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**The accumulation of soil contaminants in crops,  
location-specific calculation based on the CSOIL  
module**

Part 1 Evaluation and suggestions for model  
development

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This investigation has been performed on commission of the Ministry of Housing, Spatial Planning and the Environment (VROM), Directorate-General for Environmental Protection, Directorate of Soil Protection, in the framework of the project 711701 'Risks in relation to soil quality', subproject 711703 'Accumulation in crops', according to the RIVM Long-term Activity Programme 1996 and 1997

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Figure: Nimbus Information Systems (Engraving by Sir John Tenniel for “Alice’s Adventures in Wonderland”)

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## Preface

This research of the Soil and Groundwater Laboratory (LBG) at the National Institute of Public Health and the Environment (RIVM) has been carried out on commission of the Ministry of Housing, Spatial Planning and the Environment (VROM), Directorate General of Environment (DGM), Department of Soil Protection. Supervision was by drs. C.A.J. Denneman and dr.ir. A.E. Boekhold.

This report gives the state of the research in the current project. The project is meant to be continued and be completed in 1999. The draft report has been discussed in an advisory board consisting of:

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## Summary

Since 1991 the CSOIL exposure model has been used in the Netherlands for the calculation of the intervention values for soil and groundwater clean-up. The intervention values are based on potential risks. The CSOIL model (v7.1) has limited options for actual site-specific risk assessment but nonetheless it is used for actual risk assessment (in combination with measurements in contact media).

The CSOIL plant module is made to calculate the accumulation of soil contaminants in plants and human exposure to these contaminants due to the intake of plant material. Van den Berg (1995) already indicates that the set of formulas used for the CSOIL plant module contain large uncertainties. The calculation of the accumulation in plants with bio concentration factors (BCFs) averaged over plants and soil types, makes it less suitable for site-specific calculations. This pilot study was set up to explore possible ways to improve the plant module and focuses on the use of this module for actual i.e. site-specific risk assessment. The final aim is to develop a new module to calculate in a relatively simple way indications of the amounts of soil contaminants accumulated in consumed parts of crops as a function of location-specific soil parameters.

In a first evaluation the contaminants were classified to importance of the exposure route of plant intake in the total potential exposure as calculated with the existing CSOIL-model (v. 7.1). For many contaminants it was found that the plant-intake route contributes highly to the total exposure. This justifies the effort put into the improvement and/or extension of this module. The CSOIL formulas to calculate plant uptake of soil contaminants and their assumptions are discussed separately for three categories of compounds: metals, other inorganic compounds and organic compounds. Several possibilities and options to improve site-specific risk assessment with respect to accumulation of soil contaminants in plants are discussed. Focusing on the influence of soil parameters, the report goes briefly into the consequences of crop choice in the model. In this stage no efforts were made to obtain more realistic crop consumption rates.

As part of the study, a search was done for existing models which describe the accumulation of soil contaminants in crops, to find out which alternative models and procedures exist on this subject and to investigate if any of these models could be used for the improvement of actual risk assessment. For metals, sources like Gupta et al. (1995), Sauerbeck and Lübben (1991) and Van Gestel et al. (1992) were also investigated more closely. Models for uptake in fruits and seeds were not found.

The German UMS model seems to be a good option for the site-specific accumulation of **metals** in crops. In this model the data collected by Sauerbeck and Lübben were used and BCFs were determined for several plant species in relation to a number of metals and for various soil types. Studies of Van Gestel et al. (1992), Elzinga et al. (1997) and Janssen et al. (1996) may be useful in investigating the relation between concentrations in the soil and in porewater as influenced by different soil parameters and the influence of different soil parameters on the accumulation in plants.

No general model is available for the **other inorganic compounds**. For actual risk assessment, the rate of uptake and accumulation of these compounds in plants should be investigated for each compound separately. The realisation of this recommendation will probably be very time consuming, so we suggest focusing first on cyanides. Many other inorganic compounds are covered by separate legislation.

For **organic compounds**, the existing plant module in CSOIL for potential risks is considered as satisfactory for the plant roots, but less for the parts above the ground. The transport of contaminants to stems and leaves in the plant needs attention. This could be improved according to the generic one-compartment model for uptake by foliar vegetation in Trapp and Matthies (1995). The model is also accepted for the EUSES-model (European Uniform System for the Evaluation of Substances), but has, however, not been validated. In this study it has not been investigated how easily the necessary parameters could be obtained or how easily the model could be used for actual risk assessment.

To improve CSOIL's plant module regarding actual risks it is suggested to perform an evaluation of the solubility characteristics for inorganics other than metals and investigate pH dependencies, selective accumulation of contaminants by plants and the differences between accumulation in different plant parts such as stems and leaves .

## Samenvatting

Sinds 1991 wordt het CSOIL blootstellingsmodel gebruikt voor de berekening van de Nederlandse interventiewaarden voor bodemsanering. Deze interventiewaarden zijn gebaseerd op potentiële blootstellingsrisico's. Het CSOIL model (v. 7.1) heeft slechts beperkte mogelijkheden voor actuele lokatie-specifieke risicoberekeningen, toch wordt het daarvoor wel gebruikt (in combinatie met metingen aan contactmedia).

De plantenmodule van CSOIL is gemaakt om de accumulatie van de bodem verontreinigende stoffen in planten te berekenen en de humane blootstelling door consumptie van het plantenmateriaal. Van den Berg (1995) heeft al aangegeven dat de formules die voor de CSOIL plantenmodule zijn gebruikt grote onzekerheden bevatten. De berekening van de accumulatie in de planten met bioconcentratie factoren (BCFs) die worden gemiddeld over planten en bodemtypen, maken deze module in het bijzonder minder geschikt voor lokatie-specifieke berekeningen. Deze verkennende studie was opgezet om mogelijkheden te zoeken om de planten module te verbeteren bij het gebruik voor actuele, lokatie-specifieke risico-berekeningen. Het uiteindelijke doel is om een nieuwe module te ontwikkelen waarmee op een relatief eenvoudige manier de hoeveelheden bodemverontreinigende stoffen die in gewassen accumuleren kunnen worden berekend als functie van lokatie-specifieke bodemparameters.

Bij een eerste evaluatie zijn de verontreinigende stoffen geïnclassificeerd naar belang van de blootstellingsroute van planten opname t.o.v. de totale blootstelling zoals die met het CSOIL-model (v. 7.1) wordt uitgerekend. Het bleek dat voor vele verontreinigingen de route via opname van planten zeer veel bijdraagt aan de totale blootstelling. Dit rechtvaardigt de inspanning om deze module te verbeteren. In dit rapport worden de formules die in de CSOIL versie 7.1 worden gebruikt doorgelicht voor de 3 categorieën van stoffen: metalen, overige anorganische stoffen en organische stoffen, evenals de diverse mogelijkheden en opties om lokatie-specifieke berekeningen aan plantenopname uit te voeren.

De aandacht wordt in de eerste plaats gericht op de invloed van de bodemparameters en er wordt kort ingegaan op de consequenties van de keuze van gewassen in het model. In dit stadium zijn nog geen pogingen gedaan om meer realistische inschatting te verkrijgen van de consumptie van gewassen.

Als onderdeel van de studie is er een onderzoek gedaan naar bestaande modellen die de accumulatie van stoffen in gewassen beschrijven, om te zien welke alternatieve modellen en procedures er op dit gebied bestaan en om te bezien welke modellen bruikbaar zouden kunnen zijn voor de verbetering van de CSOIL module op het punt van de inschatting van actuele risico's. Voor metalen zijn bronnen als Gupta et al. (1995), Sauerbeck en Lübken (1991) en Van Gestel et al. (1992) nader bestudeerd. Specifieke modellen voor de opname in fruit en zaden zijn niet gevonden.

Het Duitse UMS model lijkt een goede optie te zijn om de lokatie-specifieke accumulatie van **metalen** in gewassen te beschrijven. In dit model worden de gegevens gebruikt die zijn verzameld door Sauerbeck and Lübken en in relatie tot diverse bodemtypen zijn voor een reeks van metalen BCF-waarden bepaald voor een aantal plantensoorten.

De studies van Van Gestel et al. (1992), Elzinga et al. (1997) en Janssen et al. (1996) kunnen bruikbaar zijn bij het onderzoek naar de relatie tussen de concentraties in de bodem en in het poriewater als functie van de bodemparameters en naar de invloed van de verschillende bodemparameters op de accumulatie in planten.

Er is geen algemeen model voor de **overige anorganische stoffen**. Voor ieder van deze stoffen dienen het actuele risico, de mate van opname en accumulatie per stof afzonderlijk te worden bestudeerd. De uitvoering van deze aanbeveling zal waarschijnlijk veel tijd kosten, we bevelen daarom aan om eerst de aandacht te vestigen op cyaniden. De meeste andere van de overige anorganische stoffen zijn onderworpen aan andere wetgeving (denk aan vermestings- en verziltingsbeleid).

Voor **organische stoffen** kan de huidige plantenmodule als bevredigend worden beschouwd voor de beschrijving van de opname door de planten wortels, maar niet voor het transport van de wortel naar de bovengrondse plantendelen. Het transport van de verontreinigende stoffen door de stengels en bladeren verdient extra aandacht. Dit onderdeel zou kunnen worden verbeterd met behulp van het generieke een-compartiments model van Trapp en Matthiess (1995) voor de opname door bladgewassen. Dit model wordt ook gebruikt in het EUSES-model (Europees Uniform Systeem voor de Evaluatie van Stoffen), maar is nog niet gevalideerd. In deze studie is echter nog niet onderzocht in hoeverre de benodigde parameters kunnen worden verkregen dan wel of nog aanvullende aanpassingen voor actuele risicobeschouwingen nodig zouden zijn.

Om de plantenmodule van CSOIL te verbeteren met het oog op actuele risico's stellen wij verder voor om een evaluatie uit te voeren van de oplosbaarheidskarakteristieken van de overige anorganische stoffen en in het bijzonder om aandacht te besteden aan afhankelijkheid van de pH, selectieve accumulatie van stoffen door planten en de verdeling van stoffen over de verschillende plantendelen zoals wortels, knollen, stengels en bladeren.

# 1 Introduction

## 1.1 The CSOIL model

Since 1991 the CSOIL exposure model has been used in the Netherlands for calculating the intervention values for soil and groundwater clean-up. The decisive criterion for these intervention values is the potential risk of adverse health effects resulting from human exposure to the soil contaminants [Van den Berg, 1995]. The potential exposure resulting from a certain concentration level of contaminants in the soil is calculated using CSOIL for a standardised situation, assuming a standard scenario. In this scenario based on a residential situation all possible exposure pathways are assumed to be included. The calculated exposure is then compared with the human toxicologically based Maximum Permissible Risk (MPR) to connect it with adverse health effects. The human toxicologically based intervention value for contaminated soil is defined as the soil concentration at which the exposure equals the MPR concentration. Exceeding the human toxicologically based intervention value for one or more contaminants, is considered as an unacceptable risk to humans.

## 1.2 Aim and scope of this study

The results of the CSOIL model (v. 7.1) indicate that for the considered compounds the most important part of the total exposure is contributed by three exposure routes: the inhalation of contaminated air, the intake by soil ingestion and the consumption of contaminated crops. Van den Berg [1995] indicates that the sets of formulas used for CSOIL, in the volatilisation module as well as in the plant module, contain large uncertainties. Also, neither set of formulas is suited for actual, i.e. site-specific, risk assessment. New improved volatilisation formulas have recently been incorporated in the VOLASOIL model for site-specific calculation of concentrations in indoor air [Waitz *et al.*, 1996]. The intake by soil ingestion and subsequent uptake will be the subject of a separate study. In this pilot study to recognise possible ways to improve the plant module, the CSOIL's existing set of formulas for accumulation of compounds in plants is evaluated and the possibilities for an extension to site-specific evaluation are studied.

It should be noted that to reach a stage in which decisions can be made on the risk of contaminated soils a pragmatic approach is necessary. Though efforts are done to develop a model that is realistic enough for our purpose, the report on this study does not fully reflect the state-of-the-art in plant physiology. An attempt is made, either empirically or sometimes with rough estimates, to force a decision stage, in spite of all the remaining uncertainties. This may be scientifically disappointing or even incomplete and unsound as seen from other more pure scientific disciplines but it is necessary to guide decisions on soil remediation, realising, however, that continuing improvement is necessary to reach a more satisfying stage.

The main interest in this study is the possible accumulation of harmful concentrations in crops grown on contaminated land and not the dynamics of the process of accumulation in the crops. It should also be noted that this report deals, in the first place, with the high concentration ranges in which the compounds occurring should be considered as contaminants; it does not deal with the concentration ranges in which the compounds should be considered as essential elements for the plants.

### 1.3 From potential to actual risks

As explained this report focuses on the possible extension of the modelling (formulas and data) of the CSOIL model (v. 7.1). This model is in the first place made for potential risk assessment. In this report primary we study the possibilities for constructing an extension of the modelling fit for actual risk assessments and secondary we look into possible improvements in the existing modelling for potentials risks.

Actual risks are inherent to a specific situation that can be described more accurately and in more detail than the generalised situation assumed for the modelling of potential risks. In general, this means that for an analysis of actual risks several aspects need to be evaluated in more detail. In the plant module this can be done from the viewpoint of the characterisation of the soil and the contaminants or from the viewpoint of the behaviour of various types of crops and the amounts of crop consumption:

- Soil

The soil parameters can be measured at the contaminated site and their influence on the accumulation in the crops can possibly be assessed more accurately. To be implemented in an analysis of actual risks the variation of BCF factors with soil parameters should be known. The BCF or bioconcentration factor BCF is defined as the ratio of the concentration of the contaminant in a crop to the concentration of the contaminant in the soil at the time of harvesting. Actually, if determined, such a relation could immediately be implemented in the CSOIL model, as relations with clay content and organic carbon content are usually known and also used for the calculation of the target and intervention values (also pH, conductivity of the porewater, and CEC [cation exchange capacity], and possibly the average water and nutrient content may play a role).

It should be noted that a choice has to be made for BCF values in relation to the total concentration in the soil or BCF values in relation to the concentration in the porewater. The first option is in common use. The fraction in the porewater can, however, be considered as a first estimate of the bioavailable fraction. This aspect is evaluated further (i.e. section 6.2 ).

- Crops

Field tests (i.e. on available crops) can be additionally performed as either a check on the model or instead of some of the calculations. The measurements on plants from the site can lead to a number of questions, like which types of plants should be measured (also in view of specific uptake of some plants) and in which growing phase should be measured, how are spatial variations over the site, how to correct for seasonal variations and are soil extraction procedures a good measure? In this way modelling and measuring each have their own difficulties. In this project we will try to model the accumulation in crops, on the basis of available experimental data from literature, to the needs of actual risk assessment for the decision to undertake remedial action or not. If the model appears to be not satisfactory, decisions should probably rely more on field tests. The design, standardisation and the guidelines on when to perform such field tests will not be developed in this project.

- Crop consumption

The types of crops actually grown on the contaminated or suspected site and consumed by humans (or better: grown in a specific scenario which relates with the use of the soil, see below) could be chosen for the analysis of actual risks. Since it is a risk analysis, also crops that might be expected to grow in the future on the site should play a role in the analysis of actual risks (i.e.

expressed in scenarios for future use, see also below). Section 6.6 goes into the choice of crop species. The different nature of crops affects several factors:

- ◇ Because the BCF values for metals vary considerably with plant species (see section 6.2), the BCF values should be collected for individual plant species.
- ◇ For organic substances the modelling of relevant plant processes will probably be the best way to evaluate the accumulation of contaminants in plants (see sections 4 and 6.4). More detailed modelling for several specialised categories of plants *or plant parts* should perhaps get attention.
- ◇ The consumption rates (see section 2.4) will be more accurately known in a specific situation (e.g. for a situation with home gardens). This will not be evaluated further in this study.

- Contamination

The speciation of metals (and other inorganic contaminants) can be determined for a site-specific situation. This can, for instance, be of importance for solubility and mobility of the contaminants. (The speciation relates with the source of the contamination, but is also dependent on the pH, the redox potential and the type and the age of the original contaminants.) To be implemented in an analysis of actual risks, the behaviour of the separate speciation classes / fractions of the contaminant should be known. This line of thought should, preferably, not be restricted to the plant-uptake module but be integrated in the analysis of all exposure routes to be of any use. As this is not the case the subject of speciation is also (largely) ignored in the analysis of the plant module.

- Use of the site

For the analysis of the risks in an actual situation both long-term and short-term approaches are involved. For the long-term exposure assessment an actual risk assessment also makes use of scenarios for the use of the site. The crops under consideration and the ratios of crop consumption will usually be fixed in these scenarios. The actual risk assessment is different from potential risk assessment in being site specific, which means mainly: specifying soil parameters and restricting the number of scenarios for the use of the site. The type of soil plausibly does not change very much in long- and short-term approaches.

It should be noted that though this approach may be more realistic than a potential risk assessment, it is, however, not fit for analysing acute risk situations. The analysis and prevention of acute risks is related to direct health and safety measures and continuing management of soil use. The use of the model for actual risk assessment is restricted to the decisions on the need and the urgency of remedial action.

## **1.4 Organisation of this report**

After this introduction the report is organised as follows:

**Chapter 2** briefly describes the plant module in the current CSOIL model, with the formulas incorporated included in Appendix 1.

**Chapter 3** shows the importance of the exposure route of crop consumption in relation to the other exposure routes, as calculated by the CSOIL model. The related Appendix 2 gives for 85 compounds the calculated relative contribution to the total exposure for each exposure route that is included in CSOIL.

**Chapter 4** evaluates the state-of-the-art in CSOIL for the three categories of compounds: metals, other inorganic compounds and organic compounds.

**Chapter 5** gives a short description of some other known important models for accumulation of soil contaminants in plants and evaluates the use of the results of these models for CSOIL.

**Chapter 6** evaluates the data found for metals. It focuses on the studies of Sauerbeck in relation to the UMS model and of the results of Van Gestel et al (1992 a, b). The related Appendices 3 and 4 give overviews of the numerical results from these two sources.

**Chapter 7** summarises the conclusions and recommendations of the foregoing chapters.



## **2 Exposure due to accumulation of soil contaminants in consumed plants as treated in the current CSOIL model**

The exposure is calculated from concentration of soil contaminants in the plants and estimated consumption rates of these plants. The current plant module in CSOIL calculates the concentration of a contaminant in a plant from the concentration in the soil. Two routes for uptake from soil to plant are distinguished in the module:

- uptake from the soil by the root system, particularly via soil moisture (followed by transport to other parts of the plant, distribution over plant parts and excretion and volatilisation);
- deposition on plant parts above the ground, mainly on the leaves, of soil particles dispersed in the air.

