9 Evaluation of results

According to Beck and Chen (2000), "the public has a legitimate stake in being assured of the quality of models that are used to inform the decision-making process"[23] (p. 404). Thus, especially when developing and even more so when applying a model in a decision-support context where uncertainties are even larger than in natural sciences (e.g., Morgan and Henrion, 1990; Ragas et al., 1999), it is indispensable to give an indication of how relevant the model results are, keeping in mind that "a model is essentially a hypothesis and that no model is perfect; it would presumably cease to be a model if it were" (Addiscott, 1993, p. 17). Additionally, there is reason for evaluating model results because (United States - Environmental Protection Agency, 1997c):

- uncertain information from different sources of different quality often must be combined for the assessment;
- decisions need to be made about whether or how to expend resources to acquire additional information;
- biases may result in so-called 'best estimates' that in actuality are not very accurate; and
- important factors and potential sources of disagreement in a problem can be identified.

In the following, an overview will be given on the different concepts how to evaluate exposure and impact assessment models. Furthermore, the approach will be outlined and applied by which the reliability of the results with respect to impact assessment and their valuation at the European scale shall be assessed.

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[23] Note that this does not just hold for computational models but of course also to any data: a "reported value whose accuracy is entirely unknown is worthless" according to Eisenhart (1968) (p. 1201) because the intended use of the value determines its accuracy. Care must, therefore, be taken when using the value in contexts other than the specified purpose.

9.1 Terminology

9.1.1 Validation, verification, evaluation ...

There are many notions around that describe the action to evaluate a mathematical model. Beside evaluation, these include verification, validation, corroboration, confirmation and quality assurance (e.g., Caswell, 1976; Oreskes et al., 1994; Addiscott et al., 1995; Rykiel (Jr.), 1996; Veerkamp and Wolff, 1996; Robinson, 1999; Beck and Chen, 2000; Schwartz, 2000). It shall not be tried here to give definitions of the different notions. The reader is asked to refer to the existing literature (e.g., Caswell, 1976; Sargent, 1984; Tsang, 1991; Oreskes et al., 1994; Rykiel (Jr.), 1996; Robinson, 1999).

There is broad consensus in the (natural) scientific and engineering literature (e.g., Oreskes et al., 1994; Addiscott et al., 1995; Rykiel (Jr.), 1996; Veerkamp and Wolff, 1996; Robinson, 1999; Beck and Chen, 2000; Schwartz, 2000) that the term *validation* should not be used in the sense that a model or theory is correct although this impression may prevail in the public (Bredehoeft and Konikow, 1993). The scientific reasoning basically builds on the work of Popper (e.g., Popper, 1968) arguing that the truth of a scientific theory cannot be justified, rather one can justify the preference for one theory or model due to its applicability being superior in a special context than that of a competitor. In contrast, a scientific theory can only be falsified or invalidated.

In this context, one needs to distinguish predictive models/theories from explanatory ones (Caswell, 1976). Whereas explanatory theories cannot be validated (but corroborated) the predictive models can with respect to their purpose (e.g., Sargent, 1984). As Caswell (1976) expresses it "the important point is that the truth or reality of the model is never at issue" (p. 319). Thus, the term 'valid' can be regarded to mean 'well founded and applicable' (Addiscott et al., 1995), 'acceptable for an intended use' (Rykiel (Jr.), 1996; see also Beck and Chen, 2000), or 'established legitimacy' (Oreskes et al., 1994) leading to 'increased' (Robinson, 1999) or 'sufficient confidence' (Sargent, 1984) for predictive models. In this case, it becomes clear that the validation of a model cannot be performed in absolute terms (Caswell, 1976; Addiscott et al., 1995; Veerkamp and Wolff, 1996): it is rather relative with respect to the test procedure and the existence of competing models both of which may be subject to change in the future. The test procedure in turn depends on the purpose of the model (see below). Following the legal and theological parlance, valid can also mean to be efficacious, i.e., producing the intended effect (Addiscott et al., 1995).

Another, more technical dimension of the evaluation of results is the quality assurance of the computer code (Tsang, 1991; Veerkamp and Wolff, 1996;

Schwartz, 2000) sometimes referred to as 'verification' (Rykiel (Jr.), 1996) by demonstrating that the modelling formalism is correct and complies to the conceptual model (Robinson, 1999).

It is stressed that the term 'validation' is used in its technical sense in this work so that confidence is gained into the applicability of the model with respect to external cost assessments.

9.1.2 Uncertainty

From an empirical science perspective, *uncertainty* may be defined as "a scientist's assessment of the probable magnitude of (an) *error*" (Henrion and Fischhoff, 1986, p. 792) which in turn is perceived as "the actual difference between a measurement and the value of the quantity it is intended to measure" which "is generally unknown at the time of measurement" (ibid.). Uncertainty may also just be defined as "essentially the absence of information, information that may or not be obtainable" (Rowe, 1994, p. 743).

It is a common perception that an error in a measurement (or an assessment) may stem from different sources or may consist of different components. In empirical science, the conventional distinction is between (according to Henrion and Fischhoff (1986) and literature cited therein):

- *random error* or *Category A uncertainty* due to uncontrolled variability among observations and which is evaluated by statistical means and
- *systematic error* or *Category B uncertainty* which is equal to the difference between the value to which the observed mean converges and the true value.

In the realm for instance of regulatory exposure assessment 'science' in which the focus is on deriving safeguard standards based on mathematical simulation models, a different distinction is widely accepted and used.[24] The United States - Environmental Protection Agency (1997c) classifies the *sources* of uncertainty into:

- uncertainty regarding parameter values (*parameter uncertainty*) which can be further subdivided into *uncertainty due to lack of knowledge* (about a value or its heterogeneity according to Finley and Paustenbach (1994) or in a statistical or scientific sense according to Bogen and Spear (1987)) and *var-*

[24] Note that there are more uncertainty classification or grouping schemes. These include the subdivision into Type A and Type B uncertainty with different meanings (according to Safety Series No. 100 of the International Atomic Energy Agency (1989) as quoted by Hoffman and Hammonds (1994)) vs. McColl et al. (2000). These will not be explained here.

iability arising from true heterogeneity across people, places or time termed inter-individual, spatial and temporal variability, respectively; whereas uncertainty may be reducible through further measurements, variability is usually not (Bogen and Spear, 1987; Burmaster and Anderson, 1994; Finley and Paustenbach, 1994),

- uncertainty regarding missing or incomplete information needed to fully define exposure and dose (*scenario uncertainty*) and
- uncertainty regarding gaps in scientific theory required to make predictions on the basis of causal inferences (*model uncertainty*).

Another aspect is not explicitly mentioned in the above list of uncertainty sources. This is the *decision-rule uncertainty*, a term proposed by Finkel (1990) which is referred to in several other publications (e.g., United States - Environmental Protection Agency, 1998; Hertwich et al., 1999, 2000; McKone and Hertwich, 2001). Hertwich et al. (2000) describe this type of uncertainty as the "uncertainty about whether the model output is a relevant representation of an issue of concern, and whether the model settings represent the conditions of concern" (p. 443). It arises "whenever there is ambiguity or controversy about how to quantify or compare social objectives" (ibid., p. 442) leading to "imprecise or inappropriate operational definitions for desired outcome criteria, value parameters, and decision variables" (McColl et al., 2000, p. 2-4) and involves value judgements or preferences. Following this description, also the *fundamental uncertainty* (Ragas et al., 1999)[25] and the uncertainty introduced by the *model context* (Rykiel (Jr.), 1996)[26] can be regarded as decision-rule uncertainty. The value judgements include the choice of the indicator, how to deal with timings of impacts (e.g., use of discounting or temporal cutoffs), formulation of an environmental fate model as an open or closed system, selection of summary statistics, choice of variables that express subjective value judgements in the form of utility functions (e.g., the monetary value attributed to loss of life), and also how (parameter) uncertainty is addressed (Hertwich et al., 2000; McColl et al., 2000). This list may be extended by the choice of whether to include certain exposure path-

[25] Fundamental uncertainty "stems from the assumptions underlying a model structure and its equations" and "was assessed tentatively on the basis of an analysis of (a multimedia fate model's) assumptions and equations and the results of a postal questionnaire among scientists and scientifically trained policy makers and representatives of interest groups" (ibid., p. 1857). Fundamental uncertainty is distinguished from what is called 'operational uncertainty' which is addressed by an output uncertainty analysis.

[26] "Context embodies all the assumptions, especially those that are unstated and relegated to the system environment of the model" (ibid., p. 242).

ways or not, a decision that may be affected by missing or incomplete information resulting in scenario uncertainty. Thus, scenario uncertainty can be regarded a sub-aspect of decision-rule uncertainty. The other uncertainty types distinguished by Finkel (1990) correspond to model and parameter uncertainty if the latter is distinguished into true uncertainty and (natural) variability. Due to the larger scope of decision-rule uncertainty as compared to scenario uncertainty, the uncertainty source classification by Finkel (1990) is adopted in this work.

Another uncertainty classification scheme has been provided by Rowe (1994) although not in terms of sources but *dimensions*. It is distinguished between

- *metrical*: uncertainty and variability in measurement,
- *structural*: uncertainty due to complexity, including models and their validation,
- *temporal*: uncertainty in future and past states,
- *translational*: uncertainty in explaining uncertain results (or communication uncertainty).

The metrical dimension of uncertainty is mostly related to both aspects of parameter uncertainty. Structural uncertainty tries to give indications about the 'usefulness of the model' and may, therefore, touch upon all different sources of uncertainty stated above. Past temporal uncertainty is exclusively related to the imperfect knowledge of past states of the 'world', i.e., true parameter uncertainty. More importantly, future temporal uncertainty refers to uncertain predictions of future states. This may result from incomplete scientific theory and/or from uncertain capabilities to predict future states due to the complexity of the world (e.g., temporal and spatial variability, vast amounts of interactions and feedback).[27] It is, thus, mostly related to model uncertainty although incomplete knowledge about all values of the most influential state variables will not be achieved either; if we tried we would take the chance to dramatically change the properties of the environmental system (cf. Tsang, 1991). Translational uncertainty occurs when (uncertain) results are communicated. It is due to the fact that the analyst or assessor as well as the decision-makers, professionals, stakeholders and the public have different levels of training and capability of understanding the results (Rowe, 1994). They also have different scientific perspectives with different terminologies. It is clear that this dimension of uncertainty cannot be analysed prior to the dissemination of results. What needs to be kept in mind, however, is that

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[27] The mechanistically founded deterministic believe of Sir Isaac Newton (1642-1727) that one could predict the future if one knew all present values of the state variables has disappeared at the latest since the discovery of chaos.

the assessor has to try best to put his/her results into context and be precise about the outcome so that the audience will not be misled in perceiving the results (Chapman, 2001).

Apart from the translational uncertainty dimension, there are different relevant approaches how to evaluate the different sources of uncertainty also with respect to the different dimensions which will be presented to some extent in the following.

9.2 Approaches for the evaluation of results

Prior to conducting an evaluation of a model, there appears to exist consensus that the following topics need to be specified

- the 'purpose of the model' (Caswell, 1976; Rykiel (Jr.), 1996; Scott et al., 2000; Schwartz, 2000), 'objectives' (Robinson, 1999), or 'task specification' (Beck and Chen, 2000),
- the 'performance criteria' (Rykiel (Jr.), 1996), 'demands' (Caswell, 1976), or 'statements on undesirable outcomes' (Schwartz, 2000) for which a rigorously defined 'endpoint or target of the assessment' (Hoffman and Hammonds, 1994) is necessary and
- the 'model context' (Rykiel (Jr.), 1996), i.e., the underlying assumptions and resulting contextual 'ranges of applicability' (Tsang, 1991) or 'limitations' (Veerkamp and Wolff, 1996) of the model.

Once these topics are specified, there are different ways how to evaluate mathematical models. There exist many aspects which may be considered in the evaluation of models and their results. It might, therefore, be useful to try to group the different approaches. One way of subdividing the validation process is into the components (Rykiel (Jr.), 1996): (a) operational or whole model validation, (b) theoretical or conceptual validity, and (c) data validation of which only the operational and data components can be validated whereas theory cannot (see reasons given above). *Operational validation* (or 'black-box validation', Robinson, 1999) tries to demonstrate that the model results meet the overall performance criteria (Rykiel (Jr.), 1996) according to a model's purpose or task (Schwartz, 2000). It is most often done by comparing the model results with independent results (see section 9.2.2). *Conceptual validity* "means that the theories and assumptions underlying the conceptual model are correct, or at least justifiable, and that the model representation of the problem or system, its structure, logic, mathematical, and causal relationships, are reasonable for the model's intended use" (Rykiel (Jr.), 1996, p. 234). Conceptual validity is understood here to also include the aspect whether the conceptual model is implemented into the computational model (tool) without error. *Data validation* is understood here to indicate the quality or relevance of the data used.

One may alternatively distinguish between internal and external evaluations. *Internal* evaluations provide an assessment of the "primary, theoretical material and constituent hypotheses of which the model is composed" which is "essentially a matter of making judgements about the quality of the internal properties of the model, in particular, of whether the functions ... have been 'properly' expressed and 'realistic' values assigned to the parameters ... appearing in them" (Beck and Chen, 2000, p. 406). This description of internal evaluation complies with both concepts of *conceptual* or *theoretical validity* and *data validation* given above. *External* evaluations correspond to the definition of *operational validation* given above.

The assignment of the different approaches used for the evaluation of models and their results into the categories given above is not always straightforward (Schwartz, 2000) and presumably, therefore, not attempted by some authors (e.g., Rykiel (Jr.), 1996). Some authors give lists of possible validation or evaluation approaches (e.g., Sargent, 1984; Rykiel (Jr.), 1996) of which others even recommend to apply as many of them as possible (Tsang, 1991). This is definitely beyond the scope of the present work. In the following, some simple and/or rather common approaches will be presented and reasons given to what extent these will be employed in the present exercise.

9.2.1 Minimum requirements towards uncertainty analysis of exposure assessments according to United States - Environmental Protection Agency (1997c)

According to United States - Environmental Protection Agency (1997c), the exposure assessor should at a minimum address uncertainty qualitatively by answering questions such as (adapted from pp. 2-6f, ibid.):

- What is the basis or rationale for selecting assumptions and/or parameters, such as data, conceptual or mathematical models, exposure scenarios, scientific judgement, policies or guidance of regulatory bodies, 'what if' considerations, etc.?
- What is the range or variability of the key parameters (once identified)? How were the parameter values selected for use in the assessment? Were average, median, or upper-percentile values chosen? If other choices had been made, how would the results have differed?
- What is the assessor's confidence (including qualitative confidence aspects) in the key parameters and the overall assessment (e.g., selected exposure scenarios)? What are the quality and the extent of the data base(s) supporting the selection of the chosen values?

It is clear from the context of this guidance document, that the validation exercise stops at the exposure level and does not include the components of impact assessment and valuation. However, most of the recommendations given are applicable to the other steps of the Impact Pathway Approach as well. Whereas the first bullet point which may be referred to as decision-rule uncertainty assessment is rather qualitative, especially the second one requires the identification of key parameters first. An approach how to do this is presented in section 9.2.4 below. The third bullet point deals with confidence in the overall model which may need to be built first for example by means of other validation approaches such as comparison with reported data (section 9.2.2) or the analysis of different scenarios (section 9.2.3). Thus, in order for the presented qualitative uncertainty assessment not to become overly subjective, it is recommended here to accompany it with more quantitative assessments that are not too resource-intensive (such as probabilistic uncertainty assessments, see section 9.2.5).

9.2.2 Comparison with independent data

As stated above, operational validation tries to demonstrate that the model results meet the overall performance criteria (Rykiel (Jr.), 1996) according to a model's purpose or task (Schwartz, 2000). It is most often done by comparing the model results with independent results that are the output especially of monitoring exercises (Beck and Chen, 2000) and which can be termed *traditional validation* (Veerkamp and Wolff, 1996), but may alternatively encompass results from presumably more complex or specific models (Rykiel (Jr.), 1996). Comparing modelled data to observed ones can be referred to as *history match* (Bredehoeft and Konikow, 1993) or as part of a *historical data validation* process (Rykiel (Jr.), 1996), terms superior to just stating 'validation' as they describe more precisely what is done. It shall be noted that there is agreement that this type of validation procedure, however, is not applicable to generic or screening level exposure models for which the area of potential measurements is unspecified or the substance to be assessed is not present due to the fact that it is not yet allowed to be marketed (Veerkamp and Wolff, 1996; Schwartz, 2000).

There is a dilemma with this type of validation approach which is why "(a) neutral language is needed for the evaluation of model performance" (Oreskes et al., 1994, p. 643). If there is agreement between the simulated and the independent data in terms of the pre-defined evaluation criteria the model (result) may be judged to be 'accurate' (Caswell, 1976; Schwartz, 2000), to show 'a precise or accurate fit' (Oreskes et al., 1994), to 'increase a model's credibility' (Rykiel (Jr.), 1996), or to 'increase the confidence in a model' (Robinson, 1999). Furthermore, depending on the evaluation criteria such an accuracy may be quantitative or qualitative (Rykiel (Jr.), 1996). However, validity and accuracy are related but separate concepts (Robinson, 1999). This is because the match between model results and independent data does not prove the validity of the underlying model concept, i.e., that the model is an accurate cause-effect representation of the real system (Rykiel (Jr.), 1996). There could be other conceptual models that would produce the same output, a "situation ... referred to by scientists as nonuniqueness and by philosophers as underdetermination" (Oreskes et al., 1994, p. 642). On the other hand, if a model (result) does not compare well according to the performance criteria this does not invalidate the conceptual model (Sargent, 1984). It merely indicates that something is wrong (Oreskes et al., 1994) and be it 'just' that the data fed into the (overall) model is a worse representation of 'reality' than the underlying (conceptual) model (Rykiel (Jr.), 1996).

As a consequence, it is clear that such a comparison with independent data can increase the credibility of a model or provide evidence of a model's accuracy including the used data base.

9.2.3 Scenario analysis

With respect to model validation, scenario analyses provide point estimates while changing single components and/or assumptions in order to get an overview about the possible effect on the results albeit without indicating probabilities. Different scenarios may be analysed which differ with respect to parameter values, considered processes and their formulations, boundary conditions (e.g., open vs. closed system), emissions, or spatial and/or temporal resolution etc..[28] By means of this validation approach, also aspects of the decision-rule uncertainty can be addressed which may be supplemented by parameter sensitivity analysis (see section 9.2.4) and/or probabilistic uncertainty analysis (see section 9.2.5).

In the (regulatory) risk assessment context, some authors describe scenario analysis most useful as a screening approach (Finley and Paustenbach, 1994) especially as a *bounding estimate* or as a desirable first step prior to conducting a probabilistic uncertainty assessment (Burmaster and Anderson, 1994). The bounding estimate constitutes an upper limit of individual exposure, dose or risk and is most often used only to eliminate pathways from further consideration during screening-level assessments (United States - Environmental Protection Agency, 1992). However, care must be taken when combining parameter values selected according to worst-case assumptions. Such assessments at times substan-

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[28] A rather advanced scenario analysis approach in terms of computational requirements according to Schwartz (2000) is the range/confidence estimate approach (Richards and Rowe, 1999).

tially overestimate the exposure even of high-end individuals (e.g., Finley and Paustenbach, 1994; Price et al., 1996; Mekel and Fehr, 2000) and, therefore, need to be classified as unrealistic. As a consequence, Schwartz (2000) calls them *cumulative worst case* scenarios. Also, the United States - Environmental Protection Agency (1992) stresses that the assessment according to the bounding estimate scenario is not apt to identify important or significant pathways since these should not be mistaken as actual exposure estimates.

Apart from the problem with respect to the cumulative worst case scenario whose risk is deemed small in this study, the discriminative power of scenario analyses is rather weak due to the fact that the results consist of point estimates without probability indications. Still, the scenario analysis provides indicative information about the importance of the varied component and/or assumption relative to the reference scenario. This is in line with United States - Environmental Protection Agency (1992) in that scenario analyses are proposed as a legitimate although more uncertain alternative to probabilistic uncertainty assessments. In this case, specific values are selected from the key parameter distributions with which scenario analyses are conducted.

9.2.4 Sensitivity analysis of parameters

The validation process of models with different spatial scale of applicability require different validation approaches (Addiscott et al., 1995; Rykiel (Jr.), 1996). This also holds for models that are very complex at least in terms of the number of variables and parameters contained (Caswell, 1976). For both categories of models, the definition of statistical criteria for evaluation purposes constitutes a problem (Caswell, 1976; Addiscott et al., 1995). For the validation of models operating at rather large scales, Addiscott et al. (1995) suggests to use approaches to test for efficacy which include sensitivity analysis although these are judged by some authors not to constitute a sufficient approach with respect to model validation (e.g., Schwartz, 2000). Additionally, sensitivity analyses most often are used to identify the most important parameters in terms of impact on the result on which to perform a probabilistic uncertainty assessment (e.g., Burmaster and Anderson, 1994; Price et al., 1996; Richards and Rowe, 1999; Schwartz, 2000). In this context, one needs to distinguish between important and sensitive parameters (Hamby, 1994). If a sensitive parameter is known precisely, it adds little to the variability of the output and, thus, its importance is restricted. Important parameters, in turn, are sensitive and rather uncertain.

