12 Concluding remarks

In the ExternE project series on external costs of energy, exposures and resulting impacts through contaminants present only in air were assessed and valued. In order to perform the external cost assessment in as complete a way as possible, this work has developed and applied an extension of the Impact Pathway Approach (IPA) termed WATSON ('integrated WATer and SOil environmental fate, exposure and impact assessment model of Noxious substances'). WATSON facilitates the coverage of exposures towards hazardous substances released into air, (fresh) water and soil through ingestion of various food items in a spatially-resolved pan-European setting.

12.1 The assessment framework

The approach comprises several special features that shall be recapitulated in the following.

The overall method relies on a coupled set of environmental fate models for air on the one hand and for soil and (fresh) water on the other. While the assessment for air has been adopted from the existing software tool EcoSense (European Commission, 2003d), the contaminants' environmental fate in the terrestrial and aquatic environment is described with the help of a spatially-resolved climatological box model similar to Mackay level III/IV models (Mackay, 1991). Both fate models assume long-term average conditions for the description of the environment. In line with current political concern, the focus is laid on persistent substances such as heavy metals. In particular, the trace elements arsenic, cadmium, chromium and lead are investigated. The methodological development has, therefore, focused on this substance group which had been poorly addressed previously in the realm of multimedia modelling.

Spatial aspects

The geographical coverage and degree of spatial resolution allow for a spatially-dependent assessment of the trace elements as recommended amongst other by Huijbregts (2000) and Huijbregts et al. (2001). This is also in line with for example Potting and Hauschild (1997), Krewitt et al. (2001) and MacLeod et al. (2004) in that a spatially explicit assessment is deemed more credible and informative than an a-spatial or site-generic assessment. This allows the analysis to take account of the spatial variability in landscape characteristics, food production intensities and human population densities. Furthermore, it allows the assessment to be conducted following a bottom-up approach, a main feature of the Impact Pathway Analysis.

Different spatial resolutions are employed for the air compartment (regular grid), the soil and water environment (according to catchment information), and the exposure part (according to administrative units at least at the country level), thereby taking account of the differences in terms of chemical movements in the environment and data availability with respect to exposure based on production data. Also, if trade of food is geographically constrained at all it mostly follows administrative borders. Emissions into air, water and soil are specified at least according to administrative units if not even additionally by emission sector.

Usually multi-zonal multimedia models at most distinguish the terrestrial environment into agricultural/cultivated and non-agricultural/non-cultivated land (Wania et al., 2000; Wania, 2003) whereas the presented approach may differentiate up to six terrestrial compartments (section 5.1.1). In particular the distinction of pastures from arable land appears to influence the overall result when exposure through cattle is important. If in addition the exposure through freshwater fish contributes substantially to ingestion exposures, the separate modelling of sealed surfaces also appears advisable, at least if emissions into air are investigated. According to the way these are described within the model, the impervious surfaces accelerate the transport of substances to freshwater bodies. However, freshwater fish exposure is rather irrelevant for all of the investigated trace elements for the emission scenarios analysed.

Processes and environmental characteristics considered

As regards fish exposures, a distinction between rivers and lakes is made. In particular, a physically meaningful distinction between these two freshwater body types has been introduced in terms of the dynamics of (suspended) particles. The importance of this model development has been demonstrated to increase the exposure through freshwater fish by almost one order of magnitude when spatially differentiating below the catchment level, in contrast to the treatment of all freshwater bodies as lakes as done by state-of-the-art multimedia models. A further need for improvement of the process formulation has been identified.

The terrestrial compartments are distinguished according to different land uses showing rather different susceptibilities especially to the process of water soil erosion. At least when distinguishing arable land, pastures and other soils, the water soil erosion rate has been identified to be very sensitive with respect to the exposure estimates of rather immobile substances. It could be shown for chromium that the variation of the water soil erosion rate influences the overall ingestion exposure situation even more significantly than substance-specific parameters such as the solid-water partitioning coefficient, bioconcentration factors and biotransfer factors. This finding is in contradiction to previous uncertainty and sensitivity analyses conducted with respect to exposure estimates (e.g., Hertwich et al., 1999; Huijbregts et al., 2000a) that found that environmental parameters are rather insensitive. However, their focus was on degradable substances. Recent evidence supporting the importance of the water soil erosion process with respect to exposure estimates is given by Huijbregts et al. (2003). Although the water soil erosion process has been improved towards existing multimedia models in that it is allowed to vary by compartments, further development is needed. A possible approach is suggested in section B.5.3.

