2 Assessment of human health impacts and the approach followed

Generally speaking, impacts on human health due to human activities shall be assessed and valued in the present work. Due to the spatial coverage which entails a "lack of full access ... to the phenomena of interest" (Oreskes et al., 1994, p. 644), it is necessary to perform this assessment by means of a numerical simulation model. The methodological framework to be followed consists of the *Impact Pathway Approach* which will be outlined below (section 2.2). At the onset of the present work, it focused on exposures to air pollutants via inhalation which is why the method especially needed an extension with respect to ingestion exposures via food and/or drinking water. This has brought about the necessity to introduce the media soil and water into the analysis. Furthermore, a detailed and explicit human exposure assessment needs to be set in place for the ingestion exposure route. Also for consistency reasons, the aims and requirements for the respective model development will be defined based on a general modelling review (section 2.3). The type of modelling approach to follow will then be defined. In Chapter 3, the needs for model development are formulated based on a review of existing models and according to the prioritised contaminants.

The focus is set on human health because it is known from experience that damages to human health dominate by far the external costs out of the set of receptors for which impact assessment and monetary valuation schemes are available beside global warming damages (European Commission, 1999b). It shall be noted that the evaluation of impacts on living organisms other than humans may comprise another important externality maybe even leading to a loss of species in certain settings, thereby potentially reducing biodiversity. The consideration of such impacts, however, is rather complex as one has to deal with many species showing rather different sensitivities which may even depend on the habitat in which they live. The protection of 'biodiversity' is very often formulated as a goal in the scientific as well as the political context. The definition of 'biodiversity' as an indicator, however, is rather diverse due to the fact that there are many different aspects to it (cf. Linares Llamas, 2003) all of which are quite difficult to operationalise. Examples of these aspects are diversity in genes, conservation of species where they exist or at a global level. The inclusion of impacts on other living organisms especially with respect to biodiversity is, thus, deemed a whole study area of its own which may be addressed in future investigations.

Before continuing, some definitions or considerations on some of the terms used within this study are given as follows.

2.1 Definitions and considerations of some terms

2.1.1 Nomenclature of substances of concern

In the environmental context, substances of concern are termed by notions like environmental chemical, xenobiotics, hazardous or poisonous substances, contaminants or pollutants. All of these have specific connotations. Their application shall be outlined briefly here.

Environmental chemicals, sometimes also referred to as 'man made substances' can either simply be defined as chemicals that occur in the environment (Walker et al., 2001) or as substances which enter the environment as a result of human activity and occur in concentrations or amounts that may put living organisms, in particular humans, to a risk according to Anonymous (1971) cited in Bliefert (1997) and Korte (1992). Following the second definition, environmental chemicals can furthermore be differentiated into those of natural origin and 'foreign' substances (Korte, 1992). In a strict sense, the latter are exclusively synthetic substances (Römbke and Moltmann, 1996) and are, thus, foreign to any organism, i.e., they do not play a part in their normal biochemistry. They can be termed *xenobiotics*. Environmental chemicals of natural origin may for instance be heavy metals that are enriched in the environment due to a human activity, a wide-spread example being lead in soils which has been released due to combustion of traffic fuel.

The terms *contaminant* and *pollutant* can be described separately but are often in effect synonymous. Both are used to describe chemicals that are found at levels judged to be above those that would normally be expected. Whereas the definition of 'contaminant' ends here which is simply equivalent to the second definition given for environmental chemicals above, pollution should mean contamination resulting in adverse biological effects in the environment in a scientific precise way (Chaney and Ryan, 1994; Chapman, 2001). This is, however, not an easy distinction to make. Whether or not a contaminant is a pollutant may depend on its level in the environment and the organism or system being considered

(Walker et al., 2001). Thus, one particular substance may be a contaminant relative to one species but pollutant relative to another. Even more complicated, it may be a contaminant for one individual of a population and a pollutant to a more sensitive one of the same population. Finally, the question about the existence of thresholds for an effect related to the occurrence of a contaminant is crucial. In line with the reasoning in section 7.3, in practice it is often difficult to demonstrate that harm is not being caused so that in effect pollutant and contaminant become synonymous (Walker et al., 2001). Correspondingly, the terms 'environmental chemical', 'contaminant' and 'pollutant' are used interchangeably in this work.

Some environmental chemicals are of higher concern than others. In the context of potentially toxic substances, composite terms for pollutants used in the regulatory process (e.g., of the European Union, European Commission, 2001a) are for instance: Persistent Organic Pollutants (POPs), Persistent, Bioaccumulative and Toxic (PBT) chemicals and very Persistent and very Bioaccumulative (vPvB) substances. As one can see from these notions, in any case their characteristics with respect to persistency give reason for increased attention which will have implications on the choice of contaminants on which the present study focuses (see section 3.2).

2.1.2 Nomenclature with respect to exposure

Human exposure may occur via different *routes of exposure*. The main exposure routes are inhalation of air, ingestion of food, drinking water and other matter such as soil, and dermal exposure (United States - Environmental Protection Agency, 1992, 1997c; World Health Organisation, 2000a; European Commission, 2003c). Other routes of exposure exist such as intravenous, intraperitoneal, subcutaneous and intramuscular routes (cf. United States - Environmental Protection Agency, 1994) occurring especially in the medical domain. These are, however, less important for environmental chemicals.

When assessing substances in the soil and water environment, there is no doubt that ingestion is to be included in the analysis. Inhalation may also need to be considered for instance in cases when people are exposed to substances volatilising from contaminated tap water (cf. McKone, 1993a; Finley and Paustenbach, 1994; Georgopoulos et al., 1997; Hopke et al., 2000).

Another distinction of exposures can be made according to *target populations* (e.g., workers, consumers, public, European Commission, 2003a; European Centre for Ecotoxicology and Toxicology of Chemicals, 1994). The assessment of occupational exposures as well as exposures towards consumer products is beyond the scope of the present analysis.

A third way of classifying exposures is into *direct* and *indirect*. Different definitions and distinctions are, however, made. For instance, in some regulatory risk assessment guidelines it is distinguished between direct exposures for example at the working place or through consumer products and indirect exposure via the environment, i.e., exposure via air, water, soil and food (European Centre for Ecotoxicology and Toxicology of Chemicals, 1994; European Commission, 2003a). A different distinction is made in analyses making use of environmental fate and exposure models noting that the guidelines mentioned above may involve such tools as well. In the latter context, only exposure towards exposure media that are not part of the environmental fate model is considered indirect, i.e., inhalation of air and ingestion of water are direct whereas ingestion of food is indirect (McKone, 1993a; van de Meent et al., 1996; International Council of Chemical Associations, 1998; Hertwich et al., 2000; Huijbregts et al., 2000a; Schwartz, 2000; Trapp and Schwartz, 2000). Further note that only inhalation is considered direct by United States - Environmental Protection Agency (1998) most likely because the fate analysis only covers air from which the other media's concentrations are derived.

