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Characterization of human exposure patterns to environmental contaminants:
possibilities of the USES approach

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ABSTRACT

In the framework of the research programmes "Setting Integrated Environmental Quality Objectives" (IEQO) and "Human Health and the Environment" (H&E), both initiated by the Dutch Ministry of Housing, Spatial Planning and the Environment, the applicability of the approach of the risk assessment model USES, the Uniform System for the Evaluation of Substances, for determination of multi-route exposure patterns has been studied. Determination of these patterns is required to support toxicological risk assessments and in environmental policy. The applicability of USES was tested by evaluation of 5 different model substances and comparison of the results with field data. It appeared that with minor adaptations the USES approach is suitable for IEQO project, but that it has to be supplemented with additional models to refine temporal and spatial variability in exposure for risk assessment purposes (H&E).

SAMENVATTING

De blootstelling van mensen aan stoffen in het milieu is meestal via verscheidene routes (multi-route blootstelling). Ter voorkoming van gezondheidsschade moet het milieubeleid rekening houden met verspreiding in het milieu en zodoende met multi-route blootstelling.

Ook moet voor een beoordeling van mogelijke actuele risico's, die samenhangen met milieuverontreinigende stoffen, met multi-route blootstelling rekening worden gehouden.
In twee onderzoeksprojecten "Mens en Milieu" (M&M) en "Integrale Normstelling Stoffen" (INS) speelt bepaling van multi-route blootstelling een belangrijke rol.
Bij INS moet op basis van een multi-route blootstellingspatroon een toxicologische advieswaarde voor de mens worden toegerekend naar de te onderscheiden milieucompartimenten. Daartoe is tot op heden nog geen methodiek voorhanden, omdat van de meeste stoffen blootstellingspatronen ontbreken.
Het programma M&M spitst zich toe op cumulatieve blootstelling van de mens aan milieuverontreinigende factoren. Voor dit programma is inzicht in het gedrag van milieucontaminanten interessant en de daarmee samenhangende routes van directe en indirecte blootstelling van de mens. Daarnaast is van belang de invloed die individuen middels life-style en activiteitenpatroon zelf op hun blootstelling uitoefenen. Het M&M programma vereist een beschrijving van bovengenoemde factoren op een veel hoger differentatieniveau dan voor INS vereist is omdat het aandacht besteedt aan blootstelling op nationale, regionale en lokale schaal, mede in afhankelijkheid van de veroorzakende bronnen.

Dit rapport verkent de mogelijkheden voor de ontwikkeling van een methodiek ter bepaling van multi-route blootstellingspatronen. Daarbij is uitgegaan van het USES computermodel, dat voor nieuwe en bestaande stoffen en bestrijdingsmiddelen een risicoschatting kan genereren. Tevens kan het worden gebruikt voor prioritering van stoffen. Tijdens een evaluatie berekent USES een multi-route blootstellingspatroon.
Het doel van het onderzoek, dat is beschreven in dit rapport, is vast te stellen in hoeverre de USES-benadering bruikbaar is binnen INS en M&M. Tevens is het er op gericht te signaleren waar eventueel nog aanvullingen op deze benadering nodig zijn. Dit rapport moet niet beschouwd worden als een validatie van USES.
Met USES zijn de blootstellingspatronen van 5 verbindingen doorgerekend. Deze stoffen verschillen sterk in fysisch-chemische eigenschappen en in de wijze waarop ze worden gebruikt. De berekende patronen zijn vervolgens vergeleken met actuele meetgegevens.
De USES benadering bleek geschikt om te worden gebruikt bij INS. Aanvulling is nog nodig voor blootstelling ten gevolge van contact met verontreinigde grond (bv. inslikken) en voor huidcontact met verontreinigd water.

Voor M&M zijn uitgebreidere aanvullingen nodig, omdat ruimtelijke en temporele variabiliteit in USES vrijwel niet aan de orde komen en juist dat belangrijke aspecten zijn van dat onderzoeksprogramma.

Dit rapport is opgesteld in opdracht van het Ministerie VROM, directie Stoffen, Veiligheid en Straling en directie Lucht en Energie.

SUMMARY

The exposure of humans to environmental contaminants is usually via more than one route at the same time (multi-route exposure). In order to prevent damage to health, environmental policy has to take into account dispersion through the environment and thus multi-route exposure. Additionally, for the evaluation of the risk of actual exposure to substances in the environment, recognition of multi-route exposure is a factor of relevance. For two research projects, "Setting Integrated Environmental Quality Objectives" (IEQO) and "Human Health and the Environment" (H&E) the characterization of multi-route exposure is important.
For IEQO a toxicological admissible exposure level has to be allocated to the various environmental compartments (soil, water and air), on the basis of multi-route exposure patterns. However, till now, a methodology for this procedure is not available, because exposure patterns are not available for most chemicals.
The programme H&E focuses on determination of cumulative exposures of humans to environment contaminating factors. For this programme insight in the environmental behaviour of contaminants is of interest as well as the routes of direct and indirect exposure of humans which are connected to this behaviour. Besides that, individual life-style an activity patterns and their influence on a persons exposure are of relevance. The H&E programme requires description of the aforementionted factors at a much higher level of differentiation than the IEQO programme does because H&E it looks into exposure at nation wide, regional and local scales, in relationship with the emitting sources.

This report explores the possibilities for the development of a methodology for multi-route exposure pattern determination. Starting point is the USES computer model, which can perform risk evaluations for new and existing substances and pesticides. Additionally, it may be used for prioritisation purposes. While performing an evaluation USES generates a multi-route exposure pattern. The purpose of the research described in this report, is to determine to what extent the USES approach can be applied within IEQO and H&E. It should also indicate where the USES approach needs supplementation. The report must not be considered as a validation of USES.
With USES the exposure patterns of 5 substances have been determined. These substances were highly different in physico-chemical characteristics and in the way they are used. Subsequently, the computed patterns have been compared to data derived from field studies.
The USES approach is appropriate to support the derivation of IEQOs. The approach needs to be supplemented with an estimation of exposure via soil contact (e.g. ingestion)

and dermal contact with contaminated water. For the H&E-programme more elaborate supplementation is required, because spatial and temporal variability are no major subjects in USES, while they are important aspects of the H&E research project.
This report has been prepared on behalf of the Dutch ministry of Housing, Spatial planning and the Environment, Directorate for Chemicals, External Safety and Radiation protection and the Directorate Air and Energy.

1. INTRODUCTION

The research programme Human Health and the Environment (H&E, in Dutch: "Mens en Milieu") aims at the development of a policy for the protection of the Dutch population against health effects resulting from environmental pollution. The programme has a special interest in simultaneous exposure through more than one environmental compartment (multi-route exposure) and in simultaneous exposure to more than one agent. For the H&E programme exposure-pattern characterization is required for the estimation of actual exposure, identifying in particular the extent to which exposure is dependent on the various emission source-categories. The actual exposure of a person strongly depends on his or her life style and pattern of activities. Insight in environmental behaviour of pollutants and the adherent direct and indirect exposure patterns is of interest for this programme. The H&E programme is very much interested in small scale situations e.g. a street in a build-up area, or difference between rural and city.

In a second research project, Setting Integrated Environmental Quality Objectives (in Dutch: "Integrale Normstelling") the ultimate goal is the derivation of Integrated Environmental Quality Objectives (IEQOs) for environmental contaminants of soil, water and air. The derivation of IEQOs is based on Maximal Permissible Concentrations (MPCs), which are human- as well as eco-toxicologically underpinned. These IEQOs are designed in a way that maintenance of the permissible concentration in one environmental compartment (i.e. air, water or soil) does not cause the concentration in a second compartment to exceed the permissible concentration for that second compartment. The MPCs are the starting point for the derivation of IEQOs. In this derivation process, apart from the toxicologically underpinned MPCs, economical and social aspects are taken into account as well. IEQOs are instruments in the national environmental policy and should represent safe levels of exposure for the general population. So, IEQOs are especially relevant for nation-wide scale situations.

For both projects insight in multi-route exposure, i.e. the contribution via the various environmental compartments to the total body exposure, is important. The report at hand contributes to both programmes with respect to method development for human multi-route exposure characterization. It has been prepared by request of the Ministry of Housing, Spatial Planning and the Environment for the purpose of policy development.

1.1 Background

In the memorandum "Premises for Risk-management" (1), the policy of the Ministry of Housing, Spatial Planning and Environment on the acceptability of health risks related to exposure to environmental contaminants has been laid down. The policy view is that the acceptable individual mortality risk, resulting from additional exposure to a genotoxic carcinogen, is 10^{-6}/y, maximally (approx. 10^{-4}/life time). So the MPC for such a carcinogen is that level of exposure which is associated with this additional risk on death by cancer. For non-genotoxic carcinogens and all other toxicants, the noxious action is supposed to emerge only if exposure to such a chemical exceeds a certain threshold. For humans, the threshold levels for toxic effects of (non-genotoxic) chemicals are usually not known. Therefore, in most cases human threshold levels are estimated from the results of animal experiments, usually with application of uncertainty factors. The Maximum Permissible Concentration for such a chemical is set equal to this estimated human threshold level (1). A more elaborate explanation of the Dutch system of Environmental Quality Objectives can be found in reference (2)

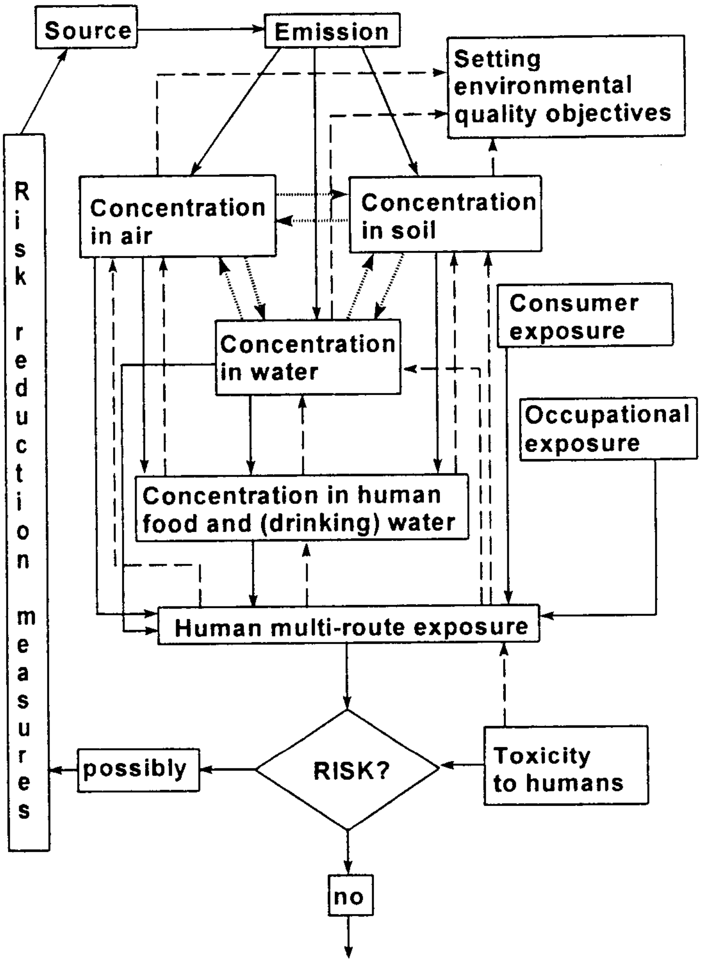

Figure 1. Diagram of multi-route exposure. Dashed arrows refer to IEQO-approach; solid arrows refer to H&E approach; dotted arrows: intercompartmental exchange.

In general, exposure to environmental contaminants may occur through more than one route. Substances may enter the human body via air (inhalatory), (drinking) water, soil (both dermally and orally) and via food (orally). Exposure may also occur through contact with consumer products and in the working environment. A schematic presentation of this multi-route exposure pattern is given in figure 1.

In this scheme (fig. 1) the H&E approach is indicated with the solid arrows.
Starting point in this approach is the emission of an individual substance and the resulting environmental concentrations. These concentrations may be determined directly in the various compartment with chemo-analytical methods or predicted/calculated from emission data using mathematical models. The spatial scale of the model calculations should vary from street-level up to nation-wide level. From the calculated or measured concentrations the contribution of the different compartments to the total human exposure can be estimated. Based on the estimation of the integral multi-route exposure a risk-assessment can be performed, which may result in actions aiming a risk reduction and/or in additional research. Knowledge of the most important routes of exposure can be used to guide the selection of the major emitting sources and the most efficient measures.

In figure 1 the IEQO-approach is indicated in with dashed arrows. Determination of IEQOs for environmental contaminants for each of the three abiotic environmental compartments requires allocation of the MPC_{human} (in mg/kg body weight/day) to each of these compartments. This allocation, resulting in three MPC_{human}, one for air, water and soil each, is based upon the contribution via each environmental compartment to the total human exposure. The allocation procedure should take into account the relevance of the individual routes of exposure for the toxicological effects to be expected, viz. the effects the integral MPC_{human} is based upon. For example: the MPC_{human} for a particular compound is 100 μg/kg b.w./d. 25% of this permissible dose enters the human body via air, 35% via soil (directly or after uptake by plants) and 40% via drinking water. Taking into account the respiration volume, uptake by plants, soil contact, drinking water consumption and intercompartmental exchange rates of contaminants, one might compute IEQOs in air, soil and water to be e.g. 1 μg/m^3 air, 10 mg/kg soil and 5 mg/m^3 (surface) water (the values mentioned are purely hypothetical and not calculated by any means).
Previous derivations of IEQOs (3) have been performed for volatile compounds, assuming that total body exposure would result from exposure via the air compartment. For compounds which can be present in relevant concentrations in other compartments as well, the derivation of IEQOs requires a more extended insight in the relative contribution via the various routes of exposure.