The extent of accumulation of the contaminants in the crops is expressed as the bioconcentration factor BCF: the ratio of the concentration of the contaminant in a crop to the concentration of the contaminant in the soil.

This study focuses on the evaluation of the route first mentioned. Different sets of formulas are used for metals, other inorganic compounds and organic compounds. For metals, the calculation is based on the total concentration in the soil. For the other inorganic compounds and organic compounds the calculation is based on the concentration in the pore water. The calculation for the organic compounds is based on empirical relations with the Kow partition coefficient (for the distribution of the compound over octanol and water).

Appendix 1 gives an overview of the formulas used in the plant module in the current CSOIL model (version 7.0). This chapter gives a short explanation on the background of these formulas.

### **2.1 General limitations of the current plant module**

To evaluate possibilities for improvement and extension it is important to recognise the limitations of the current module. It does not describe the dynamic process of uptake, but only the resulting accumulation. The general set-up of the calculation in the current plant module is based on (semi) equilibrium and stationary states and is limited in that there is:

1. no transformation and degradation of compounds,
2. a homogeneous soil,
3. the BCF-values are valid only for the high concentration ranges related with passive uptake of compounds, (active uptake can take place at low concentrations for the essential elements),
4. no crop differences with respect to lipophilicity (amounts of lipophile plant materials) and water content, only with respect to root /tuber crops and leafy /stem crops
5. no speciation of compounds (average behaviour of compounds only),
6. no differentiation in separate processes which may vary strongly over plant types, like volatilisation from leaves or uptake of gaseous compounds from the air,
7. no verification of uncertainty ranges.

In the following section CSOIL's formulas for the accumulation in plant roots is discussed in more detailed for metals, other inorganic substances and organic substances. Next the CSOIL formulas for deposition and consumption rates are discussed shortly.

## **2.2 Accumulation of soil contaminants in plant roots**

### **2.2.1 Metals**

As explained the calculation is restricted to an empirical model of the amounts accumulated in the plant, due to the limited knowledge of uptake mechanisms of these compounds in the relevant concentration ranges. Measured values for the bioaccumulation factor (BCF) are proposed as the starting-points. Sauerbeck [1988] classified a number of compounds in groups, each with a certain BCF range [ECETOC, 1990]. The geometric range average was used as the value for calculating the intervention values. (The viability of these values is discussed in Chapter 4. Table 3 in section 4.1 gives these data.)

If a measured BCF value is not available, it is calculated with the formula drawn up by Baes *et al.* [1984], giving a correlation between the BCF value and the soil-water distribution coefficient  $K_d$  (ratio of concentration in soil and in pore water):

$$\ln BCF_{pl} = 2.67 - 1.12 * \ln K_d$$

If also the  $K_d$  is not known, the BCF value should be estimated on the basis of 'expert judgement'. The general experience is, however, that at least some of these data are available.

### **2.2.2 Other inorganic compounds**

Assuming that a plant consists of approximately 80% water and that the concentration of an inorganic compound in this plant is equal to the concentration in the soil moisture, a fixed BCF value is calculated in CSOIL based on a fresh weight basis as 80% of the soil moisture concentration [Briggs *et al.*, 1982;1983, Ryan *et al.*, 1989]

As a worst-case scenario for the calculation of the concentration of inorganic compounds in the pore water, it is assumed in the CSOIL model that these compounds are completely water-soluble and thus completely available in the aqueous phase of the soil.

### **2.2.3 Organic compounds**

Based on the analysis by De Nijs and Vermeire [1990], the relations of Briggs *et al.* [1982;1983] were chosen to calculate the BCF values for subterranean (root) and aerial (leaf and stem) parts of the crop. These empirical relations are based on the concentration of a compound in the soil moisture and are functions of the  $K_{ow}$ , the octanol/water partition coefficient, of the compound considered. The used experimental data are mainly based on barley. The calculation for the aerial parts is based on the data for the stem.

## **2.3 Deposition on crops**

Due to various processes, particles of the soil in which the plant grows also occur in the air and may subsequently be deposited on crops and consumed with the crop. In CSOIL the term deposition is used only for this process of (dry) deposition of soil particles with connected contaminants on the plant and not for the additional deposition from other sources (e.g. industrial emissions) on the plant. The deposition is calculated on the basis of the Hetrick and McDowell-Boyer equation [1984], as introduced in the EPA report 'Users Manual for Tox-Screen'. Appendix 1 gives the formulas.

*This report does not focus on the evaluation of deposition on crops and the discussion in this report is restricted to the following remarks in this section:*

Washing of the crop before use would help to reduce the importance of this exposure route (see i.e. Kos *et al.* 1996), but as a worst case assumption, has not been taken into account in CSOIL. An opposing consideration is that the contaminants that are connected with the deposited soil particles may be absorbed by the crop.

Volatilisation of compounds from the topsoil may result in uptake by diffusion into the plant material in the gas phase. For compounds with a relative high  $K_{ow}$  volatilisation of the compound uptake by leaves is the major accumulation route. This process can also be considered as a form of deposition but in this report it is treated in connection with modelling the accumulation of organic substances in plants.

Another deposition route, however, not related to existing soil contamination, is by application of pesticides and fertiliser deposited mainly or partly on the plant.

## **2.4 Consumption of crops**

Exposure is effected by the consumed amounts of the contaminated crops. As already explained, the crop contaminant content is a function of both the uptake of contaminants by the crop from soil moisture and deposition of soil particles on the crop. The human exposure level depends on the crop content, but also on the amounts of vegetables and fruits consumed in the total food package and the fraction of these crops that is grown on the contaminated soil. For a calculation of the potential risks it has been assumed that the latter fraction equals the fraction of home-grown crops and that this equals 10% of the total vegetable and fruit consumption. [Linders, 1990]. It has been suggested to make this more realistic by differentiating between the group of site users that does not eat from home gardens and the group of site users that does eat (more than 10%) from home gardens.

*The focus on consumption rates of crops will be limited in this report (section 6.6).*

### 3 Relative importance of the exposure routes

In order to gain insight in the relative importance of the various human exposure routes defined for the potential risk assessment, we performed an analysis with the current CSOIL model for several compounds (see Table 1 for a summary). The complete results are given in Appendix 2. All calculations were performed at concentrations at the level of the intervention values. In the following discussion relative contributions of the exposure routes are considered, independent of the overall level of exposure for each compound. It appears in the following discussion that some assumptions for the physico-chemical parameters in the model play an important role in the results.

The distribution of the contaminants over the defined potential exposure routes is determined (mainly) by physicochemical parameters of the contaminants. The relative importance of the *actual* exposure routes is also determined by the site characterisation and the scenarios considered of importance for the site. The real exposure is also determined by the actual situation, lifestyle, age, hygienic practices (e.g. purification of drinking water), measures (e.g. entrance restrictions), plant cover, et cetera.

The analysis showed that, for almost all compounds considered, a number of the defined exposure routes contributed only marginally. These marginal exposure routes are: inhalation of soil particles, inhalation during showering, dermal contact with the soil, consumption of drinking water and dermal contact during showering. The maximum sum of contributions for these routes was found for cresols (15%). The important exposure routes, consisting of ingestion of the soil, consumption of crops and inhalation of (indoor) air account collectively for at least 85% of the human exposure to soil contamination for all contaminants considered.

**Table 1:** *The relative importance of the major exposure routes as calculated by the current CSOIL model*

Type of contaminants	Main exposure routes	Variation in contribution to the total exposure	
		Min.	Max.
Metals	crop consumption	32% (Cr)	95% (Cd)
	soil ingestion	5% (Cd)	68% (Cr)
	vapour inhalation	-	0% (all)
Other inorganic compounds	crop consumption	100%(all*)	-
	soil ingestion	-	0% (all)
	vapour inhalation	-	0% (all)
Aromatics	crop consumption	6% (benzene)	70% (phenol)
	soil ingestion	-	0% (all)
	vapour inhalation	13% (cresol)	92% (benzene)
PAHs	crop consumption	2% (benzo-ghi-perylene)	91% (phenantrene, fluorantene)
	soil ingestion	2% (naphthalene)	90% (benzo-ghi-perylene)
	vapour inhalation	0% (most)	9% (naphthalene)
Volatile chlorinated aliphatic compounds	crop consumption	0% (vinyl chloride)	16% (1,2-dichloroethane)
	soil ingestion	-	0% (all)
	vapour inhalation	82% (1,2-dichloroethane)	100%(vinyl chloride)
Chlorobenzenes	crop consumption	20% (monochlorobenzene)	97% (pentachlorofenol)
	soil ingestion	0% (several)	15% (hexachlorobifenyyl)
	vapour inhalation	0% (several)	69% (monochlorobenzene)
Drins	crop consumption	77% (aldrin)	92% (endrin)
	soil ingestion	7% (endrin)	21% (aldrin)
	vapour inhalation	-	0% (all)
Other pesticides	crop consumption	3% (maneb)	95% (atrazine)
	soil ingestion	1% (atrazine)	90% (maneb)
	vapour inhalation	0% (several)	16% (carbaryl)
Organic solvents	crop consumption	0% (octane, THF)	69% (cyclohexanone)
	soil ingestion	-	0% (all)
	vapour inhalation	29% (cyclohexanone)	99% (heptane, octane, THF)
Phthalates	crop consumption	69% (di-2ethylhexylphthalate)	91%(butylbenzylphthalate)
	soil ingestion	5% (butylbenzylphthalate)	28% (di-2ethylhexylphthalate)
	vapour inhalation	0% (di-2ethylhexylphthalate)	1% (butylbenzylphthalate)

\*: as the result of a worst-case assumption, see evaluation in section 3.2

### **3.1 Metals**

In the case of metals the only relevant routes are soil ingestion and consumption of crops. The relative contribution of the latter route is determined mainly by the bioconcentration factor (BCF value) and the fraction of total crop consumption from the homegarden (Vissenberg, Swartjes, 1996). A high BCF-value gives rise to a relatively high crop content and exposure and also to a high relative contribution of the exposure due to the consumption of crops. The assumption has been made that the BCF is independent of the total concentration in the soil and of the soil characterisation.

### **3.2 Other inorganic compounds**

In principle similar to metals, only crop consumption and soil-ingestion are involved for the other inorganic compounds. As the speciation of these inorganic compounds is not yet known, calculations have followed the 'worst-case' scenario, assuming that the substance is completely water-soluble, or in other words, that the substance is completely present in the soil moisture. As a consequence the CSOIL-model results point out that the crop consumption route remains the most important exposure route.

(If a substance is not water-soluble, deposition on the plant can still lead to a contribution of the exposure route of plant consumption. The relative importance of the soil ingestion route is determined solely by the water-solubility.)

### **3.3 Organic compounds**

#### **3.3.1. Aromatics**

For the non-hydroxyl aromatics, inhalation is by far the most important human exposure route.

In contrast, for the water-soluble hydroxyl aromatics, including chlorophenols, crop consumption is the major contributor. In these cases high log Kow values give rise to high crop content and consequently relatively large contributions of the crop consumption route.

#### **3.3.2 Polycyclic Aromatic Hydrocarbons (PAHs)**

For PAHs with high water solubility crop consumption is the major exposure route. The relatively high accumulation in the plants of dissolved PAHs is explained by the high root uptake, usually occurring at high Kow values. When the concentration in the soil-water as used in CSOIL exceeds the water solubility, intervention values found are in principle too high.

Above the level of maximum solubility in water soil ingestion gains relatively increasing importance. Only in the case of the most volatile and water-soluble PAH, naphthalene, is a small inhalatory contribution found; for the other PAHs the contribution of this route is negligible.

#### **3.3.3 Volatile chlorinated aliphatic compounds**

As expected for this group of compounds, inhalation plays a major role. The relative contributions of inhalation and crop consumption correspond with the relation between volatility and water solubility in this group of compounds.

### 3.3.4 Chlorobenzenes

For the group of chlorobenzenes an increasing degree of chlorination results in a strong decrease in the volatility. The share of inhalatory exposure decreases in that direction. Consequently, in this group of compounds the exposure by crop consumption becomes more important with an increasing degree of chlorination. Also an increasing degree of chlorination results in a decrease in the water solubility and a increase in the log-Kow value. A decrease in water solubility on its own would lead to less plant uptake, but it is overruled by the effect of decreasing volatility. For the effect of the log Kow see section 3.4.

### 3.3.5 Drins

Despite low water solubility, relatively high percentages are found for the contribution of the exposure route of crop consumption. This is caused by the high Kow values of drins, which results in high BCFs and thus high root uptake. Low water-solubility usually results in a high contribution of exposure by soil ingestion. However, compared with the volume of crops consumed the amount of ingested soil is relatively small. In this case this is mirrored by the result that crop consumption is the main exposure route for drins.

### 3.3.6 Pesticides

Crop consumption generally plays the most important role for exposure to pesticides as soil contaminants. The exposure route of inhalation is of minor importance. Because the calculations in CSOIL are based on the partitioning of compounds over the different soil phases, sometimes the calculated porewater concentration exceeds the water-solubility of a compound. This is the case for maneb, DDT and DDE (for the level of the Intervention Value) and soil ingestion becomes the most prominent exposure route.

### 3.3.7 Organic solvents

For organic solvents, the relative importance of the crop consumption route is variable due to the large variation in physicochemical properties in this group. When the vapour pressure of a compound is relatively high and the solubility is relatively low, the importance of the inhalation route becomes more prominent. This is the case for heptane, for example.

### 3.3.8 Phthalates

Crop consumption is important for this group of compounds because of their relatively high Kow values.

## 3.4 General remarks on physicochemical parameters

The Intervention Value is determined by the MPR (Maximum Permissible Risk for intake, derived from Tolerable Daily Intake) and the defined exposure pathways. The exposure pathways are closely related to physicochemical parameters: one main effect is that high water-solubility and volatility lead to higher exposure. The effect of the Kow value is more complicated. For instance, high Kow values may lead to high sorption to the soil and thus, lower exposure. However, compounds with high Kow values generally have high BCF values and lead to more uptake by the plant. In general, this may lead to ambiguous results for the relationship of crop-contaminant accumulation with the Kow value. (A complicating factor is the use of several fixed settings in the model, which especially influence calculated results for compounds with high Kow values, this is explained further in section 4.3).

### **3.5 Selection of compounds for further investigations to the exposure by crop consumption**

In Table 2 compounds are classified with respect to apparent importance of the exposure route of crop consumption. The classification in Table 2 is based on exposure calculations using the standard exposure scenario with the current CSOIL model for which the complete results are given in the Appendix 2. The main aim is to propose a classification for the compounds for which further research into the subject of accumulation in plants, is important. Other aspects which should play a role are (1) the frequency in which a substance is found as a soil contaminant and (2) the suspected errors or large uncertainties in the current model.

The compounds are subdivided into four classes in Table 2 based on the importance of the exposure route via crop consumption. These four classes contain all the inorganic contaminants and two-thirds of the 65 organic contaminants contained in the current CSOIL model. In the first two classes (80-90%, >90%) the exposure route via crop consumption is clearly dominant. We recommend to give for these classes in any way at least some renewed attention on the modelling and/or parameters used. The research into this exposure route for compounds in the last two classes (20-50%; 50-80%) is proposed to be postponed to a later phase (with some exceptions as explained later).

Note that the current model may overstress the importance of a compound as seems to be the case with sulphides, for which the assumption of complete solubility is not realistic (see section 4.3 on inorganic compounds).

As previously mentioned, beside the importance of the exposure route, the frequency of appearance of the contaminants is also relevant factor. For instance Pb, with a relatively low uptake via crop consumption is widespread as pollutant. Frequently occurring contaminants in soils are estimated to be mineral oil, PAHs, cyanides (gasworks) and the metals cadmium, lead, zinc and to lesser extent mercury, copper, pesticides (atrazin, paraquat, DDT, lindane and other HCHs) [RIVM 1996, CCRX 1995].

Most of these substances are already in the classes 1 and 2. We suggest adding at least mineral oil and the metals lead, arsenic, (for reasons specified in section 6.1) copper and mercury, and the complete set of PAHs. The latter because they form a homogeneous group of compounds are often occurring as soil contaminants.



**Table 2:** *Classification of contaminants with respect to the importance of the exposure route of human intake of soil contaminants via crop consumption based on the percentage of the total exposure (as calculated with the current CSOIL model)*

Class	Organic	Metals	Other inorganic
≥ 90 %	phenantrene fluoranthene hexaCl-benzene diCl-phenol(2,4) triCl-phenol(2,3,4) pentaCl-phenol triCl-biphenyl dieldrin endrin HCH α-HCH β-HCH γ-HCH δ-HCH carbofuran propoxur atrazine butylbenzylphthalate	Cd Zn	ammonium compounds phosphates sulphides bromides fluorides cyanides free cyanides complex thiocyanates
80-90 %	naftalene tetraCl-benzene(1,2,3,4) pentaCl-benzene monoCl-phenol(2) tetraCl-phenol(2,3,4,5) Cl-naftalene hexCl-biphenyl	Cu Mo Ni	
50-80 %	phenol cresol(p) catechol resorcinol hydrochinon chrysene triCl-benzene(1,2,4) aldrin carbaryl cyclohexanone di(2-ethylhexyl)phthalaat	As Ba Co Hg Sn	
20-50 %	xylene(m) benzo(k)fluoroanthene pyrene monoCl-benzene diCl-benzene(p) DDE styrene	Cr(III) Cr(VI) Pb	

## 4 Evaluation of CSOIL's plant-module

The description in CSOIL of the accumulation of soil contaminants in crops consists mainly of empirical relations. For organic compounds, the underlying mechanisms (uptake of contaminants by plants through soil water) have been taken into greater account. In this way the accuracy of extrapolations can be improved outside the area for which the empirical relationships have been found.

### 4.1 Metals

For CSOIL average BCF-values are collected for root crops and for leafy crops (Bockting, vd Berg, 1992). When for a compound no BCF-values are available CSOIL uses the Baes et al [1984] relation to calculate a BCF from the K<sub>d</sub>-value. An earlier version of this formula (Baes, 1982) leads to a maximum difference of a factor of 12-20 between calculated and measured BCF values. Baes *et al.* [1984] later elaborated the formula into its present form. Still, large deviations are found, generally up to a factor of 5 for the compounds considered here (but sometimes higher, i.e. for cadmium and iron, the latter deviates even by a factor of 70). In the derivation of the relation the soil properties were not taken into account. This should, however, give a possibility to introduce the soil properties. For example the recent EPA User's guide for soil screening guidance (1996) gives metal K<sub>d</sub> values (l/kg) as function of pH (for As, Ba, Be, Cd, Cr<sup>3+</sup>, Cr<sup>6+</sup>, Hg, Ag, Se, Tl, Zn and mentions non pH-dependent K<sub>d</sub> values for Sb, V and cyanide). This aspect will be evaluated in section 6.2.