It has been mentioned above that the assessment of exposures at the work place and due to consumer products which are classified as direct exposures among other by European Commission (2003a) is out of the scope of the present analysis. Therefore, the second approach to distinguish between direct and indirect exposure is followed. Indirect exposure, hence, means ingestion of food. As a consequence, direct exposure (of humans) principally occurs via inhalation of air, ingestion of drinking water and soil particles, and skin contact to air, water and soil.

Due to the model's spatial resolution (section 4.3), exposure is primarily assessed for diffuse inputs to the environment for example by multiple emission sources. However, one needs to be aware that exposure to contaminants in food and drinking water but also in ambient air can also originate from various other sources like

- accidental releases (Alloway and Steinnes, 1999; Buckley-Golder et al., 1999; Fiedler et al., 2000; European Commission, 2001b),
- tire-wear from vehicles (Councell et al., 2004),
- contamination of food for instance due to migration of substances from packaging into food (Harrison, 2001a; Watson, 2001), due to food processing (Büchert et al., 2001), or due to contamination of feeding stuff (Fiedler et al., 2000),
- natural background of trace elements (Kabata-Pendias and Pendias, 1992; Wedepohl, 1995; Reimann and de Caritat, 1998; Smedley and Kinniburgh, 2002),

- smoking for example in the case of cadmium (Chaney et al., 1999), nitroaromatic compounds (Purohit and Basu, 2000) and benzene (Hattemer-Frey et al., 1990),
- grilled food items (Purohit and Basu, 2000),
- tubing, especially for lead (Wilhelm and Ewers, 1999) but also other metals such as cadmium (World Health Organisation, 1992b),
- the working environment (Stern et al., 1984; Ewers and Schlipköter, 1991; Buckley-Golder et al., 1999), and
- those especially leading to indoor air contamination for example radon from soils (Davies, 1998; Hopke et al., 2000) and volatile organic compounds (VOCs) and polychlorinated biphenyls (PCBs) for instance from building materials (Brown et al., 1994; Bleeker et al., 1999).

Many of these exposures occur in a very localised area or only during short episodes. The spatial and temporal resolution of the environmental fate model brings about that such localised or temporary exposure assessments cannot be carried out. This means, for instance, that an assessment of the exposure of individuals cannot be conducted. This applies especially to those individuals with localised food supply that is produced on contaminated soils/feed (Tennant, 2001). The exposure scenario in which people eat only food that is produced in their vicinity (European Commission, 1996b) or even by themselves is also known as the *subsistence farmer* scenario (United States - Environmental Protection Agency, 1998). This approach can be extended to become a nested exposure assessment by exporting local food production surplus to regional and potentially to global levels (as done for radionuclides in European Commission, 1999a).

By *exposure pathways* the definition as given by United States - Environmental Protection Agency (1992) is adopted here which reads: "(an) exposure pathway is the course a chemical takes from its source to the person being contacted" (p. 7).

Exposure modelling is understood here as the "process of quantifying the mass flows of a chemical and calculating the resulting concentrations in the environment by means of mathematical expressions" (van de Meent et al., 1996, p. 103).

In general, exposure assessments most often build to rather large extents on results from environmental fate models. Therefore, some definitions with respect to environmental fate modelling shall be given here as well. Following the idea that a multimedia model includes the atmosphere, the aquatic ('water') and the terrestrial environment ('soil'), the term *medium* is reserved to these three 'environments' addressing them as a whole. The perception that media are distinguished according to their predominant phase is different from that of others (e.g., Cowan et al., 1995b) and at times complies with the definition of 'main compartments' (e.g., as distinguished by Trapp and Matthies (1998)). Biota could be considered as an additional medium.

Each of these media may be further distinguished into *compartments*. Compartments are boxes that are by definition homogeneous with respect to all of their properties (assumption of homogeneous mixing, Trapp and Matthies, 1998). Their properties may, therefore, serve as a basis in order to distinguish these. They are assumed to be at thermodynamic equilibrium internally. Following the Mackay level III/IV modelling approach as introduced by Mackay (1979), transfers between these compartments show resistances which are expressed as rates, i.e., following the processes' kinetics. Losses from the system such as chemical transformation or transport beyond the model's boundaries are also allowed for. The difference between level III and IV is that the one assesses steady-state situations assuming constant and continuous emissions while the other is also capable of investigating the temporal development of a substance's concentration in the distinguished compartments over time given a specified emission situation.

2.1.3 Considerations with respect to risk and impact assessment

Although also drawing to some extent on regulatory risk assessment methodologies, it shall be emphasized here that the present work aims at estimating impacts rather than risks. This statement can definitively be challenged since the impacts to be assessed are based on dose- or exposure-response functions that describe a statistical chance for an effect to occur (e.g., development of cancer or skin irritation occurrence) which is then combined with a severity measure such as Disability Adjusted Life Years to yield an impact (cf. section 7.3). Nevertheless there are differences in the approaches taken to assess either impacts or risks which shall be described in the following.

Many regulatory Risk Assessments (RAs) in the United States of America (US) and the European Union (EU, e.g., United States - Environmental Protection Agency, 1998; European Commission, 2003b) make use of so-called Risk Characterisation Ratios (RCRs). Such RCRs merely indicate whether there is concern or not by giving 'yes - no' answers. They are calculated by relating some effect measure such as the Predicted No Effect Concentration (PNEC) to a measure of exposure usually termed Predicted Environmental Concentration (PEC) yielded by an exposure model. For characterizing human exposure, no safety factors are introduced and the PNEC is divided by the PEC yielding a Margin Of Safety (MOS[1], European Commission, 2003b). This is then valued by experts in order to provide guidance whether to act from a regulatory body's point of view or during product development at company level. A fair degree of conservatism at least in the initial tiers of the assessment is introduced during the determination of the RCR components in order not to underestimate the risk (European Commission, 1996a; United States - Environmental Protection Agency, 1998; Organisation for Economic Co-operation and Development, 1999).

Olsen et al. (2001) point at the limited use of these rather qualitative RCRs in a context in which effects shall be assessed and aggregated according to their severity such as in Life Cycle Analyses (LCAs) and in externality valuation exercises. Still the authors conclude that "presently, there is no better method for a generally applicable, more quantitative risk characterisation" (ibid., p. 394). However, adopting conservative '(reasonable) worst case' assumptions reduces the validity of risk assessment approaches for LCA purposes (Olsen et al., 2001) although some authors consider the inclusion of safety factors for instance a strong point of risk assessments when compared to LCAs because they take uncertainties into account (Jørgensen and Bendoricchio, 2001).