According to Saltelli (2000), sensitivity analysis is "the study of how the variation in the output of a model (numerical or otherwise) can be apportioned, qualitatively or quantitatively, to different sources of variation, and of how the given model depends upon the information fed into it" (p. 3). In a more general sense, scenario analyses (see section 9.2.3) can be seen to also constitute a type of sensitivity analysis where not only the influence of variable parameters may be investigated. However, the definition of sensitivity analysis given above encompasses a variance decomposition in the results to the different influential factors ('apportioning of uncertainties') based on an uncertainty assessment for example of the Monte Carlo type (Saltelli, 2000). This is done by the class of *global sensitivity analysis* techniques (Campolongo et al., 2000b; Saltelli, 2000) which will not be considered further. The term 'sensitivity analysis' shall rather be used here in its original meaning according to Saltelli (2000) which is dealing with uncertainty in the parameters (cf. Cox and Baybutt, 1981).[29] The main purpose of sensitivity analysis in the present work is to perform a so-called *factor screening* (Campolongo et al., 2000b), i.e., the identification of 'key' (Beck and Chen, 2000), 'important' (Campolongo et al., 2000a), 'influential' (Campolongo et al., 2000b), or 'critical' (Richards and Rowe, 1999) parameters. This is done by so-called screening approaches which are computationally economical and tend to provide only qualitative sensitivity measures for instance in the form of rankings (Hamby, 1994; Campolongo et al., 2000a). Such screening approaches constitute only an early step when performing a sensitivity analysis according to its broader definition (Saltelli, 2000). Whereas scenario analyses investigate the sensitivity of factors also other than parameters (see section 9.2.3), sensitivity analysis in this context focuses on the parameter values. Their purpose is to identify the most important parameters in this work and, thus, primarily address parameter uncertainty (Hamby, 1994).

There exist a variety of screening sensitivity approaches with remarkably differing complexity (Campolongo et al., 2000a) whose appropriateness is highly model-dependent (Campolongo et al., 2000b). These can be classified as internal and as external validation tools and may allow to vary one parameter or several at a time (Schwartz, 2000). A straightforward example for those sensitivity analysis methods allowing to vary one parameter is the differential sensitivity analysis (Finley and Paustenbach, 1994; Hamby, 1994; Saltelli, 2000) which is termed 'standard one-factor-at-a-time' (OAT) or *ceteris paribus* sensitivity screening methods by Campolongo et al. (2000a). A control scenario or experiment is defined in which all parameters assume their 'nominal', 'control' or 'reference' values (Campolongo et al., 2000b; Saltelli, 2000). Then several runs are performed

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[29] Note that the distinction into input variables and model parameters is not made here. According to Campolongo et al. (2000a) among others, "(i)nput variables are directly observable in the corresponding real system, whereas parameters are not (they may be estimated)" (p. 66).

in which only one parameter is changed at a time in a defined range. Different variations from the nominal value can be found in the literature for example 5 % (Saltelli, 2000), 10 % (Finley and Paustenbach, 1994), or 20 % (Price et al., 1996). One has to keep in mind, however, that this approach should be used only when the model to be investigated is known to be linear (Saltelli, 2000) which is the case for the linear algebra approach of the Mackay-type.

Another way of performing screening sensitivity analysis is not to just assume a fix variation employed to each parameter but to either assume probability density functions (based on parametric statistics) or ranges of variation (non-parametric statistics) for each parameter (Saltelli, 2000). In order to become even more complex, the parameter (or factor) variation can be done by random sampling or experimental design in which case not just one parameter (factor) is changed (Saltelli, 2000).

In this work, the standard OAT approach is followed whose advantage Schwartz (2000) emphasises by stating that it "always results in the same sensitivity indices, irrespective of the number of investigated variables, and is easily reproducible without further software" (p. 20). It is, therefore, selected for the identification of the most influential or key parameters which is appropriate for linear models (Campolongo et al., 2000b).

9.2.5 Probabilistic uncertainty assessment

The parameter sensitivity analysis presented in the previous section constitutes a highly useful prerequisite prior to performing a probabilistic uncertainty assessment in order to economically focus the efforts (e.g., Burmaster and Anderson, 1994; Price et al., 1996; Richards and Rowe, 1999; Schwartz, 2000). Probabilistic uncertainty assessments address the variability and uncertainty in the parameter values, preferably even separately (Burmaster and Anderson, 1994) as done for instance by Price et al. (1996) and Mekel and Fehr (2000). These are carried out by defining either ranges or probability density functions to each of the prioritised parameters (United States - Environmental Protection Agency, 1992; Saltelli, 2000). In a next step, different sets of parameter values are sampled from these ranges or distributions. Many model runs are conducted in this way in order to yield not just a range of outputs but also a probability density function of these indicating the assessed likelihood of each of the model result ranges (percentiles) to occur. There is also guidance in terms of principles of good practice available suggested in the scientific literature (Burmaster and Anderson, 1994).

Examples of such probability parameter uncertainty assessments include Monte Carlo techniques, Markov chain methods and Gibbs sampling procedures (Richards and Rowe, 1999). Of these, Monte Carlo techniques are the most commonly used methods. There are also different sampling schemes (Campolongo et al., 2000b) of which simple random or Latin hypercube sampling are generally employed in risk assessments (United States - Environmental Protection Agency, 1997d).

The main advantage of the probabilistic parameter uncertainty analysis in the context of exposure assessments is that it characterizes a range of potential risks and their likelihood of occurrence (e.g., Finley and Paustenbach, 1994). This result cannot be replaced by other techniques. Its main disadvantage is the big computational effort in terms of time and hardware (Cox and Baybutt, 1981; Burmaster and Anderson, 1994; Finley and Paustenbach, 1994; Schwartz, 2000). The assignment of an appropriate probability density function to each prioritised parameter is also a topic of its own (Finley and Paustenbach, 1994; Campolongo et al., 2000b). Cox and Baybutt (1981) term the problem of the right selection of a distribution for a parameter the 'uncertainty in the uncertainty'. However, it appears as if the selection of the distribution function as such has less an influence than for instance the choice of the allowed ranges (Finley and Paustenbach, 1994; Campolongo et al., 2000b). Another disadvantage for the present work is related to the way the model to be evaluated here is implemented (see section 4.4). There are different software products for probabilistic uncertainty assessments available for spreadsheet applications like Crystal Ball® or Risk 4.5®. However, the model developed here is not a spreadsheet model. Respective off-the-shelf probabilistic uncertainty assessment plug-ins are not available. As a consequence, a tailored probabilistic uncertainty assessment would need to be implemented which has not been possible especially due to time constraints. It is, therefore, not possible to include a probabilistic uncertainty assessment of the critical parameters in the present uncertainty analysis.

9.2.6 Expert judgement

There are several evaluation approaches which involve the judgement of (independent) experts. These include face validity, Turing test and publication of the model and its results in the open-literature or other types of peer review (Sargent, 1984; Tsang, 1991; Rykiel (Jr.), 1996; Beck and Chen, 2000). These evaluations require the availability of resources, i.e., time, on the side of the respective experts to allocate to the inspection of the model. Apart from funded model evaluation exercises and review in the open-literature this will hardly be possible to achieve so that the obvious publication in the open-literature constitutes a good alternative to allow experts to have a closer look at the model and its results.

9.3 Followed approach

A thorough in-depth evaluation of the presented external cost assessment would constitute a major task in its own right. Therefore, a more semi-quantitative approach to model evaluation is followed in the present work. The basis of the uncertainty analysis constitutes the minimum requirements articulated by United States - Environmental Protection Agency (1997c) and described in section 9.2.1. This qualitative uncertainty analysis presented in section 9.3.1 is supplemented by scenario analyses as mostly a decision-rule uncertainty assessment tool (cf. section 9.2.3) and parameter sensitivity analysis (cf. section 9.2.4). In both cases, a prioritisation of the assumptions to be varied and the parameters to be included will take place (see sections 9.3.3 and 9.3.4). In order to evaluate the model's accuracy, a comparison with reported values will be presented in section 9.3.2.

Before evaluating a model, its purpose and context needs to be defined explicitly. The purpose of the Impact Pathway Approach is to provide external cost estimates that are specific to a human activity, be it a single or a collective/societal activity. This way it supports the decision-making process with respect to conducting cost-benefit analyses of different policy options. The focus of this work is to assess the external costs resulting from human health impacts that are due to indirect exposure in order to provide decision-support at the European Union level and/or its member countries including acceding and (potential) accession countries. This implies for example that the assessment be done at the European scale. Additionally, a certain degree of spatial resolution is required in order to link the different human activities to the regionally occurring impacts. The 'context' of the model describes its applicability or limitations as a result of the underlying assumptions.

9.3.1 Qualitative uncertainty analysis according to United States - Environmental Protection Agency (1997c)

As part of the qualitative uncertainty analysis of exposure assessments, it shall be outlined to what extent the addressed assumptions or choices may influence the results of the assessment (United States - Environmental Protection Agency, 1997c).

Mathematical approach, non-linearities and equilibrium partitioning

In particular the environmental fate model is formulated as an inhomogeneous system of ordinary linear first order differential equations (cf. section 4.2) according to the Mackay-type approach (Mackay, 1979, 1991). This linear formulation has been adopted due to the ease of computation, the flexibility in terms of output, i.e., steady-state and dynamic computations which can be used for different contexts (cf. section 4.4.2 on different temporal modes of operation). This is generally oversimplifying for example environmental inhomogeneities and adsorption behaviours of trace elements (e.g., Kleeberg, 1992; Addiscott, 1993; Selim and Amacher, 2001; Smedley and Kinniburgh, 2003). However, due to the data demand of alternative modelling concepts at the scale and spatial resolution required, it is deemed a valid (at least initial) approach.

When developing a multimedia model of the Mackay-type, one has principally three options how to formulate the equations. Mackay and co-workers have based their models on fugacity (Mackay, 1979, 1991; Diamond et al., 1992; Wania and Mackay, 1995; Cousins and Mackay, 2001), whereas it is also possible to formulate them based on concentrations (Brandes et al., 1996) or mass (Pennington et al., 2005). The approaches are principally equivalent although the fugacity models have problems with substances that are rather involatile ('aquivalent' approach by Mackay and Diamond, 1989 and Diamond et al., 1992). In the end, it is a matter of preference how to formulate the mass balance.

The choice of a box model of the Mackay-type brings about the assumption of homogeneity and intra-compartmental equilibrium. While the issue of homogeneity is addressed below, it shall be noted that the assumption of equilibrium is not appropriate for substances that sorb strongly to particles and/or undergo rapid transformation (e.g., Mulkey et al., 1993). For reasons given in section 3.2.3, the assumption of equilibrium is, nevertheless, made which may lead to an overestimation or underestimation of the results, depending on the effective mobility of the substance (cf. United States - Environmental Protection Agency, 1996a), its characteristics in terms of most important exposure pathways, and the time horizon analysed or the discount rate chosen.

Worst case vs. representative estimates

It has been the intention within this study to avoid the use of conservative or worst-case data and assumptions. The problem to arrive at a so-called "cumulative worst case" (Schwartz, 2000, p. 21) where the combination of several worst-case parameter values turns the assessment overly conservative (cf. Price et al., 1996) is, therefore, expected to be highly reduced. As it has been made use of expectation values (e.g., averages) the assessment is rather believed to result in reasonable or representative estimates. However, the present methodology partly draws on approaches whose objective it is to provide assistance in the regulatory safeguard standard setting process which are targeted for example at the Maximally Exposed Individual (MEI) or the Reasonable Maximal Exposure (RME, Finley and Paustenbach, 1994; Richards and Rowe, 1999; Schwartz, 2000). This is especially the case for the exposure assessment for which it has been tried to modify worst-case assumptions to meet the purpose of this work (cf. section 7.1). However, there is still a chance that at times the mean exposure is significantly higher than the point estimate based on mean parameter values (McKone and Ryan, 1989; Hertwich et al., 2000). Due to this circumstance, even the present assessment may result in overestimated exposures, impacts and external costs.

Consideration of background and speciation

At present, no background concentration of the trace elements analysed is taken into account. As long as the model is linear and no comparison with scientific or regulatory standards is to be made, there is no need for the inclusion of any background concentration information. This is because the background cancels down when performing scenario comparisons (e.g., with and without a given emission). Furthermore, it is the anthropogenically added fraction that is more (bio-) available than the naturally occurring amounts (Berrow and Burridge, 1991; Alloway and Steinnes, 1999; Greger, 1999). Not considering releases from the natural background of heavy metals is additionally supported by de Vries et al. (2004) who argue that "weathering causes only a minor flux of metals, while uncertainties of such calculations are very high" (p. 10).

Note that when keeping the assumption of linearity but still trying to consider the natural background it does not suffice to introduce it as an initial condition for steady-state analyses. This is because it is only the fluxes that matter in this situation. Rather one would have to define a new process which may be termed 'weathering' which releases the natural background into the active pool to be modelled. This is, however, a rather slow process (Colman, 1981; Drever and Clow, 1995; Greger, 1999). One could, therefore, also consider to add a new compartment into which trace elements might be transferred by 'irreversible binding' and released (again) due to weathering. For the modelling of weathering, the reader is referred to the literature (e.g., Hoosbeek and Bryant, 1992; Drever and Clow, 1995; Drever, 1997a; Lasaga, 1995; White and Brantley, 1995; Trudgill, 2000).

It is not only that the background of the same contaminant is not taken into account but also the presence of other pollutants is not considered ('mixtures'). In particular the important effect of competing ions for adsorption sites and ligands in the environment (Bolt and van Riemsdijk, 1987; Hering and Morel, 1990; Schmitt and Sticher, 1991; Borkovec et al., 1998; Jenne, 1998a; Anonymous, 1999b; Vulava et al., 2000; Aboul-Kassim and Simoneit, 2001b; Sauvé 2002) as well as for example in human bodies (Choudhury et al., 2000) is not taken into account except for protons in the different environmental media (pH-dependent partitioning). This may lead to an overestimation of the adsorption of the modelled metals (which may be regarded as non-conservative, United States - Environmental Protection Agency, 1996a) and, thus, to longer residence times. This might have a pronounced effect on the assessment of external costs especially when employing non-zero discounting schemes. The competition for ligands, in turn, might result in a higher portion of the metal that is present as a free ion which is usually more bioavailable and can, thus, act in a more toxic way than complexes with ligands other than water molecules (McBride, 1994; Wolt, 1994; Ritchie and Sposito, 1995; Schnoor, 1996). The distinction between different complexes would involve the modelling of different species which is at present not implemented in the software tool WATSON.

When talking about different ligands the issue of speciation needs to be addressed as well. As stated in section 4.2.3, speciation is not taken into account, but phase partitioning according to the linear K_d value concept (Anonymous, 1999a; Aboul-Kassim and Simoneit, 2001b). Depending on how the employed bioconcentration factors are determined, the disregard of speciation may lead to overestimations of the food concentrations since differing susceptibilities of different substance species towards transfer into living tissues are not taken into account (Berrow and Burridge, 1991; Kabata-Pendias and Pendias, 1992; Ritchie and Sposito, 1995; Markert, 1998; Helmke, 1999). The uncertainty introduced of course also depends on the substance to be assessed. For instance, it was shown that cadmium in freshwater mostly occurs as the free metal ion (Vuceta and Morgan, 1978). This, however, strongly depends on the availability of dissolved organic compounds (e.g., fulvic acids) which is also relevant for soils (Bergkvist et al., 1989; Stoeppler, 1992; Otto et al., 2001).

Furthermore, the disregard of mixtures may lead to an underestimation or overestimation of effects if two or more substances interact in a more than additive (or synergistic) or antagonistic way, respectively (Mücke 6/1995; Kroes, 1996). Thus, the different contaminants are assumed to exert their effects in a non-interactive way for example by simple similar or simple dissimilar action (Kroes, 1996). Effect assessments of mixtures still constitutes a rather open field of research (e.g., Steinberg et al. 6/1995; Escher and Hermens, 2002) which is also why current approaches to assessing the toxic effects of mixtures still treat effects as essentially being additive (Choudhury et al., 2000) in the absence of clear evidence of antagonistic effects.

Spatial differentiation into zones and compartments

Externalities are a concept within welfare economics which is mostly of interest to policy makers. For substances that undergo long-range transport by advection in air or water, externalities may not only occur in the vicinity of the source but also in rather distant places where substances arrive at (European Commission, 1999a; Barbante et al., 2001; Friedrich and Bickel, 2001a; Scheringer and Wania, 2003; Wania, 2003). These substances may, thus, undergo transboundary transport which is why such externalities are mostly of interest to national authorities and/or governments in charge of international affairs. As a result, the assessment of externalities of such substances requires the operation at rather large scales. Furthermore, they should be rather spatially-resolved in order to allow for the discrimination of effects to occur at different administrative units. This means, for instance, that releases of rather involatile substances into a freshwater body should not lead to in-stream concentrations upstream of the release site (e.g., emissions into the Dutch part of the Rhine show up in Swiss Alpine lakes within the catchment when only discriminating according to watersheds, cf. Fig. 4-2). Therefore, a rather high degree of spatial differentiation should not only be assured during the impact assessment but also during the environmental fate assessment. The influence of choosing different spatial resolutions when delimiting according to watersheds will be explored by a scenario analysis in section 9.3.3.

The geographical scope is mostly confined to Europe (cf. Fig. 4-3). The present modelling framework is set up as a system with open boundaries. Exports of substances occur via air and water flows or sediment burial. However, there is no import included since neither intercontinental water and air advection nor (re-)imports of substances (e.g., contained in food) from outside Europe are considered. Thus, substances with the potential to undergo for instance intercontinental transport via air, ocean currents or migrating species cannot properly be addressed leading to an underestimation of effects due to the open system boundaries. Such intercontinental transport is even observed for particle-bound trace elements (Church et al., 1990).

The geographical scope of WATSON has been spatially differentiated into zones according to watersheds (cf. section 4.3). Other delineation criteria exist including a regular grid (e.g., Prevedouros et al., 2004) and combinations of watersheds with other criteria (e.g., Devillers et al., 1995; MacLeod et al., 2001). The influence of the selection of one such delineation scheme on model results is not known.

WATSON allows the distinction of several compartments (sections 5.1 and 6.1). These are assumed to be internally homogeneous and to have temporally constant properties except for the substance amounts contained. The influence of the choice of which compartments to consider is evaluated by means of a scenario analysis below.

As described in section B.4.3, the water volume of a zone consists of streams and lakes fully contained in that zone. If both streams and lakes are present, this means that virtually all water entering the zone is assumed to flow through the lake(s) with longer residence times as well although only parts may actually pass through the lake (e.g., small lake to the East of lake Vänern within the Götälv catchment, Fig. 6-2). This may lead to higher concentrations in the freshwater body of this zone while reducing the input to downstream freshwater bodies. The net effect on human exposure and finally impact depends on the distribution of the freshwater fish production intensities and is, thus, not unambiguous.

Although the presented methodology is site-dependent, some non-substance-dependent properties especially influencing a substance's environmental fate and exposure are treated as if they do not vary in space. Such property values will not be representative for all locations to which they are applied. For instance, the depths of the sediment, glacier and soil compartments are invariant between zones. This may underestimate for example the root uptake by those crops whose roots reach further into the ground than the assumed soil depth, provided the substances reach this depth to considerable amounts. This may not so much affect lead, for instance, as it appears to be concentrated in the top most centimetres (Nriagu, 1978; Rickard and Nriagu, 1978) although contradicting evidence exists (Martínez García et al., 1999). The constant water volume content is deemed not to affect the results as long as the homogeneity assumption for within soils and the linear relationship between the pore water concentration and the plant concentration according to the bioconcentration factor (BCF) are maintained. However, the constant volume fraction of solids in soils has some implications when allowing the organic carbon content to vary. This will lead to varying densities of the overall solid phase in soils (cf. Eq. (B-10)). The way the equilibrium distribution coefficient is defined (cf. section A.2) this means that the smaller the solid phase density the less of the substance is associated with the solid phase. However, it is the organic matter phase which is highly relevant not only for hydrophobic substances but also for many trace elements in terms of the solid-water partitioning (Nriagu, 1978; McCutcheon et al., 1993; Aboul-Kassim and Simoneit, 2001a). A lower solid phase density will lead to a smaller fraction adsorbed and, thus, to an increased mobility and bioavailability of the respective substance in those areas where high organic carbon contents exist. As a result, the retention of the substance in the respective soil compartments is reduced which may potentially mean an earlier exposure when compared to those soil compartment with lower organic carbon contents. For the uncertainties associated with the exposure-related parameters refer to United States - Environmental Protection Agency (1998).

There are parts of the environment which are not included entirely, i.e., they are not part of or constitute own compartments. First, the marine environment is not included which brings about that exposure is underestimated due to lack of inclusion of sea fish and shellfish consumption. The same applies to exposures via drinking water which to rather large degrees originates from ground water bodies. A more detailed discussion why the marine environment and ground water, and the related exposure pathways have been excluded from the assessment is given in section 7.3. Furthermore, the inhalation exposure of farm animals is not considered. This is believed not to cause a substantial underestimation of the exposure results which is in line with Ewers and Wilhelm (1995) and Wilhelm and Ewers (1999) for cadmium and lead, respectively.