In line with the finding of the relevance of the water soil erosion process, it may be worthwhile to consider to include the process of wind soil erosion. Although it has been shown to be rather irrelevant for the environmental fate of selected semi-volatile persistent organic chemicals, i.e., PCDD/Fs, by Suzuki et al. (2000), the wind soil erosion / suspension process may be of importance. Suzuki et al. (2000) did not analyse the influence of this process in a multi-zonal context where spatial variability for example in terms of areas potentially prone to this process and distribution of food production are taken into account. Furthermore, the case may be different especially for non-degrading and rather involatile substances such as most metals (e.g., Guerzoni and Chester, 1996). However, the coupling of the air quality model (WTM) on the one hand and the soil and water environmental fate model (WATSON) on the other requires many subsequent iterative runs of both models in order to consider this process within the presented methodological approach. Furthermore, both models are mostly confined to Europe in terms of their spatial coverage. It is known that even particle-bound trace elements can undergo long-range transport between continents upon resuspension (Church et al., 1990). Thus, further developments need to be done to set up a fully integrated multimedia model with a potentially larger geographical scope that at least covers the northern hemisphere when trying to fully follow the impact pathway of hazardous substances (United States - Environmental Protection Agency, 1997b; Ryaboshapko et al., 1998; Pekar et al., 1999).

The environmental fate model developed is the first one in the realm of multimedia models to include the process of preferential flow. Its influence could clearly be demonstrated, potentially leading to a reduction in food exposures by 50 % when agricultural produce dominates the overall ingestion exposures. A process that also leads to accelerated transport especially of hydrophobic substances to the subsurface has been introduced by McLachlan et al. (2002) which is termed 'sorbed phase transport'. It may be questioned whether the processes such as 'bioturbation', 'cryoturbation' and 'erosion into cracks formed by soil drying' that are held responsible for the sorbed phase transport (McLachlan et al., 2002) are really the main processes driving the downward transport especially of lipophilic substances. Hartmann et al. (2004) have shown that even hydrophobic substances like tributyltin (TBT) reach the zone below the top soils via preferential flow when applied to soils, suggesting that this transport that also affects colloidally bound substances may be another or even the most important candidate for explaining the movement of rather insoluble substances to the subsurface. According to Flury (1996), this process is more the rule than the exception and may have different causes (Wittig et al., 1985; Helling and Gish, 1991; Steenhuis and Parlange, 1991; Schwarz and Kaupenjohann, 2000) including colloidal transport (Jarvis et al., 1999; Noack et al., 2000). It even also applies to atmospheric deposition in forests (Wittig et al., 1985; Chang and Matzner, 2000).