The existing procedure lacks the possibility to quantify the influence of relevant soil properties, like soil-pH. For this reason the current plant-module in CSOIL is not fit for the calculation of actual concentrations of accumulated soil contaminants in the plant.

Because of the deviations displayed and in view of the unreliability of the K<sub>d</sub> values, it is agreed that it is better to start from measured BCF values wherever possible. However, the standardisation of measurements is problematic. Sauerbeck [1988] collected experimentally derived BCF values and found variations with factors of 10 to 100 (see Table 3). These variations are probably originating from the impact of soil properties on the contaminant properties and may also vary with the type of crops, the level of contamination and the contaminating medium and seasonal variations. The influence of soil type on plant uptake could be corrected by means of the more common parameters of pH, organic carbon and clay content, but also other soil properties, like redox-potential may be of importance.

The BCF values given by Baes *et al.* [1984] (see Table 3) are, in general, comparable to those of Sauerbeck, but sometimes deviations occur: for mercury the BCF values exceeds far the upper boundary given by Sauerbeck [1988], for cadmium and nickel the value of Baes falls below the range of Sauerbeck.

**Table 3:** Bioconcentration factors (BCF, as (mg kg<sup>-1</sup> ds)/(mg kg<sup>-1</sup> ds) ) for a number of metals according to Baes et al. [1984], Sauerbeck [1988] and as used in CSOIL according to Bockting and Van den Berg [1992]

metal	BCF according to Baes et al. (1982)- for non-polluted soils	BCF according to Sauerbeck (1988)		BCF as used in CSOIL, van den Berg, (1995)	
	leaves, stems	potatoes	vegetables	potatoes	leave crops
As	0.04	-	*) 0.01->0.1	0.015	0.03
Ba	0.15	-	-	0.005	0.1
Cd	0.55	0.01-0.5	0.5-10	0.15	0.7
Co	0.02	-	*) 0.01-1.0	0.015	0.03
Cr	0.0075	0.01-0.5	0.01-1.0	0.002	0.02
Cu	0.4	0.5-1.0	0.01-10	0.1	0.1
Hg	0.9	-	*) 0.01->0.1	0.015	0.03
Mo	0.25	-	*) 0.1-10	0.015	0.3
Ni	0.06	0.01-0.5	0.01-2	0.07	0.1
Pb	0.045	0.01-0.5	0.01-0.5	0.001	0.03
Sn	0.03	-	-	0.015	0.03
Zn	1.5	0.01-0.5	0.5-1	0.1	0.4

\*) Not differentiating between potatoes and vegetables

For metals it is difficult to obtain mechanistic models which describe the plant uptake of metals from the soil. It is therefore necessary to focus on statistical research which links the concentration in the soil to the concentration in the plant. Very much work has been done on plant uptake in the past. This study relies completely on the available literature.

In the existing plant module in the CSOIL-model the accumulation in the plant is related to the total concentration in the soil, but for the uptake by the plant roots the concentration in the porewater is of more direct importance. It may be better to focus on the link between the concentration in pore water and the concentration in the plant. This will reduce the uncertainties in calculations of distribution of the compound over soil phases. However, in that case a characterisation of the porewater is necessary and actual concentrations in pore water do differ considerably from day to day. The actual concentrations depend among others on weather conditions, organic matter and salinity. Though average pore-water concentrations can play a role in modelling exercises, these concentrations are not good parameters for field measurements. Moreover the distribution coefficient  $K_d$  of contaminants over soil and porewater is the subject of a separate study (Koops, et al, to be published 1998).

When large uncertainties exist on the bioavailability of the soil contaminants a pseudo-characterisation of the porewater concentrations with extraction is possible, distinguishing extractable mobile metal fractions from less mobile fractions. The extraction of the soil with a standardised neutral extraction e.g. a calcium chloride solution or a leaching test from the 'Building Materials Decree' (see also Appendix 4) seems more reliable for field measurements of the average

concentrations in the porewater. Literature describing the relationships between concentrations in soil and plant extracts is available and increasing (see section 6.2). Unfortunately, the preferred method and extraction solvent are still subjects of discussion.

Extraction with  $\text{CaCl}_2$  (0,01 M) solution will probably lead to a connection with research in plant physiology; a leaching test will, however, better connect with standard field surveys on contaminated soils.

## 4.2 Other inorganic compounds

Assuming that plants consist partly of water (approximately 80%) and that the concentration of an inorganic compound in a plant is equal to the concentration in soil moisture (pore water), a fixed BCF value of 0.8 has been proposed (Briggs *et al.*, 1982 & 1983; Ryan *et al.*, 1988). Actual water contents may vary, i.e. see Appendix 4, section 2.5, but this will only result in relatively small deviations.

The main assumptions for the current CSOIL model are that:

- the compound is completely dissolved;
- the uptake of compounds by the roots is considered a passive transport process;
- the inorganic compound is distributed homogeneously over the plant organs;
- deviations that are dependent on the plant's water content are relatively small;
- the effect of transpiration on the concentration in the plant is not considered.

The assumption that the compound is completely dissolved will probably lead to overestimation of the accumulation in plants. However, the procedure described above has been used since more reliable data were lacking. Van den Berg (1995) stated that information on transport mechanism was not readily available at the moment of construction of the CSOIL model.

The differences between uptake in the stem and uptake in the leaves of a plant are not considered in the current CSOIL model. An effect that should lead to underestimation is that due to evapotranspiration from the plant the inorganic compounds in the plants could precipitate potentially resulting in a higher accumulation. Knowledge of the water balance and transport mechanism in the plant is of great importance, as a plant uses approximately 200 to 800 litres for the production of one kg dry plant material (Lexmond, personal communication 1996).

An evaluation on some specific inorganic compounds will be made in the following sections

### 4.2.1 Macronutrients (N, P)

No effort has been or will be made in the next future to derive an Intervention Value (based on *potential risk assessment*) for agriculture related macro-nutrients. This is for several reasons (Swartjes, 1997):

- The presence of nitrogen (nitrate, ammonium) and phosphate in soil is often regulated. The concentration of nitrogen is not only a result of emissions in the past but is to a large extent caused by repeated applications of fertiliser. Nitrate and phosphate are already subject to legislation in the Dutch 'vermestingsbeleid' (policy measures on eutrophication).
- Nitrogen has a short residence time in soil. This short residence time is due to leaching and modification of the N compounds by microorganisms in the soil.
- The main problem with phosphate is the eutrophication of surface waters, so legislation should evolve from interaction between the surface water quality managers and the users of fertilisers. Soil and groundwater are, however, sources of phosphate in surface waters

because of the movement of groundwater to surface waters and the decreasing buffering capacity of soils with high phosphate loads.

In soil mainly poorly soluble Ca, Fe and Al phosphates occur and to less extent phosphate bound to organic matter. As a consequence, phosphate moves only slowly and has a relatively long residence time in soils. In groundwater mainly organic P and phosphate ions like  $\text{H}_2\text{PO}_4^-$  and  $\text{HPO}_4^{2-}$  exist.

Agricultural and horticultural activities probably cause the main concentrations of macronutrients. These activities are subject to customised regulations considering concentrations of the N and P compounds. Similar to Swartjes (1997) we propose to neglect these macronutrients also for actual site specific risk analysis of soil contamination.

#### 4.2.2 Halogens (Cl, F, Br)

This chapter deals with inorganic halogen compounds. Halogen-containing compounds will be treated with the other organic compounds.

- Fluoride

In soils, the element fluorine occurs mainly as poorly soluble  $\text{CaF}_2$  especially at high pH. Fluorides sorb fairly easily on Al and Fe oxides. High fluoride concentrations are toxic for humans (kidney, bones, tooth) and for plants (stagnation of growth and damage to leaves), but the level of phytotoxicity is not well established (Will, Suter, 1995) and it is reported to be inactivated by neutral and alkaline soils (Rowe, Abdel-Maghid, 1995).

- Chloride

The toxicity of chloride for plants is highly species dependent. Some plants are more adapted to salty environments than others. Chloride is frequently found for example, in coastal regions. No large quantities of salty plants will be consumed. Salt used as food additive probably implies more risks. Separate legislation (policy measures on salination) exists for chloride, in the Netherlands this is called 'verziltingsbeleid'.

- Bromide

Bromide is used in disinfectants, bleaching agents and tranquillisers. Bromide may cause toxic effects on the nervous system and on the thyroid gland of humans. The TDI is high: 1 mg Br per kg body weight per day (Vermeire *et al.*, 1991), which means that bromide will not easily function as a restrictive factor in terms of risks.

The group of halogens is of secondary importance for the assessment of the risks of soil contamination, only fluorine, may be subject to further investigation.

#### 4.2.3 Sulphur compounds

Inorganic sulphur is found as a sulphate in oxidised environments; sulphates are generally mobile and leach easily. In a reduced environment inorganic sulphur occurs as poorly soluble sulphides. Sulphur is a necessary compound to life but could also be toxic for plant growth (both as  $\text{SO}_2$  and  $\text{H}_2\text{S}$ ) and for humans (especially as a well-soluble Mg sulphate). Sulphur is frequently found in the groundwater in the Netherlands. The main sources are atmospheric deposition, marine intrusions and manure.

Because of the interference with the policy measures on eutrophication it will be no subject of further studies in actual site specific risk analysis of soil contamination.

#### 4.2.4 Cyanides

The exposure via plant uptake for the intervention value for cyanides is probably overestimated at a high pH (cyanide occurs in free form at a high pH). The overestimation is caused by the assumption that the concentration of the compound in the plant's water is equal to the concentration in the soil porewater. Neither transformation nor degradation in soil or plant are considered in the current CSOIL model.

Now it is known that often cyanide molecules will be degraded in the plant (Meeussen, 1996; personal communication). In small amounts cyanide is easily degraded into CO<sub>2</sub> and NH<sub>3</sub> by plant cells and other cells and used as a carbon and nitrogen source for new cell elements.

Cyanides are subdivided in

- Free cyanides: CN, HCN(aq), HCN (g), NaCN and KCN, which are very mobile and easily absorbed by plants.
- Prussian blue, Fe<sub>4</sub> (Fe(CN)<sub>6</sub>)<sub>3</sub>, and other metal cyanides which are forms of an immobile cyanide complex. These complexes are not easily absorbed by the plant at low pH. Prussian blue is not toxic, only at high pH does it dissociate into toxic components.
- Thiocyanates (SCN<sup>-</sup>) are degradation products of the cyanides mentioned above.

Cyanides are frequently met as soil contaminants and for this reason alone important for further studies in actual site specific risk analysis of soil contamination. The behaviour of all these compounds is strongly pH dependent and as a consequence probably also very site specific. Unfortunately, the behaviour of cyanides in plants is hard to quantify and not well known or documented.

#### 4.2.5 Conclusions for other inorganic compounds

**Table 4** gives an overview of relevant characteristics for the discussed inorganic compounds. The purpose of the table is to extract which compounds are worthwhile looking at in more detail for a follow-up of this evaluation of CSOIL's plant module.

In the first column the compounds are ranked by estimated toxicity for humans, the first compound (free cyanide) being the most toxic. (For phytotoxicity the ranking would be different, but here human exposure is our main concern.) The column 'rate of uptake in plants' shows an estimation how easily the plants take up the compound from the soil matrix. The uptake is not always passive the plant can influence the uptake of compounds actively in the root system or it can deactivate the compound after uptake, by degradation (e.g. for cyanides) or precipitation (e.g. for metaloxalates in spinach and other plants and in which case the presence of the compound is still relevant for human toxicity).

**Table 4:** Overview of some inorganic compounds (ranked according to estimated toxicity level) and some relevant characteristics in relation to selection for further studies in actual site specific risk analysis of soil contamination.

Contaminant	Rate of uptake by plants	Mobility in soils	Occurrence in situations of soil contamination	Other relevant legislation
<b>Cyanide (free form)</b>	high, but degraded by the plant	high	often (gas production plants)	no
<b>Thiocyanides</b>	?	moderate	often as degradation products of cyanides (gas production plants)	no
<b>Cyanides complex</b>	low	low	often (gas production plants)	no
<b>Sulphides</b>	moderate	moderate/low	moderate (agriculture / atmospheric deposition)	yes
<b>Fluoride</b>	low	low	infrequent (fertiliser, natural in clay)	no
<b>Bromide</b>	moderate	high	infrequent	no
<b>Ammonium</b>	high	high	often (agriculture)	yes
<b>Chloride</b>	moderate	high	sea water, road defrosting	yes
<b>Phosphate</b>	high	moderate	often (agriculture)	yes

Because of the diverse behaviour of compounds in the group of 'other inorganic compounds' each compound will have to be evaluated separately and no general set-up is possible (as will be tried with organic compound and metals). In the framework of *actual risk assessment* for contaminated sites we will restrict ourselves to compounds for which no separate legislation exists. Primarily the 3 groups of cyanides need further evaluation. Fluorides and bromides appear to be of secondary importance.

### 4.3 Organic compounds

For the calculation of the uptake of organic compounds in plants the CSOIL model makes a distinction between root and leaf crops (see Appendix 1 section 2.3, for the formulas). Statistical relations between crop content and porewater concentration developed by Briggs *et al.* [1982, 1983] are used for the calculations for both crop types. In both cases the octanol–water partitioning coefficient (Kow) plays a main role.

Briggs *et al.* did extensive experiments with barley on the uptake by roots from solutions with non-ionised chemicals, and the subsequent translocation to shoots. The more lipophilic the chemical was the more the uptake of chemicals by the roots increased. The lower limiting value was for polar chemicals [Briggs *et al.*, 1982]. Similar to the partitioning in roots, the accumulation of the compound in the lower section of shoots can be ascribed to a process in which the compound is partitioned between the shoot and the xylem transpiration stream [Briggs *et al.*, 1983]. The experiments resulted in equations which link the concentration in the external solution, via a bioconcentration factor (BCF), to the concentration in roots and stems of barley. Briggs *et al.* claim that the reported behaviour of many pesticides in various plant species agrees with the derived relationships. In CSOIL it is assumed that the root crop concentration as calculated by Briggs's equations equals the concentration in all subterranean parts of all crops considered. Also, the concentration in the stems is assumed to resemble the concentration in aerial parts of the crops, i.e. also for the leaves.

The Root Concentration Factor (RCF, a BCF for roots) relationship used by Briggs *et al.* [1982] has been derived for the log-Kow area from -1 to 5, covering almost the entire area covered by the compounds considered here. Briggs *et al.* [1982] reported deviations between measured and calculated values of a factor of 2.

The Stem Concentration Factor (SCF) relationship used by Briggs *et al.* [1983] has been derived for the log-Kow area from -0.57 to 3.7 (with 18 chemicals), while deviations up to a factor of 3 between calculated and measured values were found. An alternative Travis and Arms [1988] relationship for the SCF (see also section 5.1.2) has been derived using data in a log-Kow area of 1.15 to 9.35 (with 28 chemicals). Travis and Arms assume a linear relation between log Kow and the SCF. Briggs assumes a bellshaped curve arguing that at low log Kow (low lipophilicity of the compound) the lipid membranes of the plant roots limit translocation from porewater to the root while at high log Kow the high lipophilicity of the compound limits the transport in the plant with internal fluids. Generally, the same trend is observed in both alternatives. Relevant differences are only at log Kow <1. Briggs's relations are preferred on theoretical grounds (de Nijs, Vermeire, 1990).

CSOIL uses the bioconcentration factors in combination with calculated porewater concentration [Van den Berg, 1995]. The porewater concentration is calculated with the assumptions of equilibrium in partitioning. Calculation of the distribution of an organic contaminant over the three soil phases is based on the fugacity theory of Mackay *et al.* [1985]. To calculate the porewater concentration, it is necessary to obtain the soil–water partitioning coefficient (Kd). The Kd value is calculated from the Koc, the organic carbon based partition coefficient as:

$$K_d = f_{oc} \times K_{oc}$$

where  $f_{oc}$  = the organic carbon fraction in the soil.



It is recommended to use measured Koc or Kd values. When necessary the Koc value can be estimated from the octanol—water partitioning coefficient (Kow), with empirical formulas like (Karickhoff, 1981):

$$K_{oc} = 0.411 \times K_{ow}$$

or

$$\log K_{oc} = 0.989 \times \log K_{ow} - 0.346$$

The Kow values can be measured in relatively simple laboratory experiments. Kd, Koc and Kow are usually in L / kg.

Polder *et al.* [1994] validated models on the uptake of organic chemicals by plant roots in relation to the use in the model USES. The model USES is comparable to CSOIL in this aspect, because both use the same equations for plant uptake. They concluded Briggs's relationship to be satisfactory for subterranean parts of the plant. For aerial parts of the plants Briggs's relationship is not satisfactory for the purpose set in the USES model. Experimental concentration factors found in literature for plant shoots appear to be higher than the values calculated by Briggs. In the leaf water is transpired, leaving the substance in the leaf tissue. The stem will equilibrate with the transpiration stream. For aerial, above-ground, plant parts, other approaches, which can be applied in the general risk-assessment of compounds need to be explored.

In the Briggs equations the accumulation of an organic compound in the plant is calculated from the concentration in the porewater. The concentration in the porewater is calculated from the soil concentration using a Kd distribution factor. The dependency of the Kd factors on location specific parameters (i.e. the organic carbon fraction, being the most important in this case) is often known. This makes the model in principle fit for assessment of site specific accumulation in crops.

From the foregoing discussion it can be concluded that to improve the CSOIL formulas for organic chemicals the main focus should be on the plant uptake into aerial plant parts. The plant uptake into subterranean parts is described satisfactorily in the current CSOIL model, also for actual, site specific, risk assessment.

## 5 Models for uptake of soil contaminants by plants

Many researchers tried to model the processes on the uptake of compounds by plants and the distribution over the plant. Models describing these processes were made for several purposes, for example to describe the mass balance of chemicals in terrestrial systems, for application in agricultural management, or as part of a general model to describe direct and indirect exposure of compounds to humans. We will focus mainly on this last application which is usually related to policy making.

A search was done for models which incorporate a module for plant uptake from soil. Plant uptake through deposition from compounds in air was not considered. This route is considered in the CSOIL model, but will not be evaluated. Here, an overview is given of some models with resemblance to CSOIL's plant-module, regarding the purpose and development status of the model and the description of the processes of plant-uptake. The possibility to use available data collected for the model is analysed.

### 5.1. Uniform System for the Evaluation of Substances (USES) *for: organic substances and metals*

#### 5.1.1 Introduction of USES

The Uniform System for the Evaluation of Substances (Jager, Visser, 1994; Linders, Jager, 1997) is a tool that can be used for rapid, quantitative assessments of the general, potential risks of **organic** substances. USES is an instrument that can be used in decision making by central governments, research institutes and the chemical industry. For organic substances the approach of plant uptake in USES is identical to the CSOIL approach.