Implications of the coupling of an air quality model to a water-soil environmental fate model

The coupling of an air quality model to an environmental fate model comprising the media soil and water limits the amount of substances that can possibly be assessed (cf. section 4.1.1). If substances are rather volatile an assessment is to be favoured in which either all media are fully integrated into one model or in which several subsequent iterative computations of both models are performed. This also holds if wind soil erosion (e.g., Nriagu, 1989; Alloway and Steinnes, 1999) leads to a substantial redistribution of substances which is at present not taken into account.

The use of air dispersion models has advantages over multimedia models of the Mackay-type if spatially differentiated emission information is available (Hertwich et al., 2000). Whether this advantage will lead to a higher or a lower estimate of the external costs largely depends on the distribution of both the substance sources and the population density.

Selection of processes and their formulation

It is not the intention here to review all processes, their formulation and the specific data used. The reasoning for the selection of the respective environmental and substance data is provided in Appendices B and C while the formulation of the processes is described in Appendix A following the mathematical approach as specified in section 2.3.2. Some of the process-related uncertainties will be addressed semi-quantitatively by means of scenario analyses and sensitivity analyses of supposedly important parameters in sections 9.3.3 and 9.3.4, respectively. Some related aspects not taken up again later will, nevertheless, be addressed in the following.

All environmental processes are kept constant in time mostly according to long-term average conditions. As a result, variability within one year and between years are not taken into account, such as changes in redox potentials, acidification, discharge, rain rates and distributions thereof, and temperature. Computations with time steps that are not full years are, therefore, not adequate in terms of resulting meaningful values for the respective period of time (e.g., seasonal, monthly, daily variations). Furthermore, predictions into the future are hampered due to the fact that "natural systems are dynamic and may change in unanticipated ways" (Oreskes et al., 1994, p. 643). Additionally, active management of the environment is not considered either. This may apply to changes in land uses, management of rivers in terms of dams as well as sedimentation and erosion control, and irrigation.

According to the *concept of geomorphic thresholds*, processes such as erosion, sediment and dissolved substance transport in rivers are not associated with average conditions but with single events (Schumm, 1977; Neal et al., 1997). These may occur even just once in a hundred or more years and may, therefore, not be contained even in long-term average data that are based on observations of a few decades time (see sections 2.3 and B.5.2). These processes will lead to a redistribution of substances from soils to surface waters and partly also to soils on floodplains.

The process of wind soil erosion is not included in the assessment for reasons given in section 4.1.1. Depending on for instance the climatic conditions, the common agricultural practice and the most relevant food item in terms of human exposure, this process may lead to higher or lower concentrations in the environmental compartments most important for human exposure. As a result, the exposure may be overestimated or underestimated due to the disregard of this process especially according to a spatially-resolved assessment that takes spatial variability for example in terms of areas potentially prone to this process and distribution of food production into account.

As stated above, irrigation is not considered. This may not be too significant in the large basins of central Europe but, for instance, in Spain (Döll et al., 2003). Similar to flooding, irrigation may influence the concentration of substances (a) in freshwater bodies due to transfer of these onto land and due to reduced water volumes and (b) in soils which receive the irrigation water.

Depending on the most important exposure pathway of the substance to be assessed, thus, the disregard of event-based (extreme) situations and the management of the environment by humans may lead to an overestimation or underestimation of the exposure.

In the case of intermittent rain, Hertwich et al. (2000) conclude that rain is important in multimedia models due to substantially prolonging the residence time in air for chemicals with a low Henry's Law constant as well as for non-volatile metals, residing only in the particle phase. When analysing the potential dose, however, the only remarkably affected chemicals were those for which most exposure occurs through inhalation (Hertwich et al., 2000). This shows that first the selection of the evaluated endpoint (here: residence time vs. potential or time-integrated dose) is relevant when assessing the importance of an assumption. Secondly, the assumption of steady rainfall in the air quality model WTM is likely not to affect the overall exposure estimate in the present assessment substantially.

The approach to model rivers differently from lakes is considered an improvement towards screening risk assessments in which water bodies are usually modelled to entirely behave like lakes, i.e., showing long residence times of water and, hence, substances (cf. section 6.1.5). This will on the one hand accelerate inputs into lakes but on the other hand also accelerate transport beyond the model's boundary downstream from lakes, i.e., into the sea. Assuming that not only the sediment particles are resuspended but also the substances present in the pore water, furthermore, increases the backflow to the water column. Although the modifications do not result in a reduced overall exposure towards freshwater fish when compared to the approach taken by screening risk assessments (cf. section 9.3.3), the way the particle dynamics in freshwater bodies is modelled as introduced here is deemed more appropriate.

As stated in section B.5.2, evaporation and consumptive water abstraction from larger water bodies are not considered due to data processing problems. This results in an overestimation of runoff and discharge. Due to the higher flushing rates, lower concentrations are estimated for some freshwater bodies which will also lower the concentration in fish following equilibrium partitioning according to bioconcentration factors.

Exposure assessment

The exposure assessment is incomplete in terms of the exposure pathways considered. Generally direct exposures in the sense of the definition used by European Centre for Ecotoxicology and Toxicology of Chemicals (1994) and European Commission (2003a) are not taken into account, i.e., exposures for example at the working place or through consumer products. This includes contamination of food and drinking water due to packaging and food preparation.

Beside exposure to drinking water and sea food, especially unintentional soil ingestion by humans is not included in the assessment which may have a substantial impact on the overall exposure situation towards rather persistent substances that are directly released into soils (Huijbregts, 1999; Huijbregts et al., 2000b). This will underestimate the overall exposure. Furthermore, food preparation such as milling, washing, peeling and cooking especially of vegetables may reduce more or less appreciably the concentrations of some substances (World Health Organisation, 1992b; Harrison, 2001a) which has not been taken into account.

The issue of worst-case assumptions has been addressed above and will not be repeated here. The application of the exposure part of a risk assessment framework developed in the US to European conditions is considered defendable. Potentially hidden obstacles may exist, for instance, with regard to different susceptibilities of the cultivated plants and/or kept farm animals in terms of uptake of substances. Also the selection of the exposure assessment scheme followed will influence the results. This will be analysed by means of a scenario analysis (cf. Table 9-4).

Receptor information is distributed according to different schemes if the data are provided at an administrative level that does not correspond to the lowest distinguished level (cf. section B.6). This means that, for instance, German annual production data on freshwater fish are distributed to the municipal level according to the freshwater volume present in the different municipalities. Depending on the resulting distribution of the food production and the assessed emissions, and the exposure pathway contributing most to the overall exposure, this may lead to an overestimation or underestimation of the exposure.

Sticking to the freshwater fish example, the freshwater volume may, furthermore, not be a good indicator for the distribution of the places where freshwater fish are kept and/or caught. It has been necessary for this particular example to additionally assume that freshwater fish are kept in water whose composition is equal to that of the freshwater bodies estimated by the fate model. This disregards the circumstance that people are not just angling in natural waters but that there is also aquaculture going on in separate lakes and/or ponds. Due to the presumably smaller water volume with potentially less inputs than into naturally occurring lake-stream networks, however, this may lead to both overestimations or underestimations of the exposure towards diffuse emissions. In the case of direct releases into freshwater bodies, the exposure assessed according to the presented approach will most likely be overestimated.

Trade is presently considered in a rather simple way. Whereas for the environmental fate assessment the system boundaries are open, these are assumed to be closed for the exposure assessment. This will overestimate the exposure within Europe that is attributable to European human activities due to export of contaminated food and import of mostly uncontaminated food, at least with respect to substances from European sources.[30] Additionally, it is obvious from trade statistics (Food and Agriculture Organization of the United Nations - Statistics Division, 2002a) that the net trade of certain food products is different between countries. This may lead to higher exposures even at the society level if the food items produced in the respective country show a higher load with respect to certain substances while at the same being exported to a lesser extent.

[30] Note that some degree of re-import may occur.

The exposure assessment allows to distinguish between different produce in terms of species. However, there are different susceptibilities not only in terms of species but also for instance in terms of different varieties of the same species and different climatic conditions (e.g., in the case of cadmium, Ursínyová and Hladíová, 2000a) beside other influences. This may over- or underestimate the presently undifferentiated exposure.

As with the environmental properties, the production, consumption and human population data are assumed to be constant with time. Whether this assumption leads to an overestimation or underestimation cannot be clearly stated. The development of the population, for instance, is uncertain. There appears to be a large probability about humankind to exist in the future (Daly and Cobb (1989) quoted in Hellweg et al. (2003)). However, the size of the population living for example in Europe is not known.

No distinction is made with respect to different population sub-groups in terms of exposure such as adults vs. children or smokers vs. non-smokers. The choice not to distinguish between different population sub-groups appears to be justified given the presently available effect information and the circumstance that a very localised exposure assessment such as towards indoor air is not intended by the developed methodology.

It shall, furthermore, be noted that the exposure via ingestion is generally more uncertain than that via inhalation in multi-pathway exposure assessments (e.g., MacLeod et al., 2004).

Impact assessment

As noted by Finley and Paustenbach (1994), the total variability in the exposure variables may be much less than the total variability in the toxicity values emphasizing the importance of reliable impact assessment approaches. Also Rabl and Spadaro (1999) and Droste-Franke et al. (2003) find that effect-related uncertainties are largest.

There is little epidemiological dose- or exposure-response information available for toxic effects following indirect exposures (cf. review by Searl, 2002). As a consequence, the use of the approach as suggested by Crettaz and coworkers (Crettaz, 2000; Crettaz et al., 2002; Pennington et al., 2002; section 7.3.1) is the only approach that is known to the author. Crettaz (2000) discusses the uncertainty sources related to the β_{ED10} slope factor approach which shall not be repeated here. Sources of uncertainty that are addressed include animal to human extrapolation in terms of effects, exposure and/or kinetics, high to low dose extrapolation (without assuming thresholds), subchronic to lifetime exposure extrapolation, inter-individual variability and data quality including experimental design (e.g., number of animals studied). In case that the true dose-response curve has a sigmoidal shape, the linear extrapolation towards lower doses will overestimate the effects (World Health Organisation, 2000b).

There is no scientific basis for assuming a threshold or no-effect level for chemical carcinogens (World Health Organisation, 2000b). This is because most agents that cause cancer also cause irreversible damage to deoxyribonucleic acid (DNA). The linear extrapolation approach can basically be justified in light of the similarities between carcinogenesis and mutagenesis as processes that both have DNA as target molecules with strong evidence for linear dose–response relationships for mutagenesis (World Health Organisation, 2000b). Furthermore, the susceptibility towards effects due to exposure to substances varies between individuals (inter-individual variability) which supports the assumption of having a non-threshold effect situation at the population level (cf. section 7.3).

The situation is different for non-carcinogens for which there exists an unresolved debate on the existence of or ability to measure thresholds and on the extrapolation of the dose-effect curve towards low doses (e.g., Krewitt et al., 2002). Therefore, external cost results due to carcinogenic effects will be stated separately from non-carcinogenic effects. Generally, if the non-threshold assumption was not tenable effects that are assessed at exposure levels below the respective threshold value would lead to an overestimation of the external costs.

The assumptions related to the human health impact indicator DALY are discussed in section 7.3. The use of the highest life expectancy at birth observed on earth is considered not to introduce an unacceptable bias when applied to the European population. The discussion on the disability weights in section 7.3.7 concludes that the disability weights for cancers as given by Crettaz et al. (2002) are considered to underestimate the years lived with a disability to some extent. In fact, the variation in the disability weight may span a few orders of magnitude depending on the method employed to determine them and the people that are involved (Schwarzinger et al., 2003). As also noted by Crettaz (2000), the uncertainties associated with the DALYp approach for non-carcinogenic effects is, furthermore, higher than for carcinogenic effects due to the subjective classification of the effects into default effect categories.

Valuation

During the valuation step different value choices are expected to be made. As noted in section 8.1, discounting is always performed when one has to deal with intertemporal effects that may or may not be valued in monetary terms. Different discounting schemes may have a profound effect on the present valuation of future impacts (Weitzmann, 1999) which may even outweigh all other influencing factors (Hellweg et al., 2003). Also the way how the Monetary value of a life year lost (VLYL) is determined may not allow for its applicability to any person living in the scope of the assessment, opening room to both ends of the scale.

As it is tried to communicate the external costs as explicitly and transparently as possible and due to the provision of sensitivity results of different discount rates, it is believed that the present study gives a good idea of the range of possible outcomes.

The potential effect of considering latency times and using a different approach to value pain and suffering-related external costs has already been discussed in section 8.2.

Miscellaneous assumptions and choices

In the following, some miscellaneous assumptions and choices shall be evaluated affecting the outcome of the assessment:

- at present the air quality model WTM provides the upper boundary condition with respect to indirect inputs into the aquatic and terrestrial environment following emissions to air (cf. section 4.1). Those portions of a substance that remain in air are not considered in the further analysis especially when assessing pulse emissions. This leads to an underestimation of impacts and, thus, external costs. The same applies to those substance amounts that are transported beyond the system boundary of the air quality model.
- information on emissions that directly enter the media soil and/or water is rather scarce and is, therefore, not considered at present. As stated in section 4.4.1, WATSON offers the possibility to distribute aggregated emission information if available. Distribution schemes according to population densities or land uses will only approximately match the true distribution of sources. Depending on the distribution of the food production and the relevant exposure pathways, this may lead to higher or lower exposure estimates.
- potentially occurring effects on organisms do not show a feedback on the environmental fate of the substance causing this effect. For instance, if concentrations became elevated enough for substantial amounts of a forest to die or causing a shift from macrophytes to phytoplankton in rivers the changes in the dynamics of these ecosystems would not appropriately be taken into account.
- due to the spatial resolution of both the air quality model used and WATSON, localised contaminations such as along roads and on floodplains cannot be assessed appropriately (cf. sections 2.1.2 and 2.3). Due to the homogeneity assumption for compartments, elevated concentrations will be diluted unless the compartment area matches the area affected by the elevated

concentrations. Diluted concentrations mean a smaller uptake into the food chain and, hence, a smaller exposure.

9.3.2 Comparison with independent data

When defining the performance criterion for the compliance of the model results with independent data, one needs to define at what stage a model whose purpose is to assess external costs from indirect exposures shall be compared to independent data. The end point of the assessment are the external costs. However, the external costs due to health impacts of a particular substance are not measured themselves; neither are the substance-induced health impacts. If they were the modelling exercise in this work would be obsolete apart from the linkage between human activity and exposure. What remains for the comparison are the exposure levels of the different routes of exposure and the concentrations in the environmental media air, soil and water further up the assessment. Thus, only intermediate results can be compared to observed values in order to operationally validate the model (Rykiel (Jr.), 1996). Such partial analysis of an overall model is also advocated by Caswell (1976).

The model to be analysed here operates at the European scale in a spatially explicit way. The spatial coverage of monitoring activities, however, is usually confined to smaller geographical scopes with an appropriate sampling density. Additionally, the investigated substances are rather persistent and have been released into the environment due to human activities for centuries (e.g., Ryaboshapko et al., 1998; Brännvall et al., 1999), however, with incomplete records. As a consequence, even if the distinction of the metals occurring in the different media into a portion of natural origin and another that was introduced by human activities is made (as tried, e.g., by the FORum of European Geological Surveys (FOREGS) project to be finalized in 2005), the match between model results and observed data will merely be indicative unless some effort is spent to set up a historical emission scenario including for example localised mining activities and past and/or present agricultural practices (e.g., arsenic-containing pesticides, Gebel, 1999, or cadmium in phosphate fertilizers, Ewers and Wilhelm, 1995). This is a task of its own and will not be tried here. Any mismatch between observed values and estimated ones, therefore, does not mean that the model itself is unreliable or inapplicable to the actual context ('operational vs. data validity' as differentiated by Rykiel (Jr.) (1996) or 'check on output space or model as a whole vs. parameter space or parts of the model' as distinguished by Beck and Chen (2000)).

As a result, only the environmental media and food concentrations as estimated by the model can be compared to independent data. When looking for observed environmental media concentrations covering large parts of Europe, it is found that the most comprehensive dataset for trace elements is available for the HELCOM area, i.e., the countries and/or river catchments adjacent to the Baltic Sea (Reimann et al., 2003). Furthermore, there is ongoing effort to extend the Convention on Long-Range Transboundary Air Pollution (CLRTAP) under the umbrella of the United Nations Economic Commission for Europe (UN/ECE) from NO_x and SO_2 emissions to also cover POPs and heavy metals (Reinds et al., 1995; de Vries and Bakker, 1998; de Vries et al., 1998a, 1998b; de Vries, 1999; United Nations - Economic Commission for Europe, 1998; Schütze, 1999; Hettelingh et al., 2002). In this frame the International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation) has issued moss monitoring results (Buse et al., 2003) which may serve to perform a pattern match between the atmospheric deposition fields as calculated by the air quality model WTM and the measured moss concentrations of the respective heavy metals and trace elements.

For food concentrations, there exist some information at the national level (Stoeppler, 1991, 1992; Ewers and Wilhelm, 1995; Wilhelm and Ewers, 1999; Ursínyová and Hladíová, 2000a, 2000b; Harrison, 2001a) and generically (Gebel, 1999; Hertl and Merk, 1999). These will be presented after the comparison of the moss survey results with the atmospheric deposition as assessed by the air quality model employed in this study.

Evaluation of atmospheric deposition fields

In order to evaluate the input data on which the WATSON model operates, the deposition fields as calculated by the air quality model WTM ('Windrose Trajectory Model') would best be compared to monitoring data. However, the data availability with respect to trace element concentrations and depositions is rather poor (cf. Droste-Franke et al., 2003). Recently a monitoring study has been published employing mosses as time-integrating collectors (Buse et al., 2003). The technique of moss analysis provides a surrogate measure of the spatial patterns of heavy metal deposition from the atmosphere to terrestrial systems. A shortcoming, however, is that moss monitoring results do not allow quantitative comparisons with real atmospheric depositions (Schmid-Grob et al., 1993) which is to a very large extent due to the non-standardisible collector 'moss'. Therefore, only a pattern match will be performed.

In the study, only naturally occurring mosses were sampled that were at most three years old and growing at least 300 metres from main roads and populated areas (Buse et al., 2003). Several countries were participating. Thus, the moss survey provides the most comprehensive dataset presently available in order to evaluate atmospheric depositions of trace elements to the terrestrial environment in Europe.

The sampling period in the moss survey was 2000/2001 (Buse et al., 2003). Therefore, the 1998 emission scenario as described and used by Droste-Franke et al. (2003) was used to conduct a pattern match between the moss monitoring results and the total atmospheric deposition fields based on the EMEP 50 x 50 km grid. Due to the uncertain nature of the emission extrapolations from 1990 to 1998 for lead, only arsenic, cadmium and chromium will be compared. Note that the units of the two datasets do not match: WTM results are given in micrograms per square metre and year whereas the moss concentrations are given in microgram per gram. Due to the fact that the integration time of the mosses for the collection of the respective samples is variable and may be as long as three years, no unit conversion is tried here.

The WTM results appear to predict elevated depositions of arsenic in the United Kingdom, Belgium and the so-called 'second black triangle of Europe' on the Czech Republic/Slovakian/Polish border in accordance with the moss survey (top of Fig. 9-1). The higher depositions in the 'black triangle' on the German/Czech Republic/Polish border may be due to a former hot spot of lignite power plants in a lignite mine area in the Czech Republic according to Buse et al. (2003) which is assumed to have been active in 1990, the year based on which the 1998 emission scenario was extrapolated (Droste-Franke et al., 2003). Buse et al. (2003) report that the main sources of heavy metals to the atmosphere in Italy are located in the north which conforms with the hot spot predicted by the WTM. However, this hot spot does not clearly show in the moss monitoring. According to the latter, the biggest hot spot occurs in the southern Romanian and Bulgarian area which only shows rudimentarily in the WTM results.

The atmospheric depositions for cadmium as predicted by the WTM comply with the moss monitoring results to a rather large degree with respect to the hot spots (bottom of Fig. 9-1): Depositions are highest in the so-called 'second black triangle of Europe', east of the Picardie area (northern France), in northern Italy and in southern Romania. The elevated moss concentrations in Portugal and in Bulgaria were partly due to forest fires and geochemical anomalies, respectively (Buse et al., 2003).

Both data sources show matching elevated depositions of chromium in the area of Belgium (metal production) and around Marseille (refineries and metallurgical industry, Fig. 9-2). As with arsenic, there is a hot spot of chromium predicted by the WTM in northern Italy which is to some extent reflected in the moss concentrations. Discrepancies between the two datasets occurring in Galicia (north-western Spain) and Bulgaria can be explained by elevated soil content and serpentine bedrock/old mines, respectively (Buse et al., 2003). The WTM results especially do not capture the area of elevated moss concentrations in Romania.

