat	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**
	IC drw						0.1		0.1	**0.5**	**1.0**	0.2
	IC grass	**1.0**	**1.0**	**1.0**	**0.7**	**0.5**		**1.0**				**0.8**
	V leaf	**-0.8**	**-1.0**	**-1.0**	**-0.7**	**-0.5**		**-0.7**				**-0.8**
	kgrowth plant	**-0.8**	**-1.0**	**-1.0**	**-0.7**	**-0.5**		**-0.7**				**-0.8**
	AREA plant	**0.8**	**1.0**	**1.0**	**0.7**	**0.5**		**0.7**				
	g plant	**0.8**	**1.0**	**1.0**	**0.7**	**0.5**		**0.7**				
	IC soil				0.3	0.5	**0.9**					
	IC air								**0.9**	0.5		
	Qtransp											**0.8**
	Flipid plant	0.2						0.3				
Substance parameters	TempMelt	**1.3**	**4.0**	**6.1**	**4.9**	**4.5**		-0.1				
	Kow	0.2					**1.7**	0.3	**1.0**			**1.1**
	ERegfirstwastewater				0.1	0.3	**1.0**		0.1	**0.6**	**0.8**	**1.0**
	Vp	-0.1	**0.8**	**0.9**	**0.7**	**0.5**	**-0.6**	-0.3		-0.2		
	ERegAir	**0.7**	**0.8**	**0.8**	**0.8**	**0.6**		**0.9**	0.2	0.2		
	kdegsoil				-0.3	-0.5	-0.1					**-0.8**
	Molw	-0.2					**-0.6**	-0.3		-0.2		
	Sol	0.2					**0.6**	0.3		0.2		
	EContAir	0.3	0.2	0.2	0.2	0.1		0.1	**0.6**	0.2		
	kdegair	-0.2						-0.5	-0.2			
	kdegwater										-0.1	-0.2
	EContfirstwastewater										0.2	
	ERegfirstwater								0.1			

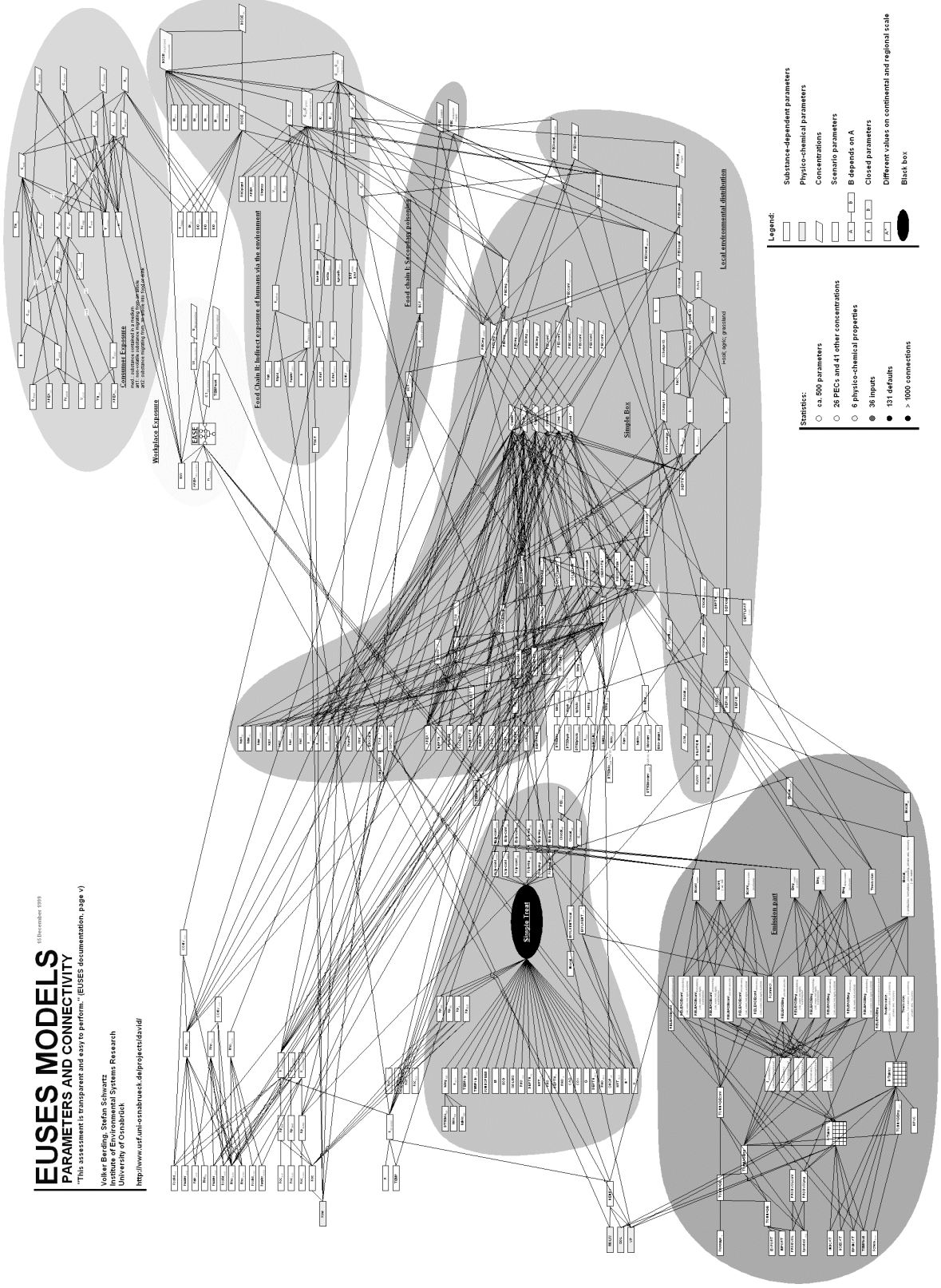
Tab. 45 Sensitivities of $DOSE_{Milk}$. Negligible parameters are not shown.