#### 5.1.2 Description of plant uptake in USES

The uptake of substances by plants is determined by the availability of the substance in the porewater, the uptake of water and substances by the roots, and the transportation from the root to other parts of a plant (Jager, Visser, 1994). This process is described in two steps. Root uptake is described according to Briggs *et al* (1982,1983) by defining the Transportation Stream Concentration Factor (TSCF), the steady state ratio of substance concentrations in xylem fluids and porewater (see also section 5.3.2). The TSCF is related to the  $K_{ow}$  (compound specific). The research of Briggs is based on barley. The concentration in the stem, relative to the concentration in the transportation stream is calculated making use of the Stem Concentration Factor (SCF), also related to the  $K_{ow}$ . SCF and TSCF combined give the relation for the partitioning between porewater and plant tissue. Briggs *et al.* (1983) also derived a relationship between porewater and root tissue (Root Concentration Factor, RCF).

In the USES document (Jager, Visser, 1994) it is noted that care has to be taken in which cases the above relations can be applied. The relations are derived from experiments with barley, only using a relatively small range of non-ionised chemicals. But at that time, no alternative for the equations were available. There is not much difference between the approach of Travis and Arms (1988) and the approach of Briggs *et al.* The approach of Briggs *et al.* (1983) is preferred in USES, since it incorporates a more plausible theoretical foundation. They found that their 'BCFroot', derived for barley, could also be used to describe the uptake of different substances in different plant species. This is an indication of the validity of their concept.

### 5.1.3 Validation of USES

In the RIVM report 'Validation of models on uptake of organic chemicals by plant roots' by Polder, et al. (1994) some conclusions are formulated:

#### *Positive conclusions*

- Is concluded that the method of calculating the *root* concentrations as derived by Briggs *et al.* (1982) is satisfactory for the purpose set in USES. It seems likely that the root concentration factor (RCF) found by Briggs is a maximum value, especially for substances with  $\log K_{ow} > 4$ .
- The experimental transpiration stream concentration factors (TSCF) found in the literature are more or less in agreement with the calculated values by Briggs *et al.* (1982).

#### *Negative conclusions*

- Several *shoot* concentration factors (SCF) found in the literature are much higher than the values calculated by Briggs *et al.* (1983). This might be due to uptake from air, non-equilibrium situations and in the case of radio-labelled substances, the SCF might be based on metabolites instead of parent compound.
- In addition Briggs *et al.* (1983) uses stem concentrations; leaves were excluded, whereas in the study of Polder, Hulzebos and Jager (1994) the SCF was based on shoots. For the SCF used in USES major adaptations are necessary.

The report includes experimental data for different organic compounds and different plant species (Tables 1 to 3 in Polder, Hulzebos and Jager, (1994)).

## 5.2. EUSES

*for: organic substances and metals*

### 5.2.1 Introduction of EUSES

EUSES is the European updated version of USES (see section 5.1)

### 5.2.2 Description of plant uptake in EUSES

For accumulation of contaminants in plants the model of Trapp & Matthies (1995) is used. This model is capable of dealing with a parameter input that is time dependent. In the framework of USES, the model output is compared with the datasets as described in the RIVM report 'Validation of models on uptake of organic chemicals by plant roots' by Polder, et al. (1994). Also in the report 'Bioconcentration of gaseous organic chemicals in plant leaves; comparison of experimental data with model predictions' by Polder, et al. (1996) a collected dataset was compared with the model of Trapp and Matthies (see section 5.3).

## 5.3. Trapp and Matthies

*for: organic substances*

### 5.3.1 Introduction to the model of Trapp and Matthies

According to Trapp and Matthies (1995) uptake of chemicals into vegetation is a major pathway for toxic substances into the food chain leading to human beings. Models that have been

developed for the uptake of chemicals by plants for agricultural purposes are often too sophisticated for the purpose of risk assessment. In their study they strictly simplify an existing model (Trapp and McFarlane, 1995) and they claim that the developed equation could be incorporated into existing models for the fate of contaminants in soil and other media. This 'simplified' model is obtaining an international status, for it was implemented in decision making instruments like EUSES and UMS (see 'EUSES' and 'UMS').

### 5.3.2 Description of plant uptake in Trapp and Matthies

The model of Trapp and Matthies (1995) considers uptake from soil; uptake from air; dilution of compounds in the plant by growth and loss of compounds by (photo)transformation. These processes are combined in a one-compartment model, consisting of one equation for the calculation of uptake into above-ground plants. The model is capable of dealing with time dependent input.

#### *Uptake from soil into roots*

Trapp & Matthies [1995] use the partition coefficient which describe the concentration ratio between two neighbouring phases in thermodynamic equilibrium and is a key property for the fate of compounds in the soil—plant—air system. For plant tissue, it can be calculated from (see also Trapp [1996] for correction)

$$K_{pw} = W_p + L_p \cdot K_{ow}^b$$

where  $K_{pw}$  is the partition coefficient between plant tissue and water (kg of substance/m<sup>3</sup> of plant per kg of substance / m<sup>3</sup> of water),  $W_p$  en  $L_p$  are the water content and the lipid content of the plant tissue (v/v),  $K_{ow}$  is the octanol-water partition coefficient of a compound [on basis of volumes: kg of substance / m<sup>3</sup> of octanol per kg of substance / m<sup>3</sup> of water (dimensionless), or alternatively as: kg of substance / kg octanol per kg of substance / L water (dimension L/kg), in the latter case substitute  $K_{ow}$  with the dimensionless ( $\rho_w K_{ow}$ ), with  $\rho_w$  the density of water kg/L] and  $b$  is a correction exponent for differences between plant lipids and octanol. The coefficient  $b$  is plant specific. Trapp & Matthies [1995] use data from barley shoots ( $b = 0.95$ ) The values of these  $b$  exponents are known for a number of other crops. For fine roots, the diffusive exchange with the soil is high, and it is assumed that near-equilibrium conditions are achieved. For thicker roots, the state of equilibrium is an upper limit for uptake, and the kinetics of uptake controls the concentration. The partition coefficient between roots and bulk soil  $K_{RB}$  is

$$K_{RB} = \frac{K_{RW}}{\rho_B K_d + \theta}$$

where  $K_{RW}$  is the partition coefficient between roots and pore water (mass /volume per mass/volume), calculated analogous to  $K_{pw}$  and the denominator is dimensionless with  $K_d$  the distribution coefficient between the bulk soil matrix and pore water (L/kg),  $\rho_B$  is the bulk soil density (kg/L), and  $\theta$  is the volumetric water fraction of the soil (v/v). The  $K_d$  is calculated from the  $K_{oc}$  and the fraction of organic carbon in the soil as discussed in section 4.3.

Trapp & Matthies [1995] concluded from a literature study that it is often sufficient as a rule of thumb to assume that the concentration in fine roots is in or below the order of magnitude of the concentration in soil.

### ***Translocation with transpiration stream***

Transport of substances from the roots to other plant parts, the translocation within the plant, is dependent on the properties of the transpiration stream. The dimensionless transpiration streamfactor (TSCF) is defined as the concentration ratio between xylem fluid and external solution (pore water in the soil). The mass transport within the xylem  $N_{\text{xylem}}$  (kg/s) is then

$$N_{\text{xylem}} = Q \cdot C_w \cdot \text{TSCF}$$

where  $Q$  is the transpiration stream (L/s),  $C_w$  is the concentration in soil water.  $C_w$  (mg/L) could be measured in the field or calculated with  $C_w \approx C_b / K_d$ , where  $C_b$  is the total concentration in (dry) bulk soil (mg/kg). The TSCF is related empirically to the  $K_{ow}$  as:

$$\text{TSCF} = 0.784 \cdot \exp \left[ \frac{-(\log K_{ow} - 1.78)^2}{2.44} \right]$$

From theoretical considerations follows that TSCF values of non-dissociating compounds should maximally be 1.

### ***Exchange with air***

Gaseous exchange between leaves and atmosphere is calculated based on the partition coefficient between leaves and air,  $K_{LA}$  is

$$K_{LA} = \frac{K_{LW}}{K_{AW}}$$

where  $K_{AW}$  is the partition coefficient between air and water (dimensionless Henry's constant) and  $K_{LW}$  is the partition coefficient between leaves and water (dimensionless). The  $K_{LW}$  is calculated like the  $K_{PW}$ .

### ***Growth***

The growth of plants depends on the stage of development. Shortly after germination, the growth is slow. Then the vegetative phase follows where the main growth takes place. Finally, during the maturation stage, the growth comes to an end.

During the exponential growth in the vegetative phase, the ratio between leaf area and volume ( $A/V_L$ ), and between transpiration and volume ( $Q/V_L$ ), is relatively constant.

## **5.4. Umweltmedizinische Beurteilung der Exposition des Menschen durch altlastenbedingte Schadstoffe (UMS)**

*for: metals, other inorganic and organic substances*

### **5.4.1 Introduction of UMS**

The UMS model is an exposure model. It has been developed in Germany (Umweltbundesamt, Fresenius Consult GmbH, focon- Ingenieurgesellschaft GmbH, 1993) for the assessment of the acute, latent and potential danger of polluted areas. The model could be applied during the preliminary phase of the research and during further study on the contaminated site.

In Germany every 'Bundesland' has one or several lists with guidelines and values for dealing with contaminated sites. Although the UMS model is not applied on a great scale yet, with the

model it might be possible for all 'Bundesländer' to follow a more or less similar procedure in the future.

In the UMS model plant parts for consumption can be contaminated through deposition of compounds from the air, through contaminated ground water and through contaminated soil. UMS differentiates between organic compounds, metals and other inorganic compounds. The soil properties and plant specific properties are indicated as important.

#### 5.4.2 Plant consumption in UMS

To be able to estimate the risk for man consuming plant material it is necessary to investigate the accumulation of contamination in plants and to find out how much plant material men consume. In the UMS model the consumer is divided into groups with different consumption rates;

- babies (no relevant exposure to plants);
- little children (90 g vegetables/d, 30% from own garden; 40 g grain/d, 25% from own garden);
- children (180 g vegetables/d, 30% from own garden; 80 g grain/d, 25% from own garden);
- teenagers (300 g vegetables/d, 30% from own garden; 100 g grain/d, 25% from own garden);
- adults (520 g vegetables/d, 30% from own garden; 180 g grain/d, 25% from own garden).

#### 5.4.3 Description of plant uptake in UMS

For each compound an individual transfer factor should be determined. This transfer factor equals the Bio Concentration Factor (BCF) used in CSOIL, being defined as the quotient of the concentration in the compound in the relevant plant part and the concentration of the compound in the soil or groundwater. The BCF's are metal specific and plant specific.

The general equation for plant uptake of all types of compounds from the soil is:

$$C_p = C_s \cdot BCF \cdot F_d$$

$C_p$  = concentration in the plant (mg/kg d.w.)

$C_s$  = concentration in soil (mg/kg d.w.)

BCF = transferfactor/ bioconcentration factor

$F_d$  = correction factor for depth of contamination

The correction factor indicates that it is also important to know at what depth a soil sample was taken. The correction factor ( $F_d$ ) used equals 1 for a depth of 0-1 meter and equals 0.2 for a depth larger than 1 meter.

#### 5.4.4 The modelling of the accumulation of metals in UMS

The *metals* discussed in the UMS document are Cd, Zn, Ni, Cu, Pb, Cr. For these metals the BCF-values of the following plant species are determined: maize, peas, oat, wheat, broadbeans, lamb's lettuce, carrots, leek, radish, celery and spinach. The data for these BCF's originate from Sauerbeck and Lübken (1991) and are mentioned and tabled in the UMS document. Additionally the metals As, Hg, Th and V are discussed shortly.

For all metals it is documented how the BCF depends on soil properties like pH, % organic carbon and clay content. This is discussed in more detail in the chapter on metals in section 6.3.

### 5.4.5 Similarity of CSOIL and UMS for other inorganic and organic compounds

Although in detail the German exposure model differs from the CSOIL-model, a lot of similarities can be found:

- 1) Since no better method was available, the concentration of *inorganic compounds* other than metals is similar to the calculation of these compounds in the CSOIL model.
- 2) The calculation of the *organic compounds* is similar to the calculation in the CSOIL-model. However, in the near future, for the calculation of plant uptake of organic compounds the model of Trapp and Matthies will be implemented in UMS (personal communication Frank Swartjes, 1996).

## 5.5. CalTOX

*for: organic compounds*

### 5.5.1 Introduction to CalTOX

CalTOX is a multimedia, multiple pathway risk assessment model, developed by the California Environmental protection agency, Department of Toxic Substances Control (DTSC). CalTOX relates concentrations of chemicals in soil to the risk of an adverse health effect for a person living or working on or near a contaminated site.

The DTSC currently makes use of a guidance document on hazardous waste-site risk assessment prepared by U.S. Environmental Protection (U.S. EPA, 1989). This document has a number of omissions. CalTOX adds time dependency, gives a range of risks or health based soil target clean up levels that reflect the uncertainty /variability of the estimates and it is available as computer spreadsheet.

Before CalTOX can be routinely used to assess risks, explicit guidance will be required. Once this guidance is developed by regulatory agencies, the use of CalTOX may improve the accuracy of the information on human health risk assessment (Computer Product Information Sheet, U.S. Department of Commerce, 1994).

### 5.5.2 Description of plant uptake in CalTOX

The model also describes ingestion. This includes ingestion of contaminated homegrown products. The model explicitly calculates the contamination of the exposure media of fruits, vegetables and grains by the chemical present in the environmental compartments of contaminated air (both the gas and water particles), water, surface soil and root-zone soil. Fruits and vegetables are divided into two groups: *exposed produce* representing above-ground edible plant parts and *protected produce* representing root crops and products protected from the environmental compartment of ambient air, such as citrus fruits. Grains are considered exposed produce.

To determine the concentrations in fruit, vegetables and grains, transfer coefficients (Tfs) are used describing the transfer of contaminants from each relevant environmental compartment to the product. Tfs are defined as the ratio's of concentrations in the product and the compartment. For the transfer from the soil the plant-soil partition coefficient from root-zone soil to roots (kg soil/kg fresh mass,  $K_{ps_{roots}}$ ) and the  $K_D$  (soil-soilwater partition coefficient) are used. Table 5 shows examples of a few parameter values in CalTOX). For these parameters is referred to the Tables I to IV of the report 'Parameter values and ranges for CalTOX' (1993).

**Table 5:** *Examples of the used parameter values in CalTOX:*

parameter description and symbol	estimation formula or symbol	CV <sup>1)</sup>	units	references
K <sub>ps</sub> (SHOOT/ROOTS) (plant/soil partition coefficient from root-zone soil to above ground plant parts),	<b>geochemical data, measured values or 7.7</b> <b>K<sub>ow</sub><sup>-0.58</sup> for org. chemicals</b>	<b>1 to 4</b>	<b>kg (soil)/kg (plant fresh mass)</b>	<b>Travis and Arms (1988)</b> <b>Bodek et al (1988)</b> <b>Bowen (1979)</b> <b>Ng (1982)</b> <b>Bennet (1981, 1982)</b>
K <sub>ps</sub> (roots/soil) (plant/soil partition coefficient from root-zone soil to roots, used for protected produce)	<b>geochemical data, measured values or 270</b> <b>K<sub>ow</sub><sup>-0.58</sup> for org. chemicals</b>	<b>1 to 4</b>	<b>kg (soil)/kg (plant fresh mass)</b>	<b>Travis and Arms (1988)</b> <b>Bodek et al (1988)</b> <b>Bowen (1979)</b> <b>Ng (1982)</b> <b>Bennet (1981, 1982)</b>
K <sub>ps</sub> (rain) (plant/soil partition coefficient for surface soil due to rainsplash)	<b>0.0034</b>	<b>1.0</b>	<b>kg (soil)/kg (plant fresh mass)</b>	<b>Dreicer et al (1984)</b>

<sup>1)</sup> CV= coefficient of variation

## 5.6. PESTLA

*for: pesticides*

### 5.6.1 Introduction to PESTLA

PESTLA is a model, developed at DLO/Staringscentrum that calculates accumulation of pesticides in the soil and the leaching of pesticides into the groundwater as a function of their sorption coefficient. It also calculates the modification of the pesticide in the soil of the agricultural land (Van der Linden en Boesten, 1989). It is used as an expertise model for the extent of use of pesticides and it only gives indicative values.

### 5.6.2 Description of plant uptake in PESTLA

The assumption is made that the concentration in the plant is 0.5 times concentration in soil water, being an empirical value for a wide range of crops and pesticides. Passive transport is assumed and no distinction between plant organs was made. These assumptions may be sufficient in relation to the goal of PESTLA, but are to simplified for our purposes. PESTLA will not be studied further in relation to the goal of the extension of CSOIL's plant module.



## 5.7. Overview of models for plant uptake

The tables in this section give an overview of different aspects of the models discussed in the previous sections of this chapter.

**Table 6 :** *Purpose, application and weak aspects of models*

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### 1. USES

purpose:	rapid, quantitative assessment of the general potential risks of metals and organic substances
application:	central governments, research institutes, chemical industries in decision making
weak aspect:	shoot concentration factors

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### 2. EUSES

purpose:	European version of USES
application:	decision making in the European community
weak aspects:	as Trapp and Matthies

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### 3. Trapp & Matthies

purpose:	simplification of models on uptake of organic chemicals in plants, for risk assessment
application:	applied in EUSES, UMS and by EPA,
weak aspects:	many parameters should be known

---

### 4. UMS

purpose:	quantification of acute, latent and potential exposure to polluted areas
application:	during preliminary phase of research and during further study of a contaminated site
weak aspects:	(organic chemicals similar to CSOIL,)
positive aspect:	metals; more specific than in CSOIL, soil specific features are incorporated

---

### 5. CalTOX

purpose:	relates concentrations of chemicals in the soil to the risk of an adverse health effect for a person living or working near the contaminated soil
application:	<i>in future</i> incorporation into regulatory decision requiring human health risk assessment
weak aspects:	laborious

---

### 6. PESTLA

purpose:	calculation of leaching and accumulation of pesticides
application:	expertise model for the implementation of pesticides
weak aspects:	not specific for compounds, simple concept.