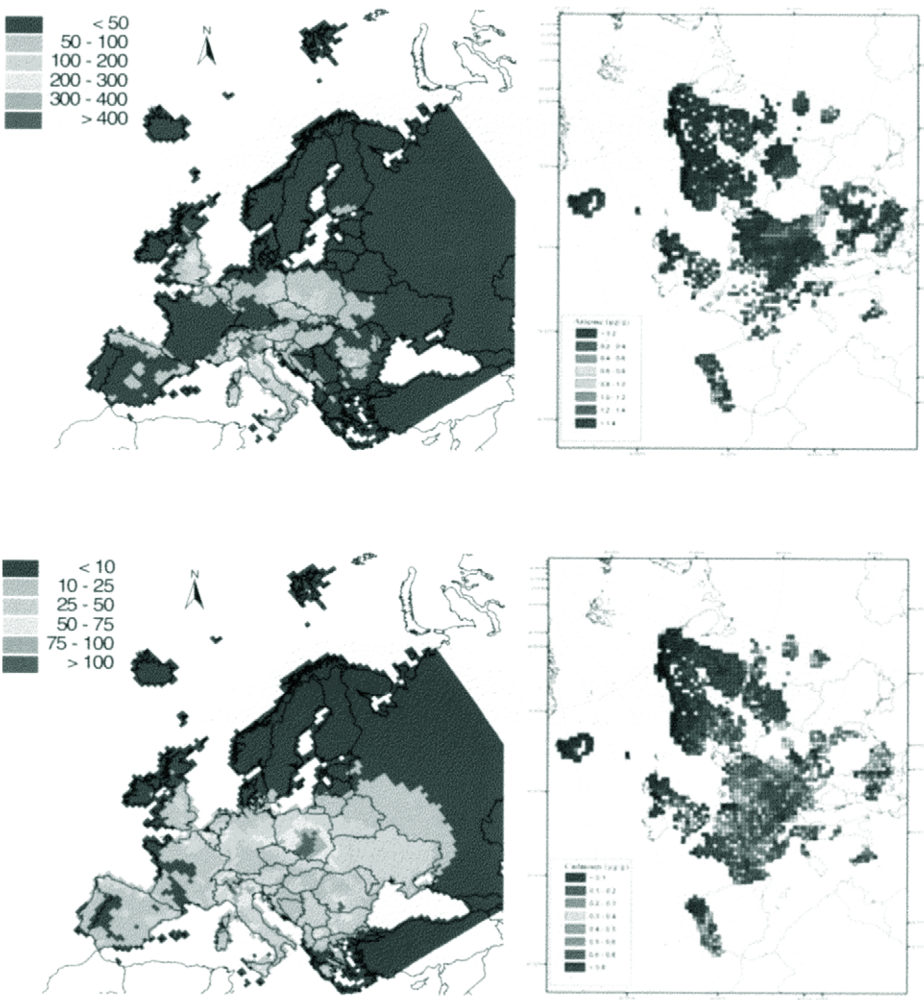

Fig. 9-1: Comparison of atmospheric depositions in micrograms per square metre and year based on air quality modelling by the Windrose Trajectory Model for emissions in year 1998 (left) and moss concentrations in micrograms per gram for the years 2000/2001 (right, taken from Buse et al. (2003) and including data for Iceland, with permission) for arsenic (top) and cadmium (bottom)

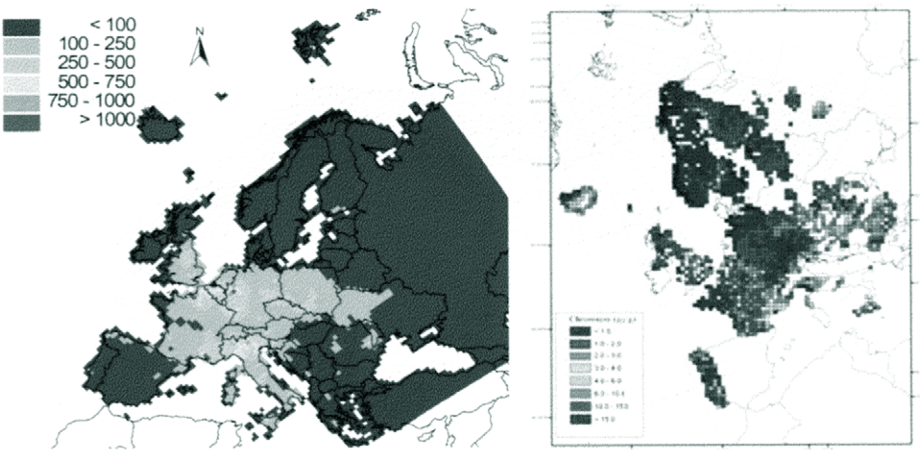

Fig. 9-2: Comparison of atmospheric depositions in micrograms per square metre and year based on air quality modelling by the Windrose Trajectory Model for emissions in year 1998 (left) and moss concentrations in micrograms per gram for the years 2000/2001 (right, taken from Buse et al. (2003) and including data for Iceland, with permission) for chromium

The comparison with the moss monitoring data reveals that the best match occurs for cadmium which is in line with a previous finding for air concentrations of these trace elements (Droste-Franke et al., 2003). This data validation exercise shows that a comparison of the environmental and food concentration data predicted by the WATSON model is only tentatively possible due to the rather poor input data in terms of atmospheric depositions. Whether this data quality issue is due to the emission data and/or the air quality model will not be further explored here.

Comparison with media and food concentration data

The concentrations in the different environmental media including food that are assessed by the presented methodology only take into account that fraction of the trace elements that was released into the environment by the human activities considered in the emission scenarios. Thus, neither the spatially variable natural background levels nor the historic anthropogenic releases and the resulting anthropogenic concentration levels can be considered. Due to lack of data, furthermore, no direct releases into the media soil and water could be included in the emission scenarios. In the absence of spatially-resolved monitoring data, merely a screening comparison of the respective media or food concentrations shall be carried out. These are compared to the concentrations assessed by the pan-European emission scenario for the year 1990 after a continuous release of 100 years (cf. section 11.2). Due to lack of a comprehensive time series on emission data, this scenario is perceived as a proxy for historic emissions since the industrial revolution. In the following, the trace elements shall be compared one by one.

The monitoring data found in the literature are given in Tables C-9, C-10, C-11 and C-12 in Appendix C for arsenic, cadmium, chromium and lead, respectively. These data serve the basis for the indication of ranges and median values to which the predicted concentrations are compared in the following Figures (Fig. 9-3 through 9-6). The predicted concentration values of soils and sediments are converted from kilogram per cubic metre bulk compartment to milligram per kilogram solid phase using spatially resolved solid phase densities and the default volume fractions of the bulk compartments that are solid phase (cf. section 5.1.3). If the lower boundary concentrations of the monitoring data are stated to be below the detection limit, half of the given detection limit is assumed as the observed concentration. In such cases, also the detection limit itself is shown in Fig. 9-3 through 9-6 (denoted as bars). Note that only values of uncontaminated samples are taken into account to the extent that these could be distinguished from contaminated ones. Further note that an attempt is made to consider the natural background. For this, the average concentrations in the upper continental crust are used as provided by Wedepohl (1995), also reproduced in Tables C-9 through C-12. These values are added to the predicted soil and sediment concentrations for the so-called '+ background' cases. In order to also assess the consequently higher concentrations in foodstuff, scaling factors are derived by relating the minimum, median and maximum values predicted in agricultural soils when considering background to those without taking background into account, respectively. These factors are then applied to the minimum, median and maximum values of all land-based produce, respectively, except for spinach. This way, it is assumed that the most contaminated foodstuff is related to the most contaminated agricultural soils and vice versa.

Without considering background, all environmental media concentrations tend to be at the lower end of or below than the (detectable) monitored concentrations (top of Fig. 9-3 through 9-6). For sediments, the discrepancy always amounts to at least two orders of magnitude. When adding the average natural background to these rather low predicted concentrations, it is not surprising that the resulting soil and sediment concentrations fall into the range of concentrations monitored for uncontaminated soils and sediments.

As regards the assessment of food concentrations, a similar picture is observed in that the predicted concentrations tend to be below those monitored without considering the natural average background in soils (bottom of Fig. 9-3 through 9-6). The difference is, however, less pronounced so that, for instance, the median spinach concentrations of lead and chromium are about the same as the expectation values of the used monitoring data. Note that there are no spinach-specific monitoring data available for lead and chromium so that the comparison can only be conducted with 'generic' vegetable monitoring data (cf. Tables C-11 and C-12). Further note that it does not become clear whether the chromium levels in several food groups including vegetables as reported by Hertl and Merk (1999) have been measured in samples of contaminated or pristine origin (cf. Table C-11). This could mean that uncontaminated samples may have smaller concentrations so that the predicted chromium concentrations also of other food groups may be in the same range as the measured concentrations.

When scaling the concentrations of those food items that are exclusively exposed via soil in order to consider the natural average background (see above), the predicted food concentrations are still lower than or compare well with those observed. The only food concentrations that are predicted to be higher than the expectation range of those measured are chromium in cereals and potatoes (cf. bottom of Fig. 9-5). The difference with respect to the median values amount to a factor of 16 and 25, respectively. In general, however, this comparison is flawed particularly by the circumstance that bioconcentration factors are used for the reactive or available portion of the trace elements in soils ("applied" contaminant, cf. United States - Environmental Protection Agency (1998) pp. A-3-18 ff.) to which by far not all of the natural background can be accounted (cf. Berrow and Burridge, 1991; Alloway and Steinnes, 1999; Greger, 1999). The '+ background' estimates, therefore, should have preferably been assessed based on the bioavailable amounts stemming from the natural background. This, however, brings about the necessity to consider the process of weathering whose modelling introduces rather large uncertainties (de Vries et al., 2004) which is why it is not done in the present study. Consequently, the comparison of the monitored levels with the predicted concentrations considering the average natural background is hampered and shall only serve illustrative purposes. Note that fish and spinach are not included in this background consideration exercise due to data availability constraints or the additional contamination pathway via air which does not allow simple scaling, respectively.

When analysing the contribution of different exposure pathways to the contamination of different food groups and in particular cattle products, it is interesting to note that soil ingestion by cattle may be rather dominant. The contribution by this exposure pathway may hold a share of at least 34 % up to 80 % of the chro-

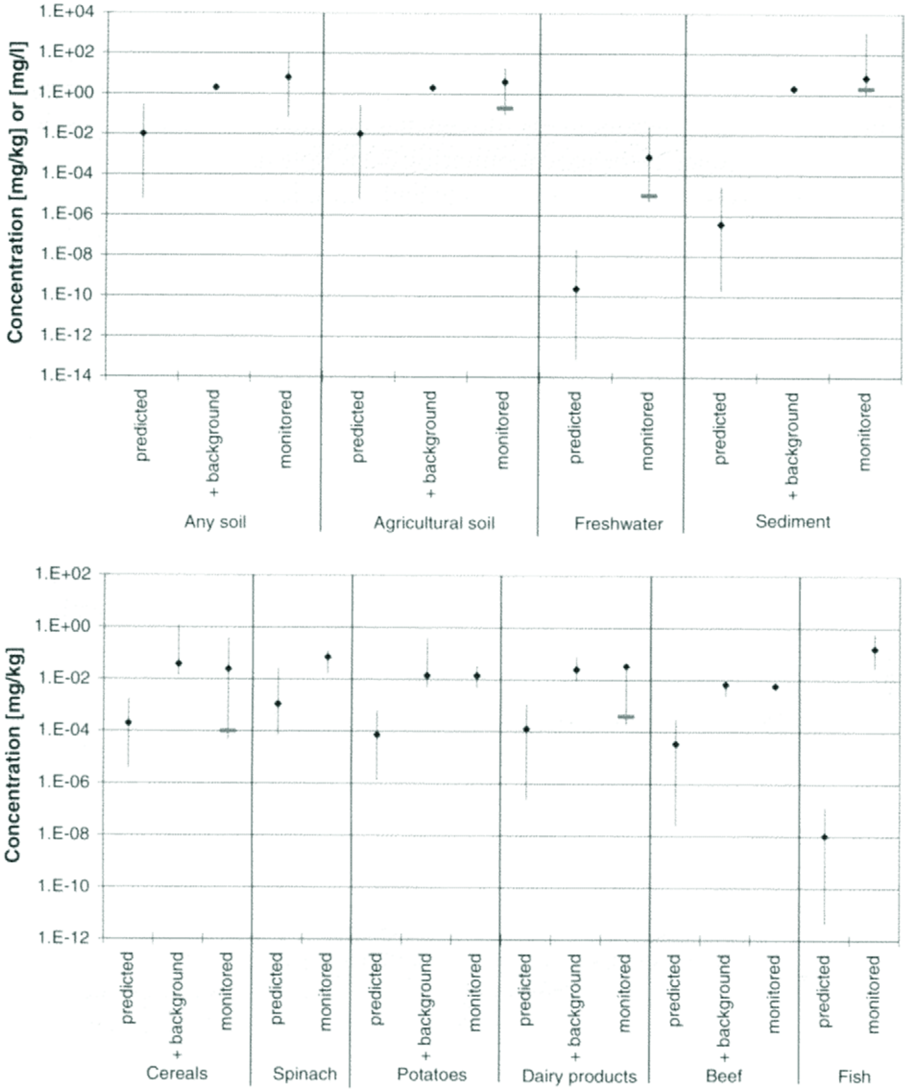

Fig. 9-3: Comparison of arsenic minimum, median and maximum concentrations as predicted by the environmental fate and exposure sub-models with reported concentrations in environmental media (top) and foodstuff (bottom, cf. Table C-9); model estimates based on the 'food removal' scenario described in Table 9-1 resulting from a 100 year continuous release according to the pan-European emission scenario for 1990 (cf. sections 11.1 and 11.2; different units; note the logarithmic scale, horizontal bars indicate reported detection limits)

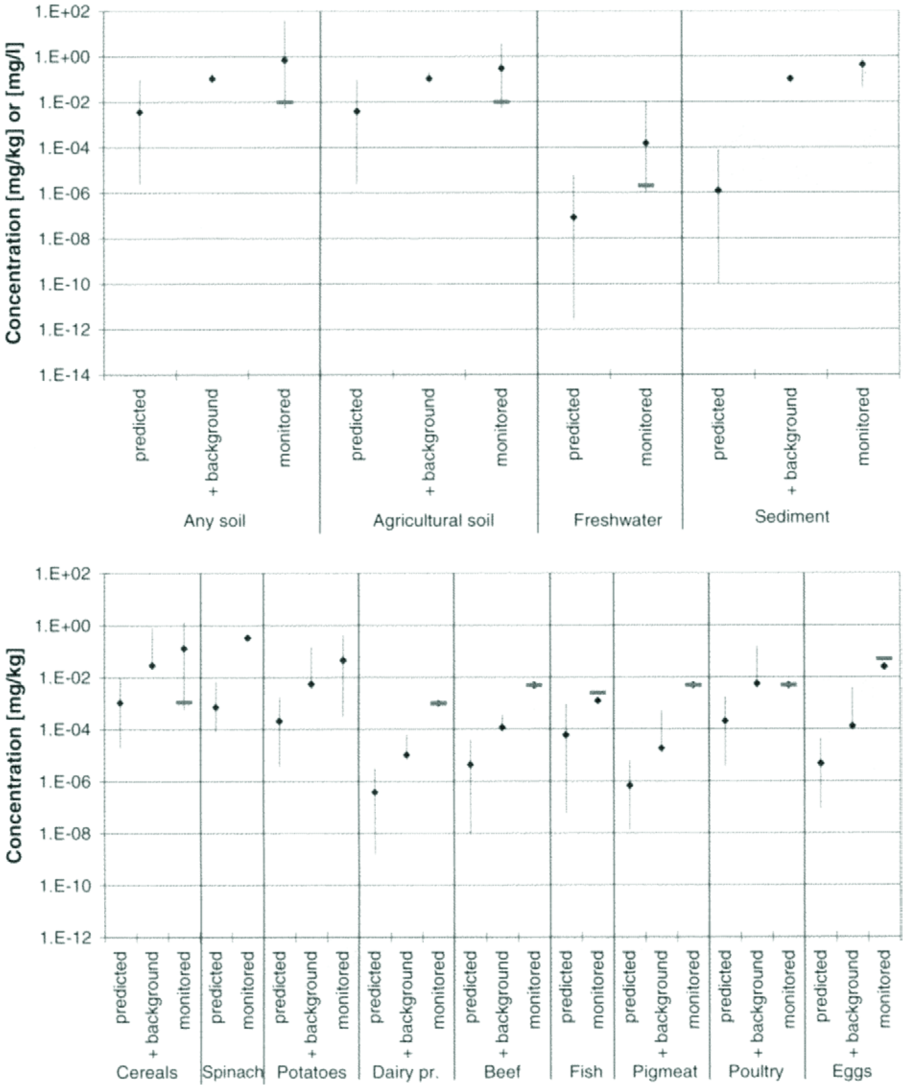

Fig. 9-4: Comparison of cadmium minimum, median and maximum concentrations as predicted by the environmental fate and exposure models with reported concentrations in environmental media (top) and foodstuff (bottom, cf. Table C-10); model estimates based on the 'food removal' scenario described in Table 9-1 resulting from a 100 year continuous release according to the pan-European emission scenario for 1990 (cf. sections 11.1 and 11.2; different units; note the logarithmic scale, horizontal bars indicate reported detection limits)

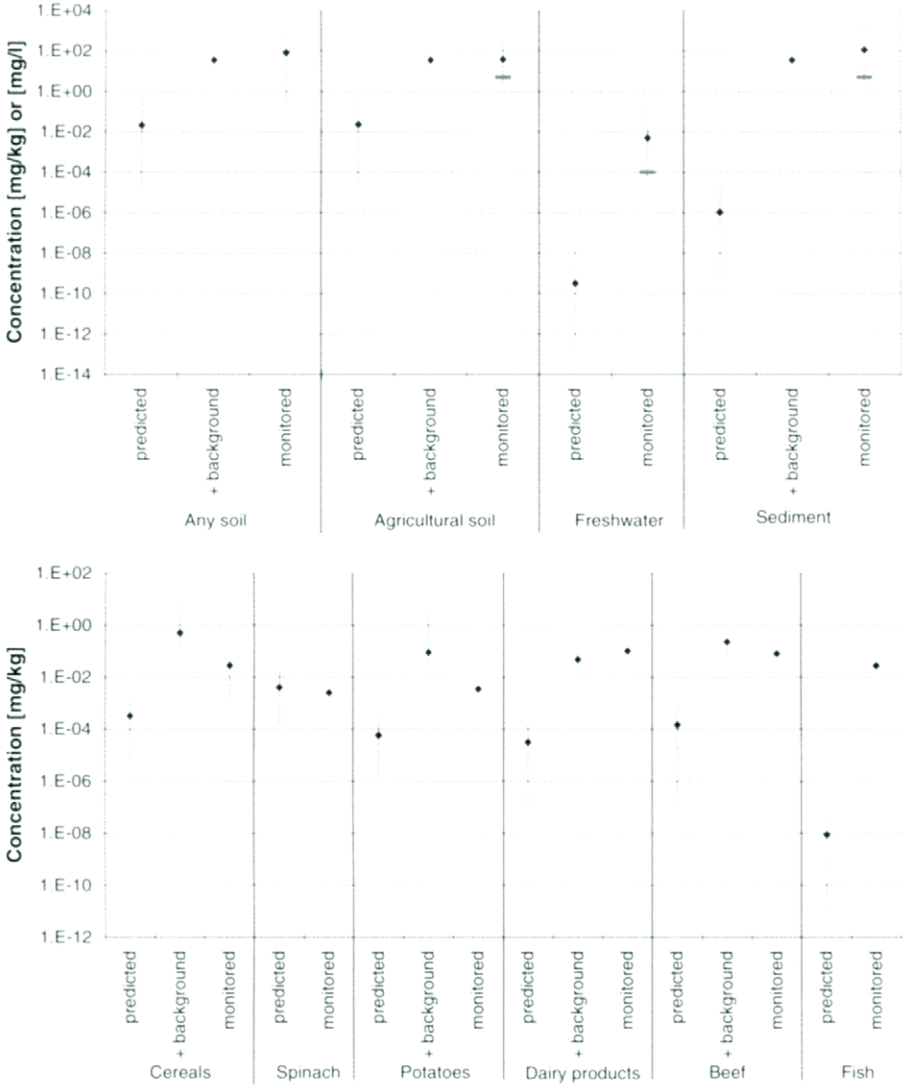

Fig. 9-5: Comparison of chromium minimum, median and maximum concentrations as predicted by the environmental fate and exposure models with reported concentrations in environmental media (top) and foodstuff (bottom, cf. Table C-11); model estimates based on the 'food removal' scenario described in Table 9-1 resulting from a 100 year continuous release according to the pan-European emission scenario for 1990 (cf. sections 11.1 and 11.2; different units; note the logarithmic scale, horizontal bars indicate reported detection limits)

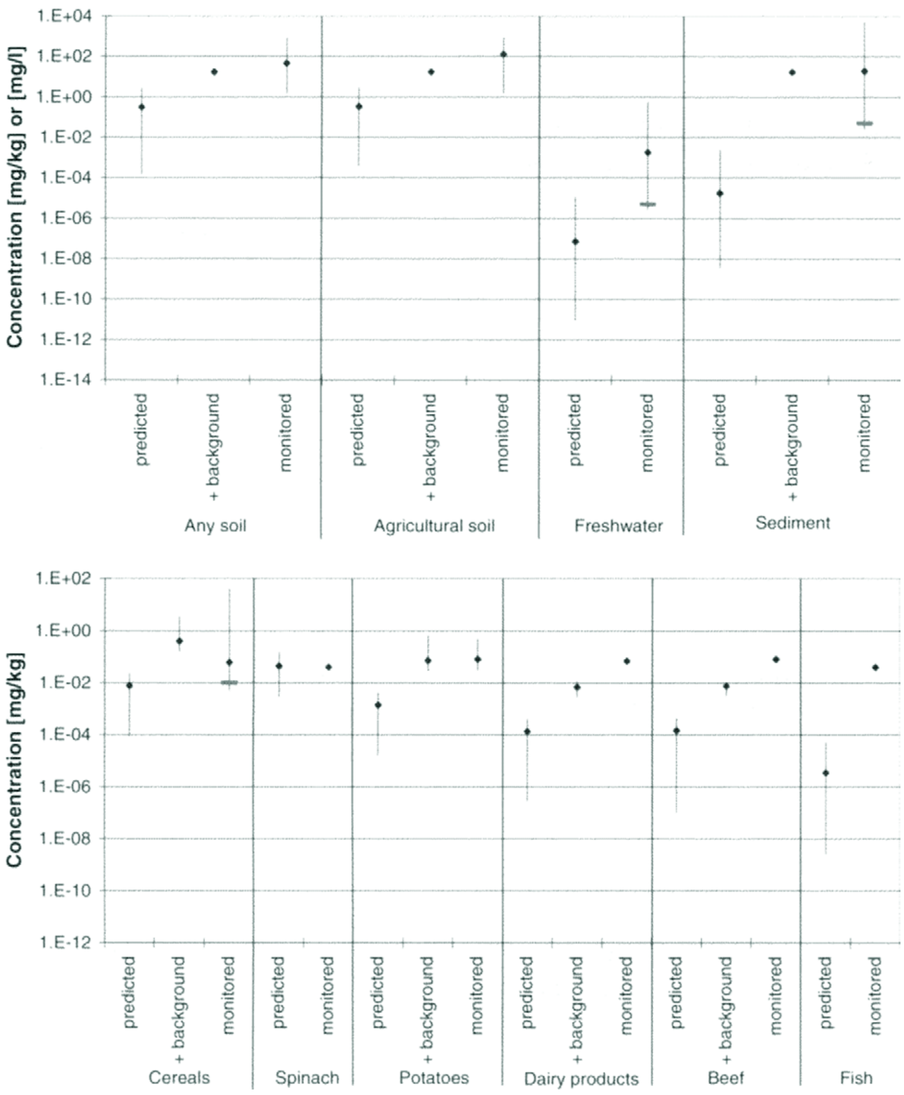

Fig. 9-6: Comparison of lead minimum, median and maximum concentrations as predicted by the environmental fate and exposure models with reported concentrations in environmental media (top) and foodstuff (bottom, cf. Table C-12); model estimates based on the 'food removal' scenario described in Table 9-1 resulting from a 100 year continuous release according to the pan-European emission scenario for 1990 (cf. sections 11.1 and 11.2; different units; note the logarithmic scale, horizontal bars indicate reported detection limits; predicted beef concentrations are compared to a measured value for pork)

mium exposure through cattle-based products, i.e., milk and beef. These shares are typically about 25 % and 50 % for arsenic exposures due to dairy products and beef, respectively. This hints at the importance of including the exposure pathway of soil ingestion by farm animals beside regular feed intake in the assessment as suggested by United States - Environmental Protection Agency (1998). It also emphasizes the need to improve the knowledge on the share of farm animals kept outdoors especially if pigs and poultry were to be included in the assessment of chromium (and arsenic). Such information is not officially available so that the assessor needs to mostly rely on expert judgement (cf. section 7.1.1).