		TCDD	PeCDD	HxCDD	HpCDD	OCDD	HHCB	DEHP	BENZ	EDC	EDTA	LAS
STP model parameters	Fraction oc raw sewage	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.7
	Input solids in raw sewage											**0.7**
	Density solids PS											0.2
	Density solids raw sewage											-0.2
	Sludge loading rate									0.1		0.1
	BOD						0.1			-0.1		-0.1
	C activated sludge									0.1		0.1
	K waterM									-0.1		
	Depth aerator									0.1		
	Depth SLS									0.1		
	HRTSLS									-0.1		
	Fraction oc activated sludge						0.1					
Regional distribution module parameters	AREA reg	-0.4	-0.5	**-0.5**	**-0.6**	**-0.7**	**-1.0**	**-0.6**	-0.3	**-0.6**	-0.2	**-1.0**
	heightAir	**-0.9**	**-0.7**	**-0.7**	**-0.6**	-0.5		**-1.0**	**-0.9**	-0.5		
	fAgric Reg		-0.1	-0.1	-0.2	-0.4	**-1.0**				-0.1	**-0.8**
	FconnectSTP				0.1	0.3	**1.0**	-0.1	-0.3	-0.3		-0.1
	ConJunge	-0.2	**-0.8**	**-0.9**	**-0.7**	**-0.5**		-0.1				
	SurfAer	-0.2	**-0.8**	**-0.9**	**-0.7**	**-0.5**		-0.1				
	Rainrate	-0.1	-0.3	-0.3	-0.1	-0.1	-0.3			-0.1	**-0.9**	
	FrunoffSoil						-0.1				**-0.8**	
	depthAgric				-0.3	-0.5	-0.1					**-0.8**
	windspeed	**-0.8**	**-0.7**	**-0.6**	**-0.6**	-0.4		**-0.5**	**-0.7**	-0.5		
	FocSoil						**0.7**					**-0.7**
	AREA EU	-0.3	-0.2	-0.2	-0.2	-0.1		-0.1	-0.4	-0.2	**-0.7**	
	Qstp						-0.2			0.1	-0.1	**-0.7**
	RhoSolid				-0.2	-0.4						**-0.7**
	FSolidSoil				-0.2	-0.4						**-0.7**
	kasl soilair						**-0.6**					
	FFlowOut Reg										**-0.5**	
	fWater Reg								-0.1	-0.5		-0.2
	Temperatur	-0.1	-0.4	-0.5	-0.4	-0.3						
	kawWater								-0.1	-0.4		
	fNatural Cont		-0.1	-0.1	-0.1	-0.1					-0.4	
	CollEffAer	-0.1	-0.3	-0.2	-0.1	-0.1						
	depthWater Reg											-0.2
	fAgric Cont										-0.2	
	FInfSoil						-0.1					
	fWater Cont										-0.1	
	RhoWater					-0.1	-0.1					
	FWaterSoil					-0.1	-0.1					-0.1
	SuspEff						-0.1					
	kawAir									-0.1		
	DepRateAer		-0.1	-0.1								
	fInd Cont										-0.1	
	fNatural Reg			-0.1	-0.1						-0.1	
	Erosion						-0.1					
Exposure module parameters	b	**3.3**	0.1	0.1			0.5	**4.8**				
	RHO plant	**-1.0**	**-1.0**	**-1.0**	**-0.7**	**-0.5**		**-1.0**				**-0.8**
	IH milk	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**	**1.0**
	IC drw						0.1		0.1	**0.5**	**1.0**	0.2
	IC grass	**1.0**	**1.0**	**1.0**	**0.7**	**0.5**		**1.0**				**0.8**
	V leaf	**-0.8**	**-1.0**	**-1.0**	**-0.7**	**-0.5**		**-0.7**				**-0.8**
	kgrowth plant	**-0.8**	**-1.0**	**-1.0**	**-0.7**	**-0.5**		**-0.7**				**-0.8**
	AREA plant	**0.8**	**1.0**	**1.0**	**0.7**	**0.5**		**0.7**				
	g plant	**0.8**	**1.0**	**1.0**	**0.7**	**0.5**		**0.7**				
	IC soil				0.3	0.5	**0.9**					
	IC air								**0.9**	0.5		
	Qtransp											**0.8**
	Flipid plant	0.2						0.3				
Substance parameters	TempMelt	**1.3**	**4.0**	**6.1**	**4.9**	**4.5**		-0.1				
	Kow	0.2					**1.7**	0.3				0.1
	ERegfirstwastewater				0.1	0.3	**1.0**		0.1	**0.6**	**0.8**	**1.0**
	Vp	-0.1	**0.8**	**0.9**	**0.7**	**0.5**	**-0.6**	-0.3		-0.2		
	ERegAir	**0.7**	**0.8**	**0.8**	**0.8**	**0.6**		**0.9**	0.2	0.2		
	kdegsoil				-0.3	-0.5	-0.1					**-0.8**
	Molw	-0.2					**-0.6**	-0.3		-0.2		
	Sol	0.2					**0.6**	0.3		0.2		
	EContAir	0.3	0.2	0.2	0.2	0.1		0.1	**0.6**	0.2		
	kdegair	-0.2						-0.5	-0.2			
	kdegwater										-0.1	-0.2
	EContfirstwastewater										0.2	
	ERegfirstwater								0.1			