When performing impact assessments, one needs to distinguish what impacts are tried to be estimated. Within the field of Life Cycle Impact Assessment (LCIA), for instance, it is common understanding that *potential impacts* are assessed. Unlike rather site-specific approaches such as Environmental Impact Assessments and higher tier Risk Assessments which try to estimate actual impacts, LCIAs try to characterize additional impacts by emissions taking place during the life cycle of a so-called functional unit (Guinée et al., 1996; Udo de Haes, 1996). These emissions, however, only have a potential to lead to different types of impacts which depends on several conditions (Udo de Haes, 1996). Heijungs (1995) describes it as follows: "(w)hether this potentiality becomes actuality is dependent on background concentrations and simultaneous synergistic or antagonistic concentrations, which are by their site-specific and product-unrelated character outside the scope of normal LCA, nor can they feasibly (be) included" (p. 223). This points at a shortcoming especially when evaluating toxic impacts within many present LCA methodologies that spatial and/or temporal information related to releases into the environment are lost during the data gathering step (Guinée et al., 1996; Nichols et al., 1996; Udo de Haes, 1996; Owens, 1997b; Krewitt et al., 2002) which is even stated as a limitation in the ISO norm (DIN EN ISO, 14042:2000). Furthermore, no information on other past or present emission activities or natural background concentrations (e.g., in the case of metals) is available.

While additionally assuming that there are no effect thresholds (Krewitt et al., 2002), this leads to a situation that may be perceived as if "all theoretically

[1] Note when evaluating pesticides according to EU legislation, the Toxicity-Exposure Ratio (TER) is defined analogously to the MOS.

possible consequences or hazards, not actual impacts or the prediction of impacts" are considered (Owens, 1997b, p. 362) extending the "worst case scenario to an impossible scenario" (ibid., p. 364).

In order to arrive at *actual impacts* of hazardous substances, it is evident that a substance must interact with an organism to exert its toxic potency leading to effects. Thus, the estimation of actual impacts necessitates information on the spatial distribution of both the change in concentration and the target organisms (Chapman, 2001; Krewitt et al., 2002) as well as their co-existence in time at the same place. One has to note that there are tendencies to make LCIAs more realistic especially in terms of the spatial distribution of releases (e.g., Potting and Hauschild, 1997; Potting et al., 1998; Nigge, 2000) partly building on the Impact Pathway Approach followed in this study (Krewitt et al., 1998, 2001; Spadaro and Rabl, 1999; cf. section 2.2).

It shall be noted that the question whether to assume threshold effect levels especially for populations will be discussed in section 7.3.

Before concluding this section, it shall, furthermore, be noted that the term 'impact' must not be understood in this document in a way to justify legal claims towards the entities responsible for the emissions investigated. To the knowledge of the author, the naming of the impact assessment step has been or was a reason why the methodology of Life Cycle Analysis has not been or was not widely used within the US.

2.2 Impact Pathway Approach

In the present work, the *Impact Pathway Approach* (IPA) is followed which has been developed within the series of ExternE Projects on 'External Costs of Energy' funded by the European Commission (1999a). It is a bottom-up approach in which the causal relationships from the release of contaminants through their interactions with the environment to a physical measure of impact (the 'impact pathway') and, where possible, a monetary valuation of the resulting welfare losses is assessed (see Fig. 2-1).

As it was the objective of the ExternE study to achieve an economic valuation of impacts, the impact assessment procedure is very much oriented to arrive at the damage level. Due to its modularity, it provides results on various intermediate levels of the environmental mechanism as well that can be used independently of any valuation methodology. According to its being a bottom-up approach, the Impact Pathway Approach strives for a high spatial resolution in order to capture the sources of the substances, i.e., human activities. Unlike regulatory risk assessments, the impacts or rather the 'risks of impacts to occur' that are assessed by the IPA are intended to be representative (so-called central or best estimate) rather than conservative or protective.

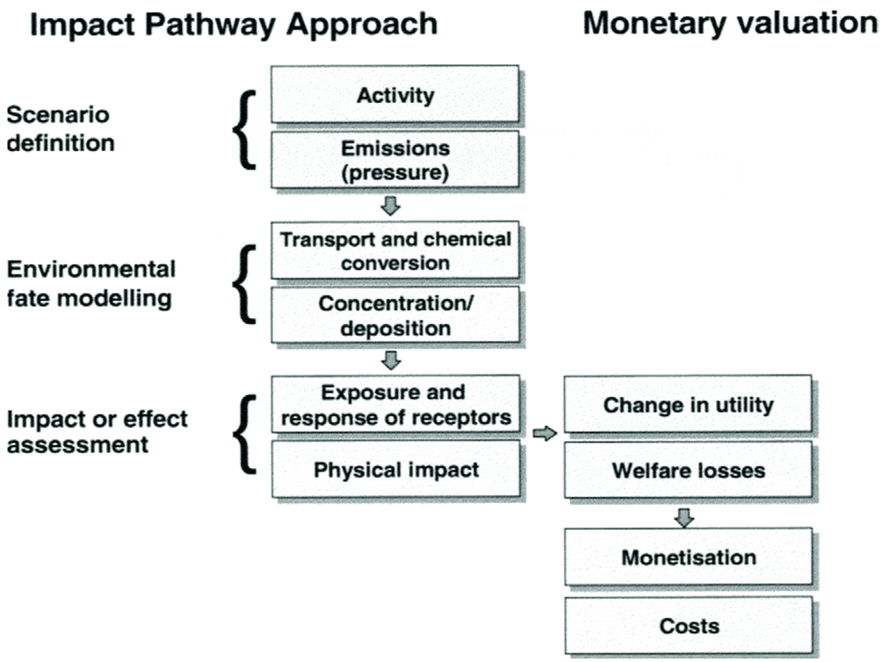

Fig. 2-1: Flowchart of the Impact Pathway Approach including monetary valuation

The Impact Pathway Approach is implemented into an integrated impact assessment and valuation tool called *EcoSense* (European Commission, 1999a). Initially, it supported the quantification of environmental impacts due to activities only at a single location such as a power plant. Further developments of the basic model led to different versions of the EcoSense model. They additionally allow the modelling of line sources and multi-sources for Europe for example from road traffic and from countries, respectively. As the emissions of the different types of sources contain different chemicals, the EcoSense transport version is capable of modelling partly different pollutants than the EcoSense single/multi source version (Table 2-1). Principally all pollutants listed in Table 2-1 (and more) can be implemented with little effort in all different EcoSense versions. Besides EcoSense Europe single/multi source versions of EcoSense have been set up for Brazil/Latin America, China/Asia, Russia and the Ukraine.