Generally, the concentrations assessed to occur in the freshwater environment, i.e., the water bodies, sediments and fish, are well below the expectation values, regularly by at least two orders of magnitude. The processes ruling the concentrations in freshwater bodies will be subject of a discussion in section 9.3.3. It will be concluded that the processes involved in the particle dynamics in freshwater bodies need further improvement beyond the development done in this work. One may argue that no input from the subsurface by means of ground water discharge is taken into account which may lead to an underestimation of the concentrations in the freshwater environment. However, this flow entrains mostly if not exclusively heavy metals of natural origin at present (e.g., Umweltbundesamt, 2000) unless one has to deal with rather localised contaminations such as mine tailings. Consequently, the disregard of this process is not considered to be the main reason for the assessed concentrations to be that low. With respect to the concentrations in the sediments, one has to note, furthermore, that only the active sediment layer is taken into account in the present assessment (cf. section 6.1.2). This may additionally lead to lower assessed concentrations as compared to measured ones.

It can be concluded from this comparison with reported concentrations that the results of the present assessment are usually within or below the ranges of expected values. However, generally a comparison with spatially-resolved monitored concentrations would be desirable. This also applies to the natural background tentatively considered in the above evaluation.

9.3.3 Scenario analysis

Scenario analysis is considered here to constitute a model evaluation method in order to address decision-rule uncertainties. This means that assumptions are varied rather than parameter values ('non-parameter-related sensitivity analysis'). The latter will be addressed by the screening parameter sensitivity analysis presented in section 9.3.4. The focus of the scenario analysis will be on those model components that are novel. An overview on the different scenarios related to the environmental fate modelling is given in Table 9-1. Note that the sensitivity analysis of the monetary valuation is part of the regular external cost assessment in Chapters 10 and 11. The end point to be investigated is the Intake Fraction and the different contributions to it (e.g., fish, cereals, potatoes, meat, milk). Additionally the influence of the employed exposure assessment on the selected food concentrations shall be evaluated. The scenario analysis focuses on the pan-European emission scenario of cadmium for the year 1990 which is described in more detail in section 11.1. The reason for selecting cadmium is basically that all exposure pathways can be investigated including those related to farm animals other than cattle. This is due to data availability reasons (cf. section C.2).

Although scenario analyses or screening sensitivity analyses primarily provide rather qualitative rankings with respect to a factor's influence (Hamby, 1994; Campolongo et al., 2000a), a varied component is considered influential here if the analysed end-point in a case scenario deviates from the corresponding reference scenario by more than 50 %.

The scenarios given in Table 9-1 shall first be described qualitatively before presenting the results in a joint analysis.

Spatial resolution

While keeping the principal criterion for spatial differentiation, i.e., according to watersheds, the number of zones will be varied (cf. Fig. 4-2 vs. 4-3). In the 'low resolution' scenario, all river basins are treated as a whole irrespective of their size. This means for instance that no tributaries are distinguished in the Danube and the Rhine catchments. In contrast, river basins are further subdivided if respective information has been available for the 'simple high resolution' scenario (and all other scenarios given in Table 9-1). The types of processes and compartments distinguished is equal for both scenarios.

Distinction of further compartments

Another aspect to spatial differentiation apart from spatial resolution into zones is the number of compartments generically distinguished in each of the zones. Principally the number of compartments should be kept as small as possible (Trapp and Matthies, 1998). It shall, therefore, be analysed to what extent the selection of the compartments to be distinguished during the assessment influences the results.

Compared to existing multi-zonal multimedia models which usually only distinguish agricultural from natural soils in the terrestrial environment (Wania et al., 2000; Wania, 2003) if at all (Devillers et al., 1995; Scheringer et al., 2000b; MacLeod et al., 2001; Woodfine et al., 2001, 2002), the database of the

Table 9-1: Scenarios evaluated with respect to the spatial resolution, the compartments distinguished and adapted processes

Scenario	Zones		Compartments distinguished			Process modifications[a]					
	low spatial resolution	high spatial resolution	four[b]	five[c]	eight[d]	lake circulation	different particle dynamics in rivers and lakes	different soil erosion rates	ice melt, overland flow on impervious soils	preferential flow[e]	food removal[f]
Low resolution	x		x								
Simple high resolution		x	x								
Lake circulation		x	x			x					
Rivers from lakes distinguished		x	x			x	x				
Eight compartments		x			x	x	x	x	x		
Five compartments		x		x		x	x	x			
Preferential flow		x		x		x	x	x		x	
Food removal		x		x		x	x	x			x

a. For processes that are included by default refer to main text.
b. Freshwater, sediment, agricultural soil and other land uses.
c. As with the four compartment setting but distinguishing arable land from pastures.
d. As with the five compartment setting but further distinguishing (semi-) natural ecosystems, non-vegetated soils/rocks, impervious land and glaciers.
e. Related processes as described in sections A.3.7 and A.6.4.
f. Related processes as described in section A.3.8.

WATSON tool contains information on land uses such as glaciers/permanent snow, urban or impervious areas, non-vegetated land and pastures as distinguished from arable land. The scenario 'eight compartments' makes use of all of the terrestrial compartment information available (cf. section 5.1). This shall be compared to the 'rivers from lakes distinguished' scenario (see below). Similar to the 'high resolution' scenario, the area shares of the compartments glaciers/permanent snow, impervious soils or sealed areas, and non-vegetated land are added to the semi-natural ecosystem compartment while assuming the properties of the latter for the 'rivers from lakes distinguished' scenario. The same applies to pastures which are merged with the arable land compartment. The processes considered by both scenarios are basically the same. However, impervious soils and glaciers need special processes due to their properties so that neither matrix leaching nor soil erosion occurs with both compartments. Instead, all runoff water is assumed to undergo overland flow in the case of impervious soils (cf. sections A.3.4 and A.3.6) whereas glaciers only show the process 'ice melt' (section A.3.5). Additionally, soil erosion rates are a function of the compartment in the scenario 'eight compartments' leading to varying velocities of this process on different compartments which are affected by this process (cf. section B.5.3). The comparison of the 'eight compartment' scenario to the 'rivers from lakes distinguished' scenario will primarily elucidate the influence of different erosion rates on the overall exposure results.

In order to find out to what extent the distinction of pastures from arable land explains the changes caused by the introduction of all of the compartments according to the 'eight compartment' scenario, another scenario is investigated. While aggregating all other terrestrial compartments into the semi-natural ecosystem compartment (as in the scenario above), arable land and pastures will be treated separately in the 'five compartments' scenario rather than being aggregated into a generic agricultural soil compartment whose properties correspond to those of the arable land compartment.

Inclusion of processes

WATSON offers the opportunity to switch processes on and off (see section 4.4) rather than assigning unrealistic values to parameters (e.g., Guinée et al., 1996; European Commission, 2003b). Many processes considered in the environmental fate module of the WATSON model are common to multimedia models. By default, the following processes are included: soil erosion without distinguishing between compartments, discharge, matrix leaching, overland flow, sedimentation, resuspension and sediment burial without distinguishing between streams and lakes, diffusive exchange between freshwater and sediment.[31] In some scenarios, these may be replaced by newly developed processes (see above and below). However, some processes have been introduced that could not be found in other models. This may be due to the fact that the degree of spatial differentiation followed is rather different from other spatially-resolved or a-spatial multimedia models. For instance, glaciers and, thus, ice melt or the distinction of large lakes, i.e., freshwater bodies that show also 'upstream' flow as part of the lake circulation process have not yet been included elsewhere. The distinction of glaciers only makes sense if also ice melt is considered. Thus, the evaluation of this process will not take place here but is part of the 'eight compartment' scenario described above.

At least three more processes are novel in this context. The first one is circulation in large lakes which has been deemed necessary to be included due to the separation of large lakes into zones (cf. Fig. 6-2 in section 6.1.5). The results of this scenario will be compared to the 'simple high resolution' scenario. Also the influence of the preferential flow process will be investigated by a scenario having the same name. This process describes a non-equilibrium flow (Schwarz and Kaupenjohann, 2000) leading to an accelerated transfer of the substances contained in wet deposition and/or dissolved or suspended in colloidal form in the soil pore water (Jarvis et al., 1999; Noack et al., 2000) to below the surface soil (cf. section A.3.7). The third newly introduced process is a combination of plant exposure via root uptake or atmospheric deposition and plant removal due to harvest (cf. sections A.3.8 and A.6.5). An analogous removal process is also formulated for (freshwater) fish. The 'preferential flow' and the 'food removal' scenarios are compared to the 'five compartments' scenario.

Formulation of processes

Apart from the evaluation of processes by means of their inclusion or exclusion, one can also vary their formulations. The freshwater compartments in multimedia models are usually treated as if they behaved like lakes (e.g., Brandes et al., 1996). This may be justified since the freshwater volume contained in lakes is more than 40 times larger than that of freshwater streams at the global scale (Korzun et al., 1974 cited in Baumgartner and Liebscher, 1990). Considering the difference in dynamics between lakes and streams, it is also obvious that lakes will rather substantially prolong the residence time of substances contained in them. Assuming, however, that all freshwater bodies behave like lakes is rather conservative in a spatially-resolved multimedia model since the removal by freshwater flow is

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[31] Note that partitioning is dependent on zonally-variable pH in the respective compartments except for freshwater and sediment

highly underestimated in areas where hardly any still waters exist. Under still water conditions the processes of sedimentation and resuspension will be slower or faster than under flowing water conditions, respectively. The degree of conservatism introduced will be investigated by comparing the scenario 'rivers from lakes distinguished' towards the 'lake circulation' scenario described above. The sedimentation rate in any freshwater body used in the 'low resolution', 'simple high resolution' and 'lake circulation' scenarios is set to pure lake conditions as described in Table 6-4 according to the alternative process formulations as stated in sections A.3.11, A.3.12 and A.3.13.

Results of the scenario analysis

The aggregated results of the scenario analysis for the emission situation in Europe in 1990 are shown in Fig. 9-7 in terms of the time-integrated (effective) Intake Fraction of cadmium for the ingestion exposures considered. Overall, one can note that the differences between the scenarios are rather small and mostly lie within a factor of two when comparing the aggregated Intake Fractions due to ingestion for the respective time horizons (see discussion on the special case of the 'preferential flow' scenario below). Also when looking at the contributions of the different food items to the overall result, the differences are rather small (Tables 9-2 and 9-3). The Intake Fraction of ingestion is dominated by the uptake of cereals. Generally, the cereal-related exposure amounts to about 81 % of the Intake Fraction due to ingestion assessed to occur for a given time horizon in the case of cadmium. For this heavy metal, also exposure through potatoes in general is substantial, i.e., about 18 % of the Intake Fraction due to ingestion. It shall be noted that also spinach, dairy products and beef are especially important for arsenic and chromium (see Chapters 10 and 11). The contribution of freshwater fish is generally insubstantial when compared to the other food items. Exposure via aboveground exposed produce becomes more relevant for cadmium when including the combined atmospheric deposition and harvest removal process especially in the short term when analysing pulse emissions (see value for spinach for scenario 'food removal' in Table 9-3). This has implications especially on the valuation results when dealing with shorter integration times and also when performing non-zero discounting.

All investigated scenarios show similar dynamics (bottom of Fig. 9-7). However marked differences exist between the scenario with and those without the process of preferential flow. The 'preferential flow' scenario leads to ingestion exposures that are about half of the those for the other scenarios at steady-state. The inclusion of this process can, therefore, be considered influential. In all cases, the development over time sets on with a steep change in the Intake Frac-

Table 9-2: Contribution of the different food items to the Intake Fraction (last row) of cadmium for time-integrated ingestion exposures according to the sensitivity scenarios (pan-European emissions to air in 1990)

Contribution to Intake Fraction	Low resolution	Simple high resolution	Lake circulation	Rivers from lakes distinguished	Eight compart-ments	Five compart-ments	Preferential flow	Food removal
Cereals	80.6%	81.5%	81.5%	81.5%	81.5%	81.5%	81.2%	81.5%
Potatoes	18.8%	18.0%	18.0%	18.0%	17.9%	17.9%	18.2%	17.9%
Spinach	0.087%	0.080%	0.080%	0.080%	0.081%	0.081%	0.099%	0.14%
Dairy products	0.038%	0.036%	0.036%	0.036%	0.039%	0.039%	0.038%	0.039%
Beef	0.030%	0.029%	0.029%	0.029%	0.031%	0.031%	0.031%	0.032%
Fish	0.0017%	0.00065%	0.00065%	0.0056%	0.0058%	0.0055%	0.0083%	0.0056%
Pork, poultry, eggs	0.38%	0.39%	0.39%	0.39%	0.40%	0.40%	0.40%	0.40%
Intake Fraction [$kg_{ingested}$ / $kg_{released}$]	$1.04 \cdot 10^{-3}$	$1.26 \cdot 10^{-3}$	$1.26 \cdot 10^{-3}$	$1.26 \cdot 10^{-3}$	$1.27 \cdot 10^{-3}$	$1.26 \cdot 10^{-3}$	$5.57 \cdot 10^{-4}$	$1.24 \cdot 10^{-3}$

Table 9-3: Contribution of the different food items to the Intake Fraction (last row) of cadmium for ingestion exposures after 25 years according to the sensitivity scenarios (pan-European emissions to air in 1990, only taking place in the first year)

Contribution to Intake Fraction	Low resolution	Simple high resolution	Lake circulation	Rivers from lakes distinguished	Eight compart-ments	Five compart-ments	Preferential flow	Food removal
Cereals	81.7%	81.5%	81.5%	81.5%	81.5%	81.5%	81.5%	80.9%
Potatoes	17.7%	17.9%	17.9%	17.9%	17.8%	17.8%	17.8%	17.7%
Spinach	0.12%	0.12%	0.12%	0.12%	0.12%	0.12%	0.12%	0.95%
Dairy products	0.044%	0.042%	0.042%	0.042%	0.042%	0.042%	0.042%	0.041%
Beef	0.036%	0.036%	0.036%	0.036%	0.035%	0.035%	0.035%	0.035%
Fish	0.0031%	0.0017%	0.0017%	0.012%	0.014%	0.012%	0.012%	0.011%
Pork, poultry, eggs	0.41%	0.41%	0.41%	0.41%	0.40%	0.40%	0.40%	0.40%
Intake Fraction [$kg_{ingested}$ / $kg_{released}$]	$8.80 \cdot 10^{-5}$	$8.97 \cdot 10^{-5}$	$8.97 \cdot 10^{-5}$	$8.98 \cdot 10^{-5}$	$9.02 \cdot 10^{-5}$	$9.02 \cdot 10^{-5}$	$8.98 \cdot 10^{-5}$	$9.09 \cdot 10^{-5}$

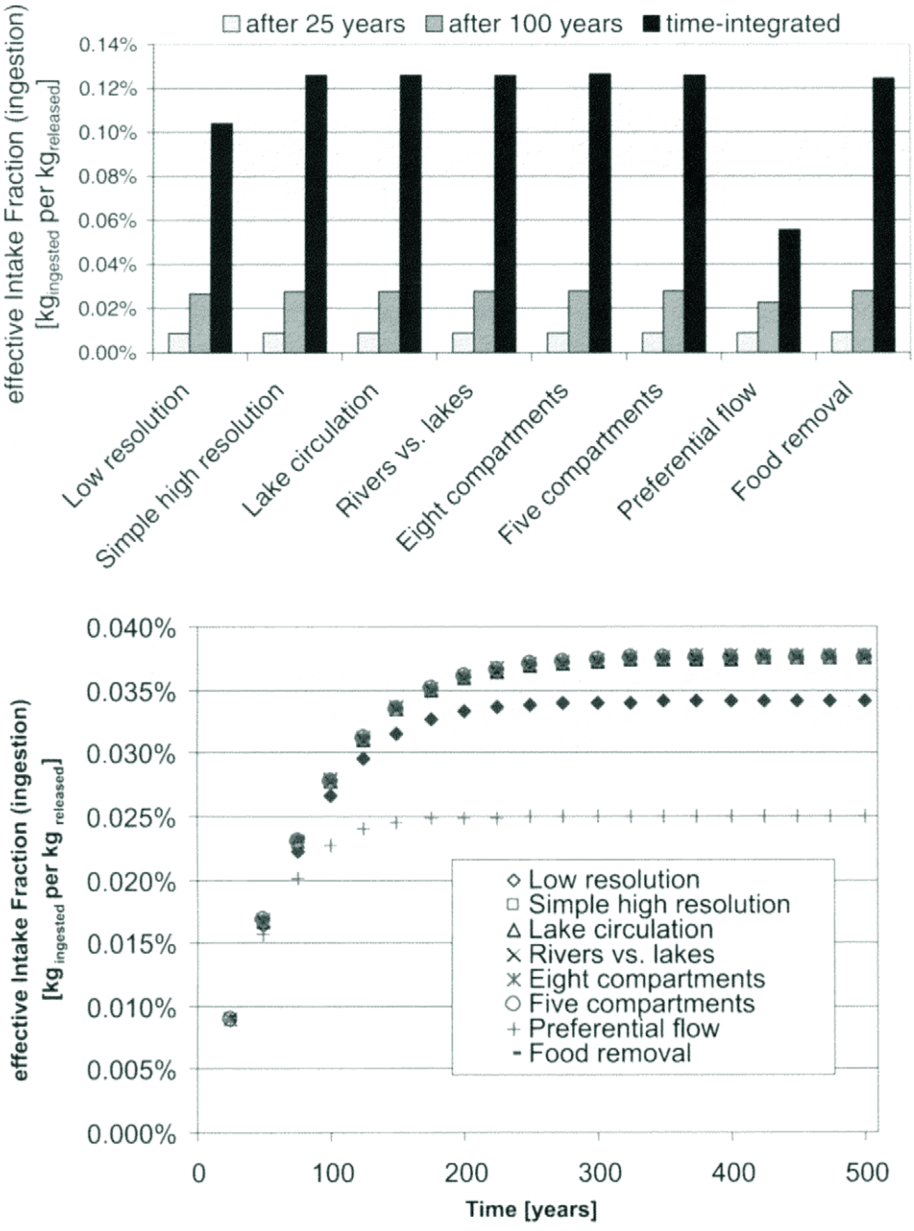

Fig. 9-7: Effective Intake Fraction for cadmium due to the ingestion of food according to the sensitivity scenarios after 25 years, 100 years and at steady state (top) and the development within the first 500 years after the pulse emission (bottom); pan-European emissions to air in 1990

tion followed by a slow approximation towards the aggregated result at steady-state. The duration of the initial fast Intake Fraction accumulation lasts about 100 and 120 years with and without the process of preferential flow, respectively. The 'preferential flow' scenario will, thus, lead to smaller exposures and damages not only in the long-run but also in the short term. The impact of preferential flow on the results will be borne in mind when concluding the case study results (section 12.3.2).