Recently, for the risk-evaluation of chemical substances the computer programme USES (Uniform System for the Evaluation of Substances) has been published (4). This computer model has been designed as a tool for the rapid and quantitative screening of the potential risk of organic substances to humans and to the environment. As such it may act as a decision support system for the central government in the evaluation of new and existing chemicals, of plant protection products and of biocides. Besides this, USES can also be applied in the prioritisation of substances for policy development.

When a risk assessment for a particular substance is carried out, the USES model generates an estimation of the multi-route exposure profile for the chemical under study. Additionally, USES predicts environmental concentrations and human exposure levels (PECs: predicted environmental concentrations). Subsequently, USES compares these PECs with reference values, NECs (No Effect Concentration), NOAELs (No Observed Adverse Effec Levels) or TDIs (Tolerable Daily Intake), thus producing PEC/NEC-ratios or Margins of Safety. Therefore the USES output provides an indication of the presence of a risk for humans or the environment, which may be associated with use and/or emission of that substance. The prediction of environmental concentration is based on estimations of the emission pattern of the chemical under study. These emission patterns are derived from data on production technology and volumina, use and physico-chemical properties of the particular substance.

Crucial in the USES risk assessment is the assumption that PECs are estimated at the 95-percentile value via a "realistic worst case" approach. Whether this is truehas to be tested in validation studies and uncertainty analyses.

1.2 Aim of the present report

In the text above it has been stressed that knowledge of multi-route exposure patterns is a prerequisite for both the IEQO and H&E programmes. Although not particularly developed for this purpose, the USES programme could be a useful tool to generate estimations of the multi-route exposure profiles of many organic chemicals. The present report describes the results of a study on the general applicability of the USES approach within these two programmes.

This study is the first step in the development of a strategy for the determination of multi-route exposure patterns. This strategy will be incorporated into a general procedure for derivation of IEQOs in future. This report also pays attention to the applicability of the USES approach within the framework of the programme "Human Health and the Environment". For this it aims at the determination of hitherto not-incorporated routes of exposure in the USES approach and at description of other limitations, which may need alleviation to make USES applicable within the H&E programme. Alternatively, supplementary models will have to be developed which deal with aspects which are not particularly relevant for the purpose of USES itself, but are highly relevant for either the H&E or the IEQO programme, or for both.

It should be noted that application of USES for characterization of multi-route exposure for standard setting is beyond the scope the programme has initially been developed for. This report must not be interpreted as the result of a validation study of the USES programme. The present document focuses merely on the characterization of multi-route exposure. The way USES performs the final risk evaluation has not been taken into consideration in-depth.

The relevance of the research described in this report lays in its contribution to the development of a general idea about determination of multi-route exposure patterns, rather than an explicit testing of a particular computer model. USES has merely been chosen as a starting point, because multi-route exposure assessment is part of the USES procedures.

2. METHODS

2.1 The USES computer model

2.1.1 general features

The following text and illustrations are based upon the USES report (4). The reader is referred to this report for a more detailed explanation of the USES programme.
The USES model has been developed in a cooperation project between the Ministry of Housing, Spatial planning and the Environment (VROM) and the Ministry of Welfare, Health and Cultural Affairs (WVC). It provides a means for a rapid general risk assessment and priority setting for so called new and existing chemicals and pesticides. It can be applied in the process of governmental decision making and prioritisation of substances. The programme performs risk evaluations, i.e. it calculates hazard coefficients (i.e. PEC/NEC ratios), for humans, aquatic and terrestrial ecosystems, for micro-organisms in a sewage treatment plant (STP) and for top predators. USES is a so called "generic" model, which can perform a risk assessment with e.g. data as those submitted in an EU base set (4).

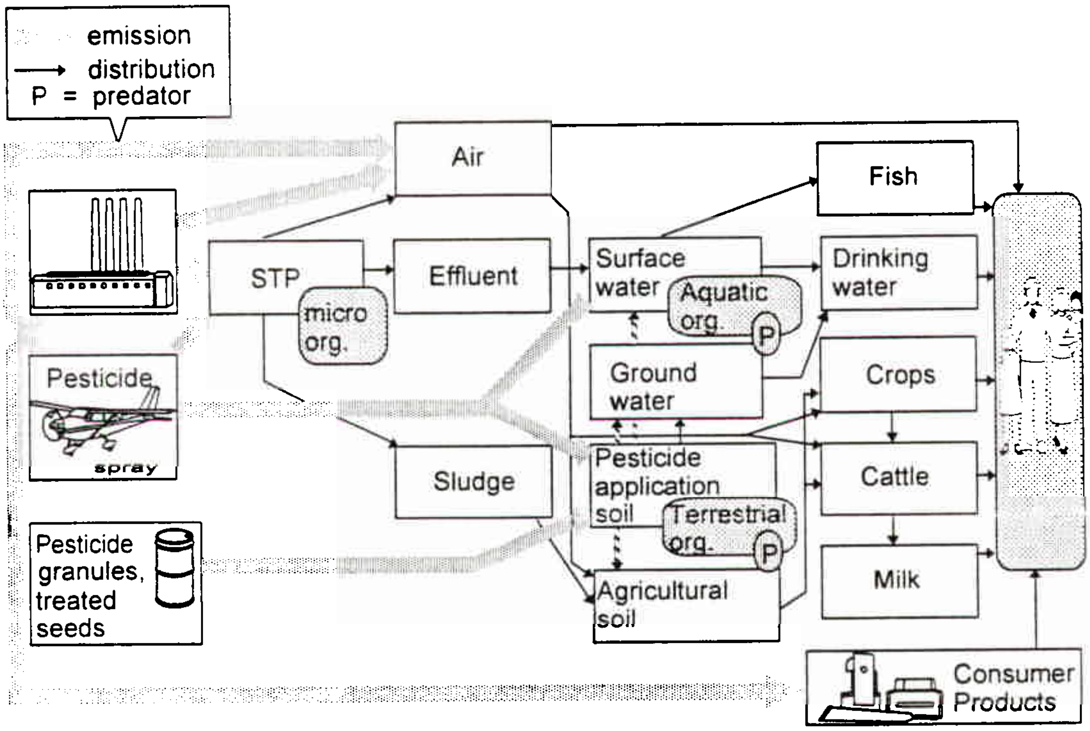

Figure 2. Summary of the exposure routes in USES.

The programme consists of a number of mathematical functions which predict concentrations of substances in the various environmental compartments. Calculations are carried out for different spatial scales: the programme distinguishes between local scale (i.e. near a source), regional scale (i.e. national scale) and continental scale (i.e. Western Europe). The results of the continental scale calculations are used as "background" concentrations for the regional scale concentrations.

2.1.2 multi-route exposure profiles

USES considers a number of exposure routes (figure 2) via which humans and "environmental organisms" are exposed to chemicals. Emissions released by industries or diffuse sources like households or agriculture are assumed to disperse into the environment. Emissions to water are (optionally) supposed to occur after treatment in STPs, or in case of e.g. pesticides occur directly to surface water or soil. From the primary compartments (air, water and soil), substances travel through, and may be taken up in humans and environmental food chains. Human uptake results from consumption of or contact with more or less severely contaminated food products, (drinking) water and air, or after contact with substances intentionally or unintentionally released by consumer products. The programme calculates human average daily intakes and an over-all average daily internal body dose, based on assumed uptake efficiencies for the various routes.

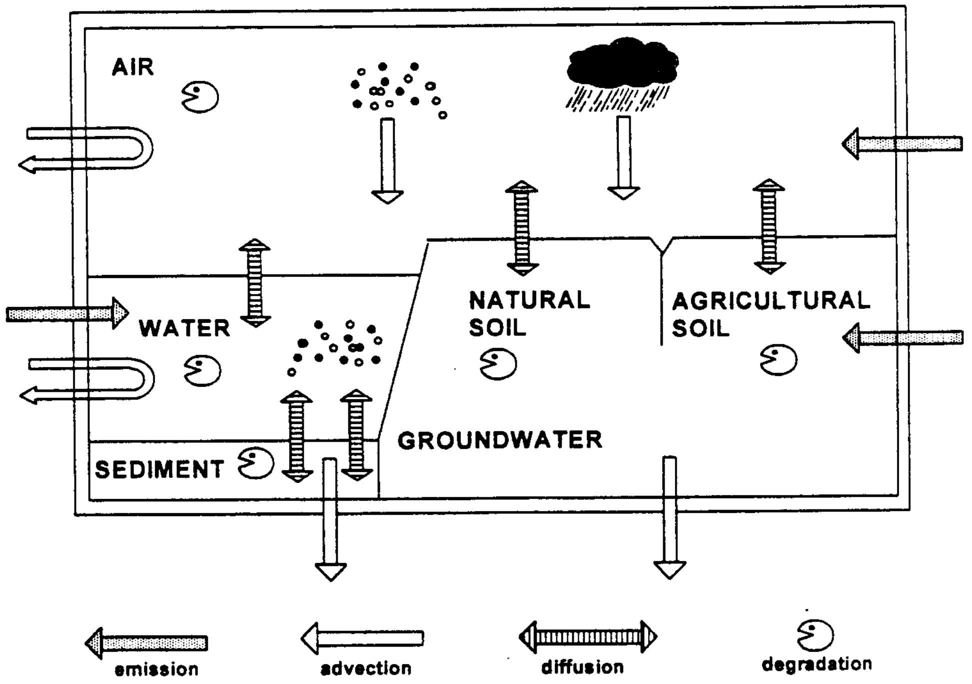

Figure 3. Regional scale environmental compartmentation in USES.

2.1.3 environmental dispersion

At the regional scale the mathematical formulas represent a Mackay type model of a standard environment, in which 8 different (sub-)compartments are assumed (figure 3). These compartments are considered to be homogenous. For air and soil, sub-compartments (e.g. aerosol in air or water phase in soil) are assumed to be in equilibrium with the other sub-compartments (thus air with rainwater or soil air with soil mineral phase). For the water sub-compartments no such equilibrium is assumed. The estimated "bulk-concentrations" in air, soil and water are steady state concentrations. This means that in order to maintain the steady state, net flows of substances may continuously occur as compensation for for instance photo- or biodegradation. In the regional scale evaluation, emissions are supposed to have a strongly diffuse character. For the regional scale estimations, concentrations and human exposure refer to long-term average situations. The regional approach is considered to be not very appropriate if in real life emissions originate from one or a very small number of sources.

For local scale calculations local sources i.e. "precisely defined" sources are looked at. For these calculations a number of special local scale models and assumptions have been incorporated into USES. These comprise for instance models for the drift of pesticides during application or the dispersion of gaseous chemicals in air near a point source such as a chimney. For local scale evaluations USES takes into account neither biodegradation in surface water and soil nor losses from the air compartment due to deposition and atmospheric degradation.

2.1.4 USES' modular design

USES has been constructed as a modular computer programme. The various modules are presented in figure 4. All modules contribute to the USES output. Similarly, the output of each module is regarded as input for the next one.

In the input module information on the physico-chemical characteristics, production, use, emissions during life cycle steps and toxicity data should be entered into USES. If physico-chemical data are incomplete, lacking data are estimated from physico-chemical constants which are known, e.g. solubility in water from lipophilicity and melting point. USES also needs data on production (volumina and technological processes) and use of chemicals, because guided by this information and by a compound's volatility and solubility in water, USES selects emission factors from a table and determines emission patterns and volumes.

The estimations of the total emissions of a compound are input data for the distribution models, the results of which are starting points for the intake models. By comparing intake results with toxicity data USES may provide an indication of the existence of a risk for ecosystems or humans.