The impact assessment is performed in a spatially-resolved way. Principally one may distinguish site-generic from site-dependent and site-specific assessments (cf. Hauschild and Potting, 2003). In site-generic assessments, all sources

Table 2-1: Summary of the pollutants currently considered in different EcoSense Europe versions

Pollutants	EcoSense Europe version		
	Single source	Multi-source	Transport
SO_2	x	x	x
NO_X	x	x	x
NH_3	-	x	x
PM_{10} (primary particles)	x	x	-
Suspended particulates (particle class differentiated)	-	-	x
Non-methane volatile organic compounds (NMVOCs)	x	x	x
CO	x	-	x
As, Cd, Cr, Hg, Ni, Polycyclic Aromatic Hydrocarbons (PAHs), Pb[a], PCB[a], PCDD/F	x	-	-
Benzene, benzo(a)pyrene, 1,3-butadiene, ethene, formaldehyde	-	-	x

a.Exposure-response functions are not implemented at present.

are considered to contribute to the same generic receiving environment while a moderate to high degree of spatial differentiation in terms of emission sources and/or receiving environment is employed for site-dependent and site-generic approaches, respectively. In order to cover different substances and different scales, the EcoSense single/multi source version for Europe provides three air quality models completely integrated into the system (Table 2-2). In order to allow for this site-dependent and/or site-specific assessment, EcoSense provides a comprehensive set of relevant input data for the whole of Europe. Based on the European CORINAIR emission database, the definition of emission scenarios takes into account emission reduction measures in specific countries or more specific administrative units as well as in industry sectors.

Table 2-2: Air quality models implemented in EcoSense

Model	Application	Type	Reference
Industrial Source Complex Model (ISC)	Local transport of air pollutants from point sources (site-specific)	Gaussian plume model	Brode and Wang (1992)
ROADPOL	Local transport of air pollutants from line sources (site-specific)	Gaussian plume model	Vossiniotis et al. (1996)
Windrose Trajectory Model (WTM)	Regional (long-range) transport and chemical reaction (site-dependent)	Climatological trajectory model	Trukenmüller and Friedrich (1995) and Trukenmüller (1998) based on work done by Derwent and co-workers (Derwent and Nodop, 1986; Derwent et al., 1988)
Source Receptor Ozone Model (SROM)	Regional assessment of ozone concentrations (site-dependent)	Episodic trajectory model (country-to-grid matrices)	Simpson and Eliassen (1997), Simpson et al. (1997)

For the impact assessment and valuation step, the initial version of EcoSense already includes a large number of exposure-response functions and monetary values that were compiled and thoroughly reviewed within the ExternE projects (European Commission, 1995, 1999a).

The Impact Pathway Approach can be regarded as a particular example of Life Cycle Analysis (LCA) which is why in the following many concepts from this field of research are drawn from.

2.3 Model aim and requirements

According to Veerkamp and Wolff (1996), "(b)efore selecting a model, the fundamental problem is to define precisely the question a model is intended to answer and the level of accuracy required" (p. 94). The main aim of the present work is to extend the existing human health impact assessment and valuation approach (cf. section 2.2) to substances that reach human beings through the media soil and water. The final indicator to be estimated are the external costs

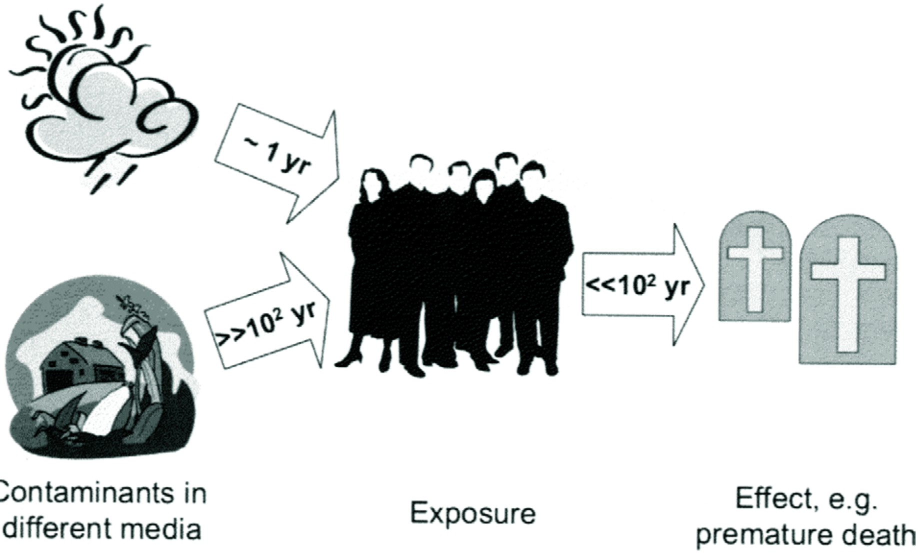

Fig. 2-2: Maximal time scales between contamination of different media leading to exposures via inhalation and/or ingestion and impacts on human health (cliparts by Corel Corporation, 1999, 2002)

related to a human activity. Due to the extending nature of the work, the methodology presented and used here needs to take into account the guiding principles and assumptions that had been followed during the series of ExternE projects for consistency reasons. According to European Commission (1999a), the guiding principles of the Impact Pathway Approach are (a) transparency, (b) consistency and (c) marginal approach.

The guiding principle of transparency is addressed by documenting precisely what was done and how in addition with an indication of the related uncertainties and methodological completeness of the assessment (cf. Chapter 9). Furthermore, the EcoSense tool has been designed to allow for any changes of the underlying data and equation formulations with respect to the impact assessment and monetisation by the (knowledgeable) user. This was achieved by the usage of a database for the storage of data as well as the equation definition (cf. section 4.4).

Consistency means that the assumptions between the different components of the Impact Pathway Approach are in line with each other. These assumptions need to apply to all of the evaluated cases (or scenarios) as well in order to allow for valid comparisons. One sub-aspect of consistency are the spatial and temporal scales that are looked at. Within the ExternE-methodology impacts are attempted to be estimated over the whole temporal and spatial scale, focusing on impacts occurring in Europe. Depending on a chemical's environmental behaviour, the lifetime between emission and exposure to a receptor may vary considerably (cf. Fig. 2-2). Whereas for example sulphur compounds in air have a residence time in the order of days (Seinfeld, 1986), persistent substances such as heavy metals may reside in soils or sediments for many years leading to rather delayed exposures to human beings (Hellweg, 2000; van den Bergh et al., 2000; Huijbregts et al., 2001). Also the time elapsed between the exposure to a pure air pollutant and an apparent corresponding impact may be in the order of decades, for instance for chronic mortality due to the exposure to fine particles (Pope et al., 1995).[2] However, the delay between emission, inhalation exposure and effect usually is at most about one generation due to the restricted residence time in air[3] of substances exerting quantifiable effects on human beings. Thus, the consideration of exposure routes due to ingestion implies the coverage of longer time horizons in order to fully assess the effects of long-lived substances. This also leads to the question how effects occurring at a very distant point in time can be valued in terms of the present value of money (cf. section 8.1 on the issue of discounting). In any case, the uncertainty about the predictability of the future is an issue that needs to be kept in mind.