Despite similar dynamics and the small difference in terms of the overall Intake Fraction due to ingestion, there are slight variations between the scenarios not considering preferential flow. The 'low resolution' scenario leads to a smaller overall Intake Fraction for cadmium than the other non-preferential flow scenarios. This is due to lower assessed exposures via many food groups in absolute terms (multiplying the overall Intake Fractions with the corresponding food group shares in Tables 9-2 and 9-3). Most notably, freshwater fish exposures are higher by a factor of about two than in the 'simple high resolution' and 'lake circulation' scenarios which will be discussed below. When comparing the environmental concentrations of cadmium of the exposure-related media agricultural soil and freshwater of the 'low resolution' scenario with those of the 'simple high resolution' scenario, for instance, different extensions of elevated concentrations combined with different peak concentrations become obvious at steady-state (Fig. 9-8 and 9-9). The differences become more apparent when relating the corresponding media concentrations to one another. The factors thus derived are given in Fig. 9-10 for freshwater and agricultural soil to the left and to the right, respectively. The category with factors between 0.98 and 1.02, i.e., practically no change, mostly contains those areas for which no information on subdivisions below the drainage basin level are provided. These are predominantly located along the sea coasts. The areas with lower factors show smaller concentrations for the 'low resolution' scenario.

The difference in terms of exposure is the result of the interplay of the distribution of elevated depositions (Fig. 9-12), the spatial distribution of the food production (cf. Table B-16) and its allocation to agricultural soils (added area shares of arable land and pastures in this case, left and right Fig. 5-1, respectively) or freshwater bodies amongst others. The elevated cadmium concentrations assessed to occur especially in central Poland for the 'simple high resolution' scenario are averaged over the entire catchments of the Odra and Wisla rivers in the 'low resolution' case. The aggregation of catchments, thus, allows higher depositions to occur over less intensively cultivated parts of the country. Similar situations result for the catchments of the Po and Rhine river. Another factor adds to the setting of the Dniepr and Don river in the eastern Ukraine and adjacent Russian areas. The effect of lower runoff on parts of the catchments (cf. Fig. B-3)

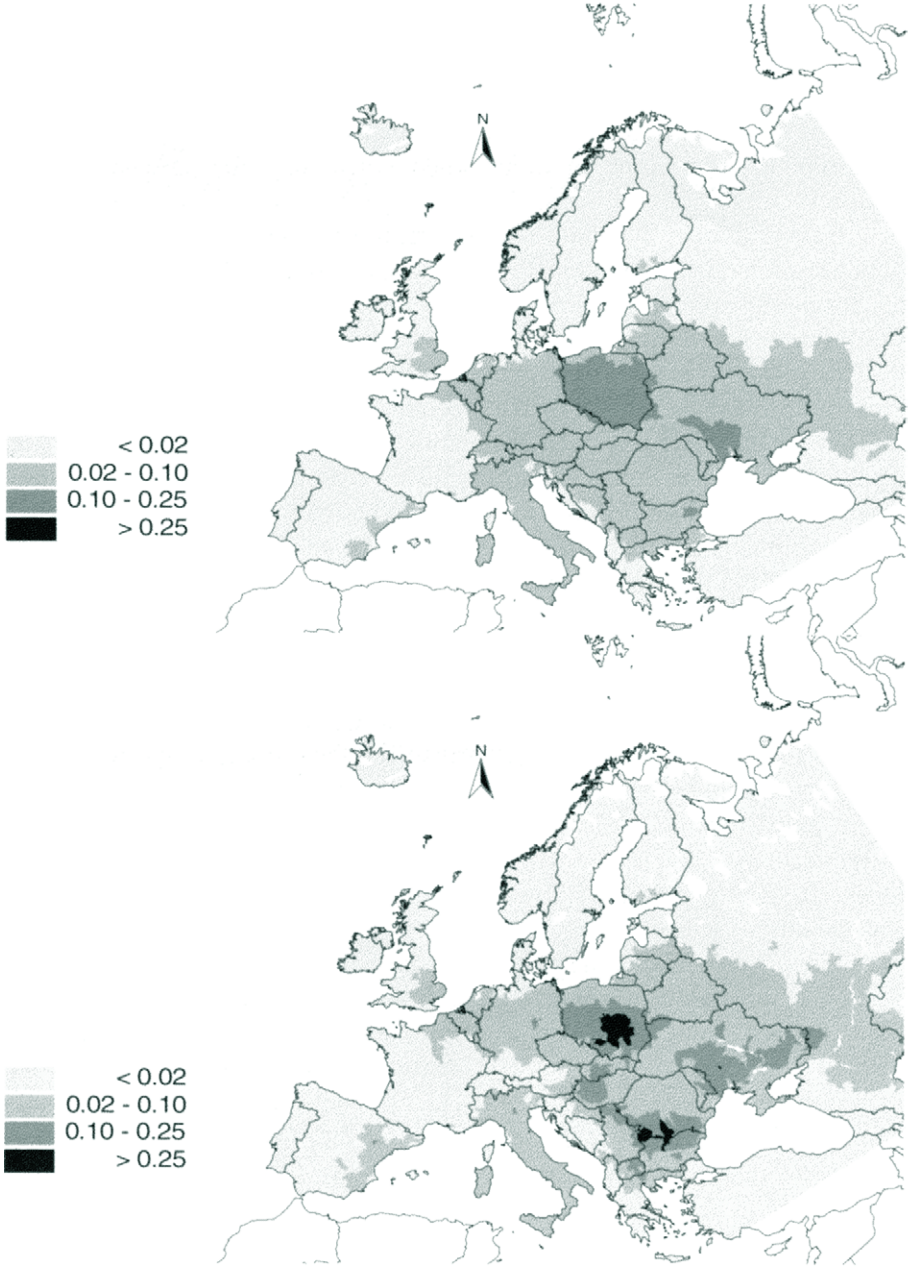

Fig. 9-8: Concentration distribution of cadmium in agricultural soil at steady-state due to 1990 pan-European emissions to air according to the 'low resolution' (top) and 'simple high resolution' (bottom) scenarios [mg/kg]

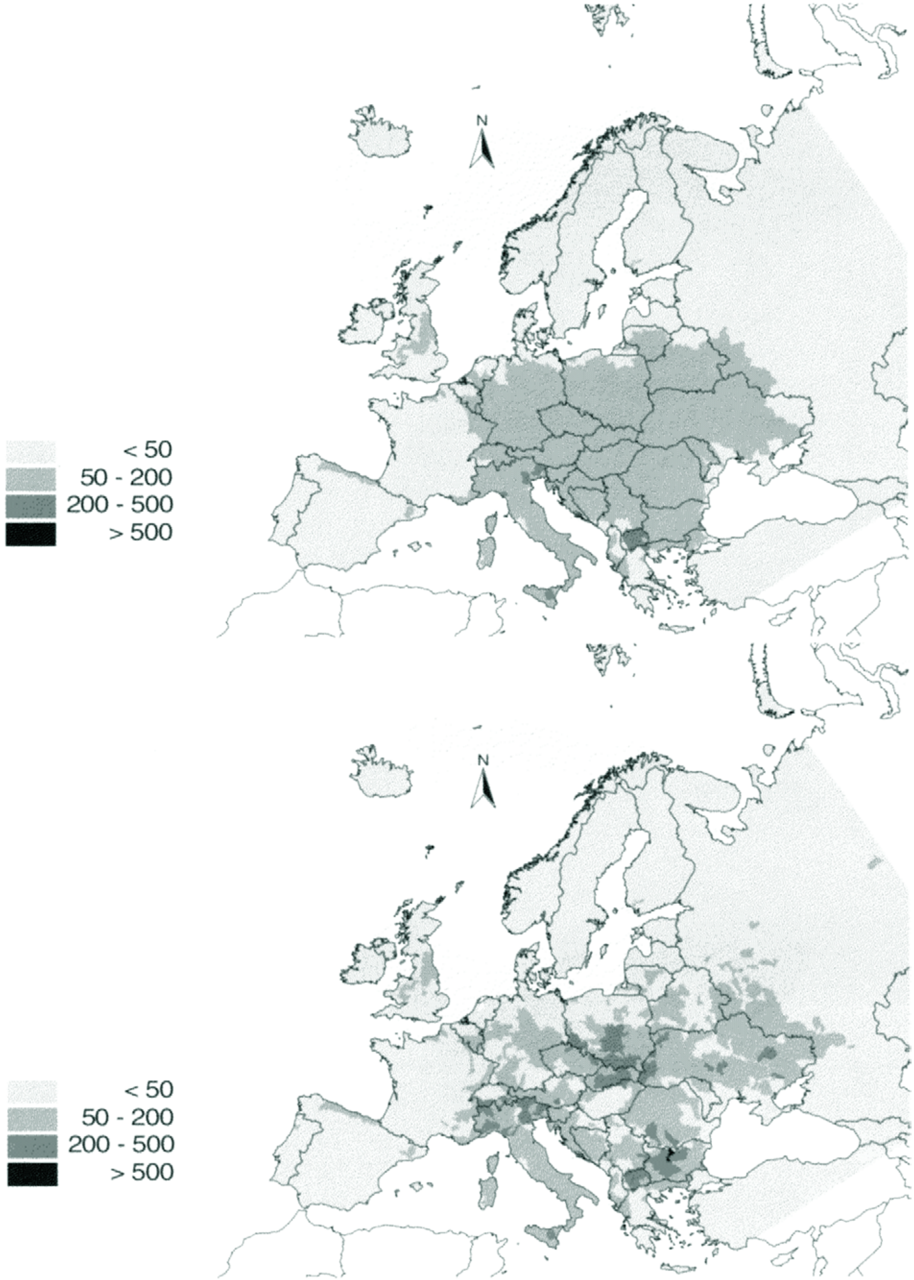

Fig. 9-9: Concentration distribution of cadmium in freshwater at steady-state due to 1990 pan-European emissions to air according to the 'low resolution' (top) and 'simple high resolution' (bottom) scenarios [mg/l]

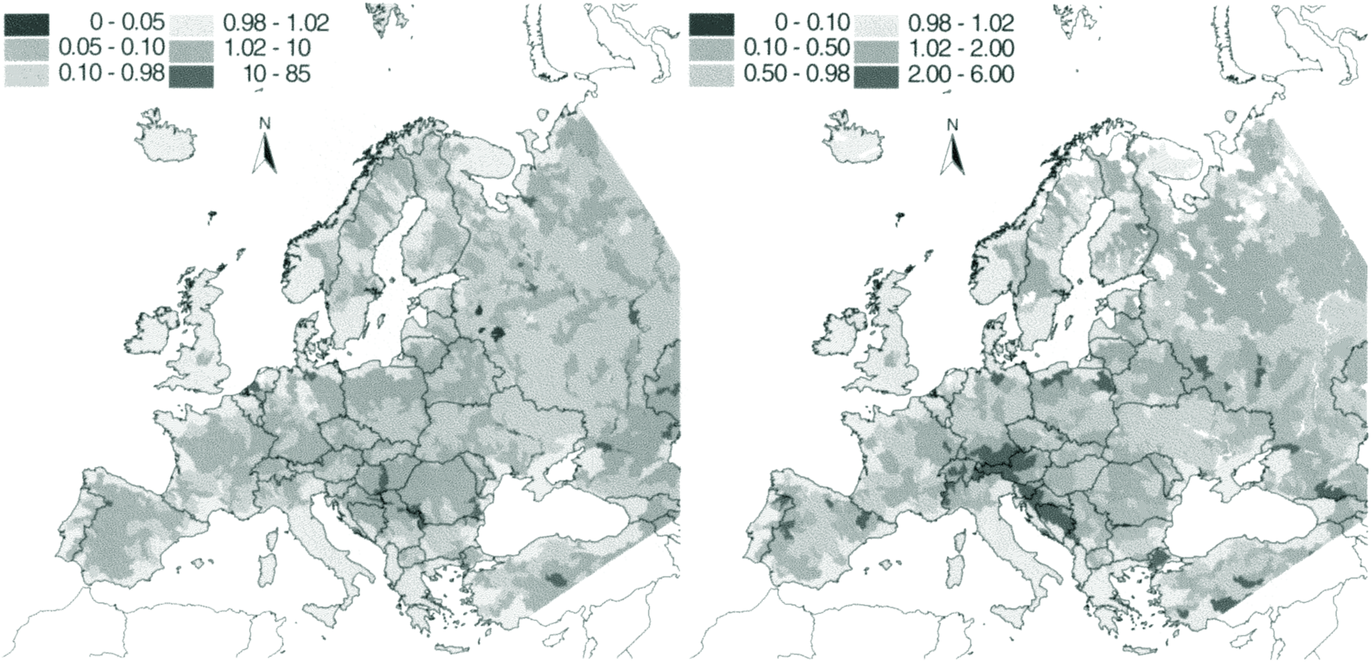

Fig. 9-10: Factors obtained by relating the concentrations of cadmium according to the 'low resolution' scenario to those assessed by the 'simple high resolution' scenario at steady-state in the freshwater (left) and agricultural soil compartments (right) [-] (pan-European emissions to air in 1990)

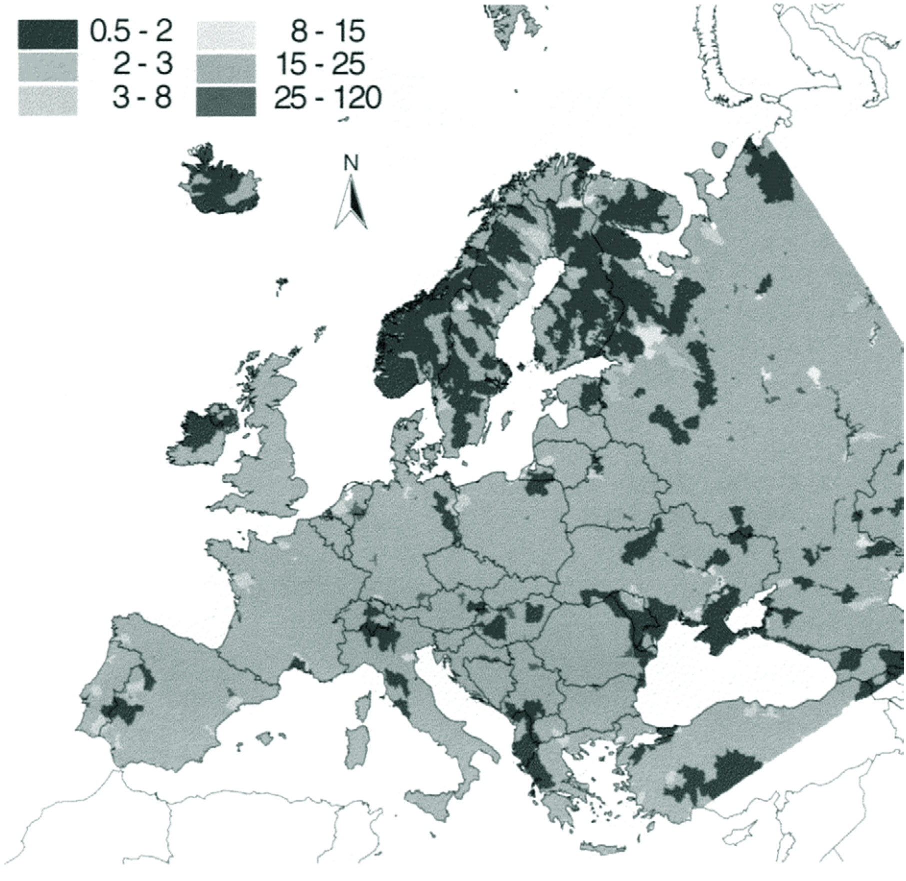

Fig. 9-11: Factors by which the freshwater concentration of cadmium according to the 'rivers from lakes distinguished' scenario deviates from those assessed by the 'simple high resolution' scenario at steady-state [-] (pan-European emissions to air in 1990)

which are not averaged over the entire drainage basin leads to higher concentrations in the 'simple high resolution' scenario in those areas with a high density of agricultural soil which is accompanied with a high production of cereals and to some degree potatoes.

There is a decrease in the contribution of freshwater fish exposure by about a factor of two when changing the environmental fate model from the low to the high spatial resolution setting. This decrease is attributable to higher concentrations predicted in many of the lakes in the 'low resolution' scenario (manifest in

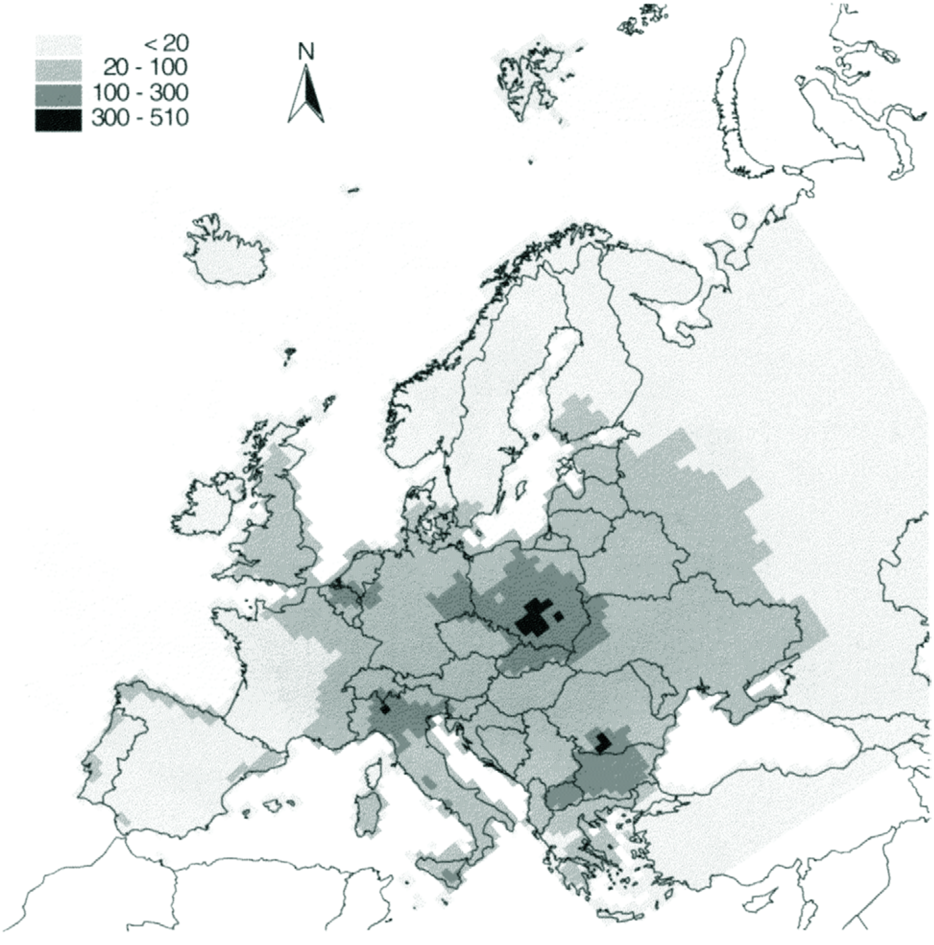

Fig. 9-12: Atmospheric deposition of cadmium according to the pan-European emission scenario for 1990 [$\mu g/m^2/yr$]

the higher exposure) that are due to the circumstance that depositions occurring in one part of the catchment are homogeneously distributed across the drainage basin. For instance, the distribution of the atmospheric deposition (Fig. 9-12) shows that especially the western parts of the Volga and Don rivers receive higher inputs than the rest. However, these depositions also reach to larger freshwater bodies in parts of the catchments that under (physically correct) downstream flow conditions are not affected by these inputs. This applies to the Rybinsk reservoir of the Volga river and the Proletarskoye reservoir of the Don river as well as for other parts of the geographical area covered such as the Rhine river and the Tagus river. In the case of the 'low resolution' scenario, however, these water bodies also receive higher inputs. As described in section B.6.1, the national fish production figures (cf. Table B-16) are distributed according to a distribution scheme that takes water volumes into account. Due to the fact that lakes hold most of the freshwater among the surface freshwater bodies, it is not surprising that higher concentrations in lakes also lead to higher fish exposures. As a result, the deviations in terms of concentrations and exposure in the short term are mostly due to the upstream transport of inputs to catchments caused by the homogeneous mixing within compartments of large sized zones.

The 'lake circulation' scenario does not lead to notable changes towards the 'simple high resolution' scenario indicating that the inclusion of circulation in large lakes is rather irrelevant for larger scale emission scenarios without direct releases into water.