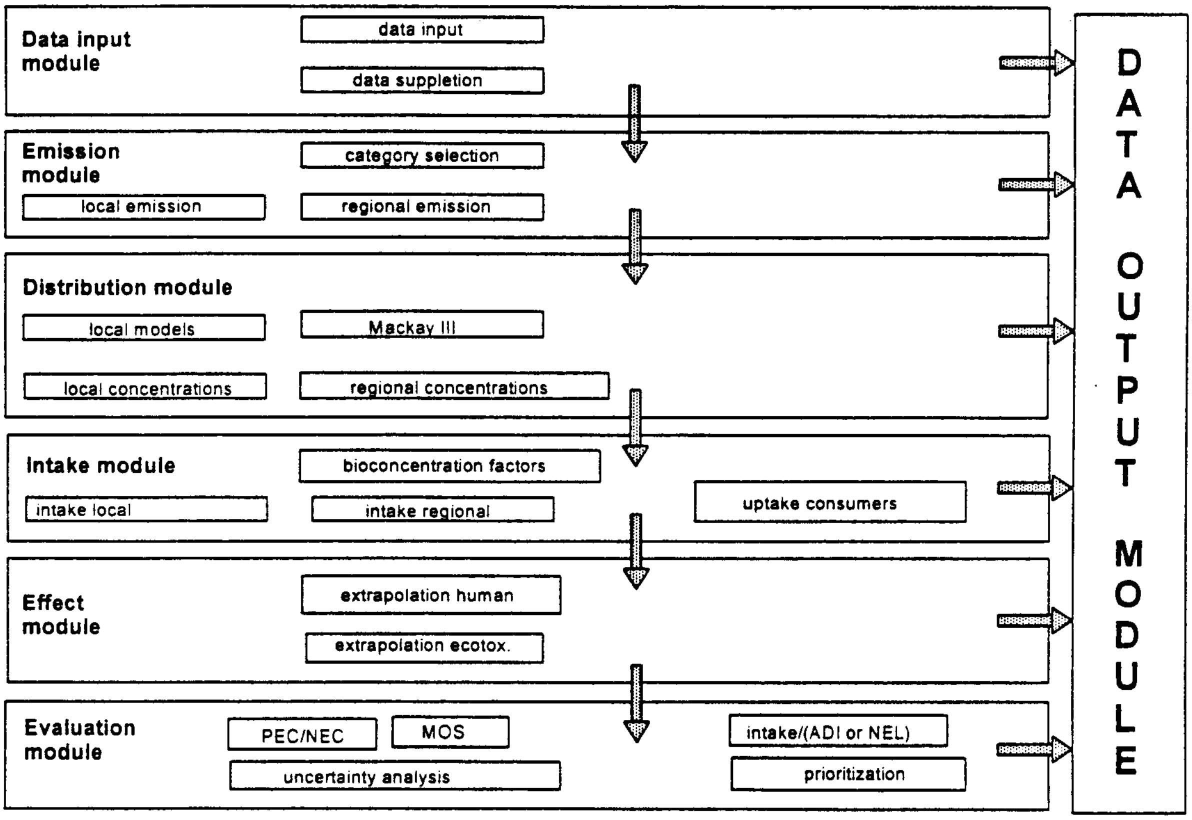

Figure 4. The structure of the USES computer program. Horizontal arrows: output data flow; vertical arrows: internal data flow.

2.1.5 Intervention in a programme run.

A USES evaluation procedure can be carried out either in one continuous process called "direct evaluation" or stepwise called "intermediate evaluation". While performing a direct evaluation the "operator" has no possibilities to influence the flow of intermediate results from one programme module towards the next one. This flow of intermediate results is roughly indicated in figure 4 with the vertical arrows. During an intermediate evaluation, the programme interrupts the evaluation process after each module and it presents the

intermediate results on the computer screen. After each module the "operator" has the opportunity to overrule the intermediate results and replace them by real-life data.
The study of the applicability of USES in multi-route exposure characterization has been performed for a variety of model-compounds. All evaluations have been carried out using the "intermediate evaluation" option. Intervention in the program's intermediate results was done for instance if real-life emission data were available, or if USES made assumptions which were not in concordance with actual situations.

2.2 Selection of model compounds

For testing the applicability of the USES model a limited number of substances has been selected. The selected compounds differ widely in physico-chemical and toxicological properties. Differences in use- and production pattern of the various chemicals may further enhance diversity in the various (expected or calculated) exposure patterns. In order to be able to perform an exposure pattern characterization for a particular compound with the USES programme information on production and use of chemicals is needed, or, alternatively, emission data for each environmental compartment are required. Table 1 shows for each of the compounds the considerations for its selection. For most of the selected compounds a Dutch Integrated Criteria Document is available. These documents have been composed for a great number of priority substances and present data on among others use, emissions, environmental chemistry, (bio)degradation and toxicological profiles. It was intended to get a wide variability within the group of selected compounds. Then detection of limitations in the USES approach is more likely. Additionally, the results may then also indicate whether it is possible to make generalisations with respect to exposure patterns for chemicals with physico-chemical properties similar to the selected model compounds. Alternatively, but less preferable, for each individual chemical to be evaluated in future, its own mapping of the exposure routes has to be performed.

Table 1. Chemicals for which a multi-route exposure pattern will be generated with the USES programme.

CHEMICAL	CHARACTERISTICS
Cadmium	heavy metal, local and systemic toxicity, not volatile at ambient temperatures. (5)
Lead	heavy metal, systemic toxicity, differences in sensitivity between children and adults, not volatile at ambient temperatures.

Benzo[a]pyrene	hardly volatile; air-born exposure through aerosols, poorly soluble in water, systemic and local toxicity, highly lipophilic, not readily biodegradable. (5)
Benzene	volatile, moderately soluble in water, systemic toxicity, readily biodegradable. (5)
Formaldehyde	gaseous, local toxicity, highly soluble in water, highly (bio)-degradable, indoor/outdoor exposure; very high natural emission.
Dichloromethane	volatile, highly soluble in water, systemic toxicity, consumer- and workers exposure.
Tetrachloroethylene	volatile, moderately soluble in water, systemic toxicity, exposure of dry-cleaning workers.
1,4-dichlorobenzene	volatile, moderately soluble in water, systemic toxicity, indoor (consumer) exposure.
Atrazin	hardly volatile, moderately soluble in water, systemic toxicity, pesticide.

2.3 Data acquisition

For the evaluation of the environmental fate of an environmental contaminant and estimation of the exposure to such a substance, USES requires data on physico-chemical characteristics, (bio)degradability, bioaccumulation and qualitative as well as quantitative data on its production and use in The Netherlands and Europe. For this preliminary survey data were abstracted from reviews on the individual compounds such as HEDSET (Harmonized Electronic Data SET; prepared for EU, DGXI, latest updates available), Dutch Criteria Documents, WHO/IPCS Environmental Health Criteria and an inventory of technical data sheets on candidate list I substances (directive 76/464/EEC; ref. 6). An extensive on-line search has not been performed. For atrazin no Integrated Criteria Document was available, but information was obtained from an extensive pesticide evaluation report (7) and the Dutch TOXBANK-data base. Additionally, for the evaluation of this compound data were extracted from the environmental research programme "Watersysteemverkenningen" (Watersystem Survey; ref. 8). No special effort was made to check on the data used. However, if various sources provided conflicting information, chemical handbooks were used to reach a decision.
If for a particular parameter a range of values was found, a final value (USES only permits input of point estimates) was obtained from the range boundaries (usually the lower value), unless additional information allowed for a better estimate. It should be

noted that usually for physico-chemical data only a narrow range in values was found. Thus accepting one of the range boundaries as final value is a meaningful procedure for obtaining a applicable estimate. In all other cases in which no proper values from experimental research could be found, the standard defaults or the results of standard estimation procedures incorporated in USES were used.
No attempt has been made to collect experimentally determined solid matter (i.e. soil, sediment, suspended matter)/ water partition coefficients. According to the USES report the estimation procedures in USES provide fairly accurate estimations of these. For non-ionic organic substances, it is believed that any deviation of the estimated values from the true values is not large enough to result in major changes in the model output (Jager, personal communication, sept. 1994).

The USES programme determines emission factors from information on the technology of production, formulation and processing, within the programme referred to under the heading "main category" e.g. "continuous production" or "wide-dispersive use". However, for many substances such information is not readily available. In case of absence of such knowledge (realistic) worst-case default "main categories" are applied. These default choices may result in emission patterns (i.e. ratios between emission to air/emission to water/emission to soil), which differ from the actual situation. Information on chemical use patterns and production volume, physico-chemical and other numerical data for the model compounds are presented in Appendices I to V.
Although an extensive comparison of actual environmental concentrations is beyond the scope of this survey, a superficial inventory of such concentrations has been made to get a coarse impression of the reality value of the USES calculated results. In addition a survey of environmental concentrations may draw attention to sources of emission and routes of exposure which are not identified by the USES-computer model. Information on environmental concentrations has been drawn mainly from the Dutch Criteria Documents and from the "Watersysteemverkenningen" (Watersystem Survey; ref. 8).

3. RESULTS

In this chapter the results of the characterization of the multi-route exposure patterns for the various model substances are presented. For each chemical date are given according to the following scheme:

- a presentation of general information
- a brief explanation of a table with computed results
- a description of the calculated exposure pattern
- a discussion of the calculated pattern in the light of actual environmental concentrations.

In the data tables only estimates of exposure are given. Estimates of environmental concentrations can be found in a appendix VI.

3.1 Application of USES

Normally, USES estimates the emissions of substances related to production, formulation, processing, private use and recovery at the regional and continental scales in one single run. If substances are formulated or processed in one country but produced in another country (i.e. there are continental emissions as a result of production, but only regional emissions as a result of e.g. processing) USES cannot calculate regional emissions and resulting environmental concentrations in one run. Then, evaluation can only be performed using a multi-run scheme. Such situations exist for instance with atrazin (not produced, but processed in The Netherlands) and 1,4-dichlorobenzene (formulated in The Netherlands and used in private households, but produced abroad). For such substances, in a first run continental emissions and concentrations should be calculated and in subsequent runs calculations for the regional situation should be carried out.
However, in a single-run approach, USES considers the continental environment concentrations as back-ground concentrations in the regional evaluation by taking into account trans-border migration of chemicals. If continental emissions and regional emissions are evaluated in separate runs, the programme cannot properly do so. In order to optimize uniformity, for all model substances continental emissions and concentrations have been neglected and the exposure pattern assessments have been based on regional emissions, only.
Apart from leaving out continental emissions, for reasons described in the previous two paragraphs, as many as possible actual data were entered into USES. Thus in all evaluations overruling intermediate programme results has occurred to a smaller or larger extent (cf. section 2.1.5).

3.2 Atrazin

3.2.1 general information

Atrazin is an agricultural triazine herbicide, which is sprayed on the soil once a year. The quantities used vary form 1 to 2 kg of active ingredient/ha in maize and asparagus culture to up to 7.5 kg of active ingredient/ha in borders of arable land and on permanently fallow land. The volume of the application varies between 50 and 1000 l/ha. Atrazin is hardly volatile and is only slightly lipophilic (7,9).

Production of atrazin takes place in at least 2 plants in Italy (10) in a total quantity of about 17000 tonnes/y. Atrazin is also produced in the UK and in Switzerland (8), but quantitative data are lacking. According to EURECO (6) production in the UK has ceased since 1988 and production in Italy amounts only 8000 tonnes/y. The HEDSET data (10) are from a more recent source than the EURECO-report (6). More information regarding the nature of the production process is not available. According to EURECO (6) an amount of 6000 tonnes/y is used in the EU as a herbicide.
Atrazin is not produced in The Netherlands, but it is formulated by one firm into several trade products. According to Ordelman et al. (8) atrazin is formulated in a batch process using dedicated equipment, which is cleaned twice yearly. Additionally it is assumed that the entire Dutch market (200 tonnes of atrazin/year) for atrazin preparations is served by this one firm. It is unknown how much atrazin the firm formulates each year. Its maximum capacity is 1000 tonnes/year. Assuming that its full capacity is handled, some 800 tonnes of formulated atrazin will be exported from The Netherlands, yearly.
USES automatically assumes always that chemicals are handled by more than one firm. Thus, the fact that only one firm is involved in atrazin formulation requires an adaptation to the USES results. Local emissions and environmental concentrations are computed for the largest company, to which USES allots 60% of the total "production". As the one firm involved handles 100% of the total amount, all local emission estimates have been multiplied by a factor of 1.67. Concomitantly regional emissions are set to 0 kg/d. While performing an "intermediate evaluation" (cf. chapter "2. Methods" section 2.1.5) these adaptations can be implemented .

Table 2. USES-estimated Multiroute Exposure profile for atrazin.

	resulting from emissions owing to			total exposure	relative importance %		
	formulation	pesticide application			owing to formulation	owing to pesticide application	
route of exposure	local^{1}	local^{1,2}	regional^{1}	regional^{1}	local	local	regional
air	54.4	< 0.1	< 0.1	< 0.1	0.44	0.06	0.03
drinking water	0.4	32.3	67.8	67.8	< 0.01	80.34	62.21
fish	< 0.1	2.3	0.5	0.5	< 0.01	5.75	0.41
plant shoots	12360	5.6	29.2	29.2	99.48	13.85	26.79
plant roots	7.1	0.1	11.5	11.5	0.06	0.01	10.55
meat	1.4	< 0.1	< 0.1	< 0.1	0.01	< 0.01	< 0.01
milk	1.4	< 0.1	< 0.1	< 0.1	0.01	< 0.01	< 0.01
total	12424.7	40.2	108.9	108.9	100.00	100.00	100.00

In The Netherlands only one firm is involved in atrazin formulation. Therefore regional emissions are set to 0 kg/d. As a result of this first adaptation, nation-wide exposure to atrazin as a result of formulating activity is calculated to be abscent.

[1] These columns contain absolute exposures in ng/kg bw/d.