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**Table 7 :** *Correspondence with CSOIL of the discussed models, with respect of the site-specific calculation of the accumulation of compounds in crops.*

aspect	CSOIL	Trapp & Matthies	USES	EUSES	UMS	CalTOX	PESTLA
metals	0	#	-	-	+	0	#
other inorganics	0	#	-	-	0	0	#
organics	0	+	0	+	0	0	0
roots/shoots	0	+	0	0	0	0	-
pH dependency	#	0	0	0	+	-	-
org% dependency	0	-	-	-	+	(1)	0
status	0	0	0	0	0	-	-
processes	0	+	0	+	0	+	-
BCFs	0	0	0	0	+	-	-

0 = similar or equal to CSOIL,

# = not available in model

+= superior to CSOIL for our purpose,

- = inferior to CSOIL for our purpose

(1) = CalTOX uses standard organic fractions for residential, commercial and industrial sites in California

**Table 8:** *Short overview of the aspects of the discussed models that are relevant for the application of site specific calculation of the accumulation of compounds in crops*

Model	Complexity	Implementation/ Acceptation	Purpose ~ CSOIL potential/actual	data available
Trapp & Matthies	moderate	in EUSES in UMS by EPA	potential and actual risks, organic chemicals	maybe in underlying literature
USES	low	national policy	potential risks, organic chemicals	in: Polder et al, 1994
EUSES	low	European policy	potential risks, organic chemicals	in: Polder et al, 1994 and 1996
UMS	low for org chemicals higher for metals	not implemented yet	actual risks, inorganic and organic chemicals	Sauerbeck and Lübken, and other literature
CalTOX	high	not implemented in policy	actual risks	no empirical data on plant uptake
PESTLA	low	widely used	potential risk	no data

## **5.8. Conclusion**

From the above evaluation of the available models we conclude that for *organic compounds* the model of Trapp and Matthies, with its wide acceptance, may be the most useful for further development of the CSOIL module for plant uptake. A drawback of the model of Trapp and Matthies, however, is that the values of many parameters should be collected. We recommend to do some effort to optimise the model on this aspect and try to simplify the model before actual implementation.

For *metals* we consider the UMS model based on extensive research on experimental BCF-values as the most useful of the reviewed approaches. Adapting for the Dutch situation is recommended. Also the model and data collection are still in development and can be improved. Chapter 6 goes in more detail into the possibilities for improvement and extension for metals.

For *other inorganic compounds* (e.g. cyanides) no general models for the uptake in plants were found and a customised approach seems necessary.

## 6 Possibilities for site-specific calculations for metals

In section 5.8 it was concluded that on the topic of accumulation of *metals* in plants the UMS-model looks promising for implementation in the CSOIL model. In this chapter the relations between UMS and the CSOIL module for plant-uptake of metals and the data from UMS and other possible sources are reviewed more extensively.

### 6.1 *The data on accumulation of metals in plants as used in CSOIL*

According to Bockting and van den Berg (1992) differences between experimental BCF-values for metals obtained from open field experiments and those from laboratory experiments with potted plants are remarkable. The latter experiments usually give higher BCF's. Possible causes for this difference are that the laboratory experiments result in :

- forced nutrition from the polluted zone,
- a higher root mass per volume and
- usually neglect ageing of the pollutants,
- neglect of or changes in temperature effects and deposition on the leaves.

As a consequence in cases where field experiment results were available, the results of laboratory experiments were excluded.

In some cases the phytotoxicity of the plants provide a natural protection for the consumers, because plants with visible deficiencies will be less attractive for consumption.

The EPA User's guide for soil screening guidance (1996) states that the data on phytotoxicity suggest that with exception of arsenic (and cadmium), metal concentrations that are considered toxic to plants are well below the levels that may impact human health through the soil-plant-human pathway. This limits the amounts that will be transferred to humans. However, bioavailability and as a consequence phytotoxic effects are usually very site-specific and it is recommended to evaluate this pathway and also ecological impacts of plant-uptake on a site-specific basis, regarding soil type, pH, plant types and chemical forms of the contaminants. The soil-plant-human pathway for sites with soils contaminated with cadmium or arsenic may be of particular concern.

The metal uptake by a plant is related to many parameters which are not taken in account in CSOIL:

- the soil characteristics (organic matter, pH, clay content, redox potential) and
- the ageing of the pollution,
- the difference in species of plants and between crop races and their resistential behaviour,
- the growing phase of the plant,
- the water management,
- concentration dependency of BCF-values.

The total metal concentration in older, full-grown plants is often lower than in younger and not fully grown species. Fruits only slightly accumulate metals and are therefore not taking into account as they are to be considered of minor threat to human exposure (Bockting, vd Berg, 1992).

The BCF values used in CSOIL are derived mainly from field experiments with a variety of vegetables carried out on different soil types polluted with sewage sludge by Sauerbeck (1988). The results presented by Sauerbeck are differentiated to plant species. The BCF values derived for potatoes and vegetables appear to differ considerably.

BCF values measured on slightly contaminated soils can be expected to be higher than the BCF's from polluted soils. Because the risk assessment is applied to serious soil pollution only experiments with polluted soils are taken into account.

The BCF-values calculated for CSOIL (see tables 3, 11) and based on reported literature results can give only an indication of the possible range. According to the Bockting and van den Berg it is not worthwhile to go more detailed into the relations between soil and plant contents, because there are too many factors possibly effecting the calculated BCF's.

## **6.2 Additional data on plant-uptake from literature**

In Switzerland and in the German Bundesland Baden Württemberg a legislation is proposed based on the contents and mobilities of metals in the soil (Gupta, et al. 1995). To assess and manage risk, in relation to the soil-plant-(animal-)human exposure routes the application of a three level evaluation system is proposed, incorporating the degree of metal contamination. Pseudo total metal fractions are introduced, subdivided in: mobile, potentially mobile and immobile and measured with a standard extraction procedure.

For lettuce and ryegrass the BCF values for some metals with respect to soil extracts are described in empirical equations as a function of the pH, CEC (cation exchange capacity) and mobile or to mobilise metal concentrations. The equations correlate rather well for Cu, Zn and Cd, but not for Pb (Aten, Gupta, 1996). This approach seems promising but still much work and research has to be done.

Several authors studied the suitability of metal soil extract concentrations in predicting the metal uptake of plants (Symeodis en Mcrae, 1977; Gruppe and Kuntze, 1988; Houba et al., 1992 and 1996; Novozamsky et al., 1995). In this approach the dependency of the BCF's on the soil characteristics is incorporated in the difference in the metal concentrations in the various soil extracts. Soil extract concentrations and plant contents often correlate rather good, but the relations are depending on the plant types and the selected parts of the plant. Especially the relatively mobile metals like Cd, Ni and Zn give rather good results, while Pb and Cu give poor results.

The results of comparable studies do not always show clear and uniform results. Besides, a lot of the reported studies are conducted in laboratory (vessel) experiments or with artificial polluted (spiked) metal solutions.

## **6.3 UMS: Studies of Sauerbeck**

The UMS model makes use of the experimental studies of Sauerbeck and Lübben (1991). These studies contain a rather comprehensive research to the effects of sewage sludge on soil, soil-organisms and plants. Several authors contributed to the report, in which the effects on a great variety of crops are described of long-time application of sewage sludge on a number of soil types at different locations.

Some major conclusions are:

- The effect of the soil characteristics on plant uptake is not uniform for all metals. Lowering of the pH resulted in a higher mobility of Cd, Ni and Zn in the soil, most likely (but not exclusively) resulting in a higher plant uptake. Within the pH range for normal agricultural circumstances pH variations had no significant effects on plant uptake of Cu, Pb and Cr.
- Soil extracts can be used to predict the uptake of Cd, Zn and Ni by plants.
- The experiments with laboratory vessels result in the same magnitude range for BCF's as comparable field experiments. No significant deviations are observed for laboratory and open

field experiments. Note that the described “laboratory vessel experiments” are conducted in the open field and with soil taken from the polluted field area. These experiments should therefore be regarded as semi-lysimeter experiments rather than laboratory vessel experiments.

- Differences in locations of field experiments, weather and climatic circumstances (deposition rates) can largely effect the results.

Arge Fresenius/ focon (unpublished, 1993) reported research to the calculation of the BCF-values for plant-uptake differentiated to soil characteristics and plants. This report was mainly based on the results of Sauerbeck and Lübben. For the metals (Cd, Cr, Cu, Mn, Ni, Pb and Zn) 1700 to 2350 experimental BCF-values were found for various plants and crops.

From these data completed with other literature data a calculated overall BCF (printed in bold in Table 9) is derived by a customised averaging expression. In this expression the contribution in the total human consumption of potatoes and a number of vegetables and fruits is taken into account.

$$BCF_{(total)} = 0.75 \times BCF_{(vegetables)} + 0.25 \times BCF_{(fruit)}$$

with

$$BCF_{(vegetables)} = (3/7) \times [(BCF_{(a)} + BCF_{(b)} + BCF_{(c)} + \dots) / n] + (4/7) \times BCF_{(potatoes)}$$

$$BCF_{(fruit)} = f \times BCF_{(vegetables)}$$

$BCF_{(x)}$  = BCF-values of specific vegetables

$n$  = number of vegetables

$f$  = fraction, value depending on metal, usually 0.1

Remarks to these equations:

- In the overall BCF calculation the fruit consumption is considered to be 25 % of the total consumption.
- The consumption of potatoes is 4/7 (60%) of the total vegetable consumption.
- In the BCF for potatoes the consumption of the whole potato including its peel is taken into account. The accumulation of metals in the peel is, however, considerably higher than the accumulation in the potato interior (differences up to a factor 10 are found between peeled and unpeeled potatoes).
- The BCF for fruits is calculated as a fraction of the BCF for vegetables.
- These rules can easily be adapted to the Dutch situation.

BCF values calculated with this procedure were adjusted to the effects of the soil characteristics on plant-uptake. The assumption is that the BCF's are basically dependent on the parameters pH, C-organic- and clay-content. The data are presented in a matrix and differentiation is restricted to regions defined in the matrix. The adjustment of the BCF's to soil characteristics by Arge Fresenius/focon is based on literature data and scientific expert knowledge. Unfortunately the process of adjusting the BCF's according to the effects of the soil characteristics is not well defined at the moment.

In Table 9 an example is given for the result for cadmium. In these tables the adjusted BCF's related to pH, C-organic and clay contents are given. The BCF for cadmium ranges from 0.1 to 3 depending on these soil characteristics.

Table 9 shows that for cadmium lowering the pH makes the overall BCF value increase.

Especially below pH= 5.5 the plant uptake is rather high, a factor 3 - 10 higher than for a

corresponding soil with a pH > 6.5. Increasing the clay and/or C-organic contents lowers the plant-uptake. These effects are most pronounced for soil with a pH > 6.5.

**Table 9 :** Overall BCF-values (based on German crop consumption packages) for Cd uptake in relation to soil characteristics

	pH <5.5			pH 5.5 -6.5			pH >6.5		
	C-org			C-org			C-org		
clay	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	3	2.5	2	1.6	1.2	0.8	1.2	0.8	0.5
15-40 %	2.5	2	1.5	1.2	<b>0.8</b>	0.5	0.8	0.5	0.3
>40 %%	2	1.5	1	0.8	0.5	0.3	0.5	0.3	0.1

Corresponding tables were made for Cr, Cu, Pb, Ni, Zn, As, Hg, Th, Se, V (and given in Appendix 3). In general the BCF range for the individual metals is comparable but smaller than the ranges found in the study for CSOIL (Bockting, van den Berg). This could be expected, as the results of the Arge Fresenius/focon study is based on a number of experiments all carried out with the same polluting matrix (sewage sludge) and similar general settings.

The pragmatic approach and the substantial dataset (1700-2350 ) provides a good bases for improving and extending the BCF-data set in the current CSOIL-model.

Note that it is not clear if this aggregation to an overall BCF-value does agree with the Dutch circumstances. In the literature it is clearly demonstrated that individual plant species (i.e. vegetables) may differ largely in the BCF-values. Table 10 shows that different choices of crops (similarly to variations in soil properties) can lead to large variations in the calculated BCF-values. The incorporation of fruits results in a reduction of approximately 20 % in the calculated overall BCF-values. For the Dutch circumstances the use of adapted consumption rates is recommended.

**Table 10:** BCF-values for uptake of Cadmium in various crops (from: Arge Fresenius / focon). The bold overall BCF-value for vegetables potatoes and fruit (in the tables 9 and 10) was calculated from several crops (not including maize and wheat) with the formula explained earlier in this section.

CROPS	BCF-value
Maize	0.08
Wheat	0.3
<b>Vegetables, potatoes and fruit</b>	<b>0.8</b>
- Beans	0.3
- Peas	0.3
- Leek	2
- Celeryloof	4.5
- Lettuce (cut)	4.5
- Carrots	0.75
- Radish	2
- Lamb's lettuce	0.3
- Spinach	4.5
- Potatoes	0.06

The figures show that variations due to crop types may be larger than due to soil types. For cadmium the maximum BCF-values are found for spinach, cellery and lettuce. For other metals the BCF values for other crops may be the highest (e.g. lead in radish, thallium in cabbage). This makes the choice of crops in the composition of the consumption package used in the calculations of the overall BCF value essential. Section 6.6 gives a first proposal.

In Table 11 an overview is given of the range of BCF-values as derived from Sauerbeck (1991) by Arge Fresenius /focon and the BCF-values as used in CSOIL by Van den Berg (1995). The average BCF-values of Arge Fresenius (1993) are generally higher than the corresponding BCF-values used in CSOIL. The upperlimit of the ranges can be substantially higher e.g. for Hg the upperlimit of the BCF is even a factor 33 higher than the BCF used in CSOIL.

**Table 11a** Ranges of BCF-values for a number of metals according to the CSOIL-model and according to Arge-Fresenius (value for standard soil and range in brackets)

Metals	BCF CSOIL potatoes	BCF CSOIL leave crops	BCF-range Arge Fresenius (1993) (veg., potatoes,/fruit)
As	0.015	0.03	0.05 (0.03 -0.07)
Ba	0.005	0.1	
Cd	0.15	0.7	0.8 (0.1 -3)
Co	0.015	0.03	
Cr	0.002	0.02	0.02 (0.001 -0.06)
Cu	0.1	0.1	0.3 (0.01 -0.8)
Hg	0.015	0.03	0.05 (0.002 -1) inorganic bound 0.15 (0.02 - 1) organic bound
Mo	0.015	0.3	
Ni	0.07	0.1	0.15 (0.01 -0.8)
Pb	0.001	0.03	0.02 (0.005 -0.08)
Se			1 (0.5 -2)
Sn	0.015	0.03	
Tl			2.3 (0.1 -7)
V			0.3 (0.3 -0.3)
Zn	0.1	0.4	1.1 (0.1 -2.9)

#### 6.4 Studies of Van Gestel et al.

At RIVM an experimental study to BCFs was performed as part of the project "Testing contaminated soils" (Van Gestel, 1992; De Wilde 1992, 1993). In this project the content and mobility of metals in various types of soils was studied by the analysis of contents, by leaching tests and by bioassays. The project contributed to the reference values for classification of building materials in the "Building Materials Decree". As a reference basis also the content and mobility of metals in soils from undisturbed sites was studied. Soils from 19 undisturbed natural reserves were sampled and extensively characterised.

In the studies of Van Gestel also BCF-values were measured in bioassays with radish and lettuce. These crops were chosen for the bioassay studies after a preliminary study into representability for plant-uptake behaviour with a larger number of crops. The principle of bioassays is that



standardised experiments are performed in such a way that they are more or less representative for the field behaviour. A complete mimicking of field tests is not tried. Advantage is that special disturbing factors in field tests as extreme weather conditions do not influence the results and less experimental noise can be expected. The standardisation of growing conditions was an important issue in the development of the bioassays. (A similar strategy has been followed with the leaching tests in the same project.)

Table 11b shows some results for the BCF values. Ranges of magnitude are generally similar to values of Arge Fresenius /focon. Averages of Van Gestel are generally higher. Table 10 shows that lettuce and radish take up relatively more Cd than most other crops. The low concentration of the undisturbed soils give rise to generally higher BCF values and this shows clearly the concentration dependency of the BCF. The BCF values of Zn and Cd are generally very close to each other.

**Table 11b** Some BCF ranges of Arge Fresenius based on consumption package and Van Gestel for radish and lettuce in contaminated and undisturbed soils (undisturbed soils in brackets)

Metal	BCF-range Arge Fresenius (1993)	BCF-range Van Gestel radish root	BCF-range Van Gestel radish leaf	BCF-range Van Gestel lettuce	Concentration ranges soil Van Gestel [mg/kg ds]
Cd	0.1 - 3	0.6 -2.4 (0.4 -26)	1.7 - 3.1 (1.3 -36)	1.5 - 4.3 (1.5 - 97)	4.5 -6.5 (<0.02 - 1.3)
Cu	0.01 - 0.8	0.6 - 1 (0.1 -30)	0.25 - 2.2 (0.2 - 10)	0.006 -1.6 (0.2 -5)	8 -50 (0.4 -27)
Pb	0.005 - 0.08	0.1 - 0.4 (0.01 - 3)	0.026 -0.1 (0.02 -0.2)	0.015 -0.1 (0.04 -0.5)	17 - 170 (<1 - 95)
Zn	0.1 - 2.9	0.9 - 2.6 (0.2 - 35)	2.8 -4 (0.5 -95)	0.4 -2 (0.2 -120)	48 -1005 (4.4 -127)

Appendix 4 gives an overview of the results of the project that are useful in relation to the study of the uptake of contaminants by plants.

- The ranges of BCF-values found for radish and lettuce in soils from *undisturbed* sites are given in Appendix 4 Table 1 (with respect to the concentrations in the soil) and Table 2 (with respect to the concentrations in the porewater).
- The ranges of BCF-values found in several *contaminated* soils, *contaminated and cleaned* soils and *contaminated, cleaned and matured* soils are found in Table 4 of Appendix 4.

These 3 tables in Appendix 4) can be compared with the collection of results of Sauerbeck, Baes and Bockting and Van den Berg as given in Table 3. Generally the results of Van Gestel et al. seem to contain larger variations (in the Tables only boundary values are given, no averages). It should be noted however that the soils studied by Van Gestel et al. cover a wide range of soil properties and consequently can be used very well to study the relations between soil parameters and BCF-values, as is shown below.