A.7 Mathematica® 3.0 Source code of the TGD exposure module

```
(* TGD exposure module (without water purification table), Mathematica 3.0 version *)

model := (

        (* plant model *)

        TSCF = 0.784 * Exp[(-1*(Log[10,Kow]-1.78)^2) / 2.44];

        (* TSCF =  0.0378; *)         (* for logKow > 4.5 *)
        (* TSCF =  0.0931; *)         (* for logKow < -0.5 *)

        Croot =(Fwaterplant + Flipidplant * Kow^b) * CAgricporew / RHOplant;

        Cleaf =((CAgricporew*TSCF*Qtransp / Vleaf +
                        (1-fPa) * Cair * gplant * AREAplant / Vleaf)) /
                (((AREAplant * gplant) /
                        ((Fairplant + (Fwaterplant + Flipidplant * Kow^b)/
                        (HENRY / (8.314 * TEMP))) *
                        Vleaf) + kplant * 24 + kgrowthplant) * RHOplant);

        (* fish model *)

        BCF = 10^(0.85 * Log[10,Kow] - 0.7 - 3);
        (* BCF = 10^(-0.2 * Log[10,Kow]^2 + 2.74 * Log[10,Kow] - 4.72 - 3); *)
                                    (* for logKow > 6 or log Kow < 10 *)
        (* BCF = 0.00141; *)          (* for logKow < 1 *)
        (* BCF = 0.479; *)            (* for logKow > 10 *)
        Cfish = CWater * BCF;

        (* cattle model *)

        BAFmeat = 10^(-7.6 + Log[10,Kow]);
        (* BAFmeat = 0.000000794; *)  (* for logKow < 1.5 *)
        (* BAFmeat = 0.0794; *)       (* for logKow > 6.5 *)
        BAFmilk = 10^(-8.1 + Log[10,Kow]);
        (* BAFmilk = 0.00000794; *)   (* for logKow < 3 *)
        (* BAFmilk = 0.0251; *)       (* for logKow > 6.5 *)

        Cmeat = BAFmeat * (Cleaf*ICgrass + Cgrassland*ICsoil + Cair*ICair + Cdrw*ICdrw);
        Cmilk = BAFmilk * (Cleaf*ICgrass + Cgrassland*ICsoil + Cair*ICair + Cdrw*ICdrw);

        (* human dose *)

        dose = 1/BW * (Cdrw*IHdrw + Cfish*IHfish + Cleaf*IHleaf + Croot*IHroot +
                        Cmeat*IHmeat + Cmilk*IHmilk + Cair*IHair * BIOinh/BIOoral)
)

values := (

        IHfish=0.12; IHmeat=0.3; IHmilk = 0.56; IHdrw = 0.002; IHleaf = 1.2;
        IHroot= 0.38; IHair = 20; BIOoral = 1; BIOinh = 0.75; BW = 70;
        ICgrass=67.6;ICsoil= 0.41; ICair=122; ICdrw=0.055; TEMP=285; RHOplant=700;
        kgrowthplant=0.035; Vleaf=0.002; b=0.95; Flipidplant=0.01; Fwaterplant=0.65;
        Fairplant=0.3; gplant=86.4; AREAplant=5; Qtransp=0.0011;
        (* HENRY, Kow, fPa, kplant, Ci depend on the substance *)
)

(* Calculate sensitivity for parameter x *)

values; Clear[x]; model; s = D[dose,x] * (x/dose); values; Print[s];
```