Although the approach originally had been described as marginal, i.e., small additional or incremental human activities leading to emissions and, thus, effects are evaluated, also analyses of whole economies have been performed in the meantime (European Commission, 2003d).

The Impact Pathway Approach principally constitutes a methodology which can be applied to any situation/location on the globe. However, it was in the first place developed for Europe (cf. section 2.2). It is also this part of the world for which the implementation of the IPA is most advanced. Because of this and due to the fact that the present work was supported by several EC-funded projects (see Acknowledgements), the tool to be described will focus on the geographical scope of the European EcoSense versions (see Fig. B-1). This also means that the environmental fate and exposure/impact assessment to be developed needs to comply with the assumptions of the models used for the inhalation impact assessment (cf. Table 2-2). In the case of the regional air quality model

2 If premature death occurs in the long run (so-called chronic mortality) one may additionally distinguish between (apparent) latency times, a period with health impairments (morbidity) and years of not realized life expectancy (e.g., Years Of Life Lost, European Commission, 1999a; Hurley and Miller, 2001; cf. sections 7.3.8 and 8.2).

3 Note that the substance may be deposited to the surface and volatilise once or many times again.

WTM which is implemented in all different EcoSense versions, one main assumption in this regard is that it operates on meteorological data that are taken as representative for a one year period (section 4.1). Furthermore, the model to be developed needs to allow for a bottom-up analysis of impacts. A spatially-resolved modelling framework is adopted in order to be able to perform site-dependent impact and external costs assessments for example to identify the contribution from different countries to the overall external costs. Spatial differences were shown to be significant in terms of exposure (e.g., Krewitt et al., 2001; Nigge, 2001) although the authors focused on inhalation exposure. Hertwich et al. (1999) found that substance-specific and exposure parameters are more sensitive to the overall exposure assessment result. However, they suggested to explore the informativeness of spatially-resolved models which is also subject of the present study.

As regards the level of accuracy required, it may be obvious that the ambition of an impact assessment methodology operating at the spatial resolution and for the geographical scope outlined above cannot be as high as in a localised impact or risk assessments for instance (Hunsaker et al., 1990). Furthermore, as is discussed in Chapter 9 the assessment endpoint, i.e., the external costs defies its monitoring. Nevertheless, expectation estimates are striven for. Already the present work as such is an improvement towards more knowledge about the magnitude of the external costs occurring due to human activities as hardly any (if at all) information on the external costs for exposure routes other than inhalation had been available prior to this effort. In line with European Commission (1999a), the external costs and the exposure leading to the related impacts will be analysed at the population level, not below (e.g., individuals).

Furthermore, the model development needs to obey the mass conservation principle in order neither to miss nor to fabricate substance amounts. It has to be noted, however, that the air quality model based on which the model development will take place (cf. section 4.1) does not fully comply to this criterion.

The extension of the Impact Pathway Approach involves the four components shown in Fig. 2-1: (a) emission scenarios, (b) environmental fate modelling, (c) exposure and impact assessment, and (d) monetary valuation. The emission scenarios are subject to the cases investigated and are, thus, part of Chapters 10 and 11. Likewise, the monetary valuation will be based on the state-of-the-art suggested by latest ExternE follow-up project(s) (cf. Chapter 8). In contrast, the environmental fate analysis on the one hand and the exposure and impact assessment on the other need to be set in place. In many risk assessments, the suggested schemes and tools do not integrate these two components but follow a modular approach by first performing an analysis of the environmental fate and then assessing the exposure and potentially the impacts (cf. United States - Environmental Protection Agency, 1998, 1999b; International Atomic Energy Agency, 2001; McKone and Enoch, 2002; European Commission, 2003c). The exposure analyses, thereby, usually assess the transfers from the environmental fate media into the exposed organisms such as humans, plants and/or animals by assuming equilibrium conditions (e.g., by employing bioconcentration, bioaccumulation, or root concentration factors). Depending on whether they intend to perform a generic assessment (e.g., International Atomic Energy Agency, 2001; European Commission, 2003c) or a regionalized assessment (e.g., United States - Environmental Protection Agency, 1998, 1999b; McKone and Enoch, 2002), the exposure assessments show different degrees of complexity. This is related to the extent to which conservative assumptions are made or protective purposes are followed.

Due to the fact that the exposure assessments follow similar, equilibrium-based computational approaches, the following section 2.3.1 will focus on the different possibilities how to design an environmental fate model.

2.3.1 Modelling framework

In the following, an overview of different existing modelling approaches is given in order to elaborate which approach is most suited for the present work, concluded in section 2.3.2. The overview is structured into:

- mechanistic versus functional/box models,
- coverage, spatial scope or model extent,
- spatial aspects other than a model's spatial scope, and
- temporal aspects.

The findings influenced the compilation of Table 2-3 which tries to demonstrate in what way properties and release patterns of the substances potentially to be included in the assessment influence the model design.

The left hand side of Table 2-3 describes a chemical's characteristics and release patterns which vary to the indicated degree (e.g., a substance's persistence can vary from absolutely persistent to readily degradable). These features have an impact on the model design, as indicated on the right hand side of the Table (e.g., non-linear dose-response information for a substance brings about the need to assess the absolute concentrations and not just their increases in the environmental medium of concern).

Table 2-3: Attempt to structure the implications of different substance properties, reaction chemistry and modes-of-entry on model design

Substance characteristics and release pattern					Design of environmental fate and human exposure model			
Long range transport	not significant	vs.	significant	⇒	small scale	vs.	large scale	Spatial scope / extent
Chemical mode-of-entry	point source	vs.	multiple point sources ('diffuse emission')	⇒	small to large scale[a]	vs.	large scale	
	non-constant releases (e.g., intermittent pulses, in- or decreasing)	vs.	Continuous and constant emission	⇒	quasi dynamic[b,c]		steady-state	Temporal scope[d]
Persistence	readily degradable	vs.	persistent	⇒	quasi dynamic[e,b]	vs.	steady-state[f]	
Properties changing due to temporally varying conditions[g]	significant	vs.	not significant	⇒	('true') dynamic (if cannot be time averaged, e.g., as for rainfall in steady-state models)		steady-state appropriate, depending on application	

Table 2-3: Attempt to structure the implications of different substance properties, reaction chemistry and modes-of-entry on model design

Substance characteristics and release pattern					Design of environmental fate and human exposure model			
Sorption and reaction behaviour	approximately linear	vs.	non-linear	⇒	linear differential equations	vs.	non-linear differential equations	Formulation
				⇒	no background data needed to estimate marginal changes in concentration or exposure	vs.	background needed to estimate absolute concentrations or exposures	Background[h]
Dose-response relationship	(pseudo) linear (at least above any biological thresholds)	vs.	non-linear	⇒				
Intermedia transfer	negligible	vs.	important	⇒	single compartment[i]	vs.	several (integrated or coupled) compartments	Number of compartments
Media via which most species exposure occurs	medium of release	vs.	another medium or several media	⇒	single compartment[a]	vs.	several (integrated or coupled) compartments	
Effect of reaction products	no concern	vs.	concern	⇒	single species[j]	vs.	multi-species	Speciation or chemical forms