Harvest removal of substances entrained in food items is considered in the environmental fate model. This is in line with Severinsen and Jager (1998) and TRIM.FaTE (United States - Environmental Protection Agency, 2002b) for aboveground plant parts. In the presented approach, additionally harvest removal of belowground produce and 'catch removal' of freshwater fish are included. In particular with respect to plants, the removal is modelled only for the edible portions. The other plant parts are not explicitly addressed. This implicitly leads to the assumption that the non-edible plant parts are added after harvest to those soils on which the produce was grown. The question, however, arises where the trace elements removed from the environment by harvest would re-enter it, for example after being released again from or by the human body. Some portions will reach the subsurface from cemeteries or enter the sewer systems from households. From the sewer system, the substances may reach surface water bodies when entrained in the effluent and/or may be trapped in the sewage sludge and could subsequently be applied to agricultural soils. Not allowing for the harvest removal processes would mean that the trace element amounts present in the respective compartments stay in these compartments and may contribute to exposure in subsequent time steps for which the assessment is conducted. Thereby, the substances may potentially be double-counted at two or more rather close points in time. Thus, the harvest removal processes prevent the double-counting of exposure towards substances for example within a generation while disregarding potential re-exposure of future generations. This may be compensated to some degree by the inclusion of discharges to soil and water if related information was available to the extent necessary. Owing to the substantial contribution of intercepted atmospheric depositions to human exposure, however, the inclusion of harvest removals does not lead to substantial reductions in the human exposure situation when air emissions are analysed, as could be shown in a scenario analysis (section 9.3.3). This exposure pathway may tend to be overestimated because rinsing of aboveground exposed produce as part of a regular kitchen practice is not explicitly taken into account. However, the ingestion exposure assessment only includes one food item of this group of produce, i.e., spinach. Due to the incomplete assessment in terms of other produce that are classified as aboveground exposed produce such as many green salads and fruits, the contribution of this produce group is, therefore, not deemed to be overestimated. It needs to be noted, however, that the exposure at the individual level and maybe even at the national level may, nevertheless, be affected due to differing preferences in terms of 'aboveground exposed produce' consumption by different people.

The inclusion of harvest removals has implications on when exposures and impacts are to be assessed. One may, therefore, consider not including these harvest removals from a Life Cycle Impact Assessment point of view where temporal information especially on emissions usually are not available or when zero-discounting is performed and one tries to cover all potential effects. However, whether the additional exposures would then be assessed in a correct way due to the missing consideration of redirected substances is another open field for discussion.

The environmental fate model takes pH-dependent or organic carbon-dependent partitioning of contaminants into account (section C.1.1). Furthermore, the derivation of the dimensionless Henry's law constant is also modelled in a temperature-dependent way (section C.1.2). A scenario analysis of either letting the solid-water partitioning coefficient depend on a zonally variable pH or one that varies only by compartment has shown that this new element in a multimedia model is influential by increasing the time-integrated effective Intake Fraction due to ingestion of chromium by more than 80 % (cf. section 9.3.4). This influence may even be more significant if solid-water partitioning coefficients became available that are not only given for example for three different pH values as provided by United States - Environmental Protection Agency (1998) but additionally on organic matter or clay content, for example.

Exposure assessment

The proposed methodological framework takes exposures through inhalation and food ingestion into account. The assessment of the latter is more complex due to both the variety of food items to which human beings might be exposed and the spatial distribution of the food production. The estimation of ingestion-related exposures builds on the site-specific risk assessment approach recommended by the US-EPA for hazardous waste combustion facilities (United States - Environmental Protection Agency, 1998), thereby striving for representative rather than for protective estimates. As noted above, the exposure is assessed at the level of administrative units for which food production and population data are usually provided. Unlike other multimedia exposure models, human exposure towards food items is assessed according to nationally variable food supply figures. A correction is included that considers losses not taken into account by this statistical measure.

Unlike most other multimedia exposure models, the food items are assumed to undergo trade within the geographical scope of the assessment framework which can be regarded an extension of the environmental fate of the investigated contaminants. This leads to homogeneous contaminant levels in all traded goods prior to consumption by humans and/or farm animals. By means of this homogenization the actual amounts of food produced at different administrative levels which may be as low as at the municipality level are taken into account in order to weigh the assessed contaminant concentrations. In order not to overestimate exposures towards European residents, the self supply of the respective goods is, furthermore, taken into account also in order to follow the mass conservation principle. However, principally bilateral information between countries is ideally needed in order to more appropriately consider the influence of trade. One such information source is the comprehensive and commercially available United Nations Commodity Trade Statistics Database (UN Comtrade).

The exposure of farm animals via feed and ingestion of soil particles requires very detailed information on the way the animals are kept. In particular the extent to which the animals are kept outdoors will significantly influence their exposure via soil ingestion towards those pollutants that do not directly contaminate the feeding stuff, i.e., that are dispersedly released. For instance, the ingestion of soil particles by cattle may contribute more than 50 % to the human exposure towards chromium stemming from particular areas according to the present assessment.