The experimentally found BCF-values were empirically related with the metal content in the soil and porewater, conductivity of porewater, pH, clay content, organic carbon content and CEC-values of the soil. In this part of the study the main attention was on undisturbed soils. For instance for Cadmium was found :

BCF factors for lettuce:

$$\log \text{BCFs}(\text{Cd}) = 5,6 - 0,04 \cdot \text{Corg} - 0,5 \cdot \text{pH}$$

$\log \text{BCFw}(\text{Cd}) = \text{not significant}$

BCF factors for radish root:

$$\log \text{BCFs}(\text{Cd}) = 6,9 - 0,01 \cdot \text{L} - 0,09 \cdot \text{Corg} - 1,1 \cdot \text{pH}$$

$$\log \text{BCFw}(\text{Cd}) = 5,2 + 0,5 \cdot \text{pH} - 0,0008 \cdot \text{G}$$

BCF-factors for radish leaf:

$$\log \text{BCFs}(\text{Cd}) = 4,4 - 0,001 \cdot \text{L} - 0,05 \cdot \text{Corg} - 0,7 \cdot \text{pH}$$

$$\log \text{BCFw}(\text{Cd}) = -1447 + 735 \cdot \text{pH} - 0,4 \cdot \text{G}$$

(Additionally) used symbols:

BCFs = bioconcentration factor with respect to soil, in undisturbed soils

BCFw = bioconcentration factor with respect to pore water, in undisturbed soils

G = conductivity of the pore water

L = clay content [%]

Corg = organic carbon content [%]

More examples are given in section 2.3 of Appendix 4. It should be noted that not all metals give significant results for the low concentrations found in the studied soils from undisturbed sites.

To make the results comparable with those of Sauerbeck (as presented in table 9) below a similar table is presented of ranges of variations for Cadmium-uptake in radish-root and radish-leaf, as calculated with the relations found by Van Gestel et al.:

**Table 12:** BCF of Cadmium in radish root (with respect to concentrations in the soil):

	pH =5.5			pH = 6			pH =6.5		
	C-org			C-org			C-org		
clay	1 %	2 %	3%	1 %	2 %	3%	1 %	2 %	3%
15 %	4	3.5	2.5	1.2	0.9	0.8	0.3	0.3	0.2
27.5 %	3	2.5	2	0.9	<b>0.7</b>	0.6	0.2	0.2	0.2
40 %	2.5	2	1.5	0.7	0.5	0.4	0.2	0.2	0.1

**Table 13:** BCF of Cadmium in radish leaf (with respect to concentrations in the soil)

	pH =5.5			pH = 6			pH =6.5		
	C-org			C-org			C-org		
clay	1 %	2 %	3%	1 %	2 %	3%	1 %	2 %	3%
15 %	3	2.5	2.5	1.4	1.2	1.1	0.6	0.5	0.5
27.5 %	3	2.5	2.5	1.3	<b>1.2</b>	1.1	0.6	0.5	0.5
40 %	3	2.5	2.5	1.3	1.2	1.0	0.6	0.5	0.5

The results of Sauerbeck for a range of vegetables, potatoes and fruits weighted on rate of consumption were given in Table 9. These values for Cadmium are in the same range as those presented in the Tables 12 and 13, usually between the values found for radish-roots and radish-leaf. (Radish was selected by Van Gestel et al. because of experienced representativity of this crop

for metal uptake.) Van Gestel similarly investigated the relations for a very large range of metals: As, Ba, Ca, Cd, Co, Cr, Cu, K, Mg, Mo, Na, Ni, Pb, Sb, Se, Sn, V, Zn, Hg. These results can be valuable for incorporation in the model for site-specific calculation of soil contaminants in crops, preferably in relation and in addition to the results of Sauerbeck.

### **6.5 Remarks on the relation of BCFs with concentration levels and on the speciation of metals**

A point of attention is the relation of the BCF-values with the level of contamination or the concentrations of the metals in the soil and porewater. For low concentrations metals can be useful and beneficial for the plant, for high concentrations phytotoxicity can occur. I.e. lead and zinc may interact with calcium and change the uptake rate and control of essential calcium in the plant (Russell, et al., 1995). If the plants die or appear unattractive they usually will not be consumed. The range of importance for risk analysis of human uptake is in between these levels. In this range it can be expected that many plants do have some mechanism to adapt to the overdose of metals, like: local extensions and reductions of the root system, reducing the entrance into the root system, precipitation and storage as immobilised oxalates in the leaves, or by fluid excretion.

Beside the level of concentrations of course also the speciation and chemical forms of the contaminating metals can be of importance. Probably this can be related with pH and redox-potential (which consequently can be useful field parameters for actual risk analysis). In the project "Testing contaminated soils" the mobility of the metals is more closely studied with leaching tests (see section 5.2 in Appendix 4), but also tests better designed for the study of the relations with plant-uptake of metals might be useful (i.e. extraction with calciumchloride, see Lagas, Groot, Koops et al (1996) who used this extraction technique to assess the bioavailability of metals in several types of soils in agricultural use and Andrews, Town, et al. (1996), who found that this technique gave the best estimate of plant availability of cadmium).

### **6.6 Crop differentiation**

In the UMS-model the BCF-values are calculated by performing a weighted average over a choice of crops (weighted in relation to the contribution to the average consumption). The procedure is outlined in section 6.3. The differences that are found between BCF-values of different crops, as shown in Table 10, make clear that the choice of crops can influence the results to a large extend.

To manage the first literature searches a limitation is necessary and suggested below. Wegener-Sleeswijk, Kleijn [1993] reported research on consumption of vegetables nursed in homegardens (originating from Hulshof, 1988). From these data and some estimations a classification is derived in Table 14 of crops to be studied with priority because of the estimated importance for the human uptake via crop consumption.

**Table 14:** Classification of crops with respect of the estimated importance for uptake via crop consumption [after Wegener-Sleeswijk, Kleijn, 1993].

Type	Crop	Frequently nursed in homegardens	Estimation of rate of contaminant uptake from soil	Rate of consumption [1]	Class
Greens	<i>Lettuce</i>	+	++	++	1
	<i>Spinach</i>	+	++	++	1
	<i>Endive</i>	+	++	++	1
	Cabbages	+	+	0/-	2
	Cauliflower	+	+	+?	2
	Curly kale	+	+	+	2
	<i>Cucumbers</i>	+	++	++	1
Tubers	<i>Potatoes</i>	+	++	+++	1
	<i>Carrots</i>	+	++	+	1
	<i>Red beetroot</i>	+	++	+	1
Bulbs	Onions	+	+	+	2
	Leek	+	+	(0)	2
Stems	<i>Rhubarb</i>	+	++	+	1
Beans	Peas	+	+	0	2
	Haricots	+	+	+	2
	French bean	+	+	0	2
	Horse bean	+	+	(-)	2
	Apples	+	-	(++)	2
Fruits	Pears	+	-	(+)	3
	Strawberries	+	+	(+)	2
	Red currants	+	0	(0)	3
	<i>Tomatoes</i>	+	+	++	1 ?
	Paprika	-	+	-	3
Grains	Wheat	-	0	(++)	4
	Corn/Maize	+	0+	(0)	3
	Barley	-	0	(+)	4

Rate of contaminant uptake is estimated high (++) for fast growers and consumed parts with intense soil contact and estimated low (-) for fruits from trees. Rate of consumption: estimation of the authors when bracketed.

If it is necessary and possible to differentiate for crops in the assessment of the actual risks, it is proposed that crops from class 1 from Table 14 (indicated in italic) will be considered first.

Maybe in Table 14 the sensitivity for uptake is underestimated in some cases, because of specific uptake in plant parts. Leafy materials may have high metal contents (as oxalates), starchy materials like beans and grains might be more sensitive for uptake of organic contaminants. (Also carrots can easily obtain organic compounds in their oil-cells, but carrots fall already in class 1). It is known also from the results of the UMS model, that fruits hardly take up contaminants from the soil (except possibly by deposition for low growers). To obtain some information on such aspects it is suggested to add various crops with lower priority, like for instance cauliflower, haricots, (apples,) onions and corn.

## 7 Conclusions and recommendations

The general conclusion from this study is that several possibilities and options were found to make site-specific calculations of the accumulation of soil contaminants in crops.

The CSOIL model is built for the analysis of potential risks. The formulas can, however, be made useful for estimations of actual risks, by calculating site-specific BCF-values. The building of a model for more complete site-specific risk can be successful using the current state of the art. It should be noted that an increased level of detail is unavoidable with the consequence of higher complexity and development costs.

More specific recommendations for future research are given in the following sections in relation to types of crops, contaminants and soils.

### 7.1 Crops

As shown, plant uptake is very crop-specific and the choice of crops considered is important for the results. The consequence of this specificity is also that for potential risk analysis, the selection of crops used in the calculations affects the result considerably.

The selection of crops to be considered can be made in relation to the average consumption rates or in relation to susceptibility for accumulation in the crops. The choice between these two can be a matter of policy. The UMS model gives us an excellent example how to work it out for average consumption rates (see sections 6.3 and 6.6). The choice of vegetables, potatoes and fruits and the weighting in the calculation of overall BCF values in the UMS-model may, however, be less representative for the Dutch situation, but it can be adapted. For site specific risk analysis the choice of crops used in calculations can also be made similar to that of the crops grown in the current situation for assessment, but only for an assessment made for a short timespan. For long-term assessment estimates, considering also foreseeable changes, crop consumption could be made similar to those of potential risks (dependent on the scenario for use of the soil).

To make implementation possible data should be collected for specific plants. From the current literature data for various plants are available for at least a number of metals. Collected data can also be used to pinpoint crops with a high uptake of contaminants.

### 7.2 Metals

The results of the German UMS model could, to a large extent, be used for extension of CSOIL's plant module to actual risk assessment. The UMS model used studies of Sauerbeck and Lübben, which contain much useful data. These data allow differentiation with respect to acidity, organic carbon content and clay content of the soil. The data are presented in a matrix and differentiation is restricted to regions defined in the matrix.

The UMS model uses statistical methods to construct empirical relations. As a consequence of the recommendation to use UMS, we do not recommend a mechanistic plant model for metals.

The studies of van Gestel *et al.* (1992a, 1992b) and others may also be useful as being complementary, especially to implement the influence of different soil parameters. The results of Van Gestel *et al.* lack, however, in crop differentiation as they tried here to make use of 'representative' crops. The results of Van Gestel *et al.* indicate that the general value for water content of crops used in the plant module needs reconsideration.

For field surveys the extraction of soil with a calcium chloride solution or with a standard leaching test can give useful additional information. The reported literature concerning relationships of concentrations in the extracts and in the plant is unfortunately still little.

Speciation of metals is a subject that needs attention; however, it cannot be considered for the plant module only and when considered it should be integrated in other parts of the CSOIL model too.

### **7.3 Other inorganic compounds**

A few suggestions were made to achieve better modelling of the plant uptake of the other inorganic compounds:

For the calculation of potential accumulation in plants the worst case assumption is used that all inorganic contaminants are completely solved. By evaluating the solubility characteristics of the inorganic compounds overestimation of risks can be reduced, especially for sulphates and sulphides.

The transport of the compound in the plant is of major importance when considering accumulation of these inorganic compounds in the consumed plant parts. A literature study concerning the transport mechanism in the plant might lead to a better model concept for some inorganic chemicals. It is recommended that knowledge (from agricultural sources) on the average evaporation rates and water content in plants should be incorporated into the model.

The behaviour of the different pH-dependent cyanide forms in the plant should be the subject of more literature studies, especially the degradation of cyanides in the plant. The thesis of Meeussen (1992) might be a helpful tool here.

We propose to give no priority to the macro-nutrients, nor to Cl, for plant N and P because they are covered by other legislation.

It is concluded that the uptake of these compounds in plants should be investigated for each compound separately in the framework of actual, site-specific risk analysis. We propose to focus primarily on cyanides as a frequent occurring contamination. Other inorganic compounds are covered by other legislation or have a very low toxicity level. Additional suggestions for further research are:

- evaluation of the solubility characteristics of cyanides, fluorine, sulphates and sulphides to avoid overestimation of risks;
- evaluation of evaporation rates and water content in plants;
- investigation of the different pH-dependent cyanide forms in plants;
- investigation of the difference between uptake in stem and leaves;
- investigation of selective uptake or 'filtering'.

### **7.4 Organic compounds**

In chapter 5 was concluded that the plant uptake into the roots is described satisfactorily in the current CSOIL model and that the formula is also useful for the site-specific calculation of accumulation of soil contaminants in plants. However, the CSOIL plant module for translocation to the stem and the leaves could be improved according to the generic one-compartment model of Trapp and Matthies [1995] for uptake of organic chemicals by foliar vegetation. Trapp and Matthies consider the following processes: translocation to shoots, gaseous deposition on leaves, volatilisation from leaves, metabolism and degradation processes and dilution by exponential growth. The model is, however, not (completely) validated. The model may be applied for site-specific actual risk assessment with differentiation of soil parameters (especially pH and organic carbon fractions) and plant parameters.

For translocation to consumed plant parts other than leaves, i.e. fruits and seeds, no model seems to be available. A complicating factor is that the model contains many variables that are difficult to trace.

The necessary variables should be obtained from the literature or investigations. We have not yet investigated how easy it is to obtain the necessary parameters.

To be able to make a list for substances for which further research is needed, a classification in relation to the importance of the exposure route of plant uptake has been made. Also frequency of appearance as soil contaminants and drawbacks in the model for specific substances have been studied and looked into as arguments for this selection.

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## Appendix 1. CSOIL (v. 7.1) formulas for deposition and crop content.

### 1. Deposition

The contribution of crop deposition is incorporated in the total crop content (see section 2) and calculated as follows:

$$Cdp = \frac{TSPo * DRo * frs * Cs * [ fin / (Yv * fEi) ] * \{ 1 - (1 - \exp [- fEi * te]) / (fEi * te) \}}{1}$$

Cdp : crop content compared to deposition [mg subst.kg<sup>-1</sup> dw crop]

TSPo : concentration particles in outdoor air = 0.07 [mg.m<sup>-3</sup>]

DRo : deposition velocity = 1 [cm.s<sup>-1</sup>] = 864 [m.d<sup>-1</sup>]

frs : fraction particles in soil = 0.5 [-]

Cs : concentration of contaminant in the soil [mg subst.kg<sup>-1</sup>.soil]

fin : fraction interception by crop = 0.4 [-]

Yv : crop yield = 0.28 [kg dw.m<sup>-2</sup>]

fEi : weathering constant = 0.033 [d<sup>-1</sup>]

te : growing period of a crop = 180 [d]

With the indicated fixed values of the model substituted this results in:

$$Cdp = 1.089 * 10^{-3} * Cs \quad [mg.kg^{-1} dw crop]$$

### 2. Crop content

#### 2.1 Metals

**root crops:**

$$Cpr = BCFr * Cs$$

Cpr : content of the root crop [mg subst.kg<sup>-1</sup> dw crop]

BCFr : bioconcentration factor root [(mg subst.kg<sup>-1</sup> dw crop)/(mg subst.kg<sup>-1</sup> dry soil)]  
(see Table 3 in the main report)

**leafy crops:**

$$Cps = BCFs * Cs + Cdp$$

Cps : content of the leafy crop [mg subst.kg<sup>-1</sup> dw crop]

BCFs : bioconcentration factor crop [(mg subst.kg<sup>-1</sup> dw crop)/(mg subst.kg<sup>-1</sup> dry soil)]  
(see Table 3 in the main report)

When no BCFr or BCFs are known:

$$\ln BCFr/s = 2.67 - 1.12 * \ln Kd$$

Kd : soil-water partitioning coefficient of the substance

*The Kd value can be estimated with the procedure shown in section 4.3 of the main report.*

## 2.2 Other inorganic compounds

*based on high water-solubility: concentration in pore water = concentration in plant moisture*

### **root crops:**

$$C_{pr}' = C_{pw} * (1 - fdwr)$$

$C_{pr}'$  : content of the root crop [mg substance.kg<sup>-1</sup> fresh weight crop]

$C_{pw}$  : concentration in pore water [mg. substance kg<sup>-1</sup> water]

$fdwr$  = ratio dry weight-fresh weight root crops [0.202 kg dw.kg<sup>-1</sup> fresh weight]

$$BCFr' = (1 - fdwr)$$

$BCFr'$  : bioconcentration factor root [(mg subst.kg<sup>-1</sup> fresh crop)/(mg subst.dm<sup>-3</sup> soil moisture)]

### **leafy crops:**

$$C_{ps}' = C_{pw} * (1 - fdws) + C_{dp} * fdws$$

$C_{ps}'$  : content of the leafy crop [mg subst.kg<sup>-1</sup> fresh weight crop]

$fdws$  = ratio dry weight -fresh weight leafy crops [0.117 kg dw.kg<sup>-1</sup> fresh]

$$BCFs' = (1 - fdws)$$

$BCFs'$  : bioconcentration factor stem [(mg subst.kg<sup>-1</sup> fresh crop)/(mg subst.dm<sup>-3</sup> soil moisture)]

## 2.3 Organic compounds

### **root crops:**

$$BCFr' = 10^{(0.77 * \log Kow - 1.52)} + 0.82$$

$BCFr'$  : bioconcentration factor root [(mg subst.kg<sup>-1</sup> fresh crop)/(mg subst.dm<sup>-3</sup> soil moisture)]

$$C_{pr}' = BCFr' * C_{pw}$$

$C_{pr}'$  : content of the root crop [mg subst.kg<sup>-1</sup> fresh weight crop]

### **stems:**

$$BCFs' : [10^{(0.95 * \log Kow - 2.05)} + 0.82] * [0.784 * 10^{(-0.434 * (\log Kow - 1.78)^{2/2.44})}]$$

$BCFs'$  : bioconcentration factor stem [(mg subst.kg<sup>-1</sup> fresh crop)/(mg subst.dm<sup>-3</sup> soil moisture)]

$$C_{ps}' = BCFs' * C_{pw} + C_{dp} * fdws$$

$C_{ps}'$ : content of the leafy crop [mg subst.kg<sup>-1</sup> fresh weight crop]

## Appendix 2: Relative contributions of exposure routes to the total human exposure from a contaminated soil

The exposure has been calculated on the basis of the CSOIL (v7.1) set of formulas, with a concentration  $C_s$  [mg.kg<sup>-1</sup>] in the soil equal to the proposed human toxicological Intervention Value. The indication \*\* behind some compounds names means that given this concentration in the soil, water-solubility is exceeded.