Table 2-3: Attempt to structure the implications of different substance properties, reaction chemistry and modes-of-entry on model design

Substance characteristics and release pattern					Design of environmental fate and human exposure model			
Effect of parent and/or of transformation substance	independent of location	vs.	dependent on location	⇒	zero dimensional	vs.	one to two dimensional	Spatial resolution[k]

a. This depends on the long range transport capabilities of the receiving medium or of the media into which intermedia transfers occur, for example.
b. 'Quasi' denotes that only the concentration of the substance varies in time (cf. Brandes et al., 1996).
c. The relationship between the steady-state solution of a linear Mackay-type multimedia model and the time-integrated exposure assessment of pulse emissions is, however, acknowledged (cf. Heijungs, 1995).
d. Suggestion: decades would be a meaningful temporal scope for today's society when computing dynamically; this could be increased significantly for sustainability considerations and when addressing intergenerational equity.
e. Dynamic approaches are suggested for substances with quick transformation and/or adsorption rates (cf. Mulkey et al., 1993; Wania and Mackay, 1999).
f. In the case of very persistent substances, it may be desirable to at least give an indication of the time horizon for the development towards the steady-state (Cowan et al., 1995a; Trapp and Matthies, 1995), for example by means of level IV calculations in the case of Mackay-type multimedia models ('response time', Mackay, 1991).
g. Like vapour pressure etc.; it shall be noted that also environmental properties or states including target organisms vary in time, potentially requiring the use of 'true' dynamic models termed 'structurally dynamic models' or 'variable parameter models' (Jørgensen and Bendoricchio, 2001, p. 315 and pp. 382ff; see main text for further explanations).
h. If (varying) background concentrations need to be taken into account due to non-linear fate mechanisms or effect measures (cf. sections 4.2.3 and 7.3, respectively), the model's scope needs to be large when not just assessing subsistence farmer exposure scenarios (cf. section 7.2) regardless of whether the substance has only localised sources and is very immobile. Depending on the variability of background concentrations and/or the characteristic travel distance of the respective substance, either a nested model set-up (like SimpleBox version 2.0, cf. Brandes et al., 1996) or a global model (e.g., GLOBOX, Wegener Sleeswijk, 2005) could be used. Furthermore, the background potentially also of reactants and competing substances needs to be included in the assessment.

i. If emission takes place into different compartments, all receiving compartments need to be considered even if no intermedia transfer occurs.

j. If reverse reaction is negligible.

k. 'Lateral spatial resolution' or 'dimensionality' according to van de Meent et al. (1996); see main text for further explanations; the nested approach followed in the SimpleBox model version 2.0 (Brandes et al., 1996) might be classified differently, as the different scales vary in their spatial resolution (note: whether a model has also vertical subdivision, e.g., layers, is not of importance here).

Mechanistic versus functional/box models

Any fate and exposure model makes the assumption of homogeneity[4] in the distinguished elementary spatial units for which balances are computed. The size of those elementary spatial units and, hence, the model formulation is what makes the difference between a mechanistic and a functional or lumped parameter model. In contrast to functional models, mechanistic models are based on rate constants and not on capacities (Hoosbeek and Bryant, 1992). Mechanistic models use ordinary (one independent variable like time) or even partial differential equations (more than one independent variable; e.g., additionally x, y and z location coordinates) and are, hence, relatively more and/ or absolutely highly data demanding. The mechanistic models which use partial differential equations would only be favoured if such a high information density on environmental state variables as well as on emissions could be provided more or less readily. This will presently at best only be the case for very localised emissions with little to no dislocation of the substances of concern (local spatial scope). However, the present work focuses on an impact assessment methodology at the European scale which is why functional models or simple mechanistic models with ordinary differential equations are to be favoured primarily due to environmental and emission data availability reasons. Examples for the latter are the multimedia models of the Mackay-type (e.g., Mackay, 1991). Despite their simplifications, functional models seem likely to be increasingly advantageous also with respect to their performance when the physical scale of the modelling exercise increases (Addiscott, 1993).

Coverage, spatial scope or model extent

Depending on a substance's mobility in and/or its diffuse release into the environment, a fate and exposure model may need to cover up to the whole world (Table 2-3). For instance, mercury has a residence time in air in the order of months to years (Lindqvist and Rodhe, 1985; United States - Environmental Protection Agency, 1997b) in which it could travel around the globe several times. Nevertheless, the appropriate spatial modelling resolution may not only be a function of fate, but also the importance of exposure levels at different locations remote to the source. Also, depending on the available information on where emissions take place which may vary from site-generic over site-dependent to site-specific, the spatial scope of the assessment needs to be adjusted (Organisation for Economic Co-operation and Development, 1999;

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4 Be it just homogeneity in terms of degree of variability or similar stochastic behaviour.

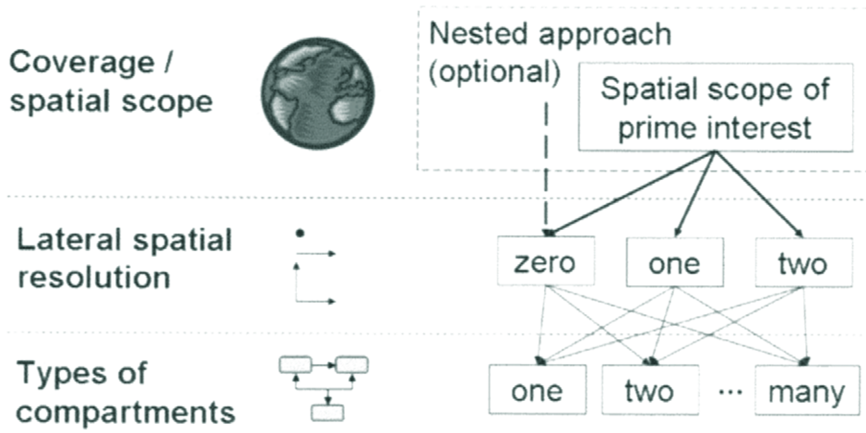

Fig. 2-3: Options for the combination of the spatial scope, lateral spatial resolution and compartmentalisation of an environmental fate (and exposure) model (clipart by Corel Corporation, 1999)

Hauschild and Potting, 2003). For instance, due to the usual lack of spatially (and temporally) resolved Life Cycle Inventory (LCI) data (e.g., Owens, 1997a), generic Life Cycle Impact Assessments should be performed at the global level. Apart from formulating a fully generic model of the whole world, there are principally two ways to take global scale distributions of chemicals into account (cf. Fig. 2-3):

- 'sub-regions interconnected by advection' (Wania, 1996): the total model's scope is divided into adjacent regions (or zones) where all regions have the same level of detail (same hierarchical level). Multimedia model examples for the global scale are the models with meridional zones described in Wania and Mackay (1995) and Scheringer et al. (2000b), and the GLOBOX model (Wegener Sleeswijk, 2005) that subdivides the whole globe by national boundaries. Many atmospheric chemistry and global oceanic models similarly exist, with various levels of complexity and demonstrated validity, and
- 'nested sub-regions' (Wania, 1996): the world is divided into areas with higher and lower levels of detail. The components with higher level of detail are contained in the ones with less details. An early example is the SimpleBox 2.0 model (Brandes et al., 1996) with a global scale represented by an arctic, a tropic and a moderate zone. There is a continental scale nested

in the latter zone, which in turn contains a regional scale. IMPACT 2002 (Pennington et al., 2005) reflects a more recent example, offering the possibility of a spatially-resolved European model nested in an a-spatial global model.