The originally published concept of the Intake Fraction is principally adopted as the exposure measure and extended in several ways. As is also done by other authors, the Intake Fraction is distinguished according to routes of exposure, i.e., inhalation and ingestion. For ingestion exposures, this measure is further differentiated according to different exposure pathways. Second, the Intake Fraction is confined to effective exposures by only taking into account the chemical form of a contaminant that may cause an adverse effect. Third, the effective Intake Fraction especially for ingestion exposures may be given for different time horizons and not just at steady-state as initially proposed by Bennett et al. (2002). This is particularly desirable when analysing rather persistent contaminants in a policy decision support context in order to give an indication of the time horizons potentially involved. Fourth, this measure may be provided for different nations and/or sub-populations allowing among other for the assessment of imports and exports of contaminants between countries as done by Droste-Franke and Friedrich (2003). As a result, the differentiation of the Intake Fraction allows a better representation of population-based exposure situations in time and space.

Impact assessment

The most limiting information with respect to the complete assessment according to the Impact Pathway Approach is that related to effects following ingestion exposures. Due to the lack of existing epidemiologically derived effect information, use is made of the approach proposed by Crettaz (2000), Crettaz et al. (2002) and Pennington et al. (2002). This derives a linear so-called β_{ED10} slope factor that represents a measure for the population-averaged excess individual risk of an effect per unit daily dose for a lifetime exposure. The linearisation is based on a non-threshold assumption. For consistency reasons, the approach is also applied to inhalation exposures.

Generally, the approach makes use of rather different effect measures in order to derive a β_{ED10} slope factor (cf. section 7.3.1). The related uncertainty in the derived dose-effect model depends amongst other on whether it is derived from dose-response information such as *slope factors* or *unit (lifetime) risks* in US-EPA (United States - Environmental Protection Agency 1996b) or WHO terminology (World Health Organisation, 2000b), respectively, or from threshold effect measures such as NOAEL or LOAEL. This emphasizes the importance of reliable effect information for which a very high need for further research is identified and recommended. More reliable dose-response information such as that based on epidemiological evidence may become available in the near future for the trace elements investigated through ongoing projects such as the EC-funded ESPREME project (Estimation of willingness-to-pay to reduce risks of exposure to heavy metals and cost-benefit analysis for reducing heavy metals occurrence in Europe, contract number: 502527).

In order to arrive at impacts, the approach proposed by Crettaz (2000), Crettaz et al. (2002) and Pennington et al. (2002) employs the Disability Adjusted

Life Year (DALY) concept endorsed by the WHO for considering different severities of the assessed effects. This severity measure is also adopted in the presented methodological framework based on most recent findings (Keller, 2005). It has the advantage of aggregating morbidity and (premature) mortality impacts as equivalents of Years Of Life Lost (YOLLs). This allows the monetary valuation to be performed as commonly done within the ExternE project series. A weak point of the adopted DALY values is the treatment of morbidity which relies on rather uncertain disability weights (cf. section 7.3). Also, the assessment of non-cancer effects which are derived from cancer-related average DALYs is based on a rough classification of these diseases. Consideration may, therefore, be given to look for alternatives with respect to this measure of severity of human health damages and to combine these with the prioritised dose-response information. However, the DALY approach is still deemed a step towards a more differentiated assessment of cancers for whose valuation only one generic monetary value for any type of cancer is used according to the latest ExternE methodology (European Commission, 2004).

Valuation

Due to the very long time horizons involved in the ingestion exposures towards the trace elements investigated, a new discounting scheme is proposed that takes the issue of intergenerational equity into account. It is termed *intergenerationally equal, positive personal discounting*. It combines intergenerational equity by assuming a discount rate of zero between generations with the observation that each individual shows a positive pure time preference. As is argued in section 8.2.4, this discounting scheme yields external costs that are about half of those when constantly discounting at a rate of 0 %.