[2] "local" exposure owing to pesticide application results from emission from a "hypothetical" small source and is estimated in a INSBOX submodule [USES manual]

USES calculates emissions from biological sewage treatment plants (STPs), which in the case of atrazin are not particularly effective, because atrazin is only slowly biodegraded. A non-biological STP, in which atrazin is absorbed to activated charcoal, however, is highly effective in scavenging atrazin from the waste water stream. Therefore, the "formulating-firm" passes all emissions to water through a non-biological treatment installation before discharging to surface waters (8). Virtually all atrazin is absorbed in the STP sludge (i.e. the charcoal bed), which is subsequently treated as chemical waste.
The actual amount of atrazin in the effluents from the non-biological STP is 1 g atrazin/day to surface water (yearly average; ref.8).
However, a non-biological STP is no part of the USES model. Thus a second set of adaptations has been made. Emissions to surface water have been set to 1 g/day, whereas the amount in the sludge stream has been calculated from the total amount of atrazin in the primary (untreated) waste water stream, of which USES itself provides an estimate. Thus while performing an "intermediate evaluation" in USES, intermediate programme results in the local STP-module have been over-ruled according to the steps described below:

1) All emissions from the local source are multiplied by a factor of 1.67 and al other regional emissions were set to 0 kg/y.
2) Virtually the entire influx of atrazin into the local STP is allotted to the sludge-stream. The waste water stream from the STP receives only 1 g/d.

Most likely the company discards the solid residues, the "sludge", from the treatment installation as chemical waste. So none of this sludge is brought onto agricultural soil. This assumption, however, is over-ruled by the USES programme, which does not allow a sludge-application less than 20 kg/ha/y. As a result of the first adaptation (i.e regional emissions are set to 0 kg/y), regional exposure to atrazin as a result of formulating activity is calculated to be absent.

The use of atrazin, "processing" in terms of USES, is wide-dispersive. In concordance with Ordelman et al. (8) it is assumed that atrazin is sprayed on the soil once a year just after plant emergence. The quantity of spraying liquid used is 400 L/ha, containing 1.5 kg active ingredient, which according to several sources (7,8,9) are no extreme estimates. For spraying on nearly bare soil a drift-fraction to surface water of 1% and a fraction reaching the soil surface of 80% of the applied quantity should be chosen for running USES (4). An overview of input data for atrazin has been given in appendix I.

3.2.2 construction of the table with results for atrazin:

Running USES for atrazin produces the exposure estimates presented in table 2. The values represent internal exposures through the media mentioned.
Under total exposure (regional) values are given which represent exposures resulting from regional emissions only. Summation of local exposure estimates is unwanted because *local* exposure resulting from formulation emissions is spatially separated from *local* exposure resulting from atrazin application. For formulation a real point source can be recognised, with releases of atrazin in local air, surface water and soil. A real local emission source cannot be recognised for application of atrazin (agricultural chemicals in general). For application (processing of pesticides in general) , USES assumes a hypothetical "point-source", which emits a defined very low percentage of the entire annual use. This emission results in local environmental concentrations in air (at 100 m distance from the "point-source"), groundwater and agricultural soil.

3.2.3 exposure pattern for atrazin

Owing to application, at a regional scale about 60% of the total body exposure to atrazin originates from intake through drinking water. The remainder is almost completely made up for by consumption of vegetables. At local scale due to formulation, however, the importance of consumption of vegetables is far more important than exposure via drinking water. Moreover, the human intake of atrazin as a result of consumption of vegetables at the local scale (formulation) exceeds the intake by other routes by at least three orders of magnitude. While computing the exposure pattern for formulation, input data for regional emissions have been set to zero. Hence, estimated atrazin concentrations in human intake media at the regional scale are nought. So, at regional scale, human intake of atrazin, resulting from formulation is negligible.
Computations with USES result in predicted regional total exposure levels below the µg/kg b.w./d range. People living close to the formulation plant, however, are expected to be exposed to levels in the µg/kg b.w./d range.
The resulting patterns do not greatly depend on the adaptation made to the USES input data with respect to the STP. If this correction is left out USES still estimates the local exposure (owing to formulation) to atrazin in plant shoots to be 98 % of the total exposure. Moreover, even the absolute amount to which humans are exposed via this route is hardly changed when the adaptation is not performed. Preliminary test-runs with USES indicate that exposure via plants is strongly dependent on the atrazin emission to air. A ten-fold change (either increase or decrease) in emissions to air results in an about ten-fold change in plant shoot concentrations.

3.2.4 atrazin: comparison of measured verses predicted exposure pattern:

USES-generated estimates of concentrations of atrazin in the human environment are presented in appendix VI.

Ordelman et al. (8) have not provided an extensive overview of environmental concentrations of atrazin but these authors have given analytical results for various water bodies. In state fresh water bodies in The Netherlands (6 locations) atrazin concentrations from 80 (river Rhine at Lobith) to 430 ng/l (river Meuse at Eijsden) have been reported. In non-state fresh water bodies atrazin (28 locations) has been found in concentrations between 50 (Flevoland) and 2900 ng/l (drainage area river Aa). The concentrations mentioned here are average values. At specific locations maximum concentrations may be considerably higher, whereas at other places only a small difference between average and maximum concentrations has been found (8). The median average value from the regional water bodies is 250 ng/l. In groundwater (5 locations) the median average value is 70 ng/l. Assuming no effective purification in drinking water production facilities, USES estimates a daily intake via drinking water of about 70 ng/kg b.w./d. based on an estimated average concentration in ground water of 2.37 µg/l.

No data are available on the atrazin concentrations in air. However, in the period of maximum use, i.e. May-August, in rainwater (which is, according to USES, a part of the air compartment) average concentrations between 30 to 740 ng/l have been found. Atrazin has also been found in rainwater during periods in which the pesticide is not used and at locations where no atrazin has been sprayed (8). Concentration of atrazin in air (i.e. as a vapour) is considered to be of little relevance due to the chemical's low volatility (8). Probably the presence of atrazin in rain water is a result of spraying and drift rather than from intercompartmental redistribution. As rainwater is usually not inhaled, inhalatory exposure to atrazin is probably very low. USES also calculates the concentrations of chemicals in rain water and aerosols, but these concentrations do not appear in the USES output. As already mentioned in section 2.1.3, rain and aerosols are considered to be integral parts of the compartment air.

Apart from the intake via drinking water, intake via vegetables is a second likely route of exposure, owing to the use of atrazin as an agricultural pesticide. From the USES output it seems that wet and / or dry deposition may contribute to a considerable extent to plant atrazin concentrations.

A more complete real-life scenario of human exposure to atrazin cannot be drawn up. The exposure pattern as generated by USES does not conflict with the above data.

3.3 1,4-Dichlorobenzene

3.3.1 general information

This compound is used as an intermediate in chemical synthesis, as air refresher in toilet-blocks, as a "general insecticide", germicide and agricultural fumigant and in pharmaceutical products (11). The industrial production of 1,4-dichlorobenzene in Europe amounts approximately 49000 tonnes each year (6,10). Of these, 19600 tonnes are used as chemical intermediates and the rest is formulated into various consumer products such as the ones mentioned above. Information about the way 1,4-dichlorobenzene is produced is not available so it is assumed that a batch production process with multipurpose equipment is followed for odour agents (default USES for this use category). Formulation of consumer products takes place in multipurpose equipment (default USES). Main category at processing is "wide-dispersive" for the odour agents and non-dispersive for the intermediates (defaults USES).
Other sources of emission into the environment are burning of wood or garbage and unintended formation in other industrial processes. The total amount emitted by these sources is not known and therefore not incorporated in the study.
All emissions to water, whether from private use or from industrial sources, are supposed to flow through an STP.

1,4-dichlorobenzene is not produced in The Netherlands, but it is imported in a quantity of about 700 tonnes/y (10). More recent data in the Integrated Criteria Document Chlorobenzenes (12), however, indicate an imported quantity of about 354 tonnes over 1987, 28 tonnes of which were exported. In that year, the remainder was used virtually completely in toilet blocks, moth-balls, and urinal tablets. About 246 tonnes were sold to public lavatories, hotels, restaurants and other institutions and the rest (326 - 246 = 80 tonnes) to private households.
Toilet blocks contain 10 to 20% of 1,4-dichlorobenzene and moth-balls and urinal tablets may consist for 100% of this compound. Despite the known decline in use of 1,4-dichlorobenzene as a disinfectant, the exposure pattern mapping for this compound will be based on a total use of 326 tonnes/y (354-28). Regional industrial emissions result solely from the formulation of this quantity into the consumer products mentioned above. These emissions are assumed to result from multi-purpose equipment (default USES). For consumer exposure 1,4-dichlorobenzene is evaluated as a substance for personal and domestic, thus "wide-dispersive", use.
The exposure of consumers to 1,4-dichlorobenzene (private or public places are assumed to be identical) is assumed to be entirely due to its use in toilet blocks or urinal tablets. More detailed information about the USES input and the model parameters for consumer exposure is given in appendix II.

Table 3. USES-estimated Multiroute Exposure profile for 1,4-dichlorobenzene

	resulting from emissions owing to formulation and use as an odour agent[1]		consumer exposure[2]	total exposure		relative importance (% of "local" result of local emissions)	relative importance (% of total exposure)	
route of exposure	local[2]	regional[2]		regional[2]	consumer exposure[2,3]	local	regional[4]	consumer exposure
air	160	0.3	80360	0.3	80360.3	53.44	80.09	100.00
drinking water	17.1	< 0.1	-[5]	< 0.1	< 0.1	5.70	6.78	< 0.01
fish	22.5	< 0.1	-	< 0.1	< 0.1	7.52	1.15	< 0.01
plant shoots	50.2	< 0.1	-	< 0.1	< 0.1	16.78	6.33	< 0.01
plant roots	49.4	< 0.1	-	< 0.1	< 0.1	16.50	5.61	< 0.01
meat	< 0.1	< 0.1	-	< 0.1	< 0.1	0.03	< 0.01	< 0.01
milk	< 0.1	< 0.1	-	< 0.1	< 0.1	0.03	< 0.01	< 0.01
total	299.4	0.3	80360	0.3	80360.3	100.00	100.00	100.00

Calculations with USES do not account for continental or regional emissions resulting from use in consumer products and the adherent exposure.

[1] evaluation as chemical for personal/ domestic use, use category odour agents.

[2] these columns contain absolute exposure in ng/kg bw/d.

[3] this column represents summed values of "regional" and consumer exposure, a simillar column may be constructed for "local" and consumer exposure.

[4] To keep the table clearly structured the data in this table are rounded. This column is based on absolute exposure data as given in the column "resulting from emissions owing to formulation and use as an odour agent". The figures in the latter column are rounded. 0.3 ng/kg bw/d via air comes from 0.276 and 0.3 ng/kg bw/d (total) comes from 0.345. Hence air exposure represents 80% of the total exposure and so on.

[5] Use of 1,4-dichlorobenzene dous not result in exposure via these media.

3.3.2 Construction of the table with results for 1,4-dichlorobenzene

Table 3 shows the amounts of 1,4-dichlorobenzene which are absorbed via the indicated routes of exposure. These amounts have been calculated for regional environmental pollution in The Netherlands.

All 1,4-dichlorobenzene applied products for "personal/domestic use" will finally reach the environment. This is taken into account in the USES model. However, estimates of this emission and the resulting exposure through the environment are not visible in the USES output, but are incorporated into the data for regional exposure under the column "resulting from emissions owing to formulation and use as odour agent".

3.3.3 exposure pattern for 1,4-dichlorobenzene

According to the computations generated by USES (table 3), exposure of the general population ("regional") is mainly through inhalation. Only some 20 % of the total body intake originates from other routes. For people living in the neighbourhood of 1,4-dichlorobenzene-handling industries a considerable part of the total body exposure may result from intake via vegetable foodstuffs if these are grown on local soil (assumed to be supplemented with local STP-sludge). To a lesser extent consumption of fish caught in local surface waters (usually fresh water) and drinking water, produced from local sources, may contribute to the total body exposure as well.

Consumer exposure resulting from the indoor use of products containing 1,4-dichlorobenzene greatly exceeds the exposure via all other routes. This is even so for consumers living near 1,4-dichlorobenzene-handling industries.

3.3.4 1,4-dichlorobenzene: comparison of measured verses predicted exposure pattern:

USES-generated estimates of concentrations of 1,4-dichlorobenzene in the human environment are presented in appendix VI.

Concentrations of 1,4-dichlorobenzene in food materials and drinking water have not been determined. Some data have been given for concentrations in air (indoor as well as outdoor) and fresh water (ground water and surface water; ref. 12).

The average concentration in outdoor air is 24 ng/m^3 and in indoor air 7.2 µg/m^3. The latter value is probably strongly influenced by the indoor use of consumer products containing 1,4-dichlorobenzene. For air in living rooms values up to 300 µg/m^3 have been found, whereas the air concentrations in toilets may be as high as 1000 µg/m^3. The uptake resulting form exposure to the outdoor concentration mentioned above (24 ng/m^3) can be calculated with the USES algorithms. The resulting value, 5.1 ng/kg b.w./d., can be

considered as a fairly realistic indication of exposure of people who do not use 1,4-dichlorobenzene at home.
Drinking water levels are below the detection limit (10 ng/l). At the detection limit exposure via drinking water amounts to about 0.6 ng/ kg b.w./d. This value is low as compared to the inhalatory exposure. The integrated criteria document (12) does not give indications for actual concentrations of 1,4-dichlorobenzene in food.
The actual indoor concentrations as mentioned above mean that the inhalatory exposure resulting from consumer products is indeed of much greater relevance than exposure to 1,4-dichlorobenzene resulting from other sources. The consumer exposure scenario which has been assumed for running USES is probably too pessimistic with respect to the absolute exposure. Recalculation using the actual indoor air concentration in toilet rooms (300 $\mu g/m^3$) would result in a daily intake of about 1600 ng/kg b.w./d. The difference between these values and those in table 3 results from over-estimation of the rate of release of the chemical from toilet blocks in the USES model (cf. Appendix II).
A better scenario for consumer exposure for chemicals released from a solid matrix is not available in USES. However, a more realistic scenario for consumer exposure would have altered the conclusion that inhalation of 1,4-dichlorobenzene is the major route of exposure by no means. Thus real-life data do not deny the calculations by the USES model.