According to van de Meent et al. (1996), the nested approach could be used to combine different types of models (e.g., functional models at the larger scale with mechanistic models at the local scale). This of course depends on whether a chemical is released at only one site or diffusely at many sites and whether background concentrations need to be considered (see Table 2-3). Advantages of nesting even spatially-resolved regional models into a global model include that all the chemical releases are taken into account and that the importance of exposures outside of the modelled region can be estimated.

Both approaches apply to scales below the global scale as well. Whereas type 1 is more data demanding, the nested approach would allow to have a generic, however to some degree spatially-resolved, broad scale environmental fate and exposure model. In the context of representative impact and external cost assessments, this would bring us only a small step closer to the assessment of actual rather than potential impacts. In a comparison of regional distribution models, on the other hand, Märker et al. (2000) stress the demanding task of modelling at intermediate or regional scales. This is due to the fact that heterogeneities at intermediate scales cannot be as appropriately accounted for as at smaller scales since data is most often not available. However, the properties do not average out like at the global scale. The main question resulting from this is whether one wants to model only a portion or the full extent of the model's scope in more detail. This is again dependent on the kind of substance one wants to assess (cf. Table 2-3).

Spatial aspects other than a model's spatial scope

"Different processes and connectivities emerge as dominant as we move from the plot scale to catchment or regional scales" (Kirkby et al., 1996, p. 396) or put the other way around, as one moves from generic assessments such as status quo Life Cycle Analyses (LCAs) to local risk assessments for instance. Upscaling, and in particular downscaling, are problems that one is confronted with when developing a fate and exposure model with different spatial resolutions for a given spatial scope. From a soil science perspective, Wagenet (1998) cited in Addiscott (1998) commented that at different scales different variables are often needed to describe similar processes. Addiscott (1998) reflects further whether different models are needed, too, given the present limitations of our understanding of the processes. In line with this, van de Meent et al. (1996) recommend to use (a-spatial) multimedia box models only for screening purposes. Predictions of effects at specific times and places, on the other hand, may require the use of a more sophisticated dynamic, two or three-dimensional air, water or ground water quality model.

Some authors suggest to use (pseudo) linear formulations with parameters showing little variability when modelling at larger scales (Addiscott, 1993, 1998). Prominent examples of (pseudo) linear models in the area of environmental fate models are the multimedia box models of the Mackay-type (Mackay, 2001) that have already been used in the field of Life Cycle Impact Assessment (LCIA, e.g., Huijbregts et al., 2000b; McKone and Hertwich, 2001; Hertwich et al., 2002; Jolliet et al., 2003). As opposed to these fully integrated multimedia model mathematical solutions, it is also possible to directly link single-medium models (Margni, 2003; an example for a mechanistic very localised model: Whelan et al., 1992; Margni et al., 2004). The usage of single-medium models is also to be favoured if a pollutant does not escape from the medium into which it is exclusively released (van de Meent et al., 1996). Klepper and den Hollander (1999) come to the conclusion that the value of using single-medium models is dependent on the type of medium when dealing with chemicals that are not true multimedia substances ('multi-hop'). Whereas the applied multimedia model gives fair estimates for air and soil, a single-medium spatially-resolved model should be used to assess a substance's concentration in the water compartment in order to improve the assessment (ibid.).

In line with Hertwich et al. (2002), one of the underlying principles guiding these reflections is that the properties of the considered substances highly influence the model design related to spatial aspects in order to meaningfully assess a substance's interaction with the environment, including human beings (see Table 2-3). Here, *spatial aspects* comprise the questions about the spatial scope, the so-called *lateral spatial resolution* (or 'dimensionality' according to van de Meent et al., 1996), as well as the number of environmental compartments to be distinguished. By 'lateral spatial resolution' the different ways how to differentiate a model's geographical scope into zones is meant. Examples are many typical multimedia models without spatial differentiation into zones. A lateral spatial resolution of zero means that there is only one zone distinguished. When this resolution is unity one has to deal with a line or cascade model where one zone follows the other. Example are the GREAT-ER model (European Centre for Ecotoxicology and Toxicology of Chemicals, 1999) and the single-medium water model with sequential water stretches as applied in Trapp et al. (1994). A model with a lateral spatial resolution of two consists of zones that are added one to another to fill the entire area of a model's geographical scope by several zones. Examples are the global model with nine separate climatic zones as described by Wania and Mackay (1995), TRIM (United States - Environmental Protection Agency, 1999a), the POPCYCLING-Baltic model (Wania et al., 2000), BETR (MacLeod et al., 2001), EVn BETR (Prevedouros et al., 2004), IMPACT 2002 (Pennington et al., 2005) and GLOBOX (Wegener Sleeswijk, 2005).

One reason for conducting impact assessments in a spatially-resolved way is that the concentrations of a substance and/or the susceptibilities of target organisms can vary substantially in space. Wania (1996) differentiates between two primary causes why there is spatial variability of chemical concentrations in the environment (beside temporal variations):

- type 1 variability is due to spatial differences in source strength and the inevitable incompleteness of mixing processes; this variability is highest for immobile and reactive chemicals with localised emission patterns and
- type 2 variability is caused by the variability of the environment resulting in different intensities of various fate processes in different locations.

Apart from the release mode, hence, the main criteria for the decision whether and to what degree to model in a spatially-resolved way are a substance's dislocation behaviour and interaction with the environment.

The distinction of these two types of variability was made on the background of environmental fate modelling. For an impact assessment context, it is stressed here that type 2 should be explicitly extended to also comprise exposure, particularly in the case of humans. This variability is caused by the variability in exposure patterns resulting in different intensities of food and water supply on the one hand and population density on the other. Additionally, there might be cases where also the effect side needs to be taken into account when determining the spatial importance in a fate and exposure model. This is especially the case if sensitivities against a poisonous chemical vary substantially, for instance for different varieties of species at different locations or for the same species under different environmental conditions (e.g., temperature, salinity/hardness of waters).