Implementation

A special feature is also the way in which the methodological approach for the assessment of ingestion exposures and their valuation is implemented. In contrast to the majority of multimedia fate and exposure models which are implemented as spreadsheet models, for instance EUSES (Vermeire et al., 1997), USES-LCA (Huijbregts, 1999), CalTOX (McKone, 1993b) and IMPACT 2000 (Pennington et al., 2005), the software tool WATSON is coded in C++ (MS Visual Studio 6.0) and uses a LINUX-based Oracle database version 8.1.6i. This facilitates a flexible definition of process formulations and combinations as well as the use of different environmental settings allowing the modelling of different substances (e.g., Trapp and Schwartz, 2000). Unlike many existing multimedia models, WATSON's mass balance is based on concentrations (like SimpleBox, Brandes et al., 1996). The software tool also offers the possibility of running in different modes of operation for the investigation for example of continuous and constant emissions over time and at steady-state or of a pulse emission and its consequences in terms of exposure over different time horizons. Storing the data in a database means that the data are kept separately from the simulation code (Robinson, 1999) which facilitates their changeability (Veerkamp and Wolff, 1996) at the expense of computation time however.

12.2 General limitations of the assessment

Although comprising many innovations and providing a reliable exposure and impact assessment approach, the main constraints with respect to the presented methodological framework shall be noted.

The methodology developed is at present limited with respect to substances that are to some extent volatile, i.e., only substances with a low vapour pressure can be assessed.

The assessment of ingestion exposures is incomplete. In particular, no exposure through seafood, drinking water or soil ingestion by humans is at present included in the assessment. Furthermore, several crops classified as aboveground exposed produce are missing. Exposure pathways related to dermal contact, occurring at the working place or through consumer products are also not taken into account due to the assessment being conducted at the regional scale (see next point) and/or for disperse emission sources. This should not be interpreted as implying that transfers from other environmental media through these or other pathways are unimportant.

Exposures occurring in a very localised area or only during short episodes cannot be addressed adequately. The spatial and temporal resolution of the environmental fate model does not allow such localised or temporary exposure assessments to be carried out. This means 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).

Due to the use of long-term average data for the description of the environment, computations with time steps that are not full years do not give adequate results in terms of meaningful values for the eventually desired period of time (e.g., seasonal, monthly, daily variations).

Especially with respect to the long time frames involved, until a certain percentage of the steady-state concentration is reached, such long-term analyses face the problem of justifying the assumption of constant long-term environmental and societal conditions (e.g., hydrological cycle, pH, population) which are subject to changes such as climate change, acid rain and shifts in birth and death rates. However, trying to predict these changes is also highly speculative.

As discussed in section 2.3, substances with rather short residence times in the environment require dynamic modelling approaches that do not rely on long-term average conditions. Thus, such substances should be assessed cautiously with the help of the presented approach, if at all.

The quantification of impacts and external costs are related to impacts on human health. These are assessed for the stated contaminants. Thus, impacts of other pollutants and on other receptors such as biodiversity still need to be assessed in order for the analysis to be as comprehensive as desired for a well-informed policy decision-making.

A problem especially related to naturally occurring contaminants is that background levels may need to be taken into account. This is particularly the case if there are non-linear relationships considered either in the environmental fate, exposure or impact assessment which is not the case at present. Not only background levels would be required when there are non-linearities considered but also past and present emission information. Information on present releases of contaminants into water and soil but also of past emissions into all media is incomplete so that for instance local hot-spots due to metal mining and processing, sewage sludge and fertilizer amendments, and local depositions on roadside soils can hardly be included in the assessments, partly owing to the regional spatial resolution. As just indicated, information on releases to soils or waters in particular are identified as not yet existing for the spatial scope required.

Although the present modelling tries to take into account the persistent nature of the trace elements for instance by the distinction of several compartments differing in the rate of advective flows and by the consideration of their pH-dependent partition behaviour and, therefore, comprises a step forward in large scale trace element assessments, it could potentially be improved for example by the inclusion of speciation apart from considering only the effective fractions of the pollutants in food items and air by means of the effective Intake Fraction. However, this is not feasible at the spatial scale needed for national or even pan-European assessments especially due to data availability constraints with respect to transformation rates and concentrations of reaction partners or competing ions in the respective media.

Inactivation processes such as irreversible binding are not explicitly considered but deemed to be part of the solid-water partitioning coefficient that is recommended to be provided for aged samples.