The distinction of rivers from lakes in terms of the processes related to particle transport leads to smaller exposures through freshwater fish in the long run by about one order of magnitude ('rivers from lakes distinguished' vs. 'lake circulation' or 'simple high resolution' scenarios in Table 9-2). Fig. 9-11 shows the influence of the inclusion of this process on the freshwater concentrations of cadmium at steady-state. The colours dark green, light green and brown indicate areas where the deviation amounts to about a factor of two, five and twenty, respectively. While a factor of two is considered tolerable, the other two factors are due to the volume shares of stagnant waters assumed to occur under pure river conditions and in zones of large rivers that pour into the sea with 5 and 21 vol.-% of stagnant waters, respectively (cf. section 6.1.4). Surprisingly, the sedimentation process rather than the discharge seems to dominate the dynamics of freshwater streams with respect to even rather soluble trace elements such as cadmium as compared to lead, chromium and arsenic (cf. Table C-1). Otherwise the reduction in the sedimentation rate particularly in rivers would not have had such a pronounced effect on the concentrations assessed to such a large extent. Thus, a formulation of this process which is even more adequate to running waters than the attempt undertaken here appears necessary. Nevertheless, the process formulation as suggested and used in this study leads to higher contributions of the freshwater fish consumption to the overall ingestion-related Intake Fraction by at least about a factor of seven for the emission scenario analysed.

When comparing the 'rivers from lakes distinguished' to the 'eight compartments' scenario, again a revaluation of some of the less contributing food items takes place. The contribution of those food items related to pasture-based food chains (especially meat and dairy products) increase by about 10 % in the long-run when distinguishing more compartments. This can be attributed to the reduced water soil erosion rate on pastures assumed to prevail (cf. introduction to section 5.1) which leads to a slower removal out of this compartment and, thus, a slower decline in concentration. The increase in exposure via fish occurring in the short term is especially explicable by the fast transport from sealed surfaces to the freshwater environment. This is supported by the lower fish exposure of the 'five compartments' scenario when compared to that with eight compartments. This is the only notable difference when distinguishing impervious land, glaciers and non-vegetated land from natural soils for the emission scenario analysed. Owing to the observation that exposure through dairy products and beef is considerable for the trace elements arsenic and chromium (cf. Fig. 11-6), the distinction between pastures and arable land is made in the following.

When comparing the 'five compartments' scenario to the 'food removal' scenario the main difference lies in the contribution of the aboveground exposed produce, i.e., spinach, to the overall ingestion-related Intake Fraction in the short run. The inclusion of food removal can be considered influential in the case of spinach which increases by more than 50 % in the short term. Due to the fact that harvest is modelled as an ultimate removal process from the environment, the time-integrated ingestion-related Intake Fraction even slightly decreases by about 1 %.

Having analysed cadmium in more detail, the question arises to what extent the exposure towards the other three trace elements is affected by the variation of the respective components. As particularly arsenic and chromium are less mobile than cadmium, it is expected that the consideration of processes involving the particle-bound fraction of the trace elements in soils will have a more pronounced on the overall exposure and the most contributing food items. This will most notably be the case for the distinction of different erosion rates on different land uses and the preferential flow process. Allowing lower erosion rates to occur on pastures than on arable land will increase the contribution of cattle produce to the aggregated effective Intake Fraction for arsenic and chromium. The steady removal by means of preferential flow will lead to a more substantial reduction of the effective Intake Fraction for arsenic and chromium than for cadmium, given the long time scales involved when integrating the exposure over time (e.g., Fig. 11-5). For lead, the difference to the scenario analysis results presented for cadmium are not expected to be remarkable.

Variable results due to the employed exposure assessment scheme

Another question is related to the influence of the employed exposure assessment scheme. Two exposure assessment schemes especially with respect to heavy metals/trace elements have been encountered: one by the International Atomic Ener-

Table 9-4: Maximum concentrations in agricultural produce at steady-state for air emissions in 1990 according to the exposure assessments as given by International Atomic Energy Agency (2001) and United States - Environmental Protection Agency (1998) for Europe

Food concentration [mg/kg FW]	IAEA	US-EPA
Cadmium		
cereals	0.19	0.03
beef	0.025	0.0002
Lead		
cereals	1.30	0.65
milk	0.11	0.04
beef	0.20	0.04

gy Agency (2001) and another by the United States - Environmental Protection Agency (1998). These vary in terms of process formulation (cf. section A.7), parameter values and exposure pathways considered. Additionally, the IAEA exposure assessment framework is meant to be used at a screening level whereas the US-EPA's is more advanced/detailed, i.e., less conservative (cf. Chapter 7).

An analysis of these two exposure assessment schemes has been undertaken taking into account the food items cereals, beef and dairy products for cadmium and lead (Table 9-4). As this has been done early in the course of the development of the present methodology some components of the assessment could not be taken into account. The following parameter must, therefore, be regarded to be set to unity: $fr\_w_{effective/total}$, $fr\_w_{not\ consumed/food\ supply}$ and $fr\_w_{self\text{-}supply}$. For their meaning, refer to section A.7.

It can be seen that the IAEA exposure assessment overestimates the exposure to the respective food items by about an order of magnitude as compared to the one by the US-EPA. This is, in fact, not surprising due to its intended use for screening purposes. From this rough comparison, it appears justified to follow the exposure assessment as suggested by the United States - Environmental Protection Agency (1998) unless more realistic exposure assessments become available.

9.3.4 Sensitivity analysis of the parameters

As stated above, the sensitivity analysis of the parameters to be presented in the following constitutes a screening parameter sensitivity analysis. This is used in order to identify the most important parameters in the sense of Hamby (1994).

It remains to be defined when a parameter is judged to be important. This can be done in different ways. When constantly increasing one parameter after another by 20 %, Price et al. (1996) defined a change in the result of 1 % as an indication that a parameter "significantly contributed to the uncertainty and variation of the dose rate estimates" (p. 265). The relationship between model output and parameter variation is adopted as the evaluation criterion according to:

$$\text{abs}\left(\frac{\frac{(IF_{\text{sens}} - IF_{\text{ref}})}{IF_{\text{ref}}}}{\frac{(x_{\text{sens}} - x_{\text{ref}})}{x_{\text{ref}}}}\right) \geq \frac{0.01}{0.2} = 0.05 \tag{9-1}$$

where

IF : Intake Fraction due to ingestion of single food items and/or the aggregated ingestion exposures according to the reference sensitivity case ('ref') or a parameter variation case ('sens') [kg_{ingested} per kg_{released}]

x : parameter value as used in the reference sensitivity case ('ref') or during a parameter variation case ('sens') [variable units].

There are many parameters included in the assessment whose comprehensive evaluation is beyond the scope of the present study. A prioritisation of the parameters for which a sensitivity analysis shall be carried out is, therefore, performed according to findings in the literature.

Generally, substance-specific properties appear to be most relevant (Blanchard and Lerch, 2000; Huijbregts et al., 2000a) especially when releases into air are investigated (Hertwich et al., 1999). These may be related to the environmental fate, exposure, or effect/impact assessment. For degrading substances, the persistence in the release compartment appears to be most important (Hertwich et al., 1999; Huijbregts et al., 2000a). However, chemical transformation is not considered at present for the trace elements investigated. The solid-water partitioning coefficient (K_d) is, thus, deemed to be the most important parameter with regard to the environmental fate of non-volatile substances (Blanchard and Lerch, 2000; Huijbregts et al., 2000a).

Table 9-5: Assigned pH values in the 'compartmental pH variation' sensitivity case

Compartment in this study	pH	Compartment according to Huijbregts (1999)
Freshwater	7	Continental freshwater bodies
Sediment	7	Continental freshwater sediments
Glaciers[a]	7	Continental freshwater bodies (analogy assumption)
Impervious areas[a]	6	Continental industrial soils
Non-vegetated land[a]	6	Continental natural soil
(Semi-) natural ecosystems	6	Continental natural soil
Arable land	7	Continental agricultural soil
Pastures/grassland	7	Continental agricultural soil

a.Compartment not analysed when only distinguishing five compartments.

The influence of the solid-water partitioning coefficient on the overall results shall be analysed in two ways: first, by varying the parameter values themselves and secondly, by varying the pH values of the compartments upon which the solid-water partitioning coefficient is allowed to depend. In the proposed external cost assessment framework, several parameters vary in space (cf. Appendix B) to which the pH of the different compartments belongs. It is a key variable influencing the partitioning of non-volatile trace elements (Wolt, 1994; United States - Environmental Protection Agency, 1998; Anonymous, 1999b) and their mobility in watersheds (e.g., Scudlark et al., 2005). Although the substance-specific partitioning coefficient may also vary by orders of magnitude for a certain pH range (e.g., Anonymous, 1999b) which will be investigated by the first sensitivity analysis related to the K_d value, the influence of assuming variable pH values especially for soils will be analysed. In most scenarios, the pH value of the compartments are allowed to vary by zone[32] and compartments whereas it only varies by compartments for the sensitivity case 'compartmental pH variation' mostly according to the values specified in Huijbregts (1999) that are given in Table 9-5. However, the latter sensitivity case merely serves illustrative purposes since no change in the parameter values can be computed according to the denominator on the left hand side of Eq. (9-1). Thus, this sensitivity case will be analysed in the same way as the sensitivity scenarios of section 9.3.3.

[32]Except for freshwater bodies and corresponding sediments.

Exposure-related parameters are especially influential for releases into water and for the indirect exposure routes according to Hertwich et al. (1999). According to the emission scenarios analysed in Chapters 10 and 11, considerable contributions to the time-integrated exposure stem from cereals, potatoes, dairy products and beef for the contaminants investigated. Therefore, the sensitivity of the results towards the respective bioconcentration factors and biotransfer factors shall be analysed.

Depending on the particular situation, the uncertainty related to effect information may dominate the overall uncertainties (Finley and Paustenbach, 1994; Rabl and Spadaro, 1999; Huijbregts et al., 2000a; Droste-Franke et al., 2003). The impact assessment of both ingestion and inhalation exposures is performed in a site-generic way, i.e., using the same dose-response relationships and severity measures throughout the model's geographical scope and for all individuals exposed. Its influence is qualitatively discussed in section 9.3.1 above and will be taken into account during the presentation of the case study results (Chapters 10 and 11) and the conclusions drawn.

The scenario analysis presented above (section 9.3.3) has shown that some variations in the formulation of the environmental fate model changes the relative importance of the food items contributing to the overall exposure. In particular the influence of the water soil erosion process shall be analysed which constitutes one criterion according to which the terrestrial compartments have been distinguished from each other (cf. section 5.1).

The sensitivity analysis of the parameters focuses on the pan-European emission scenario of chromium for the year 1990 which is described in more detail in section 11.1. The reason for selecting chromium is that different food items substantially contribute to its overall exposure. Furthermore, it shows the most pronounced variability in terms of its solid-water partitioning coefficient in the pH range prevailing at least in agricultural soils (i.e., a pH between 5 and 7, Scheffer and Schachtschabel, 1989) among the trace elements analysed (Table C-1).

Results of the sensitivity analysis of the parameters

The sensitivity analysis of the parameters is performed based on the environmental settings of the 'food removal' scenario (see section 9.3.3) which will be used for the assessment of external costs in Chapters 10 and 11. The development of the effective Intake Fraction for ingestion over time is displayed in Fig. 9-13 for all sensitivity cases analysed. The differences appear to occur only in the intermediate to long run except for the 'BCF above -90%' scenario due to the reduced contamination of cereals (cf. Table 9-6) dominating in the short run (cf. Fig. 9-14).

The assessed values of the ingestion-related effective Intake Fractions after 25 years and time-integrated are given by food item in Table 9-6. In order to identify significant changes, the numbers resulting according to the term in brackets of Eq. (9-1) are presented in Table 9-7. Note that exposure through pork, poultry and eggs cannot be assessed for chromium due to data availability constraints (cf. Table C-5).

The indicator values for the aggregated effective Intake Fractions for ingestion ('sum' in Table 9-7) show that all parameters significantly influence the exposure situation of the human population at least in the long run. It needs to be noted, however, that if the K_d value of chromium is increased by one order of magnitude this significance does not show in the aggregated effective Intake Fraction for ingestion but only in that for cattle exposure and fish (parameter sensitivity 'Kd +900%'). This can be attributed to the very high K_d values of chromium at higher pH values (cf. Table C-1). In contrast, the lower K_d values will only prevail in non-agricultural areas due to the minimal pH values allowed to occur owing to management practices in pasture soils which is 5.5 whereas for arable land it is 6.0 (cf. section B.5.1). It appears as if the behaviour of chromium is only highly affected in those compartments that have pH values about or below 6. This is the pH range where the most substantial changes in the pH-dependent partitioning behaviour of chromium occurs. At or below a pH value of 6, a higher share of chromium is, therefore, present in the soil pore water. In this dissolved state, chromium may increasingly undergo the processes of matrix leaching and/ or overland flow leading to a quicker decline of its concentration in soils with lower pH values and, thus, to a lower exposure through food produced on these soils. As a result, also the exposure via fish increases. When decreasing the K_d values of chromium by one order of magnitude, in turn, the effect becomes significant at the aggregated exposure level. This also indicates that it does not seem to make a remarkable difference whether a substance's K_d value is in the order of 10^3 or 10^4 m^3 per kg.

In the short term, i.e., after 25 years, only the variation in terms of the bioconcentration factors of aboveground ('BCF above -90%') and belowground produce ('BCF below -90%'), and the biotransfer factor for milk cattle ('BTF dairy -90%') leads to significant changes in the aggregated effective Intake Fraction for ingestion. This is due to the marginal contribution of beef to the overall ingestion exposure in the short term (top vs. bottom of Fig. 9-14). The overall higher contribution of cattle produce in the long term is attributable to the reduced water soil erosion rate on pastures compared to arable land (cf. Table 5-7) leading to a slower removal of the trace elements out of this compartment and, thus, to a higher exposure of terrestrial food items in the long run. In general, the influence of parameters mostly or exclusively involved in the environmental fate assessment,

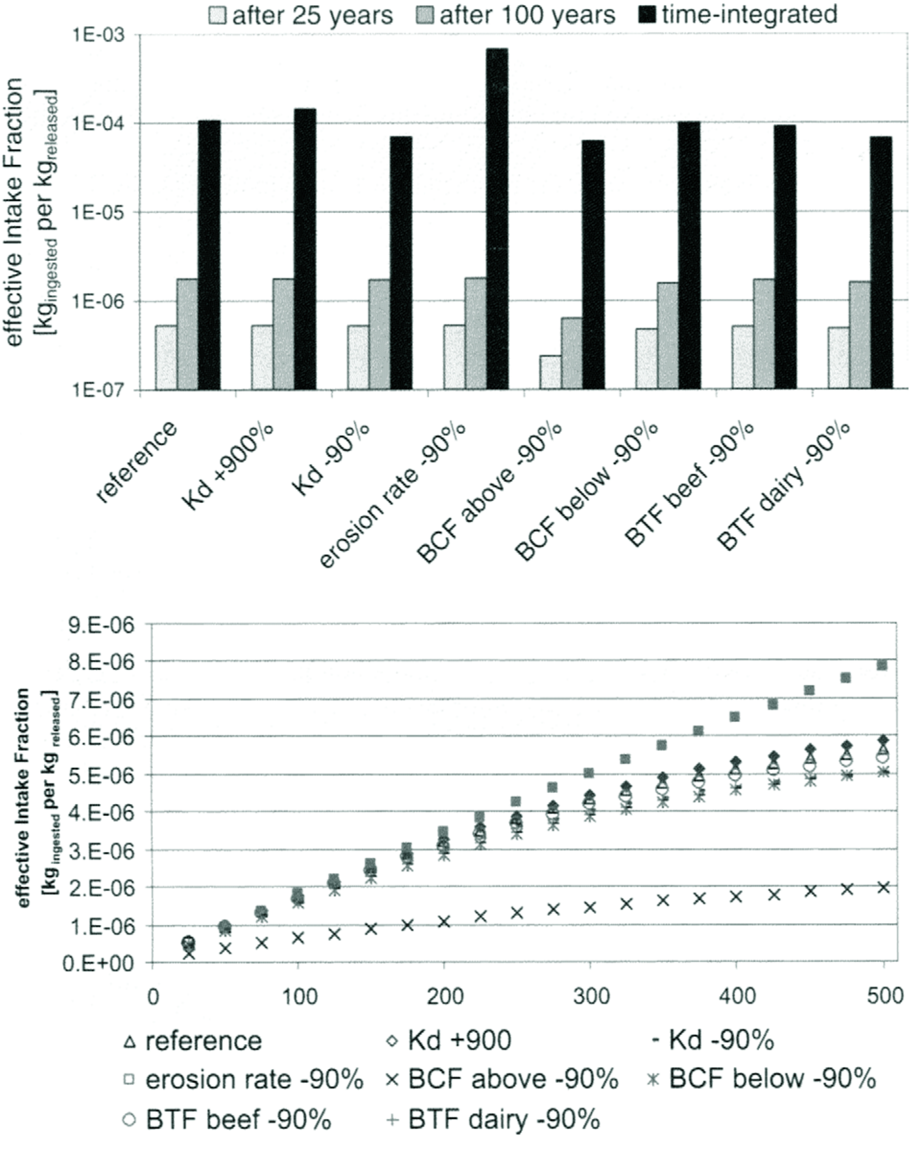

Fig. 9-13: Effective Intake Fraction for chromium due to ingestion of food according to the sensitivity analysis after 25 years, 100 years and at steady state (top) and the development within the first 500 years after the pulse emission (bottom); pan-European emissions to air in 1990

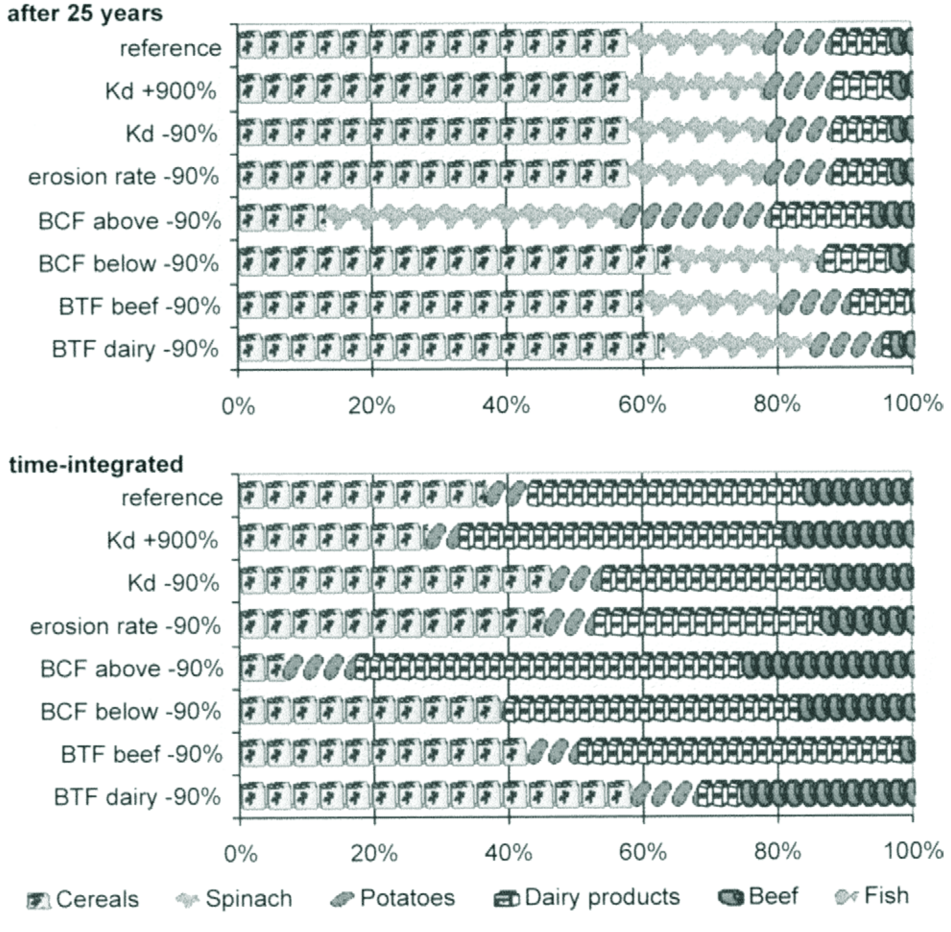

Fig. 9-14: Relative contribution of the different food items to the effective Intake Fraction (ingestion) of chromium after 25 years (top) and time-integrated (bottom) for a one year pulse emission according to the pan-European emission scenario to air in 1990 (cliparts by Corel Corporation, 2002)

i.e., the solid-water partitioning coefficient K_d and the soil erosion rate, only occurs to be significant in the long run. This indicates that the dynamics of chromium in the environment are rather slow. Thus, only the parameters involved in the exposure assessment of those food items remarkably contributing in the short term (cf. reference case in Fig. 9-14) show significant changes in the aggregated effective Intake Fraction for ingestion when integrating over shorter time horizons.