The effect side also raises the question about considering transformation products of an emitted chemical. This is because the transformation product might be more toxic than the parent compound and again the organisms might show different sensitivities towards the transformation product at different locations. The issue of speciation is only taken up in Table 2-3 but will not be explicitly addressed in this work.

Temporal aspects

As regards the temporal aspects, release patterns and substance properties with respect to degradability and other environmental fate behaviour such as volatilisation may play a role in the design of an environmental fate and exposure model (cf. Table 2-3). There are principally two different temporal scopes: steady-state and dynamic.

Steady-state is the situation in which the fluxes into a spatial unit for which a mass balance is calculated equal the fluxes out of it. As a result, the inventory and, thus, the concentration in this balance unit do not change in time. It is the final situation which would occur if for example a society or a human activity proceeded to emit a substance into the environment at a given level. To assess this situation may be relevant for sustainability-related questions (e.g., European Commission, 2003d). The occurrence of a steady-state may be assumed for non-short-lived substances (Wania and Mackay, 1999) and when the assessment is applied for a short time period (Mackay, 1991). Another application is related to the assessment of pulse emissions in the context of Life Cycle Impact Assessments of hazardous substances. Heijungs (1995) has shown that the steady-state solution of a (linear) multimedia model can also help to assess the time-integrated exposure to pulse emissions under certain conditions which does not require dynamic computations. One has to note that the development towards a steady-state may take a considerable amount of time (e.g., several hundreds or even thousands of years in the case of metals, van den Bergh et al., 2000; Huijbregts et al., 2001; de Vries et al., 2004) given the potentially very long residence times of, for example, trace elements in soils (Alloway et al., 1996), also implying that initial concentrations do not play a role any more (only fluxes, not stocks are relevant). It may, therefore, be desirable to at least give an indication of the time horizon for the development to the steady-state (Cowan et al., 1995a; Trapp and Matthies, 1995) especially in the case of very persistent substances such as non-radioactive elements. This leads to the necessity for dynamic approaches. In the case of Mackay multimedia models, so-called level IV calculations constitute a means in order to provide such response times (Mackay, 1991).

Dynamic calculations should be favoured in cases in which (a) releases of substances are discontinuous or not constant (except for exposure assessments of constant pulse emissions, see above), (b) substances are dealt with whose fate is largely controlled by the transformation or sorption rates (Mulkey et al., 1993; Wania and Mackay, 1999) and/or (c) whose properties vary substantially in time (e.g., due to diurnal or seasonal temperature changes). One has to note that not only substance properties may change in time. Brandes et al. (1996) introduce the term 'quasi-dynamic' for calculations that only allow for the change of substance masses in the model while all other parameters are kept constant. If the dynamic behaviour also of environmental parameters shall be considered a 'true dynamic' model needs to be employed. Depending on the degree of variations in the considered parameters, such models may be classified as 'structurally dynamic models' or 'variable parameter models' (Jørgensen and Bendoricchio, 2001, p. 315

and pp. 382ff). Those models account for the evolutionary potential of ecosystems which is stochastic in nature. Stochastic processes not only play a role for organisms but can also be encountered in less complex model situations such as describing solute movements in soils (cf. Richter et al., 1996, pp. 6f).

Depending on the dynamics of the phenomena as such and also the spatial scale, different *temporal resolutions* need to be employed for dynamic approaches. These may range from below a day to annual time steps. When trying to cover a rather large geographical area with some degree of spatial resolution (e.g., regional), data availability is critical both in space and time. For geochemical processes, Drever (1997b) notes that "it is rarely possible to construct a meaningful catchment budget for a time-scale of less than a year" (p. 241). Water balance models operating at intermediate spatial scales usually operate at monthly to annual time scales ('seasonal scale', Blöschl, 1996) when not dealing with events such as floods. An annual temporal resolution is also used for climatological air quality models (e.g., Trukenmüller, 1998).

Depending on how detailed the impact assessment shall be conducted, also temporal aspects of the effect might need to be included. For instance, climatological models are not well suited to assess impacts on different development stages of an organism each of which might show different susceptibilities towards the chemical under study. This depends on many factors, including the release pattern of the chemical.

2.3.2 Conclusion with respect to the modelling framework

Beside the criteria mentioned in the introduction to this sub-chapter (2.3), i.e., striving for transparency, consistency and central estimates, assessing impacts at the population level, and following the mass conservation principle, several decisions with respect to the design of the environmental fate model need to be made. These concern:

- the modelling approach (e.g., capacity, mechanistic, stochastic, lumped parameter models),
- the geographical scope of the model and how this is subdivided ('spatial resolution' with respect to zones and/or land uses/compartments), and
- the temporal resolution.

A few of the various options presented in the previous sections are already decided upon. This is because the present work builds on an existing methodological framework (the Impact Pathway Approach, cf. section 2.2) with a suggested software tool (EcoSense, European Commission, 1999a). As a result, the geographical scope should be the same mostly covering Europe as presented in Fig. B-1. The Impact Pathway Approach, furthermore, constitutes a fairly detailed, site-dependent or bottom-up approach to evaluate human exposure towards contaminants. This suggests to use a rather high spatial differentiation of the geographical scope of the model, rather than employing a site-generic modelling approach. Finally, the temporal scale of the air quality model to which the soil and water model is to be connected is one year using annual average data (Trukenmüller, 1998). Another advantage of using long-term average data and annual time steps is that it facilitates the assessment of the steady-state situation. This is relevant for time-integrated exposure assessments of pulse emissions (Heijungs, 1995) and sustainability analyses of constant and continuous releases to the environment (European Commission, 2003d).

The remaining degrees of freedom are, therefore, the specific way how to spatially differentiate (which will be dealt with in sections 4.3 and 5.1) and the general modelling approach. For consistency reasons, functional models or simple mechanistic models with ordinary differential equations are to be favoured. This is primarily due to environmental and emission data availability reasons at the geographical scope at which the EcoSense model operates to which its extension shall comply. Another reason might be that these model types are increasingly advantageous for larger scale modelling (Addiscott, 1993). Examples are the multimedia models of the Mackay-type (e.g., Mackay, 1991). These have several advantages, amongst others:

- the models are "well suited for predicting average regional concentrations resulting from highly dispersed and diffused sources" (Cowan et al., 1995b, p. x); noting their use limitation to screening exposure assessments if implemented without spatial and temporal differentiation (van de Meent et al., 1996),
- the "intermediate effort and reasonable accuracy" (Tolle et al., 2001, abstract) of multimedia fate models make them well suited for Life Cycle Impact Assessments "involving comparative assertions or governmental policy decisions" (ibid.), and
- when developing the soil and water model according to this modelling approach, a potential extension towards a fully integrated multimedia model is possible in the future.

The multimedia modelling approach of the Mackay-type is, therefore, adopted here.