3.4 Benzene

3.4.1 general information

Benzene is a very important substance in chemical industry. Total production in the EU amounted 4.4 Mtonnes in 1987 (6). The largest production plant for this substance in the EU is Dow Chemical Terneuzen (NL). Benzene is used as a starting material in the production of various other substances such as ethylbenzene, cumene, cyclohexane, nitrobenzene and maleic anhydride which in their turn are used in the production of other compounds (6). Benzene is a component of car fuel (10% of the total use in The Netherlands in 1988; ref. 13). Less than 2% of the total is used as a solvent, usually as a component in mixtures. As such it may be present in (consumer) products like paint, rubber glue, paint removers and degreasers. Benzene may also be present in rubber materials and shoes (13).

Benzene is produced by hydro-dealkylation of toluene and xylene, as a by-product of naphtha cracking, in oil refineries (product of pyrolysis) and by distillation of coal tar (6). Benzene may also be liberated during combustion of coal, oil and to a lesser extent gas.

Emissions to the Dutch environment have been fairly well described for the year 1981 (13). The emission data for that year have been used in the exposure mapping at the regional scale. This has been done while performing an "intermediate evaluation", by directly inserting emissions to air, water and soil into the data-entry fields for "regional emission" in USES. For this purpose total emissions to air, water and soil, respectively, have been derived from data in the Integrated Criteria Document Benzene (13). In this derivation procedure the role of individual sources is lost. Therefore, a local scenario cannot be defined and evaluated.
The emission of benzene from automobiles (evaporation, waste gases) has been evaluated in a separate run, because this emission exceeds that of all other sources by far (7300 tonnes/y vs. 1067 tonnes/y; ref. 13). USES has no option (use category) to evaluate car fuel components such as benzene which are no additives but remain in the fuel during the oil refinery process. The use category car fuel additives which is represented in USES has been applied instead.
Other relevant data for benzene have been listed in appendix III.

3.4.2 Construction of the table with results for benzene

The results in table 4 have been obtained from regional emission data. Therefore, only regional environmental concentrations and regional exposures are given. The exposures in column "total exposure" are the summation of the exposures resulting from "car fuel additive" and "all other emissions".

3.4.3 exposure pattern of benzene

The majority of benzene enters the systemic circulation via inhalatory exposure (table 4). This route accounts for approximately 97% of the total exposure. Another, less important route is the exposure to benzene via drinking water, which accounts for about 2.4 %. Exposure via other intake media is virtually negligible.
It should be noted that due to lack of data, the exposure of consumers has not been incorporated in the table. Consumer exposure may result from inhalation of benzene vapours while filling a car's fuel tank or while driving a car. Car air contains 50 to 300 $\mu g/m^3$ benzene (5). Inhalation or dermal contact may also occur during e.g. painting. Apart from dermal contact with benzene-containing products, consumer exposure to benzene is largely via inhalation, meaning that consumer exposure will not greatly influence the general character of the exposure pattern.

Table 4. USES-estimated Multiroute Exposure profile for benzene

	resulting from emission as car fuel component	resulting from all other emissions	total exposure	relative importance (% of total exposure)
route of exposure	regional[1]	regional[1]	regional[1,2]	regional
air	42.8	7.3	50.1	97.32
drinking water	< 0.1	1.2	1.2	2.41
fish	< 0.1	0.1	0.1	0.19
plant shoots	< 0.1	< 0.1	< 0.1	0.06
plant roots	< 0.1	< 0.1	< 0.1	0.02
meat	< 0.1	< 0.1	< 0.1	< 0.01
milk	< 0.1	< 0.1	< 0.1	< 0.01
total	42.84	8.7	51.5	100.00

1 these columns contain absolute exposure in ng/kg bw/d.

2 figures in this column represent summation of exposure resulting from "car fuel additive" and "all other emissions".

3.4.4 benzene: comparison of measured verses predicted exposure pattern:

USES-generated estimates of concentrations of benzene in the human environment are presented in appendix VI.
In the criteria document (13) for various environmental compartments benzene concentrations have been mentioned. In fresh surface waters less than 100 ng/l has been found, whereas ground water may contain 3 to 30 ng/l (under "uncontaminated" soil). The concentrations in coastal sea waters are between 5-20 ng/l. Indoor concentrations in occupational air are given for situations abroad, only, which concentrations are 60 µg to 160 mg/m^3. In The Netherlands, indoor non-occupational concentrations are between 3 and 148 µg/m^3, apparently usually in the low µg/m^3-range. Average concentrations inside motor vehicles are 27 to 66 µg/m^3. Intake via cigarette smoke may be 10-30 µg/cigarette, which means that smoking 10 cigarettes a day would lead to a daily intake of 1.4 - 4.2 µg/kg b.w./ d. Ambient air concentrations depend strongly on the distance to sources. In built-up area concentrations (traffic and industrial emissions) are from 2 to 93 µg/m^3 (highest reported maximum in city centre) and reported to be higher than in rural areas: average values approx. 2 µg/m^3 and maxima 5.3 to 8.1 µg/m^3. These concentrations cannot be compared with results of the USES model, because in USES no procedure has been incorporated which estimates the environmental release of car fuel additives from exhaust pipes.
Slooff et al. (13) have estimated that the daily intake of benzene via food is probably less than 185 µg/d, corresponding to 2.6 µg/kg b.w./d. The benzene in food is probably largely originating from its natural occurrence in e.g. vegetable products. Benzene in food may also originate from its migration from packaging material, but this route of contamination is supposed to be declining (13). Intake via drinking water is probably in the low ng/d range. Thus exposure to benzene as a result of human activities (i.e. industrial and other emissions) will be largely through inhalation, whereas the routes of exposure via food and drinking water do not contribute to a large extent (i.e. exposure resulting from human activities). In line with Kennedy and Van der Schee (1986, cited in 13) one would even have to conclude that uptake of naturally occuring benzene via food is of much less relevance than indicated above. These authors assumed a lower natural benzene contents in food. In contrast to Slooff et al. (13), Wichmann et al. (5) do not consider contamination of food via packaging materials of major relevance.
The exposure pattern as estimated by USES is largely in concordance with the data above, whereas these data demonstrate the important role of traffic emissions for benzene exposure (cf. difference between built-up and rural areas). However, the estimation of the total daily uptake as calculated by USES is somewhat low. It should be noted that with the mathematical formulas in USES a continuous inhalatory exposure to 2 µg/m^3 (approx. the rural back-ground) can be calculated to correspond to a daily internal dose of about 400 ng/kg b.w./d. The total daily dose as mentioned in table 4 is only as high as 50 ng/kg b.w./d.

3.5 Benzo[a]pyrene

3.5.1 general information

Like many other polycyclic aromatic hydrocarbons, benzo[a]pyrene is a substance which is unintentionally formed during excessive heating of carbon-containing materials. These processes may be straightforward combustion of for instance car- or aircraft fuel or industrial processes like coke- and anode production for the aluminum industry. Thus benzo[a]pyrene is formed in many (industrial) processes as a waste material. As such it is usually emitted to air. The release of benzo[a]pyrene during forest-fires is not quantified and therefore not taken into account. Benzo[a]pyrene is also released while burning multi-burners, fireplaces and while smoking tobacco. In the latter case part of the emission is directly inhaled and reaches immediately its site of action i.e. the respiratory tract (14,15). The actual total formation of benzo[a]pyrene is not known. Industrial and use categories have not been assigned. The same is true for main categories at production, formulation and processing. Owing to this lack of information USES cannot perform an emission pattern estimation. The exposure pattern characterization for benzo[a]pyrene has therefore been accomplished on the basis of emission data provided by the Dutch Integrated criteria document on polycyclic aromatic hydrocarbons (14).This document provides a calculation of the total emission of benzo[a]pyrene to various environmental compartments which is based on data over several years. The authors of the document allow for an uncertainty in the emissions of a factor 2 to 10 (14).
While running USES, the choice of industrial and use categories is important. Based on these categories, apart from deriving emission estimates, USES also selects specific pathways while evaluating the environmental fate of chemicals, especially in the case of pesticides. Because no specific information was available, these categories both have been set to "others", meaning that no specific dispersion characteristics have been defined (4). Similar to the approach for benzene, emission data are directly inserted into the USES programme while performing an "intermediate evaluation". Emission to air (2.8 tonnes/y) and water (0.5 tonne/y) are assumed to occur at fairly constant rates throughout the year. The integrated criteria document (14) states only a total emission to soil (1.1 tonnes/y). For running USES it is assumed that half of this amount is released to industrial soil and the other half to agricultural soil.

Table 5. USES-estimated Multiroute Exposure profile for benzo(a)pyrene.

	resulting from emssions from an anode bakery	resulting from all sources	relative importance (% of total exposure)	relative importance (% of total exposure)
route of exposure	local[1]	regional[1]	local	regional
air	81.5	< 0.1	0.5	0.02
drinking water	0	0.5	0	0.81
fish	0	4.7	0	7.54
plant shoots	7526	1.5	45.8	2.54
plant roots	16.3	44.4	0.1	70.5
meat	4406	5.9	26.83	9.36
milk	4389	5.8	26.73	9.29
total	16420	62.9	100	100

[1] these columns contain absolute exposure in ng/kg bw/d.

In order to make an estimation of local exposure to benzo[a]pyrene, in the local emission submodule data for an aluminum anode bakery have been entered. Such a bakery releases 0.5 tonne of benzo[a]pyrene per year (14). In appendix IV an overview of other data for benzo[a]pyrene is given.

3.5.2 construction of the table with results for benzo[a]pyrene

Table 5 contains the results of the calculations for benzo[a]pyrene. No summation of data has been performed as regional exposure data are based on total benzo[a]pyrene emission and thus already include the exposure resulting from emissions of a local source (in this case an anode bakery).

3.5.3 exposure pattern of benzo[a]pyrene

The major route of exposure of humans to benzo[a]pyrene is via food (see table 5). At the local scale, about half of the daily dose is received through consumption of plants, especially the aboveground parts. Via meat and milk the rest of the daily dose is obtained. At the regional scale consumption of plant roots is the major route of exposure. Consumption of fish, milk, and meat are of considerable importance, as well, and roughly account for 10% of the total daily load.

In air and water benzo[a]pyrene is associated strongly with suspended matter. In air the free compound is easily degraded photo-chemically via reaction with OH· radicals. It is unclear to what extent photo-degradation is hampered by adsorption to aerosol particles (14). According to Baart (TNO-MW, personal communication, 1995) the $DT_{50,photodegradation}$ in air in Appendix IV is far to low (i.e. photodegradation is overestimated); which may be related to the aerosolic nature of benzo[a]pyrene in air. USES estimates the over-all DT_{50} in air for the free (i.e. not particle-associated) fraction of the chemical in air and performs further calculations with this over-all value; in other words it considers particle bound chemicals as photochemically inactive. In practice , for benzo[a]pyrene this results in virtually no photodegradation in air, because USES estimates that the free fraction of benzo[a]pyrene is very small.

3.5.4 benzo[a]pyrene: comparison of measured verses predicted exposure pattern:

USES-generated estimates of concentrations of benzo[a]pyrene in the human environment are presented in appendix VI.

Slooff et al. (14) have described the exposure pattern to polycyclic aromatic hydrocarbons (PAHs), among which is benzo[a]pyrene. Major route of exposure to benzo[a]pyrene is probably via food. However, the food components which contribute most have not clearly been specified. Exposure to PAHs via meat or fish is probably of relevance if these products are not properly prepared i.e. overheated or brought into contact with smoke of burning fat while barbecuing (14). Other real-life sources of exposure are passive and active smoking (even more than food) and indoor open fire-places. At the regional scale inhalation is quantitatively not an important factor in the exposure pattern.

Local scale data cannot conveniently be compared with actual data, as the criteria document (14) gives an estimate of the actual intake figure via air for a distance of 2000 m to the source (i.e. the anode bakery) while USES calculates exposure levels at a distance of 100 m from the source. Converting the actual intake figure according to the USES formulas results in an inhalatory uptake of about 0.3 ng/kg b.w./d, which is considerably less than the USES output. No data were provided for other routes of exposure at a local scale.

The assumption that half of the emission to soil reaches the industrial soil, while the other half reaches agricultural soil is probably an over-estimation for the agricultural soil. If the entire emission to soil is allocated to industrial soil (calculation results not shown), the total exposure of humans is decreased by a factor of two. This is largely owing to a reduced exposure via plant roots and animal products. Concentration in and exposure via air is hardly changed then and uptake via plant shoots is reduced by some 10%. If the concentration in agricultural soil is set to zero, only a minor additional decrease in the exposure via plant shoots is observed, while plant roots do no longer contribute to exposure to benzo[a]pyrene. This indicates that the overground plant parts receive most of their benzo[a]pyrene through other environmental compartments than soil and that contamination of soil with this chemical determines the extent of human exposure via plant roots. These modifications alter the exposure pattern for benzo[a]pyrene with respect to the relative importance of the various routes through food. Nevertheless, in both modified exposure scenarios routes via food are of paramount importance, with air and drinking water each representing < 1% of the total daily exposure.