The geographical scope to which the methodology can be applied at present is confined to Europe. However, the model framework exists and 'merely' needs some data acquisition and processing in order to apply it to other parts of the world. This extension of the model may even lead to a globally applicable tool necessitating an air quality model that operates at the same spatial scale (e.g., Leip and Lammel, 2004; Munthe and Palm, 2003).

12.3 Application of the assessment framework

The application of the methodological framework suggests the following conclusions.

12.3.1 Case studies

Different scenarios have been investigated addressing emissions into air from single facilities or for the whole of Europe.

Generally, exposure via staple food items such as cereals and to a lesser extent potatoes contributes substantially to the ingestion exposures of all trace elements analysed. Dairy products are additionally important for chromium and even more so for arsenic. Beef only constitutes a significant share of chromium's Intake Fraction in the long run while freshwater fish and other animal products such as pork, poultry and eggs contribute insignificantly. In line with the findings in European Commission (2000b) for cadmium and arsenic, the inhalation pathway only contributes marginally to the overall human exposure.

The investigation of the development of the human exposure via ingestion towards a one year pulse emission has revealed that not only the total amounts taken in increase slowly over time[33] but also that the food items contributing most to the time-integrated Intake Fraction vary over different time horizons. This variation may even lead to entirely different patterns in terms of the dominating food items at different integration times. For instance, the long-term exposure towards arsenic is dominated by dairy products (about 80 %) while in the short term the composition of the ingestion exposure is shared between cereals, dairy products, potatoes and spinach. Also, the exposure through aboveground exposed produce such as spinach contributes more substantially in the near future after the pulse emissions occurred while its contribution to the effective Intake Fraction may be insubstantial in the long run. The contribution of aboveground exposed produce through interception of atmospheric deposition, thus, appears to be similar to inhalation-related exposures in terms of its intertemporal significance for human exposure towards pulse emissions. Expressed inversely, exposures through the

[33] For the trace elements with slower dynamics, i.e., arsenic and chromium, less than 2 % of the fraction that leads to exposure may have reached the human population after 100 years upon a one-year pulse emission according to the present assessment.

media soil and water are most dominant in the intermediate to long term for very persistent substances although for cadmium the contribution of the ingestion pathways to the overall human exposure is already substantial in the short term. Exposures towards very persistent substances through the media soil and water are, therefore, highly relevant with respect to sustainable development especially in terms of intergenerational equity.

The dominance of the ingestion exposures is also reflected in the external costs. Inhalation-related annual external costs according to the investigated emission scenarios are negligible when compared to the related ingestion exposures when discounting at a rate of 0 %. The damage factors due to ingestion are substantially smaller when non-zero discounting is performed, owing to the long-lived nature of these pollutants (cf. Hellweg, 2000; van den Bergh et al., 2000; Huijbregts et al., 2001; de Vries et al., 2004) and associated slow dynamics. Depending on the dynamics of the respective pollutant, the effect is more (e.g., arsenic) or less (e.g., cadmium) pronounced. However, discounting the damages following ingestion exposures with up to a discount rate of 3 % does not change the ranking of the inhalation and ingestion exposures in terms of their contribution to the aggregated impact.

It has been postulated by Spadaro and Rabl (2004) that site-dependency of releases are almost irrelevant for ingestion exposures. However, it could be demonstrated that the variation in terms of damages due to ingestion between sites is similar to the inhalation-caused damages despite the homogenizing effect on food concentrations caused by the present representation of trade. Furthermore, it has been found that the impact of discounting depends on the emission scenario analysed. This can be attributed to different dynamics for the metal to reach the human population from different release sites. This stresses once more that, although trade may lead to homogeneous levels in the food items under consideration, it does not mean that the site of release is almost irrelevant. Thus, employing different discounting schemes may cause comparable damages from different release sites to become distinct. The difference is expected to increase if trade is modelled according to bilateral trade information (see above).