A.8 Structure of parameters in EUSES



A.9 Released project publications

Model validation

- TRAPP, S.; S. SCHWARTZ (2000). Proposals to Overcome Limitations in the EU Chemical Risk Assessment Scheme. Chemosphere **41** (7): 965-971
- SCHWARTZ (1997). Organische Schadstoffe in der Nahrungskette - Vorstudie zur Validierung von Expositionsmodellen. In: Reports of the Institute of Environmental Systems Science, University of Osnabrück, ISSN 1433-3805 (Ed. M. Matthies), Report No. 5, Osnabrück

Software evaluation

- BERDING, V.; S. SCHWARTZ; M. MATTHIES (1999). Visualisation of the Complexity of EUSES. Environ. Sci. Pollut. Res. **6**: 37-43
- SCHWARTZ, S.; V. BERDING; S. TRAPP; M. MATTHIES (1998). Quality Criteria for Environmental Risk Assessment Software - Using the Example of EUSES. Environ. Sci. Pollut. Res. **5**: 217-222

Comparison with monitoring data

- SCHWARTZ, S.; V. BERDING; M. MATTHIES (2000). Aquatic Fate Assessment of the Polycyclic Musk Fragrance HHCB - Scenario and Variability Analysis in Accordance with the EU Risk Assessment Guidelines. Chemosphere **41** (5): 671-679
- BERDING, V.; S. SCHWARTZ; M. MATTHIES (2000). Scenario analysis of a Level 3 multimedia model using generic and regional data. Environ. Sci. Pollut. Res. **7** (3) *(in press)*
- SCHWARTZ, S.; V. BERDING; M. MATTHIES (1999). Umweltexpositionsabschätzung des polycyclischen Moschus-Duftstoffes HHCB – Szenarienanalyse mit EUSES. Umweltmed. Forsch. Prax. **4**: 7-11

Comparison with alternative models

- BERDING, V.; F. KOORMANN; S. SCHWARTZ, J. WAGNER; M. MATTHIES (1999). Spatial Refinement of Regional Exposure Assessment. Proceedings of the NATO Advanced Research Workshop on Modelling of Environmental Chemical Exposure and Risk '99, Sofia

A.10 Test environment for the evaluation of the software

Tested software version:	EUSES 1.00 970214.
Implemented by:	TSA Group Delft bv, The Netherlands
Binaries:	6 Files (3.2 MB)
Sources:	Not available
Test platform:	80486 33, Windows 95, 32 MB Pentium® 133, Windows NT 4.0, 32 MB, Pentium® Pro 200, Windows NT 4.0, 128 MB
Graphic:	Matrox Graphics MGA Millennium (1152×864)