The pattern described above, i.e. the majority of the exposure to benzo[a]pyrene is via food and inhalatory exposure is a minor pathway, has also been mentioned by Wichmann et al. (5) on the basis of actual environmental concentrations and an exposure/uptake model which is more elaborated than the corresponding modules in the USES programme. Although the actual data do not deny the exposure pattern as predicted by USES, they do not confirm it either. The major problem is the absence of information about the relevance of various diet constituents. As the diet seems to be of major relevance in the case of benzo[a]pyrene (see table 5), at least with respect to the total exposure, one would need more information to accept or reject the predicted pattern.

It should be noted that the most important toxic effect of benzo[a]pyrene is its carcinogenicity to the respiratory tract (5,14). The amount received via inhalation, though comparatively small, is probably of far more relevance than the amount received via food. This is demonstrated by the oral and inhalatory carcinogenicity data provided by Slooff et al. (14), which indicate that oral intake of 140 µg/d or inhalation of 70 ng/d both would both lead to the same additional cancer risk of 1 per 10^4 life-time exposed persons. Thus calculation of a total daily (internal) dose, which is compared to a total body MPC, results in an underestimation of the inhalatory risk.

3.6 Dichloromethane

3.6.1 general information

Dichloromethane is an extremely important chemical in industry. The chemical is mainly used as a solvent in a great number of products (e.g. paint strippers, glues) and in industrial applications as a degreaser, an extraction fluid (decaffeination), as a solvent in the production of lacquers, resins and plastics and as a solvent/reaction medium in chemical and pharmaceutical industry. It is also used as a propellant in aerosol spray-cans as a blowing agent for polyurethane foam and as coolant/refrigerant ("freon 30"; refs. 6,16,17).
Dichloromethane is produced in quantities of over 500000 tonnes/y through-out the world. Half of the total mondial production takes place in Europe (16,17).The total production capacity in The Netherlands amounted 15000 tonnes in 1985, a quantity which was believed to increase in the years thereafter. Dichloromethane is produced by chlorination of methane or methylchloride in installations which can also produce chloroform (tricholomethane) and tetrachloromethane (6). The total amount of dichloromethane used in The Netherlands is about 35000 tonnes/y; 19780 tonnes are imported each year (situation 1984), 500 of which are in formulated products. According to Slooff and Ros (16) about 27000 tonnes of dichloromethane are exported from The Netherlands each year. However, owing to rather large inaccuracies, the difference between inland import/production and export cannot be used to determine the emission of dichloromethane in The Netherlands (16).

Because of its wide-spread use and its many applications in either open or closed systems it is inconvenient to base an evaluation with USES on production and use quantities. Moreover, as more than half of the quantity used/produced in The Netherlands is exported subsequently in whatever from, these quantities may result in unrealistic emission estimates. Characterization of human multi-route exposure has therefore been performed

on the basis of the emission estimates as presented by Slooff and Ros (16). These estimates provide an indication of the annual regional environmental load with dichloromethane. Regional industrial emissions of dichloromethane amount to 3853 and 68 tonnes/y to air and water, respectively. Industrial emissions to soil are negligible. Diffuse sources (e.g. resulting from consumer products) amount to 2910 and 10 tonnes/y (air and water). Slooff and Ros (16) have assumed that owing to application of paintstripper in private households some 100 tonnes of dichloromethane are emitted to soil (not specified with respect to industrial, or agricultural use). In reality this amount is probably dumped on household waste-deposits or incinerated. In order to take this amount into account in USES it will be assumed to be dumped on industrial soil, thus allowing for evaporation to air and leaching to groundwater, while excluding direct uptake by crops and pasture, which would occur if emissions would have been allocated to agricultural soil.
The evaluation at the local scale has been performed for the (only) production plant in The Netherlands (not necessarily the largest environmental source). Emission data for this plant are taken from the built-in emission tables in USES. It is assumed that the chemical is produced in dedicated equipment which requires limited cleaning. Plant emissions have been assumed to occur at "production" only.
Owing to its wide-spread use and its applications, exposure to dichloromethane is very likely to occur in occupational situations. At the Maximal Accepted Concentration (350 mg/m^3; ref. 18), daily inhalatory absorption of dichloromethane would be about 21 mg/kg b.w./d (weekly average; continuous exposure during 5 working days of 8 hours/week). Assuming 200 working days/y at this level (350 mg/m^3), the yearly average uptake can be calculated to be 11.5 mg/kg b.w./d.

In USES a scenario for inhalatory exposure to consumer products has been incorporated and this scenario is not suitable for the calculation of the exposure to dichloromethane. This scenario has been primarily developed for rapidly volatizing or gaseous compounds or small particles and not for chemicals which escape from a solid or liquid matrix. It has nevertheless been applied for 1,4-dichlorobenzene (a solid) and dichloromethane in paintstripper (jelly) with the restriction that rate of the volatisation has to be estimated or rather guessed.

An indication of consumer exposure to dichloromethane may be calculated for a private person applying 1 kg of paint stripper in a room of 20 m^3. Paint strippers may contain as much as 50% dichloromethane (18). For other exposure parameters the reader is referred to appendix V.

Table 6. USES-estimated Multiroute Exposure profile for dichloromethane.

	resulting from a production plant	resulting from all diffuse sources	resulting from all industrial sources	total exposure	relative importance (% of total exposure)	relative importance (industrial plus diffuse sources; % of total exposure)
route of exposure	local[1]	regional[1]	regional[1]	regional[1,2]	local	regional
air	24460	18.5	24	42.5	56.98	98.8
drinking water	16300	< 0.1	0.4	0.46	37.97	1.08
fish	1434	< 0.1	< 0.1	< 0.1	3.34	0.09
plant shoots	479	< 0.1	< 0.1	< 0.1	1.12	0.03
plant roots	254	< 0.1	< 0.1	< 0.1	0.59	0.01
meat	< 0.1	< 0.1	< 0.1	< 0.1	< 0.01	< 0.01
milk	< 0.1	< 0.1	< 0.1	< 0.1	< 0.01	< 0.01
total	42930	18.6	24.4	43.06	100	100

[1] these columns contain absolute exposure in ng/kg bw/d.

[2] this column contains summed data from both other

The present configuration of the consumer exposure module does not accept the consumer exposure data for dichloromethane as given above and presented in appendix V. It cannot properly deal with low exposure incidence and large quantities of product. A possible consumer exposure scenario for dichloromethane has therefore been worked out by hand. Based on the exposure scenario as mentioned above and in appendix V and assuming rapid evaporation of dichloromethane from the paint stripper with no ventilation, a concentration of 250 mg/m^3 can be calculated. Estimation the uptake applying the same formulas and defaults as USES gives an exposure of 5.6 mg/kg/d on the days the paint stripper has been used. The annual average, whether appropriate or not, corresponding with this value is 31 µg/kg b.w./d, a value which may be compared with the other results obtained for dichloromethane.

3.6.2 construction of the table with results for dichloromethane

The table with the results for dichloromethane (table 6) provides exposure data for local and regional exposure to this substance. For this chemical two types of regional sources have been distinguished. Thus, two regional exposure scenarios have been evaluated. The results of these two scenarios have been summed and the results are presented in the column "total exposure".

3.6.3 exposure pattern of dichloromethane

From table 6 it can be concluded that (at least for the regional exposure estimations) virtually all dichloromethane enters the body via air. This is not very surprising for such a volatile chemical. The local exposure scenario, which is based on a USES-generated emission pattern, indicates that in the proximity of an industrial source a considerable part of the total body exposure may enter the body via water. This difference between regional and local exposure patterns arises from the comparatively low emissions to surface water in the regional pattern, whereas in the local model half of the total emission flows into the waste water stream. The efficiency of the local STP in the USES model in reduction of dichloromethane flowing to surface waters is not as high as in reality. The model calculates 74% of the dichloromethane input in the STP to volatilise to air. In reality this percentage may be as high as 95% (16). For consumers the inhalatory exposure to dichloromethane exceeds the total exposure to dichloromethane at regional scale by far. Workers are exposed to levels even higher than locally exposed people.

3.6.4 dichloromethane: comparison of measured verses predicted exposure pattern:

USES-generated estimates of concentrations of dichloromethane in the human environment are presented in appendix VI.

In the integrated criteria document Slooff and Ros (16) have estimated exposure levels of the general population to dichloromethane. The majority of the Dutch population experiences an outdoor concentration of 600 ng/m^3 in air corresponding to 128 ng/kg b.w./d (calculated with the USES algorithms). This value is about three times higher than predicted by the USES model (one should compare 128 ng/kg b.w./d with the sum of both regional exposure levels). Slooff and Ros (16) do not provide exposure data to dichloromethane in food but indicate that in drinking water the concentration is usually less than 100 ng/l. In the USES model this would lead to a daily uptake of 3 ng/kg b.w./d. These data support the USES prediction (see table 6) that inhalation of dichloromethane is the prime route of exposure. It should be noted that exposure to dichloromethane resulting from either consumer products or occupational sources has not been incorporated in the calculations. Furthermore, Slooff and Ros (16) show that indoor air and "local air" may be more severely contaminated with dichloromethane than regional ambient air.

3.7 Formaldehyde and tetrachloroethylene

Considering the results for the previous compounds it is concluded that it is likely that for these two chemicals the major route of exposure will be inhalatory.

It should be noted that dermal contact with and uptake of formaldehyde cannot be excluded because formaldehyde is an allowed constituent in cosmetics (e.g. shampoo). Extra indoor exposure to formaldehyde may result from incomplete combustion of methane while preparing meals, from smoking, or from slow release by resins in e.g. chipboard (20).

Like with benzo[a]pyrene formaldehyde has its major effects at the site of body entrance (i.e. the respiratory tract; ref. 20). It can thus be concluded that a calculation of the total body dose is not very relevant.

The major source of exposure of the general population to tetrachloroethylene is believed to result from the release of this chemical into air after use in dry-cleaning shops and laundries (21). Extra (occupational) exposure will take place in such places as well. Another source of emission of tetrachloroethylene to air is the metal industry in which it is used as a degreasing agent. Taking into account the high volatility of tetrachloroethylene, and its major emission to air (about 90% of the total emission; ref. 21), evaluation with USES will most certainly produce an exposure pattern similar to that of dichloromethane.

3.8 Lead and cadmium

These two chemicals have not been further evaluated with the USES programme. For reasons discussed in Section 4.2 such a calculation does not provide a meaningful result.

4. DISCUSSION

This report focuses on the applicability of the USES risk assessment approach for determination and characterization of human multi-route exposure patterns to chemicals. The applicability has been tested with a small number of model substances for which exposure evaluations have been carried out by means of the USES computer model. The research project aimed at the recognition of limitations of the USES approach, which may hamper its applicability within the framework of Setting Integral Environmental Quality Objectives (IEQO) or the programme Humans and Environment (H&E). While analyzing the model performance, various limitations have been encountered, part of which have been mentioned in the USES report, too. These limitations are subdivided into two classes "operational": referring to working with the programme itself, and "theoretical", which are connected to the scientific background of the results.

Despite these limitations, which will be discussed in more detail in sections 4.1 and 4.2, for the chemicals for which an exposure evaluation could be carried out, the estimated multi-route exposure pattern did not deviate to a large extent from the patterns which could be derived from field data. The differences between the field data derived patterns and the USES generated patterns are probably well within the uncertainty limits of either USES or the field data or both (personal communication Jager, 1995). This means that if the USES approach is supplemented to alleviate some of the limitations mentioned, the extended approach is suitable to derive IEQOs based on human toxicity data. This is even more so if as many as possible actual emission data are used as starting points. At least exposure through direct uptake via contaminated soil should be incorporated into the procedure.

It should be noted that the degree of similarity between the predicted and actual exposure patterns seems higher than the degree of similarity between actual and predicted environmental concentrations. This might indicate that prediction of environmental concentrations, but not the ratios between them, has higher uncertainty than the prediction of the resulting patterns of multi-route exposure, which patterns depend on the ratios between the environmental concentrations rather than their actual numerical values. This is especially ture for the regional exposure patterns. Local exposure patterns, which may differ from the regional ones, could not be compared with actually determined data.

The derivation procedure might follow the lines of a "trial and error" approach, in which in a first step a preliminary estimation of a total daily body exposure is produced from physico-chemical data and, among others, emitted quantities. The ratio of the estimated exposure versus a tolerable exposure level is the factor with which environmental concentrations in the USES model should be altered to arrive at an exposure which does not exceed the tolerable level. These concentrations could be considered as the $\text{MPCs}_{\text{human}}$ for the various environmental compartiments.

For the H&E programme, USES can provide an indication of a multi-route exposure pattern for the general population. As this programme focuses also on sub-populations and on spatial and temporal variation, next to USES new models or approaches have to be developed or existing ones have to be implemented.
An extension which allows USES to accommodate metals and other substances which cannot be evaluated up to now, is a major one. Such an extension is too complicated to be implemented in the short run. Therefore, for such chemicals the USES approach is not appropriate, and complementary models should be used.