Table 9-6: Components of the effective Intake Fraction for chromium due to the ingestion of different food items according to the considered sensitivity cases after 25 years and time-integrated for the pan-European emission scenario to air in 1990 (emissions only take place in the first year) [$kg_{ingested}$ / $kg_{released}$]

Sensitivity scenario	Cereals	Spinach	Potatoes	Dairy products	Beef	Fish	Sum
after 25 years							
reference	$3.04 \cdot 10^{-7}$	$1.05 \cdot 10^{-7}$	$5.29 \cdot 10^{-8}$	$4.52 \cdot 10^{-8}$	$1.63 \cdot 10^{-8}$	$1.33 \cdot 10^{-14}$	$5.24 \cdot 10^{-7}$
Kd +900%	$3.04 \cdot 10^{-7}$	$1.05 \cdot 10^{-7}$	$5.29 \cdot 10^{-8}$	$4.52 \cdot 10^{-8}$	$1.63 \cdot 10^{-8}$	$9.54 \cdot 10^{-16}$	$5.24 \cdot 10^{-7}$
Kd -90%	$3.04 \cdot 10^{-7}$	$1.05 \cdot 10^{-7}$	$5.29 \cdot 10^{-8}$	$4.46 \cdot 10^{-8}$	$1.60 \cdot 10^{-8}$	$4.82 \cdot 10^{-13}$	$5.23 \cdot 10^{-7}$
erosion rate -90%	$3.04 \cdot 10^{-7}$	$1.05 \cdot 10^{-7}$	$5.29 \cdot 10^{-8}$	$4.52 \cdot 10^{-8}$	$1.63 \cdot 10^{-8}$	$5.10 \cdot 10^{-15}$	$5.24 \cdot 10^{-7}$
BCF above -90%	$3.04 \cdot 10^{-8}$	$1.05 \cdot 10^{-7}$	$5.29 \cdot 10^{-8}$	$3.52 \cdot 10^{-8}$	$1.46 \cdot 10^{-8}$	$1.33 \cdot 10^{-14}$	$2.38 \cdot 10^{-7}$
BCF below -90%	$3.04 \cdot 10^{-7}$	$1.05 \cdot 10^{-7}$	$5.29 \cdot 10^{-8}$	$4.52 \cdot 10^{-8}$	$1.63 \cdot 10^{-8}$	$1.33 \cdot 10^{-14}$	$4.77 \cdot 10^{-7}$
BTF beef -90%	$3.04 \cdot 10^{-7}$	$1.05 \cdot 10^{-7}$	$5.29 \cdot 10^{-8}$	$4.52 \cdot 10^{-8}$	$1.63 \cdot 10^{-9}$	$1.33 \cdot 10^{-14}$	$5.10 \cdot 10^{-7}$
BTF dairy -90%	$3.04 \cdot 10^{-7}$	$1.05 \cdot 10^{-7}$	$5.29 \cdot 10^{-8}$	$4.52 \cdot 10^{-9}$	$1.63 \cdot 10^{-8}$	$1.33 \cdot 10^{-14}$	$4.84 \cdot 10^{-7}$

Table 9-6: Components of the effective Intake Fraction for chromium due to the ingestion of different food items according to the considered sensitivity cases after 25 years and time-integrated for the pan-European emission scenario to air in 1990 (emissions only take place in the first year) [$kg_{ingested}$ / $kg_{released}$]

Sensitivity scenario	Cereals	Spinach	Potatoes	Dairy products	Beef	Fish	Sum
time-integrated							
reference	$3.88 \cdot 10^{-5}$	$1.48 \cdot 10^{-7}$	$6.71 \cdot 10^{-6}$	$4.35 \cdot 10^{-5}$	$1.71 \cdot 10^{-5}$	$1.70 \cdot 10^{-12}$	$1.06 \cdot 10^{-4}$
Kd +900%	$3.98 \cdot 10^{-5}$	$1.49 \cdot 10^{-7}$	$6.92 \cdot 10^{-6}$	$6.97 \cdot 10^{-5}$	$2.69 \cdot 10^{-5}$	$1.80 \cdot 10^{-13}$	$1.44 \cdot 10^{-4}$
Kd -90%	$3.19 \cdot 10^{-5}$	$1.36 \cdot 10^{-7}$	$5.33 \cdot 10^{-6}$	$2.28 \cdot 10^{-5}$	$9.12 \cdot 10^{-6}$	$1.47 \cdot 10^{-11}$	$6.93 \cdot 10^{-5}$
erosion rate -90%	$3.05 \cdot 10^{-4}$	$4.07 \cdot 10^{-7}$	$5.11 \cdot 10^{-5}$	$2.28 \cdot 10^{-4}$	$9.12 \cdot 10^{-5}$	$1.44 \cdot 10^{-12}$	$6.75 \cdot 10^{-4}$
BCF above -90%	$3.90 \cdot 10^{-6}$	$1.09 \cdot 10^{-7}$	$6.74 \cdot 10^{-6}$	$3.58 \cdot 10^{-5}$	$1.57 \cdot 10^{-5}$	$1.71 \cdot 10^{-12}$	$6.22 \cdot 10^{-5}$
BCF below -90%	$3.88 \cdot 10^{-5}$	$1.48 \cdot 10^{-7}$	$6.72 \cdot 10^{-7}$	$4.35 \cdot 10^{-5}$	$1.71 \cdot 10^{-5}$	$1.70 \cdot 10^{-12}$	$1.00 \cdot 10^{-4}$
BTF beef -90%	$3.88 \cdot 10^{-5}$	$1.48 \cdot 10^{-7}$	$6.71 \cdot 10^{-6}$	$4.35 \cdot 10^{-5}$	$1.71 \cdot 10^{-6}$	$1.70 \cdot 10^{-12}$	$9.08 \cdot 10^{-5}$
BTF dairy -90%	$3.88 \cdot 10^{-5}$	$1.48 \cdot 10^{-7}$	$6.71 \cdot 10^{-6}$	$4.35 \cdot 10^{-6}$	$1.71 \cdot 10^{-5}$	$1.70 \cdot 10^{-12}$	$6.70 \cdot 10^{-5}$

Table 9-7: Values of the parameter sensitivity evaluation measure (term in brackets of Eq. (9-1)) for pan-European emissions of chromium to air in 1990 for time horizons of 25 years and time-integrated (emissions only take place in the first year)

Sensitivity scenario relative to reference	Parameter variation	Cereals [%]	Spinach [%]	Potatoes [%]	Dairy products [%]	Beef [%]	Fish [%]	Sum [%]
after 25 years								
Kd +900%	9	0	0	0	0.003	0	-10.3	0
Kd -90%	-0.9	0.005	0	0.005	1.58	1.8	-3917	0.20
erosion rate -90%	-0.9	-0.014	0	-0.014	-0.001	-0.001	68.5	-0.010
BCF above -90%	-0.9	100	0.32	0	24.6	11.04	0	60.6
BCF below -90%	-0.9	0	0	100	0	0	0	10.1
BTF beef -90%	-0.9	0	0	0	0	100	0	3.10
BTF dairy -90%	-0.9	0	0	0	100	0	0	8.62

Table 9-7: Values of the parameter sensitivity evaluation measure (term in brackets of Eq. (9-1)) for pan-European emissions of chromium to air in 1990 for time horizons of 25 years and time-integrated (emissions only take place in the first year)

Sensitivity scenario relative to reference	Parameter variation	Cereals [%]	Spinach [%]	Potatoes [%]	Dairy products [%]	Beef [%]	Fish [%]	Sum [%]
time-integrated								
Kd +900%	9	0.30	0.14	0.34	6.70	6.43	-9.94	3.91
Kd -90%	-0.9	19.7	8.4	22.8	52.8	51.7	-849	38.57
erosion rate -90%	-0.9	-762	-196	-734	-471	-483	17.5	-595
BCF above -90%	-0.9	99.9	28.7	-0.48	19.6	8.96	-0.36	46.0
BCF below -90%	-0.9	-0.06	-0.023	100	-0.001	0	-0.04	6.30
BTF beef -90%	-0.9	0	0	0	0	100	0	16.07
BTF dairy -90%	-0.9	0	0	0	100	0	0	41.0

The number of food items on which the varied parameters may have a significant influence varies from one to all. Due to the fact that the biotransfer factors (BTFs) are not involved in the environmental fate assessment, their variation only influences the exposure towards dairy products and beef. Given that the BTFs are included in all related exposure pathway computations in a linear way (cf. sections A.7.9, A.7.10 and A.7.11), their variation translates one to one into the exposure assessment of the respective food item (100 % in Table 9-7).

A similar finding is observed for the bioconcentration factor of belowground produce where merely the exposure towards the analysed crop is significantly influenced (i.e., potatoes). Due to the inclusion of the harvest removal process in the environmental fate model according to the 'food removal' environmental setting, however, the chromium concentrations are increased in those arable land compartments with joint cultivation of aboveground and belowground produce. The reduced uptake by potatoes in the 'BCF below -90%' sensitivity case leads to a higher exposure through aboveground produce at least in the long run (bottom of Fig. 9-14).

The variation of the bioconcentration factor of aboveground produce, in turn, significantly influences the human exposure towards several food items. Cereals as well as spinach classify as aboveground produce. Furthermore, farm animals are fed cereals to differing degrees. According to the 'BCF above -90%' sensitivity case, thus, the contribution not only of cereals and spinach but also of dairy products and beef to the human ingestion exposure is significantly influenced when varying the bioconcentration factor of aboveground produce. While cereals are regarded as being protected towards atmospheric depositions, spinach leaves are exposed. The contribution of spinach to the effective Intake Fraction is not significantly affected in the short term when reducing the BCF of aboveground crops due to the inclusion of the exposure pathway directly via the atmosphere. This stresses the importance of the atmospheric exposure of leafy vegetables which also shows in Fig. 9-14 and in the case studies presented in Chapters 10 and 11.

The 'Kd -90%' and the 'erosion rate -90%' sensitivity cases affect all food items significantly in the long run. Only the freshwater fish Intake Fraction after 25 years already indicates substantial changes in the short term for these two sensitivity cases. However, freshwater fish exposure is insubstantial (cf. Fig. 9-14). The decreased water soil erosion rate leads to smaller transfers of pollutants from the terrestrial compartments that show water soil erosion to the freshwater environment (section A.3.3), thereby increasing the concentrations in the arable land and pasture compartments and decreasing those in the freshwater bodies. This is reflected in the signs of the sensitivity evaluation measure in Table 9-7. This finding supports the need to appropriately represent the water soil erosion process for non-degrading and non-volatile substances, at least for long term assessments. This has already been postulated in the present work with respect to the spatial differentiation in terms of compartments (see section 5.1). The exposure through freshwater fish is also inversely correlated to the decrease in the K_d values. This result is due to two effects. First, the share of dissolved chromium in the freshwater environment is increased which leads to higher fish exposure. Second, also the total amount of chromium present in the freshwater environment is larger which can be attributed to a higher influx from the terrestrial compartments via the process of overland flow (section A.3.4) which depends on the amount of chromium present in the soil solution. The shift in the equilibrium distribution coefficient relating the aqueous phase concentration to the bulk concentration due to the change in the K_d value also translates into the process of matrix leaching. This again leads to a faster reduction of the chromium concentration in permeable soils (section A.3.6) and, thus, to a lower exposure through terrestrial plants and farm animals.

The influence of different assumptions with respect to the pH values of terrestrial compartments on the partitioning behaviour of chromium has also been investigated (Table 9-8). The effect of letting the K_d value depend on either a zonally variable pH value or just on a compartmentally variable pH value mostly shows in the long run for this metal. Two aspects can be noted. First, the aggregated effective Intake Fraction for ingestion exposures increases by 84 % when comparing the 'compartmental pH variation' to the 'zonal pH variation' scenario. This change fulfils the evaluation criterion with respect to the scenario analysis performed above where the variation in one component is classified influential if it leads to a change by more than 50 % with respect to a reference scenario (cf. section 9.3.3). Second, the contribution of different food items to the aggregated effective Intake Fraction for ingestion also changes remarkably in the long run. While there are higher exposures through all food items at least by a few percent, the exposures through dairy products and beef increase by more than a factor of two when integrating over time for the 'compartmental pH variation' scenario. These additional exposures appear to occur only in the long run as the effective Intake Fraction due to ingestion are practically equal for both sensitivity cases analysed after 25 and 100 years. The increases in the absolute value of this exposure measure are only 0.004 % and 0.4 %, respectively. The remarkable increase in the exposures through pasture-related food items can again be explained by the minimal pH values occurring in this compartment when allowing the pH to zonally vary. The invariant pH values representing neutral conditions (cf. Table 9-5) lead to a slower decline of the chromium concentrations in pasture soils and, thus, to a higher overall exposure through dairy products and cattle (see also the discussion above).

Table 9-8: Contribution of the different food items to the Intake Fraction (last row) of chromium for ingestion exposures after 25 years and time-integrated according to the sensitivity cases with respect to pH variability (pan-European emissions to air in 1990, only taking place in the first year)

Contribution to Intake Fraction for ingestion	After 25 years		Time-integrated	
	Zonal pH variation[a]	Compartmental pH variation	Zonal pH variation[a]	Compartmental pH variation
Cereals	58.1%	58.1%	36.5%	20.5%
Potatoes	10.1%	10.1%	6.3%	3.6%
Spinach	20.1%	20.1%	0.14%	0.077%
Dairy products	8.6%	8.6%	41.0%	55.2%
Beef	3.1%	3.1%	16.1%	20.7%
Fish	$2.5 \cdot 10^{-8}$	$1.8 \cdot 10^{-8}$	$1.6 \cdot 10^{-8}$	$9.3 \cdot 10^{-9}$
Intake Fraction [$kg_{ingested}$ per $kg_{released}$]	$5.24 \cdot 10^{-7}$	$5.24 \cdot 10^{-7}$	$1.06 \cdot 10^{-4}$	$1.95 \cdot 10^{-4}$

a.Previously termed 'reference' sensitivity case.

Above, significant sensitivities of different parameters have been identified. However, if the sensitive parameters are known rather precisely these parameters' sensitivities are not considered important (Hamby, 1994) which shall be discussed next.

In section B.5.3, it is discussed that the water soil erosion rate is a function of several site-specific parameters such as rainfall erosivity and crop management amongst others. The absolute value of the water soil erosion rate 'naturally' varies by two orders of magnitude across Europe (Milliman and Meade, 1983; Walling and Webb, 1983) when taking the sediment yield as an approximate indicator to the net erosion process which is in line with Morgan (1999). When operating at the regional scale and employing long-term average values, the water soil erosion rate may, therefore, be known rather precisely. However, it merely depends on compartments according to the methodological approach proposed here. Thus, the erosion rate contributes rather substantially to the overall uncertainty of the assessment. Whether the impacts according to the scenarios analysed in

Chapters 10 and 11 are overestimated or underestimated depends on the interplay of the spatial variability of several parameters such as atmospheric deposition, food production and erosion-prone areas and needs further investigations.

The concept of the solid-water partitioning coefficient K_d relies on a linearisation of the sorption behaviour of a chemical compound (Anonymous, 1999a; Aboul-Kassim and Simoneit, 2001b). The partitioning of a substance may, however, be influenced by several conditions of the environmental system for which a substance's behaviour shall be assessed. Influential factors may be the binding capacity of the solid phase, the total concentration of the substance present in the system, the pH, the organic carbon content especially in colloidal form and potential reaction partners amongst others. Although the present assessment allows to consider a pH-dependency of the partitioning of substances, the K_d value of a substance and a given pH range may still vary by one order of magnitude or more (Anonymous, 1999b). The introduction of pH-dependency could be demonstrated to decrease the sensitivity of the K_d value in the case of chromium at least when increasing its values. However, the consideration of further dependencies appears to be necessary to more reliably assess the considered substances. As a result, the K_d needs to be classified as contributing substantially to the overall uncertainty.

The transfer of substances into plants and/or animals according to a linear relationship between soil or feed concentrations on the one hand and living tissue concentrations on the other is deemed to be especially uncertain (e.g., by Vermeire et al., 1997). The reasons for this are similar to those related to the K_d concept in that the linearisation does not take into account all relevant aspects on which the BCFs and BTFs may depend such as plant or animal species variety, diet, solid and/or climatic conditions, age of the plant or animal (e.g., McLachlan, 1994). Due to the strong dependency of the exposure results on these parameters even already in the short term, they also contribute to the uncertainty of the estimated exposure.

Having analysed chromium in more detail, the question arises to what extent the exposure towards the other three trace elements is affected by the variation of the respective parameters. The solid-water partitioning coefficient of chromium in the neutral to alkaline range is the highest among the trace elements investigated (cf. Table C-1). According to the reasoning given above, it is evident that not only a decrease but also an increase of the respective K_d values will have a pronounced effect on the results for arsenic, cadmium and lead. The behaviour of chromium upon variation of its solid-water partitioning coefficients additionally suggests that the time-integrated Intake Fraction of the more mobile trace elements cadmium and lead will not only be lower but will also approach its final value quicker when decreasing the K_d values due to accelerated removal of these metals from the arable land compartment through which most of the human exposure occurs (cf. Fig. 10-3, 10-4 and 11-6). A pronounced effect will also be observed for all of the other substance-dependent parameters, provided these are related to food items through which a considerable amount of the exposure occurs. For instance, the exposure through animal products is rather small for cadmium and lead according to the case studies investigated (cf. Fig. 10-3, 10-4 and 11-6). Consequently, the variation of the biotransfer factors are expected to exert a less pronounced effect on the aggregated ingestion exposure estimates for these two heavy metals. The impact of the water soil erosion rate on the environmental fate at least of cadmium and lead is expected to be less significant, owing to their higher mobility reflected in their smaller K_d values. The higher mobility of these two metals also shows in the percentage of the ingestion-related effective Intake Fraction estimated for the case studies to occur after 10 and 100 years. These are at least a factor of four higher for cadmium and lead than for chromium and arsenic (cf. sections 10.3 and 11.4).

9.4 Concluding remarks on the evaluation of results

Overall, there exist many uncertainties associated with the presented exposure assessment methodology as presented in section 9.3. The uncertainties related to the impact assessment tend to be even larger (Finley and Paustenbach, 1994) especially due to the unavailability of reliable epidemiological dose-response functions. The appropriate evaluation of large scale multimedia exposure and impact assessment models is generally hindered due to the process of policy decision-making related to their application (Ragas et al., 1999), lack of data (Hunsaker et al., 1990) and/or adequate evaluation criteria (Rykiel (Jr.), 1996) at the respective scale.

Data quality is one of the major limiting factors in model development and model quality assurance (e.g., Addiscott et al., 1995; Beck and Chen, 2000). Regarding particularly the spatially distributed parameters, the selection of appropriate values could not always be guided by data quality but was rather constraint by data availability. Therefore, the influence of choosing other datasets e.g. for the hydrological cycle could not be evaluated.

Other uncertainty analyses have shown that especially emission information is crucial for site-dependent assessments of environmental media concentrations (Price et al., 1996; United States - Environmental Protection Agency, 1998; Vink and Peters, 2003). The comparison of the atmospheric deposition data with moss monitoring data points into the same direction but may also partly be due to the employed air quality model. It is, furthermore, supported by an EU position paper (European Commission, 2000b) emphasizing that one of the presently best available pan-European emission estimates for 1990 (Berdowski et al., 1997) in parts considerably overestimates emissions.

Trapp and Matthies (1998) have formulated as one goal in the modelling of compartment systems to keep the number of compartments as low as possible. It has been shown in the conducted scenario analysis that the distinction of pastures from arable land leads to an increase of exposure due to pasture-based food chains such as dairy products and beef. Depending on the emission scenario and the substance investigated, this may be important. Also the distinction of rivers from lakes especially with respect to the particle mass balance has a pronounced effect on the freshwater fish exposure estimates. In contrast to the differentiation of the agricultural soils into arable land and pastures, the distinction of flowing from stagnant water bodies in the way it is performed here, however, does not increase the number of compartments but merely changes compartmental characteristics. The most significant increase in compartments is related to the spatial differentiation of the model's geographical scope into zones. It appears as if the lower spatial resolution does not change the overall exposure estimates substantially for the pan-European emission to air scenario analysed. However, analyses of more localised emission sources including direct discharges into water may reveal more pronounced differences as discussed above.

The scenario analysis performed addressing different environmental settings in terms of spatial differentiation, process formulations and process inclusions supports the findings by Hertwich et al. (2000) in that the variation in the spatial resolution and/or spatial differentiation does not affect the overall exposure results of large scale contamination scenarios to a substantial degree, for instance by more than a factor of two. However, these may cause a shift in the relative importance of the different food items contributing to the overall human exposure.

The sensitivity analysis of the parameters has identified the water soil erosion rate as one non-substance-specific parameter that is significantly influential on the exposure results at least of rather immobile substances such as chromium, beside substance-specific parameters such as the solid-water partitioning coefficient, bioconcentration factors and biotransfer factors. Having used the water soil erosion as one key criterion for the distinction of compartments in an exposure assessment of rather persistent and non-volatile substances is, therefore, supported.

Regardless of the magnitude of the uncertainties, the present work as such is an improvement towards more knowledge about the magnitude of the external costs occurring due to human activities. Before this contribution hardly any (if at all) information on the external costs for exposure routes other than inhalation had been available. Therefore, the uncertainty is considered acceptable given the model's purpose (Eisenhart, 1968; Scott et al., 2000). Furthermore, the approach is adequate in that the model design has been orientated at the 'availability of parameter values' and the 'acceptability of the goals with the budget' (Draper and

Smith, 1966 cited in Caswell, 1976). Given that "the modeller and the users may have different thresholds for confidence" (Robinson, 1999, p. 68), it has been tried to document the methodological approach as explicit as possible taking account of the suggestion "that the manner in which a study is performed is more important in forming a user's quality perception than the quality (or validity) of the model and its results" (ibid., p. 68). This is in line with one of the good-practice principles to show all formula used in the model (Burmaster and Anderson, 1994; Veerkamp and Wolff, 1996) which is done in Chapters 4, 5, 6 and 7, and Appendix A of the present work.