12.3.2 Remarks on the magnitude of the external costs

The damage factors derived in Chapters 10 and 11 have been put into perspective against those for the classical air pollutants and for previously reported trace elements to the extent available. Inhalation-related quantifiable annual external costs are negligible (i.e., four orders of magnitude smaller) according to the pan-European emission scenario for 1990 of the investigated trace elements when compared to an emission scenario for the classical air pollutants of the same year. The consideration of ingestion exposures of these pollutants, however, could lead to an increase in the total quantifiable external costs at most by between 0.4 % and in the order of some 10 % when discounting at a rate of 3 % and 0 %, respectively, according to the presently available information. In this respect, effects due to elevated blood pressure caused by lead are most contributional. A similar finding could also be demonstrated for ingestion exposures to dioxins and dioxin-like substances in selected countries (Droste-Franke et al., 2003), amounting to a few percent when employing a discount rate of 3 %.

The direct use of the damage factors and (annual) external costs as estimated in this study must be performed with caution. As demonstrated in section 9.3, the damage estimates via ingestion exposure may vary by about a factor of two depending on different representations of the environment, particularly when considering the process of preferential flow. Ingestion exposure estimates are still more sensitive towards single parameters such as substance-specific solid-water partitioning coefficients, bioconcentration factors and biotransfer factors, and substance-independent water soil erosion rates. As discussed in section 9.3.1, however, the dose- or rather concentration-response information employed is considered one of the most important component of the analysis in terms of uncertainty emphasizing the importance of reliable effect information. The disregard of chemical speciation constitutes another major source of uncertainty whose impact has not been quantified.

Another problem lies in the incomplete knowledge about the effect-causing characteristic of particulate matter. In general, there is a discussion going on whether the particle size or its composition is (more) responsible for the observed adverse effects. Apparently, "(e)vidence is accumulating that metals may have a role in any toxic response to complex air particles" (Godleski et al., 2000, p. 74). However, also other constituents of particulate matter such as sulphates are considered to be responsible for its toxic effect (Gordon et al., 2000). In any case, care must be taken not to double-count the effects towards particle-associated metals via inhalation. While the effects assessed to occur due to inhalation exposure towards particulate matter are not related to carcinogenesis (cf. e.g. Friedrich and Bickel, 2001a), most of the inhalation-induced effects estimated in the present study are (cf. Tables 7-6 and 7-7). This means that the damage factors derived for the trace elements considered in this study can be added to those for particles due to different effect endpoints. Non-cancer effect information is only used for chromium. However, the quantified, non-cancer related damages via inhalation of hexavalent chromium are rather negligible (Tables 10-6 and 11-3).

12.3.3 Quantitative evaluation of predicted concentrations

A comparison has been conducted of the predicted concentrations according to a 100 year continuous release of the trace elements into air according to the pan-European emission scenario with reported concentrations. It could be demonstrated that the results of the present assessment are almost exclusively within or below the ranges of expected values.

A scenario analysis and a sensitivity analysis of the parameters was conducted in order to evaluate the novel components of the assessment framework such as the compartments distinguished in this work, the pH-dependent partitioning of trace elements, and improved or newly introduced processes (e.g., preferential flow, harvest removals, riverine suspended particle dynamics and compartment-dependent water soil erosion). It could be demonstrated that all of these components are influential on the overall human exposure. These analyses have, furthermore, revealed that the influence of the environmental fate and exposure parameters and/or settings mostly shows in the intermediate to long run. Thus, when analysing one year pulse emission scenarios and performing (regular) non-zero discounting, most of the uncertainties related to these model components become largely irrelevant.