DI	: uptake of contaminant via soil-ingestion [mg.kg <sup>-1</sup> .d <sup>-1</sup> ]
DA	: uptake of contaminant via dermal contact with soil [mg.kg <sup>-1</sup> .d <sup>-1</sup> ]
IP	: uptake of contaminant by inhaled soil particles [mg.kg <sup>-1</sup> .d <sup>-1</sup> ]
IV	: uptake of contaminant via vapours [mg.kg <sup>-1</sup> .d <sup>-1</sup> ]
i	: indoor
o	: outdoor
VI	: uptake of contaminant via crop consumption [mg.kg <sup>-1</sup> .d <sup>-1</sup> ]
DI-w	: uptake of contaminant via drinking water [mg.kg <sup>-1</sup> .d <sup>-1</sup> ]
IV-w	: uptake of contaminant by inhaling vapour during showering [mg.kg <sup>-1</sup> .d <sup>-1</sup> ]
DOSE	: lifelong average daily exposure to contaminants [mg.kg <sup>-1</sup> .d <sup>-1</sup> ]
l	: lifelong exposure

Relative contributions of the various life long exposure routes (in percentages):  
(tables in Dutch)

### 1. Metals

Cs	DI-l	DA-l	IP-l	IV-li	IV-lo	VI-l	DI-wl	IV-wl	DA-wl	DOSIS	naam
6.78E+02	49	0	0	0	0	51	0	0	0	2.10E-03	arseen
4.26E+03	32	0	0	0	0	68	0	0	0	2.00E-02	barium
3.48E+01	5	0	0	0	0	95	0	0	0	1.00E-03	cadmium
2.25E+03	68	0	0	0	0	32	0	0	0	5.00E-03	chroom(III)
3.15E-01	68	0	0	0	0	32	0	0	0	7.00E-07	chroom(VI)
4.52E+02	49	0	0	0	0	51	0	0	0	1.40E-03	cobalt
1.57E+04	17	0	0	0	0	83	0	0	0	1.40E-01	koper
1.97E+02	49	0	0	0	0	51	0	0	0	6.10E-04	kwik
1.46E+03	61	0	0	0	0	39	0	0	0	3.60E-03	lood
9.11E+02	14	0	0	0	0	86	0	0	0	1.00E-02	molybdeen
6.58E+03	20	0	0	0	0	80	0	0	0	5.00E-02	nikkel
6.46E+05	49	0	0	0	0	51	0	0	0	2.00E+00	tin
5.65E+04	9	0	0	0	0	91	0	0	0	1.00E+00	zink



## 2. Other inorganic compounds

Cs	DI-l	DA-l	IP-l	IV-li	IV-lo	VI-l	DI-wl	IV-wl	DA-wl	DOSIS	naam
5.71E+02	0	0	0	0	0	100	0	0	0	1.70E+00	ammonium verbindingen
3.36E+02	0	0	0	0	0	100	0	0	0	1.00E+00	bromiden
1.68E+01	0	0	0	0	0	100	0	0	0	5.00E-02	cyaniden vrij
4.36E+00	0	0	0	0	0	100	0	0	0	1.30E-02	cyaniden complex
2.35E+01	0	0	0	0	0	100	0	0	0	7.00E-02	fluoriden
2.35E+04	0	0	0	0	0	100	0	0	0	7.00E+01	fosfaten
36E-01	0	0	0	0	0	100	0	0	0	1.00E-03	sulfiden
3.69E+00	0	0	0	0	0	100	0	0	0	1.10E-02	thiocyanaten

## 3. Aromatics

Cs	DI-l	DA-l	IP-l	IV-li	IV-lo	VI-l	DI-wl	IV-wl	DA-wl	DOSIS	naam
1.09E+00	0	0	0	92	0	6	2	0	0	4.30E-03	benzeen
2.44E+02	0	0	0	80	0	15	2	0	3	1.36E-01	ethylbenzeen
7.41E+01	0	0	0	30	0	70	0	0	0	6.00E-02	fenol
1.17E+02	0	0	0	18	0	66	13	0	2	5.00E-02	cresol(p)
3.39E+02	0	0	0	87	0	10	1	0	1	4.30E-01	tolueen
2.56E+01	0	0	0	74	0	20	2	0	3	1.00E-02	xyleen(m)
2.28E+01	0	0	0	36	0	61	3	0	0	4.00E-02	catechol
1.04E+01	0	0	0	37	0	60	3	0	0	2.00E-02	resorcinol
1.08E+01	0	0	0	40	0	57	3	0	0	2.50E-02	hydrochinon

## 4. PAHs

Cs	DI-l	DA-l	IP-l	IV-li	IV-lo	VI-l	DI-wl	IV-wl	DA-wl	DOSIS	naam
2.90E+04	88	0	1	0	0	5	0	0	0	5.00E-02	antraceen **
1.12E+04	84	0	1	0	0	9	0	0	0	2.00E-02	benzo(a)anthraceen **
7.56E+03	57	0	0	0	0	38	0	0	0	2.00E-02	benzo(k)fluoraantheen **
1.11E+03	84	0	1	0	0	10	0	0	0	2.00E-03	benzo(a)pyreen **
4.20E+02	32	0	0	0	0	66	0	0	0	2.00E-03	chryseen **
6.61E+02	5	0	0	1	0	91	0	0	3	2.00E-02	fenantreen
1.07E+03	8	0	0	0	0	91	0	0	0	2.00E-02	fluoranteen
1.18E+04	89	0	1	0	0	4	0	0	0	2.00E-02	indeno(1,2,3cd)pyreen **
1.20E+04	90	0	1	0	0	2	0	0	0	2.00E-02	benzo(ghi)peryleen **
6.63E+03	50	0	0	0	0	46	0	0	0	2.00E-02	pyreen **

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6.03E+02	2	0	0	9	0	84	2	0	4	5.00E-02	naftaleen
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## 5. Volatile chlorinated aliphates

Cs	DI-l	DA-l	IP-l	IV-li	IV-lo	VI-l	DI-wl	IV-wl	DA-wl	DOSIS	naam
3.86E+00	0	0	0	82	0	16	1	0	0	1.40E-02	1,2-dichloorethaan
1.89E+01	0	0	0	88	0	10	1	0	0	6.00E-02	dichloormethaan
9.21E-01	0	0	0	96	0	3	0	0	0	4.00E-03	tetrachloormethaan
3.89E+00	0	0	0	96	0	3	0	0	0	1.60E-02	tetrachlooretheen
8.86E+00	0	0	0	89	0	9	2	0	0	3.00E-02	trichloormethaan
3.03E+02	0	0	0	91	0	7	1	0	0	5.40E-01	trichlooretheen
7.72E-02	0	0	0	100	0	0	0	0	0	3.50E-03	vinylchloride

## 6. Chlorobenzenes

Cs	DI-l	DA-l	IP-l	IV-li	IV-lo	VI-l	DI-wl	IV-wl	DA-wl	DOSIS	naam
5.20E+02	0	0	0	69	0	20	6	1	4	3.00E-01	monochloorbenzeen
1.15E+03	1	0	0	50	0	41	3	1	5	1.90E-01	p-dichloorbenzeen
9.04E+00	3	0	0	22	0	70	1	0	4	5.00E-04	trichloorbenzeen(1,2,4)
1.80E+01	5	0	0	2	0	89	0	0	2	5.00E-04	tetrachloorbenzeen(1,2,3,4)
2.37E+01	7	0	0	3	0	88	0	0	1	5.00E-04	pentachloorbenzeen
2.68E+01	8	0	0	0	0	90	0	0	0	5.00E-04	hexachloorbenzeen
1.40E+01	1	0	0	16	0	83	0	0	0	3.00E-03	monochloorfenol (2)
3.25E+01	2	0	0	7	0	91	0	0	0	3.00E-03	dichloorfenol (2,4)
5.63E+01	3	0	0	2	0	92	1	0	2	3.00E-03	trichloorfenol (2,3,4)
1.83E+01	1	0	0	7	0	86	5	0	1	3.00E-03	tetrachloorfenol (2,3,4,5)
7.98E+01	0	0	0	0	0	97	1	0	2	3.00E-02	pentachloorfenol
9.12E+00	3	0	0	10	0	84	1	0	3	5.00E-04	chloornaftaleen
5.52E+00	9	0	0	0	0	90	0	0	0	9.00E-05	trichloorbifenyyl (2,5,2')
8.72E+00	15	0	0	0	0	84	0	0	0	9.00E-05	hexachloorbifenyyl

## 7. Drins

Cs	DI-l	DA-l	IP-l	IV-li	IV-lo	VI-l	DI-wl	IV-wl	DA-wl	DOSIS	naam
1.38E+01	21	0	0	0	0	77	0	0	0	1.00E-04	aldrin
5.45E+00	8	0	0	0	0	91	0	0	0	1.00E-04	dieldrin
4.36E+00	7	0	0	0	0	92	0	0	0	1.00E-04	endrin

## 8. Pesticides

Cs	DI-l	DA-l	IP-l	IV-li	IV-lo	VI-l	DI-wl	IV-wl	DA-wl	DOSIS	naam
1.13E+04	85	0	1	0	0	8	0	0	0	2.00E-02	DDT **
7.83E+03	59	0	0	0	0	36	0	0	0	2.00E-02	DDE **
2.48E+01	4	0	0	2	0	93	1	0	0	1.00E-03	d-HCH
1.82E+01	3	0	0	3	0	93	1	0	0	1.00E-03	HCH
4.23E-01	3	0	0	2	0	93	1	0	0	2.00E-05	b-HCH
2.11E+01	3	0	0	2	0	93	1	0	0	1.00E-03	g-HCH
4.61E+02	7	0	0	16	0	77	0	0	0	1.00E-02	carbaryl
4.35E+02	7	0	0	0	0	92	0	0	0	1.00E-02	carbofuran
6.80E+02	5	0	0	1	0	93	0	0	1	2.00E-02	propoxur
2.98E+04	90	0	1	0	0	3	0	0	0	5.00E-02	maneb **
2.10E+01	1	0	0	1	0	95	3	0	0	5.00E-03	atrazin

## 9. Solvents

Cs	DI-l	DA-l	IP-l	IV-li	IV-lo	VI-l	DI-wl	IV-wl	DA-wl	DOSIS	naam
5.29E+02	0	0	0	99	0	1	0	0	0	3.10E+00	heptaan
3.71E+02	0	0	0	99	0	0	0	0	0	3.10E+00	octaan
9.12E+03	0	0	0	29	0	69	2	0	0	4.60E+00	cyclohexanon
7.76E+02	5	0	0	1	0	91	1	0	1	2.50E-02	butylbenzylftalaat
4.63E+03	28	0	0	0	0	69	0	0	0	2.50E-02	di(2-ethylhexyl)ftalaat **
1.06E+00	0	0	0	87	0	9	1	0	2	1.00E-03	pyridine
2.49E+02	0	0	0	62	0	29	3	1	5	7.70E-02	styreen
3.99E-01	0	0	0	99	0	0	0	0	0	1.00E-02	tetrahydrofuran
9.39E+01	0	0	0	97	0	2	0	0	1	1.80E-01	tetrahydrothiofeen

### Appendix 3: Overview of BCF-values for metals in relation to soil characteristics as derived by Sauerbeck

**Table 1:** BCF's for **Cd** uptake in relation to soil characteristics

	<b>pH &lt;5.5</b>			<b>pH 5.5 -6.5</b>			<b>pH &gt;6.5</b>		
	<b>C-org</b>			<b>C-org</b>			<b>C-org</b>		
<b>clay</b>	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	3	2.5	2	1.6	1.2	0.8	1.2	0.8	0.5
15-40 %	2.5	2	1.5	1.2	<b>0.8</b>	0.5	0.8	0.5	0.3
>40 %%	2	1.5	1	0.8	0.5	0.3	0.5	0.3	0.1

**Table 2:** BCF's for **Zn** uptake in relation to soil characteristics

	<b>pH &lt;5.5</b>			<b>pH 5.5 -6.5</b>			<b>pH &gt;6.5</b>		
	<b>C-org</b>			<b>C-org</b>			<b>C-org</b>		
<b>clay</b>	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	2.9	2.6	2.3	1.7	1.5	1.1	0.9	0.8	0.7
15-40 %	2.3	2	1.7	1.3	<b>1.1</b>	0.9	0.6	0.5	0.4
>40 %%	1.7	1.4	1.1	0.9	0.7	0.5	0.3	0.2	0.1

**Table 3:** BCF's for **Ni** uptake in relation to soil characteristics

	<b>pH &lt;5.5</b>			<b>pH 5.5 -6.5</b>			<b>pH &gt;6.5</b>		
	<b>C-org</b>			<b>C-org</b>			<b>C-org</b>		
<b>clay</b>	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	0.8	0.7	0.6	0.22	0.2	0.18	0.09	0.08	0.07
15-40 %	0.6	0.5	0.4	0.17	<b>0.15</b>	0.13	0.06	0.05	0.04
>40 %%	0.4	0.3	0.2	0.12	0.1	0.08	0.03	0.02	0.01

**Table 4:** BCF's for **Cu** uptake in relation to soil characteristics

	<b>pH &lt;5.5</b>			<b>pH 5.5 -6.5</b>			<b>pH &gt;6.5</b>		
	<b>C-org</b>			<b>C-org</b>			<b>C-org</b>		
<b>clay</b>	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	0.8	0.65	0.5	0.5	0.4	0.3	0.2	0.15	0.1
15-40 %	0.65	0.5	0.35	0.4	<b>0.3</b>	0.2	0.15	0.1	0.05
>40 %%	0.5	0.35	0.1	0.3	0.2	0.1	0.1	0.05	0.01

**Table 5:** BCF's for **Pb** uptake in relation to soil characteristics

	pH <5.5			pH 5.5 -6.5			pH >6.5		
	C-org			C-org			C-org		
clay	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	0.08)*	0.06	0.04	0.06	0.04	0.04	0.2	0.015	0.01
15-40 %	0.06	0.04	0.02	0.04	<b>0.02</b>	0.01	0.015	0.01	0.005
>40 %%	0.04	0.02	0.01	0.02	0.01	0.05	0.01	0.005	0.005

)\* corrected

**Table 6:** BCF's for Cr uptake in relation to soil characteristics

	pH <5.5			pH 5.5 -6.5			pH >6.5		
	C-org			C-org			C-org		
clay	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	0.06	0.05	0.04	0.04	0.03	0.02	0.03	0.02	0.01
15-40 %	0.05	0.04	0.03	0.03	<b>0.02</b>	0.01	0.02	0.01	0.005
>40 %%	0.04	0.03	0.02	0.02	0.01	0.005	0.01	0.005	0.001

**Table 7:** BCF's for As uptake in relation to soil characteristics

	pH <5.5			pH 5.5 -6.5			pH >6.5		
	C-org			C-org			C-org		
clay	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	0.07	0.06	0.05	0.07	0.06	0.05	0.07	0.06	0.05
15-40 %	0.06	0.05	0.04	0.06	<b>0.05</b>	0.04	0.06	0.05	0.04
>40 %%	0.05	0.04	0.03	0.05	0.04	0.03	0.05	0.04	0.03

**Table 8:** BCF's for Hg (*inorganic bound*) uptake in relation to soil characteristics

	pH <5.5			pH 5.5 -6.5			pH >6.5		
	C-org			C-org			C-org		
clay	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	1	0.8	0.6	0.08	0.06	0.06	0.008	0.006	0.004
15-40 %	0.9	0.7	0.5	0.07	<b>0.05</b>	0.03	0.007	0.005	0.003
>40 %%	0.8	0.6	0.4	0.04	0.04	0.02	0.006	0.004	0.002

**Table 9:** BCF's for **Hg (organic bound)** uptake in relation to soil characteristics

	<b>pH &lt;5.5</b>			<b>pH 5.5 -6.5</b>			<b>pH &gt;6.5</b>		
	<b>C-org</b>			<b>C-org</b>			<b>C-org</b>		
<b>clay</b>	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	1	0.8	0.6	0.21	0.18	0.15	0.08	0.06	0.04
15-40 %	0.9	0.7	0.5	0.18	<b>0.15</b>	0.12	0.07	0.05	0.03
>40 %%	0.8	0.6	0.4	0.15	0.04	0.09	0.06	0.04	0.02

**Table 10:** BCF's for **Tl** uptake in relation to soil characteristics

	<b>pH &lt;5.5</b>			<b>pH 5.5 -6.5</b>			<b>pH &gt;6.5</b>		
	<b>C-org</b>			<b>C-org</b>			<b>C-org</b>		
<b>clay</b>	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%	7	6	5	3.7	3	2.3	2	1.5	1
15-40 %	6	5	4	3	<b>2.3</b>	1.6	1.5	1	0.5
>40 %%	5	4	3	2.3	1.6	0.9	1	0.5	0.1

**Table 11:** BCF's for **Se** uptake in relation to soil characteristics

	<b>pH &lt;5.5</b>			<b>pH 5.5 -6.5</b>			<b>pH &gt;6.5</b>		
	<b>C-org</b>			<b>C-org</b>			<b>C-org</b>		
<b>clay</b>	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%									
15-40 %		0.5			<b>1</b>			2	
>40 %%									

**Table 12:** BCF's for **V** uptake in relation to soil characteristics

	<b>pH &lt;5.5</b>			<b>pH 5.5 -6.5</b>			<b>pH &gt;6.5</b>		
	<b>C-org</b>			<b>C-org</b>			<b>C-org</b>		
<b>clay</b>	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%	<1 %	1-3 %	> 3%
< 15%									
15-40 %		0.3			<b>0.3</b>			0.3	
>40 %%									

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## **Appendix 4: Review of useful data for metals from the project "Testing contaminated soils"**

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### **1. Introduction to the project "Testing of contaminated soils"**

The project "Testing contaminated soil" was organised to test and value concentrations of metals and PAHs in soil. Contaminated, cleaned and supposedly clean soils were sampled and analysed in several ways. The project was in the first place meant to develop a better framework for the soils that were treated with wet soil cleaning techniques. It was organised because the suspicion had risen that though the concentration of contaminants can be lowered with these techniques, in the same time the mobility of the residual fraction (which is often still a 2 digit percentage of the original amount) could have been increased.

Additionally leaching tests for soils were tried out and developed further in the project and the project was used as a pilot for the development of bioassays. The uncertainty in the concentrations of metals in soils and leaching percolates (introduced by factors that deregulate the regular chemical analysis) were examined more closely and methods for chemical analysis were consequently adjusted.

The results appeared also to be useful for the 'Regulation on Building Materials', especially to support the development of concentration boundary values for classes of building materials. The reference levels for the leaching of metals from clean soils were defined from results of the project. The bioassays were meant to support these reference values by studying the concentrations in plants and earthworms in the same normal "healthy" soil.

The used samples of clean soils were taken at 19 supposedly clean sites from the topsoil and the subsoil. The sites were normally not accessible nature reserves with undisturbed soil (most of these sites were sampled earlier by Edelman or a study into background values of metals). Concentrations in the soil were measured for metals (As, Ba, Ca, Cd, Co, Cr, Cu, K, Mg, Mo, Na, Ni, Pb, Sb, Se, Sn, V, Zn, Hg) and PAHs (16 EPA), and also soil parameters were determined (pH, clay content, organic carbon content, CEC). Additionally leaching behaviour of metals was determined. Bioassays were developed and performed with lettuce, radish (root and leaf), earthworms and 'Microtox'. An OECD-reference soil was added to the samples in all experiments.