The limitations as indicated above will be discussed in the following paragraphs. Between square brackets the relevance for either IEQO or H&E will be indicated in the text. Finally, over-all conclusions will be presented.

4.1 Operational limitations

While testing the USES model, several technical limitations have been encountered which had to be solved before meaningful evaluation of exposure patterns could be carried out. The recognition of these limitations originates from an abundance rather than a lack of data. This is especially so when substances are used for many purposes and for all different applications quantitative date are available. In case of such an abundance, one would like to perform an evaluation on the basis of real-life information. However, USES does not always conveniently permit the input of real-life data.
Especially for compounds with a complex use pattern e.g. dichloromethane or benzene, it is difficult to generate an "over-all" emission pattern. Ideally for each type of use the amount involved is known and an "over-all" emission pattern can be generated in a number of separate runs. The various emission patterns obtained in that way can be added up to a total emission pattern. As environmental concentrations depend linearly on the emission volumina, it is even possible to add up the final environmental concentrations resulting from each use (at least if these concentrations are annual averages; not if they represent e.g. emission episode averages in local scale surface water scenarios).
Conveniently, for all example chemicals in the present study, over-all emission patterns could be found in literature and the predicted exposure routes are more or less a result from actual data. For chemicals, for which emission data are lacking, the emission patterns have to be estimated by USES via a realistic worst-case approach. The predicted exposure

patterns for such chemicals may be less reliable than the ones based on actual emission data [limitations for H&E and IEQO].
For at least two test substances (atrazin and 1,4-dichlorobenzene) production does only take place abroad and does therefore not result in regional emissions (i.e. within The Netherlands). Thus within the USES framework, production leads to continental emissions, whereas formulation and use may lead to both continental and regional emissions. It is, however, not possible to evaluate such a situation in a single run in USES. One might consider to evaluate such complex situation in separate runs, one at continental scale and one at regional scale: the multi-run approach. However, in a single-run evaluation, the calculated continental environmental concentrations are used within USES to estimate continental back-ground concentrations and to calculate trans-border migration of substances. This trans-border migration contributes eventually to regional environmental concentrations. In a multi-run approach the trans-border transport of substances is not properly accounted for.
For the moment it is impossible to correct regional environmental concentrations for trans-border migration, for instance by adding continental concentrations to the regional ones, which are each calculated in separate runs. Such a summed value is not equal to the regional concentration in for instance air when calculated in a single run. This problem may be circumvented by determining continental environmental concentrations in a first run, followed by an "intermediate evaluation" for regional scale exposure during which continental environmental concentrations are entered. For none of the test-substances discussed in this paper, this alterative approach has been followed. It is expected that the exposure pattern based on regional emissions, only, will not differ greatly from an exposure pattern based on both regional and continental emissions [Vermeire, personal communication, 1994]. However, if continental emissions are left out, USES assumes continental concentrations to be zero and it calculates with this value. Trans border migration thus becomes a "continental sink". This is in particular relevant for the compartments air and water [limitation for H&E rather than IEQO].

Another technical problem arises when a chemical is produced or/and imported in the region under study and/or (partly) exported. USES cannot assign separate volumina to commercial traffic. One could consider import and export as special cases of the technical problems described above for trans-border migration. In case of import one is confronted with the situation of production abroad and use at home, while export might be considered as a separate "use category" with no domestic emissions. The latter situation gets even more complicated if a chemical is e.g. formulated before shipment. However, import and export of chemicals will probably influence absolute exposure levels to a higher extent

than the exposure profile. So for the derivation of IEQOs and determination of multi-route exposure patterns in general, export and import may not be very relevant.
The conclusion from the paragraphs above is that it is far more convenient to enter emission data into USES than to follow the present approach via which USES generates emission pattern estimates guided by production or other data. Application of realistic emission data further improves the quality of the final results, as emission volumina and calculated average environmental concentrations are linearly linked. For new substances, for which only a base set of data but no realistic emission data are available the USES approach provides a means to fill the various data gaps.

A number of other important operational limitations encountered during the research are given in table 7. Some of these limitations may affect the program's results, while others may hamper a full understanding of the meaning of the results.

Table 7: List of operational limitations of USES

limitation	relevant for
The estimation of local exposure is based only on local emissions and not on local + regional emissions. In other words USES does not take into account regional back-ground concentrations at the local scale. Possibly this limitation does not influence the estimated exposure pattern to a great extent.	H&E rather than IEQO
Data available cannot always be conveniently entered into the program: e.g. a DT_{50} in air may be the result of various processes such as direct photolysis or chemical reaction with OH· radicals or ozone (resulting from photochemical activity). If only reaction rates with OH· radicals are known formally no data can be entered here, though these reactions can be very efficient.	H&E and IEQO
USES provides only final (steady state) concentrations in the output. However, knowledge of inter-compartmental distribution might be of benefit in emission reduction decision making; e.g it is not very efficient to reduce emissions to water if the majority of concentrations in plants originates from emissions to air.	H&E
The association of chemicals in sub-compartments, e.g. rain water or suspended particulate matter is not visible in the USES output.	H&E
The emission module has no possibility to generate an emission pattern for chemicals to which industrial or and/or use categories cannot be assigned. Neither is it possible to define a chemical as a "waste product". e.g.: benzo[a]-pyrene. Thus such chemicals can only be evaluated if actual emission data are available.	H&E and IEQO

From the report it is very difficult to determine whether results are estimations of an "average" exposure scenario or of "realistic worst case" calculations and assumptions. In general the model aimes at of "realistic worst case" estimates. It would enhance insight in the value of the results, if for the many calculations and defaults the status "avareage value" or "realistc worst case calculation" was made more explicit.	H&E
The actual exposure profiles do not appear in the *printed* output. This is not really a limitation but in the light of IEQO or H&E it is quite inconvenient.	H&E and IEQO
For local exposure scenarios only the life-cycle step with the largest emission to water is taken into account. If a full evaluation of a local exposure situation is desired, the separate life-cycle steps and their respective emissions have to be evaluated in separate runs, the results of which have to be added up manually.	H&E

4.2 Theoretical limitations of the USES models

Apart from the technical limitations mentioned above a number of limitations have been encountered which may reduce the applicability of the programme results in various aspects.

For people who are indirectly exposed, USES generates exposure estimates at two scale levels, one local and one regional. According to the USES report (4) the programme is not suitable for the evaluation of regional exposure to chemicals which are emitted from a small number of sources. Sophisticated local models are then probably more appropriate [relevant for H&E].

Within USES, regional and continental environmental compartments are considered as homogenous boxes. However, for most chemicals strong concentration gradients occur. This is for instance the case with 1,4-dichlorobenzene in water in the river Rhine (12). For other chemicals e.g. benzene or benzo[a]pyrene (13,14), which are very diffusely emitted still recognisable differences in exposure may exist between various locations; in these cases rural versus built-up areas. The application of agricultural pesticides is related to the growth of specific crops, which in the case of atrazin are dominantly maize or asparagus. These crops are grown in southern and eastern parts of The Netherlands, mainly and thus in these parts atrazin environmental concentrations are higher than in other parts of the country (8). These examples indicate that the assumption of homogenous environmental compartments is rather hypothetical and that spatial variation in environmental concentrations is an important source of inter-individual variability in exposure levels [relevant for H&E].

Apart from spatial variation in exposure temporal variation should be recognised as well. Pesticides like atrazin are usually applied during a distinct period, in the case of atrazin shortly after plant emergence, giving rise to peak levels on a usually comparatively low back-ground level (cf. ref. 8) Currently, with respect to human exposure this kind of variability cannot be studied with the USES programme [relevant for H&E and for specific substances (e.g. inhalatory irritants) for IEQO, as well].

The local and regional exposure patterns are supposed to be representative for continuous exposure (4) It should be noted that exposure to one continuous concentration e.g. in air is hardly realistic. Even changes in wind direction may affect concentrations, especially at the local scale.

Another reason for temporal variation, which is linked to spatial variation is human behaviour. One could consider this at both short-term e.g. diurnal or at long-term e.g. life. A short-term pattern for exposure to nitrogen dioxide has been described by Ryan (22) who shows that daily exposure to NO_2 is characterized by two peaks which result from preparing meals and from commuting. A similar temporal variability has been found for benzene, benzo[a]pyrene and 1,4-dichlorobenzene, for which even marked interindividual exposure pattern differences exist (cf. smokers vs. non smokers, commuters vs. non-commuters, consumers vs. non-consumers) (5,12,13,14). The procedures which averages intermittent exposures in time, assuming that the product of exposure and time gives a constant toxicological response, is, though it is the most convenient available, not necessarily scientifically sound (23).

Both life-time and short term variability have been accounted for by Wichmann et al. (5), who have modelled a person's life by sub-dividing it in seven different age-stages, each stage having its own behavioral and other personal characteristics. Such an age-related differentiation of spatial and temporal variation might be of relevance for the absolute exposure data and it may shed light upon variability between the exposure patterns related to the different life stages. The system of Wichmann et al. (5) has only been used to determine average life-time exposure and has not been used to make a detailed inventory of these life stage exposure patterns. In the C-soil model (24) a person's life is sub-divided into only two stages. This model predicts that, in the case of soil pollution, oral ingestion of soil contributes to the exposure of children to a large extent, at least for compounds with low volatility.

As age-related differentiation is based upon (new) assumptions with their own uncertainties the outcome of such an "improved" model need not necessarily be more accurate than the outcome of the present models i.e. USES. It may be more beneficial to perform an exposure assessment taking into account various sources of variability and their uncertainties in order to generate a probability distribution of exposure over the population (cf. ref. 25).

According to the USES report (4) not all chemicals can be properly evaluated with this computer programme [limitation for H&E as well as IEQO]. The system is not directly suitable for the risk assessment of inorganics, surfactants and ionized/ionizing compounds. These problems are among others connected to the fact that for these chemicals no simple K_{ow} can be defined. Additionally, metals and ionizing compounds may occur in the environment in more than one form, each having its own physico-chemical properties. This means that USES is not applicable for at least two of the selected chemicals (lead and cadmium).
Some of this limitation may be overcome by entering the K_p value. Direct input of partition coefficients (e.g. between water and soil or water and suspended matter) may be applied to USES, but than at best, the programme can provide meaningful estimations of concentrations in water and soil, only. However, inter-compartmental exchange of metals and ionizing chemicals is still incompletely modelled [personal communication, Van de Meent, 1994] and the distribution of chemicals over the various sub-compartments is not visible in the USES output. In addition, submodules which are entirely depending on K_{ow}, such as those on bioaccumulation, will still produce erroneous results if K_p values are used instaed of K_{os}s. Conversely, if a chemical occurs predominantly in the non-ionized status at environment pH, which is for instance the case with atrazin (pKa = 1.7; ref. 9), an evaluation with USES is very well possible [personal communication, Vermeire, 1994].
A rough impression of the pattern of exposure to metals can be obtained by evaluation of data generated by the soil-pollution evaluation programme C-soil, which shows that for metals and compounds with low volatility in general, ingestion of soil may be an important route of exposure (24).
The fundamental differences between C-soil and USES is that USES is designed to provide a uniform means for risk evaluation, whereas the C-soil model is rather designed to generate soil pollution intervention values and C-soil, being a generic model, is not suitable for risk assesment for a specific situation of soil contamination. C-soil does not take into account industrial emissions but assumes a local soil contamination as the infinite source of exposure. Based on equilibrium calculations and estimations of inter-compartmental exchange-rates the pattern of the total exposure of humans (children and adults, separately) is determined. It should be noted that the environmental compartments in C-soil are very small scale indeed, even smaller than the local scale compartments in the USES programme.

Apart from the limitations mentioned above, some other problems were encountered, which have been summed up in table 8 without further discussion. Some of these can be found in the USES report, too (4).

Table 8. List of theoretical limitations of the USES program

limitation	relevant for
USES produces estimates of the exposure to the primary compound, only. The model does not account for possible (persistent) biotransformation or degradation products. This is especially important if such products have a relevant biological activity.	H&E rather than IEQO
In- and uptake via ingestion of soil and dermal absorption of substance out of (drinking) water have not been incorporated into the programme. These routes may be of greater importance than assumed up to now (24,26,27).	H&E and IEQO
The programme estimates only the exposure for an adult person, who is fairly inactive. The default inhalation rate of 20 m^3 air/d is intermediate between the value for resting and the value for light activity (5).	H&E
Local emission of car fuel additives cannot be properly evaluated. Emissions at the production plant can be assessed, but not emissions from traffic itself.	H&E
The computation of a total internal daily dose is not per definition relevant. The present approach in USES can be valid for systemic effects, only (cf. ref. 5). Thus, the calculated multi-route exposure patterns should be evaluated in the light of the most relevant toxicological effect. For some of the selected chemicals the relevant effect depends only on exposure via one single route. For such chemicals and effects detailed knowledge of multi-route exposure is redundant. This the case for some inhalatory carcinogens.	H&E and IEQO
Exposure via substances in air has only been modelled for ambient air. For non-consumers, the concentrations of a substance in indoor and outdoor air are implicitly assumed to be equal. Only for consumer products indoor scenarios have been implemented.	H&E
The model estimates exposure to substances through fish on the basis of fresh water concentrations. This is not particularly relevant in the Dutch situation in which predominantly sea fish is consumed.	H&E and IEQO
The concentration in fish is calculated from the concentration in surface water, defined as 32 x diluted STP-effluent. Dilution of STP-effluent may vary within 5 orders of magnitude.	H&E and IEQO
Human food intake is a reflection of the average consumption pattern. This may lead to underestimation of the exposure of certain population groups e.g. milk consumption in children is higher than in an average person, especially when related to body weight (4,28).	H&E

In this first approach no special attempt has been made to quantify the uptake of individual compounds in the human body. The total exposure data in tables 2 through 6 refer however to internal exposure. The internal exposure has been calculated using the default absorption coefficients from USES (0.75 for inhalatory exposure, unity for oral exposure routes). These values have been taken from an overview of harmonised model parameters (28). It is very well possible that compounds to which one is exposed via air for a very short period are absorbed with much higher efficiency (up to 90%) while for continuous inhalatory exposure much lower values (40 to 50%) are probably appropriate [personal communication, Van Veen, 1994; Wichmann et al., 1993]. Thus for continuous exposure, the inhalatory absorption coefficient of 0.75 might be too high. Likewise, the coefficient of unity for oral exposure is probably rather 'worst-case" for many chemicals. Additionally, in the regional and local scale models, USES does not take into account that a substance may be absorbed to particulate matter, which may greatly influence its bioavailability.