12.4 Applicability of the approach to other contexts

In Chapters 10 and 11, more detailed results have been shown at different levels of the assessment according to the Impact Pathway Approach which may serve different purposes. For instance, the indicator *Intake Fraction* may be used in the context of risk analysis or Life Cycle Impact Assessment (LCIA) as done by Bennett et al. (2002), Bodnar et al. (2002) and Evans et al. (2002) among others. Similarly, the indicator Disability Adjusted Life Years (DALYs) has also been applied in LCIAs in order to characterize a contaminant with respect to an impact indicator ('characterization factor', e.g., Goedkoop et al., 1998; Hofstetter, 1998; Crettaz, 2000; Crettaz et al., 2002; Pennington et al., 2002; Jolliet et al., 2003) although some reservations exist with respect to its use due to the inclusion of value choices (Krewitt et al., 2002, see also section 8.2.3). In many cases, the analysed energy-related emissions lead to the highest impacts within a life cycle study with respect to the toxicity impact category (e.g., Beck et al., 2000; Saouter et al., 2002). Due to the model development undertaken, the presented LCIA-related metrics for trace elements typically released by energy conversion processes constitute a step towards a better assessment of these contaminants within Life Cycle Analyses (LCA). The use of the DALY concept also offers the possibility to compare the assessed impacts to health reports issued by the WHO (e.g., World Health Organisation, 2002) although noting that the DALYs used by the WHO most likely will be derived considering age-weighting and discounting, rendering a direct comparison of the numbers less readily feasible.

The valued impacts may, furthermore, be used in contexts where cost-benefit analyses (CBAs) are conducted. Owing to the monetisation of external effects that are not taken into account when deriving private costs of profit-oriented enterprises, such analyses are typically conducted by public bodies especially at the national or the supranational level. National accounting frameworks that take environmental aspects explicitly into account are one such example (cf. European Commission, 2003d).

12.5 Outlook and closure

Previous prioritisations within the ExternE project series especially of the classical air pollutants, thus, appear justified in the sense that the selected substances have high damage factors and are emitted in rather large quantities. However, the amount of contaminants added by the present work to the list of substances for which external costs can be estimated is small compared to those that remain unaddressed. Their monetised impacts may add up to substantial shares of the so far quantifiable external costs. The main obstacle on the way to a larger substance coverage is seen in the lack of effect information in terms of dose- or exposure-response functions. The appropriate consideration of hydrophobic and (semi-) volatile substances for instance may bring about the need to develop a fully integrated spatially-resolved multimedia model which includes an air compartment and potentially even a plant compartment (Simonich and Hites, 1994; Wagrowski and Hites, 1997; Bennett et al., 1998; McLachlan and Horstmann, 1998; Severinsen and Jager, 1998; Cousins and Mackay, 2001). Furthermore, many substances of public concern are rather long-lived in air and, therefore, require at least hemispheric if not global models for their appropriate assessment (United States - Environmental Protection Agency, 1997b; Ryaboshapko et al., 1998; Pekar et al., 1999). Another issue is that mercury for example is not only long-lived in air but human exposure towards its methylated and most toxic species mostly occurs through fish and especially marine fish (United Nations Environment Programme, 2002). A cost-efficient abatement strategy for this heavy metal which is desirable for setting a strategy on mercury at the EU level[34] for instance would require a link between emissions and exposures that consequently not only comprises the whole medium air but also the marine environment including the organisms relevant in the final human exposure towards mercury. This involves not

[34] See e.g. http://europa.eu.int/comm/environment/chemicals/mercury/index.htm as of August 2005

only transport of mercury according to wind or water currents but also entrained in migrating fish and finally trade (United Nations Environment Programme, 2002). Such a global model on fish migration and chemical environmental fate for the marine environment is still to be developed. Thus, further research is needed not only with respect to improving or rather identifying reliable effect information.

The approach taken is a balance between the ambitious goal to be able to address trace element contamination at a continental scale with a regional resolution on the one hand and state-of-the-art in modelling of trace elements on the other. When interpreting the results obtained in decision-making, it is, therefore, important to acknowledge the practical state-of-the-art, the uncertainties and the need for ongoing scientific advances. According to the 'ethos of applied demography' (Essink-Bot, 1998), however, it is better to provide the best possible estimates based on, at times, poor data, than providing no estimates at all. In this sense, the present work constitutes an improvement towards more knowledge about the magnitude of the external costs occurring due to human activities. Before this contribution, hardly any information (if at all) 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). Several issues of further development have been named for which hopefully further funding can be obtained.

In supporting the assessment of releases of hazardous substances into air, (fresh) water and soil at the European scale in a spatially-resolved way, the present work is an improvement towards more knowledge about the magnitude of health impacts and external costs occurring due to human activities as hardly any information (if at all) particularly on the external costs for exposure routes other than inhalation had been available prior to this effort.