Contaminated and cleaned soils were sampled during the processing at 3 types of wet soil cleaning plants (flotation, acid extraction, high pressure scrubbing) from input and output of the process, considering the passage time in the soil cleaning plant. For these soils only the contaminating metals: As, Cd, Cu, Pb and Zn were analysed (with respect of concentrations, leaching behaviour and bioassays). Also the effect of maturing of the cleaned soil was considered. For the properties of the sampled soils see section 5.

### **2. Experimental results for BCF-factors of plants in soils from undisturbed sites**

#### **2.1 BCF- values**

BCF factors of metals were measured for radish root, radish leaf and lettuce in supposedly clean soils as ratios of the concentration in the plant parts to the concentrations in the soil and to the

concentrations in the pore water. The following BCF-factors are measured with respect to concentrations in soil (from measurements in 15 -20 different soil samples):

**Table 1:** BCF-values with respect to the concentrations in the soils from undisturbed sites

	<u>Radish root</u>	<u>Radish leaf</u>	<u>Lettuce</u>
Ba	0,09 - 11	0,22 - 4,9	0,042 - 3,5
Ca	0,53 - 42	0,9 - 57	0,60 - 43
Cd	0,39 - 26	1,3 - 36	1,5 - 97
Co	-	-	0,35 - 40
Cu	0,12 - 43	0,012 - 5,7	0,030 - 2,8
Cr	-	-	0,18 - 4,9
Hg	0,079 - 12	0,13 - 3,5	0,31 - 4,2
K	18 - 258	20 - 140	31 - 312
Mg	0,25 - 23	0,48 - 40	0,11 - 24
Na	2,1 - 337	3,0 - 106	0,74 - 88
Ni	0,067 - 4,9	0,054 - 1,2	0,016 - 3,0
Pb	0,01 - 2,8	0,015 - 0,2	0,004 - 0,52
Zn	0,24 - 35	0,46 - 95	0,5 - 122

The following BCFw-factors are measured with respect to concentrations in pore water:

**Table 2:** BCFw-values with respect to the pore-water in the soils from undisturbed sites

	<u>Radish root</u>	<u>Radish leaf</u>	<u>Lettuce</u>
As	-	-	57 - 852
Ba	18 - 732	68 - 497	21 - 358
Ca	7 - 184	52 - 853	11 - 172
Cd	63 - 1531	109 - 2708	632 - 9244
Co	-	-	111 - 2555
Cu	16 - 541	1,1 - 108	18 - 79
Cr	-	-	27 - 1470
K	143 - 3645	137 - 3705	168 - 4574
Mg	15 - 94	28 - 248	2,6 - 201
Mo	-	-	204 - 1582
Na	22 - 457	33 - 262	2,2 - 312
Ni	20 - 404	25 - 212	13 - 379
Pb	44 - 1048	8 - 189	4,9 - 481
Zn	57 - 438	175 - 884	54 - 990

## 2.2 Relations of BCF-factors with soil parameters

The following relations were evaluated:

- (a) for radish root, radish leaf and lettuce: the BCF-factors of Ba, Ca, Cd, Cu, K, Mg, Na, Ni, Pb, Zn as functions of the concentrations in
  - the pore water and in relation to contents of organic carbon, clay content and pH-H<sub>2</sub>O
  - and



- the soil and in relation to contents of organic carbon, clay content and pH-H<sub>2</sub>O
- (b) Concentrations in pore water as functions of the concentration in
  - the soil and in relation to organic carbon, clay content and pH

### 2.3 Examples of relations of the BCF-factors with soil parameters

A selection of the derived relations, for Cd, Cu, Zn (see the original report of Van Gestel, Dirven-van Breemen and Kamerman, RIVM rapnr. 216402004 (1992) for the other relations):

#### *Used symbols:*

- [X] = concentration X in mg/kg, in soil from undisturbed site
- [X]<sub>medium</sub> = concentration X in mg/kg, in other medium as soil from undisturbed site
- BCF<sub>g</sub> = bioconcentration factor with respect to soil in soil from undisturbed site
- BCF<sub>w</sub> = bioconcentration factor with respect to pore water in soil from undisturbed site
- K<sub>p</sub> = partition coefficient concentration X in pore water and in soil from undisturbed site
- L = % clay content
- OC = % organic carbon content
- pH = pH-H<sub>2</sub>O
- G = conductivity of the pore water

#### *Examples of relations for cadmium, copper and zinc:*

##### Background values

$$[Cd] = -0,4 + 0,006 \cdot L + 0,03 \cdot OC + 0,09 \cdot pH$$

$$[Cu] = -2,0 + 0,6 \cdot L + 0,15 \cdot OC + 1,0 \cdot pH$$

$$[Zn] = -31 + 2,0 \cdot L + 0,3 \cdot OC + 2,0 \cdot pH$$

$$K_p(Cd) = -1421 - 2,9 \cdot L + 26 \cdot OC + 357 \cdot pH$$

$$K_p(Cu) = +1,3 + 0,04 \cdot L + 0,04 \cdot OC + 0,4 \cdot pH$$

$$\log K_p(Zn) = -1,4 + 0,06 \cdot L + 0,02 \cdot OC + 1,0 \cdot pH$$

##### BCF for lettuce:

$$\log BCF_g(Cd) = 5,6 - 0,04 \cdot OC - 0,5 \cdot pH$$

$$\log BCF_g(Cu) = 6,3 - 0,06 \cdot OC - 1,4 \cdot pH$$

$$\log BCF_g(Zn) = 5,3 - 0,06 \cdot L - 0,03 \cdot OC - 0,6 \cdot pH$$

$$BCF_w(Cd) = \text{not significant}$$

$$BCF_w(Cu) = \text{not significant}$$

$$BCF_w(Zn) = -858 + 317 \cdot pH - 0,15 \cdot G$$

##### BCF for radish root:

$$\log BCF_g(Cd) = 6,9 - 0,01 \cdot L - 0,09 \cdot OC - 1,1 \cdot pH$$

$$\log BCF_g(Cu) = 5,2 - 0,06 \cdot L - 0,08 \cdot OC - 0,9 \cdot pH$$

$$\log BCF_g(Zn) = 7,0 - 0,06 \cdot L - 0,05 \cdot OC - 1,0 \cdot pH$$

$$\log BCF_w(Cd) = 5,2 + 0,5 \cdot pH - 0,0008 \cdot G$$

$$\log BCF_w(Cu) = 6,7 - 0,39 \cdot pH - 0,0004 \cdot G$$

$$\log BCF_w(Zn) = \text{not known, only:}$$

$$\log [\text{Zn}]_{\text{radish root}} = -1,3 + 0,8 \cdot \log[\text{Zn}]_{\text{pore water}} + 0,3 \cdot \text{pH} - 0,0003 \cdot G$$

BCF for radish leaf:

$$\log \text{BCFg}(\text{Cd}) = 4,4 - 0,001 \cdot L - 0,05 \cdot \text{OC} - 0,7 \cdot \text{pH (corrected)}$$

$$\log \text{BCFg}(\text{Cu}) = \text{not significant}$$

$$\log \text{BCFg}(\text{Zn}) = 7,4 - 0,06 \cdot L - 0,03 \cdot \text{OC} - 1,0 \cdot \text{pH}$$

$$\log \text{BCFw}(\text{Cd}) = -1447 + 735 \cdot \text{pH} - 0,4 \cdot G$$

$$\log \text{BCFw}(\text{Cu}) = \text{not significant}$$

$$\log \text{BCFw}(\text{Zn}) = \text{not known, only:}$$

$$\log [\text{Zn}]_{\text{radish leaf}} = 1,2 + 0,7 \cdot \log[\text{Zn}]_{\text{pore water}} + 0,1 \cdot \text{pH} - 0,0002 \cdot G$$

Not all relations are as accurate. Sometimes it was not possible to derive any significant relation for the mentioned metals. Beside the mentioned metals Ba, Ca, Cd, Cu, K, Mg, Na, Ni, Pb, Zn other metals were measured, but results were often too close to the detection boundaries to be able to derive a relation. In some cases only 1 of 2 measurements were significantly different from the detection boundary. This was the case with: As, Cr, Mo, Sb, Sn, Se, V (with small variations for each plant).

## 2.4 Other evaluated relations with soil parameters

Finally relations were measured of:

- (c) Background concentrations in soil from undisturbed sites of the wide spectre of metals (mentioned in section 1 of this Appendix) as a function of clay content, organic carbon content and pH-H<sub>2</sub>O
- (d) Partition between concentration in pore water and in soil as a function of organic carbon, clay content and pH

for radish root, radish leaf and lettuce relations were measured of:

- (e) Concentrations of metals in plant parts as functions of the concentrations in soil and of organic carbon, clay content and pH
- (f) Concentrations of metals in plant parts as functions of the concentrations in pore water and of organic carbon, clay content and pH

## 2.5 A remark on the water content of plants

The dry weight/fresh weight ratio varied in the bioassays in soils from undisturbed sites as:

**Table 3:** The dry weight/fresh weight ratio in the bioassays in soils from undisturbed sites

	<u>min</u>	<u>max</u>	<u>mean</u>	<u>number of measurements</u>
Radish root	0,07 -	0,16	0,116 ± 0,003	20
Radish leaf	0,09 -	0,15	0,102 ± 0,006	20
Lettuce (1)	0,06 -	0,30	0,12 ± 0,02	18
<u>Lettuce (2)</u>	<u>0,06 -</u>	<u>0,45</u>	<u>0,13 ± 0,02</u>	<u>17</u>

standard deviation of the mean

For a healthy lettuce plant the dry weight/fresh weight ratio is about 0,05. The found high values were related to inhibited growth (i.e. in soils from undisturbed sites with low pHs). Lettuce

appeared to be very sensitive and 2 bioassays were necessary. The mean water content of these consumption crops is not 80% as supposed in CSOIL, but  $88 \pm 1\%$ .

### 3. *Experimental results for BCF-factors of plants in contaminated/cleaned soils*

BCF- values (with respect to the concentrations in the soil, not with respect to the concentrations in the pore water) varied as follows for contaminated (Cont.), cleaned (Cleaned), Cleaned and matured (CleaMat.) and soils from undisturbed sites(Und.):

**Table 4:** The BCF-values in contaminated and cleaned soils

	Soil	Radish root		Radish leaf		Lettuce	
Cd	Cont.	0,6 -	2,4	1,7 -	3,1	1,5 -	4,3
	Cleaned	1,7 -	2,6	4,4 -	13	5,5 -	8,4
	CleaMat.	0,31 -	0,55	1,4 -	2,5	- -	-
	Und.	<u>0,39 -</u>	<u>26</u>	<u>1,3 -</u>	<u>36</u>	<u>1,5 -</u>	<u>97</u>
Cu	Cont.	0,6 -	0,59	0,25 -	2,2	0,006 -	1,6
	Cleaned	0,06 -	0,41	0,53 -	3,3	0,12 -	0,4
	CleaMat.	0,18 -	0,69	0,01 -	0,78	- -	-
	Und.	<u>0,12 -</u>	<u>29</u>	<u>0,19 -</u>	<u>9,9</u>	<u>0,2 -</u>	<u>4,9</u>
Pb	Cont.	0,11 -	0,38	0,026 -	0,11	0,015 -	0,12
	Cleaned	0,030 -	0,097	0,026 -	0,96	0,032 -	0,053
	CleaMat.	0,023 -	0,094	0,007 -	0,032	- -	-
	Und.	<u>0,011 -</u>	<u>2,8</u>	<u>0,015 -</u>	<u>0,20</u>	<u>0,040 -</u>	<u>0,52</u>
Zn	Cont.	0,93 -	2,6	2,8 -	4,1	0,36 -	1,9
	Cleaned	0,39 -	1,6	4,1 -	18	0,42 -	1,0
	CleaMat.	0,26 -	0,63	1,1 -	2,6	- -	-
	Und.	<u>0,22 -</u>	<u>35</u>	<u>0,46 -</u>	<u>95</u>	<u>0,2 -</u>	<u>122</u>

The higher BCF-values in the soils from undisturbed sites with their comparably low concentrations of contaminants, make clear that BCF-values depend significantly on concentrations and/or on the soil parameters (the latter conclusion can of course also be drawn from the indicated relations of BCF-values in soils from undisturbed sites).

### 4. *Bioassays as a reference*

To be able to consider a soil as normal/healthy on the basis of bioassays reference values where needed for normal concentrations in the plant parts. A literature search was performed for concentrations that are measured in plants grown in clean substrates. For example for the metals mentioned above was found:

**Table 5:** Concentrations of some metals in crops grown in clean soils/substrates (from a literature search)

	<u>radish root</u>	<u>radish leaf</u>	<u>lettuce</u>
Cd	0,08 - 4,0	0,11 - 4,8	0,17 - 14,5 [mg/kg d.s.]
Cu	1,8 - 50	0,07 - 10	<0,042- 16
Pb	≤0,25 - 37	<0,18 - 4,8	0,22 - 12
<u>Zn</u>	<u>15 - 207</u>	<u>25 - 434</u>	<u>17 - 323</u>

A reference framework was developed for testing the quality of cleaned soil based with bioassays. The boundary values are determined on basis of mortality of 10% of the earthworms, and yield and ratio dry weight/fresh weight ratio for radish. For the metals treated earlier it was found for example:

**Table 6:** Selection of proposed boundary values for valuating the results of the developed bioassays

	<u>earthworms</u>	<u>radish root</u>	<u>radish leaf</u>
Cd	10	2,0	4,0 [mg/kg d.s.]
Cu	20	50	10
Pb	25	37	5,0
<u>Zn</u>	<u>150</u>	<u>210</u>	<u>450</u>

Also it was concluded that in the Microtox-test an inhibition of max. 30 % can be considered as normal.

## 5. *Properties of the various sampled soils*

Soils of several types and qualities were sampled and evaluated in the project:

- undisturbed/natural soils,
- untreated/contaminated soils
- contaminated/cleaned soils and
- contaminated/cleaned/matured soils.

### 5.1 Soil parameters of the sampled soils

The sampled 19 l supposedly clean soils from undisturbed nature reserves varied in properties as follows

**Table 7:** Properties of the sampled soils from undisturbed sites

	<u>min.</u>	<u>max.</u>	
pH-KCl	2,5 -	6,3	
pH-H <sub>2</sub> O	3,6 -	6,8	
org.C.	2,0 -	33,1	[weight%]
clay content	0,7 -	47,0	[weight%]
perc. 2 - 38 µm	0,3 -	38,9	[weight%]
M50	50 -	220	[µm]
<u>CEC</u>	<u>3,4 -</u>	<u>78,0</u>	<u>[meq/100 g]</u>

M50= median grain distribution

The sampled 5 contaminated/untreated and contaminated/cleaned soils varied as follows in properties:

**Table 8:** Properties of the sampled contaminated and cleaned soils

	<u>min.</u>	<u>contaminated</u>		<u>cleaned</u>		
		<u>max.</u>	<u>min.</u>	<u>max.</u>		
pH-KCl		5,4 -	6,1	5,2 -	7,7	
pH-H <sub>2</sub> O	6,2 -	6,6	5,5 -	7,9		
org.C.		2,2 -	4,5	0,33 -	0,82	[weight%]
clay content		0,69-	1,93	0,09 -	0,36	[weight%]
M50		150 -	180	125 -	225	[µm]
CaCO <sub>3</sub>		<u>0,05 -</u>	<u>0,11</u>	<u>0,04 -</u>	<u>0,28</u>	[%]
As		3,6 -	12,1	2,1 -	5,3	[mg/kg d.s.]
Cd		4,5 -	6,5	0,7 -	1,4	
Cu		8 -	50	8-	24	
Pb		17 -	170	27(!) -	64	
Zn		<u>48 -</u>	<u>1005</u>	<u>64(!) -</u>	<u>300</u>	

In all cases the treatments consisted of short experimental pilot cleaning procedures, aiming in the first place at low Cd-concentrations. Not all these pilot cleaning procedures were successful (and other experiments not sampled by us have followed).

In soils from undisturbed sites these concentrations were :

## 5.2 Concentrations and leaching behaviour in soils from undisturbed sites

**Table 9:** Concentrations of (a selection of) metals in the soils from undisturbed sites

	<u>min.</u>	<u>topsoil</u>		<u>subsoil</u>		reference value standard soil
		<u>max.</u>	<u>min.</u>	<u>max</u>		
As		0,9 -	41	<0,6 -	38	29 [mg/kg d.s.]
Cd		<0,02 -	1,27	<0.02 -	0,09	0,8
Cu		0,4 -	27	<0,4 -	34	36
Pb		<1 -	95	1,1 -	21,0	85
Zn		<u>4,4 -</u>	<u>127</u>	<u>1,6 -</u>	<u>97</u>	<u>140</u>

The variations in soils from undisturbed sites can also be substantial. In the subsoil all concentrations were generally lower. In soils from undisturbed sites also the concentrations and the leaching emissions of Ba, Ca, Co, Cr, Hg, K, Mg, Mo, Na, Ni, Sb, Se, Sn, V were measured. (The metals can be immobile, for example as included in the dust of precious stones that is a natural part of the soil.)

Several reference leaching. test were proposed for the Regulation on Building Materials and developed further and tried out for soils in this project. The column leaching test mimicks natural flowthrough of rainwater in the soil and gives an impression of the (initial) leaching rate. The availability test is performed in a more rigorous way and gives an impression of the total mobile fraction of the metals present in the soil.

Examples of leaching emissions resulting from the used reference leaching tests are for the mentioned earlier metals:

**Table 10:** Leaching emissions of some metals from the soils from undisturbed sites as measured by the standard column leaching test as a measure of the initial leaching rate (upper boundary cumulative emission at a liquid/solid ratio of 10 l/kg in a flow through column)

	<u>min.</u>	<u>topsoil</u>		<u>subsoil</u>		
		<u>max.</u>	<u>min.</u>	<u>max</u>		
As		0,03	- 1,16	0,01	- 0,03	[mg/kg d.s.]
Cd		0,002	- 0,034	0,001	- 0,018	
Cu		0,07	- 0,39	0,01	- 0,06	
Pb		0,03	- 1,74	0,01	- 0,06	
<u>Zn</u>		<u>0,14</u>	- <u>4,74</u>	<u>0,02</u>	- <u>0,56</u>	

**Table 11:** Mobile fraction of some metals in the soils from undisturbed sites as measured by the reference leaching availability test (at L/S = 200 l/kg)

	<u>min.</u>	<u>topsoil</u>		<u>subsoil</u>		
		<u>max.</u>	<u>min.</u>	<u>max</u>		
As		0,6	- 0,9	0,6	- 0,6	[mg/kg d.s.]
Cu		0,2	- 0,8	0,2	- 0,3	
Pb		0,2	- 2,1	0,2	- 0,7	
<u>Zn</u>		<u>0,6</u>	- <u>29,3</u>	<u>0,6</u>	- <u>10,1</u>	