4.3 over-all conclusions

With respect to the general population, the exposure patterns calculated with USES seem to be appropriate for the determination of IEQOs (cf. chapter 1) for the type of compounds USES has been developed for. This conclusion is mainly based upon the fact that for none of the example substances which have been studied, major differences between real-life and computer-predicted patterns were found, although actual data are not always as detailed as the USES-generated patterns and do not for instance give insight in differences between local and regional exposure profiles. However, the resolution of the programs results with respect to temporal, spatial or individual variability is very limited. Therefore, USES may be applied within the frame work of derivation of environmental quality objectives aiming at life-time protection of the population at large, i.e. the "avarage Dutchman". For people who live in the proximity of a emission source, the multi-route exposure pattern can differ from the "average" situation. For such situations it may be desirable to reconsider the applicability of the IEQOs derived for the general population. For the H&E programme it can only be seen as a starting point. Complementary to USES a number of estimation procedures has to be developed in order to account for variability at the spatial, temporal or individual scales. Taken together these procedures may represent a support and decision system within the framework of determination of health risks (i.e. the H&E program) in which the exposure of particular groups or even individuals has to be taken into account.

It should be noted that as many real-life data as possible have been used to perform the exposure evaluations. As the performance of USES seems to be rather sensitive to emission input data, this may have positively influenced the program's results.

It is expected that IEQOs will have to be derived for many chemicals. Thus exposure mapping will have to be performed for many environmental contaminants. Therefore, the availability of a proper generally applicable operating procedure would be very helpful. One option to do this is by classification of substances with respect to physico-chemical characteristics followed by assigning standardized exposure patterns to the various classes. In view of the time required to perform an evaluation for a particular substance, it is not a major problem to evaluate each individual compound separately, provided that data have been collected in advance and the number of substances is limited. In absence of actual emission data the defaults emission profiles in USES, which are based on volatility,. solubility in water, production volumina and use pattern, can always be applied. The results of the present study more or less confirm the assumption that for volatile substances the major route of exposure is inhalation. In a previous study this assumption has been the starting point in the derivation of IEQOs for many such chemicals (3). However, such a generalised approach only focuses on volatility and not on solubility or emission profile. A more elaborate sensitivity analysis of USES may indicate to which extent such a generalised approach is indeed applicable and at which cut-off points a more substance-specific approach has to be followed. Moreover, whether or not a more generalized approach is finally required (and possible), can be judged more adequately after determination of multi-route exposure patterns for e.g. several tens of compounds.

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    benzene: update: 18/11/93
    benzo[a]pyrene: update: 17/1/94
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APPENDICES

Appendix I: Data for atrazin

type of data	value	source
Continental production / formulation volume: use Netherlands formulation: use:	 17000 tonnes/y 6000 tonnes/y 1000 tonnes/y 200 tonnes/y	 (1) (2) (3) (3)
use and industrial categories	agricultural chemical, pesticide,	(1,3)
main categories at: production formulation (Netherlands) processing	 batch production multipurpose equipment dedicated equipment limited cleaning wide-dispersive use	 default USES (3) default USES
melting point	173 °C	(1)
vapour pressure	4×10^{-5} Pa	(4)
log K_{ow}	2.34	(1)
solubility in water	30 mg/l	(1)
Henry's law constant / air-water partition coefficient	2.843×10^{-3} Pa·m^3/mol 1.2×10^{-7} l/l	estimate USES (4)
DT_{50} photodegradation, air	1.04 d	(1)
DT_{50} hydrolysis, water	86 d	(pH = 5-9; ref 5)
DT_{50} biodegradation water	77 d	(minimum value; ref 5)
DT_{50} biodegradation soil	50 d	(5)
DT_{50} biodegradation STP	77 d	set equal to DT_{50} biodegradation water.
bioconcentration factor (fish)	5	(5)

soil-water partition coefficient	2.608 l/kg	estimate USES
sediment-water partition coefficient	2.608 l/kg	estimate USES
suspended matter-water partition coefficient	8.992 l/kg	estimate USES

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Appendix II: Data for 1,4-dichlorobenzene

type of data	value	source
Continental		
production	49000 tonnes/y	(1)
use	27400 tonnes/y	(2)
Netherlands formulation / use	326 tonnes/y	(3)
industrial category	personal and domestic use	[Vermeire; pers. commun., 1994[1]]
use categories	odour agent	(1,2)
main categories at:		
production	intermediates: stored off-site	default USES
	odour agent: batch production multipurpose equipment	default USES
formulation	multi-purpose equipment	default USES
processing	intermediates: non-dispersive odour agent: wide-dispersive	default USES
melting point	53 °C	(1)
vapour pressure	90 Pa	(1)
log K_{ow}	3.4	(1)
solubility in water	79 mg/l	(1)
Henry's law constant /	160 Pa·m^3/mol	(4)
air-water partition coefficient	0.0675 l/l	estimate USES
DT_{50} photodegradation, air	3 d	(1)
DT_{50} hydrolysis, water	10^6 d	default USES
DT_{50} biodegradation water	3 d	estimate USES
DT_{50} biodegradation soil	12.5 d	estimate USES
DT_{50} biodegradation STP	0.009627 d DT_{50} calculated by USES, based on ready biodegradability	(1,5)

bioconcentration factor (fish)	120	derived from various sources[2]
soil-water partition coefficient	29.94 l/kg	estimate USES
sediment-water partition coefficient	29.94 l/kg	estimate USES
suspended matter-water partition coefficient	103.2 l/kg	estimate USES
extra data for consumer exposure:		
time scale of consumer exposure	chronic	assumed life time use of toilet blocks
weight fraction in product	50%	"average" value of percentage of 1,4-dichlorobenzene in consumer products
number of events	3 /day	arbitrarily estimated
quantity of product used	200 mg	arbitrarily estimated
exposure time	9 min /event	estimated toilet resident time
volume of a toilet room	5 m^3	"standard" toilet: (1.4 × 1.4 × 2.5 m)

1 HEDSET (1) categorises 1,4-dichlorobenzene in industrial category 3: chemical used in synthesis. This classification does not result in emission owing to private use. Therefore the classification as mentioned in the table has been applied.

2 Several estimates were found in the range of 50 to 200. One of the tests provided a bioaccumulation factor of about 800 (1). Based on a Log K_{ow} of approx. 3.4, USES estimates the bioaccumulation factor to be 125. In combination with the experimental data a value of 120 has been selected for running USES for 1,4-dichlorobenzene.

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Appendix III: Data for benzene

type of data	value	source
Continental		
production	3800000 tonnes/y	(1)
use	6400000 tonnes/y (1984)	(2)
Netherlands		
production	930000 tonnes/y (1981)	(1)
use	1026000 tonnes/y (1981)	(1)
industrial category	chemical used in synthesis	(1,2,3)
use categories	intermediate car fuel additive	
main categories at: classification not used; calculations for benzene have been performed on the basis of realistic emission data		
melting point	5.5 °C	(3)
vapour pressure	10^4 Pa	(3)
log K_{ow}	2.19	(3)
solubility in water	1780 mg/l	(3)
Henry's law constant / air-water partition coefficient	550 Pa·m^3/mol 0.2321 l/l	(4)
DT_{50} photodegradation, air	5.3 d	(1)
DT_{50} hydrolysis, water	10^6	default USES
DT_{50} biodegradation water	5 d	estimate USES
DT_{50} biodegradation soil	0.3208 d	estimate USES
DT_{50} biodegradation STP	0.009627 d DT_{50} calculated by USES, based on ready biodegradability	(1,5)
bioconcentration factor (fish)	7.204	estimate USES
soil-water partition coefficient	1.846	estimate USES

sediment-water partition coefficient	1.846	estimate USES
suspended matter-water partition coefficient	6.366	estimate USES
extra data for consumer exposure: consumer exposure may occur; e.g. at petrol filling stations or during painting. Detailed information is not readily available.Therefore, consumer exposure has not been estimated.		

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Appendix IV: Data for benzo[a]pyrene

type of data	value	source
Continental production use Netherlands production/ use	Precise production values are unknown, Emission data are available.	
industrial category use categories	both not assigned, assumed "others"	
main categories at: production formulation processing	not assignable, assumed "others"	
melting point	179 °C	(1)
vapour pressure	1.3×10^{-8} Pa	(2)
log K_{ow}	6.06	(1)
solubility in water	0.15×10^{-3} mg/l	(2)
Henry's law constant / air-water partition coefficient	0.0219 atm.m^3/mol 9.3×10^{-6} l/l	estimate USES (2)
DT_{50} photodegradation, air	0.067 d	(2) derived from OH· reactivity in air
DT_{50} hydrolysis, water	1×10^6 d	default USES
DT_{50} biodegradation water	1000 d	default USES
DT_{50} biodegradation soil	3.909×10^5 d	default USES
DT_{50} biodegradation STP	10^6 d	default USES
bioconcentration factor (fish)	53400	estimate USES
soil-water partition coefficient	13680	estimate USES
sediment-water partition coefficient	13680	estimate USES

suspended matter-water partition coefficient	47190	estimate USES
extra data for consumer exposure: consumer exposure may occur; e.g. while smoking, eating barbecued food or burning open fire places. Data are not readily available and therefore consumer exposure has not been estimated.		

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Appendix V: Data for dichloromethane

type of data	value	source
Continental production use Netherlands production/ use	370000 tonnes/y (1985) 15000 tonnes/y	(1) (1)
industrial category use categories		
main categories at: production formulation processing		
melting point	- 95.1 °C	(2)
vapour pressure	435 hPa	(3)
log K_{ow}	1.25	(2)
solubility in water	13 g/l	(2)
Henry's law constant / air-water partition coefficient	271 Pa·m^3/mol 0.126 l/l	(3) [estimate USES, value according to (4) : 0.13]
DT_{50} photodegradation, air	57.3 d	(5) derived from OH· reactivity in air
DT_{50} hydrolysis, water	600 d	(3)
DT_{50} biodegradation water	7 d	(2)
DT_{50} biodegradation soil	17.48 d	estimate USES
DT_{50} biodegradation STP	0.5 d	derived from (2)
bioconcentration factor (fish)	8	(2)
soil-water partition coefficient	0.212	estimate USES
sediment-water partition coefficient	0.212	estimate USES

suspended matter-water partition coefficient	0.7309	estimate USES
extra data for consumer exposure		
weight fraction in product	50%	(6)
number of events	2/y	arbitrarily chosen
quantity of product used	1 kg	arbitrarily chosen
exposure time	2.5 h	arbitrarily chosen

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Appendix VI: USES-predicted regional environmental concentrations of the model compounds.

environmental compartment	intake medium	unit	atrazin[a]	1,4-dichlorobenzene[b]	benzene[c]	benzo[a]pyrene[d]	dichloromethane[e]
air	air	ng/m^3	0.17	1.28	234	0.07	199
surface water		µg/l	0.22	< 0.01	0.09	< 0.01	0.03
pore water		µg/l	2.37	< 0.01	< 0.01	0.02	< 0.01
natural soil		µg/kg	0.49	< 0.01	< 0.01	106.3	< 0.01
agricultural soil		µg/kg	5.09	0.02	< 0.01	174.9	< 0.01
industrial soil		µg/kg	0.49	23.4	0.23	12110	4.94
	drinking water	µg/l	2.37	< 0.01	0.04	0.02	0.02
	fish	µg/kg	2.87	< 0.01	0.63	30.22	0.26
	plant shoots	µg/kg	5.10	< 0.01	< 0.01	0.27	< 0.01
	plant roots	µg/kg	6.48	< 0.01	< 0.01	25.080	< 0.01
	meat	µg/kg	< 0.1	< 0.01	< 0.01	3.423	< 0.01
	milk	µg/kg	< 0.1	< 0.01	< 0.01	1.082	< 0.01

a: owing to "processing as a pesticide"; b: owing to "formulation + use as an odour agent"; c: owing to "car fuel additive" + "all other emissions"; d: owing to "all industrial sources"; e: owing to "diffuse" + "industrial" sources.