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Human health risks due to consumption of vegetables from contaminated sites
Towards a protocol for site-specific assessment

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Rapport in het kort

Gezondheidsrisico's bij consumptie van groenten gekweekt op verontreinigde bodems. Naar een protocol voor locatie-specifieke beoordeling.

Om te kunnen bepalen of de consumptie van groenten die zijn gekweekt op verontreinigde bodems gezondheidsrisico's met zich meebrengt, heeft het RIVM een procedure ontwikkeld. De procedure is stapsgewijze opgezet om een degelijke onderbouwing van de gegevens en een efficiënte werkwijze te waarborgen. Het principe is: simpel indien mogelijk, complex indien noodzakelijk. Als in een eerdere stap een risico kan worden uitgesloten, kan de procedure worden gestaakt. Zo niet, dan is een vervolgstap nodig. Daarin wordt de beoordeling steeds specifieker op de omstandigheden van de locatie gericht. Op die manier leidt de procedure tot een realistischere uitkomst, maar hij wordt ook tijdrovender.

De procedure, die uit vier stappen bestaat, is als volgt. In stap 0, die voorafgaat aan de reken- en meetstappen, wordt de *mogelijkheid* ingeschat of de consumptie van groenten op een bepaalde locatie schadelijk kan zijn voor de gezondheid. In Stap 1 worden vervolgens de bodemgehalten van schadelijke stoffen gemeten en vergeleken met zogeheten *Kritische bodemgehalten* (de waarden waarbinnen groenten telen veilig is). Stap 2 omvat een *gedetailleerde bepaling van het locatie-specifieke gezondheidsrisico, op basis van berekening.* Ten slotte is in Stap 3 *een gestandaardiseerd meetprotocol* beschreven. Dat is een richtlijn om op een locatie te bepalen welk typen groenten, en de hoeveelheid daarvan, het beste kunnen worden bemonsterd als indicatie voor de gezondheidsrisico's.

Trefwoorden: plantopname, CSOIL, cadmium, gezondheidsrisico's, bodemverontreiniging

Abstract

Human health risks due to consumption of vegetables from contaminated sites. Towards a protocol for site-specific assessment.

RIVM has developed an approach which allows human health risks of vegetable consumption from contaminated sites to be assessed. A tiered approach was used to guarantee the scientific basis and efficient use in practice. The underlying principle is: simple when possible and complex when necessary. If the risk can be eliminated in an early step, the assessment can be stopped. If not, assessment continues in the next tier, becoming more site-specific with each tier. This results in a more realistic, but also more time-consuming, assessment. The approach consists of four tiers that are laid out as follows. Tier 0, which precedes the calculation and measurement tiers, investigates the *possibilities* for experiencing adverse human health effects due to vegetable consumption. Subsequently, the soil concentrations of pollutants are measured in Tier 1 and compared with so-called *Critical soil concentrations* (for which vegetable consumption from contaminated sites is safe). Tier 2 offers the possibility for a *detailed assessment of the site-specific risks for human health on the basis of calculation*. Finally, Tier 3 shows *a standardized measurement protocol*. This protocol offers guidance and advice on the type and amount of crops that can be sampled in the field, thereby providing an indication for human health risks.

Key words: plant uptake, CSOIL, cadmium, human health risks, soil contamination

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Samenvatting

Met het doel om zowel op degelijke onderbouwde als op efficiënte wijze de risico's te kunnen bepalen van het consumeren van groenten gekweekt op verontreinigde bodems is een stapsgewijze procedure ontwikkeld. In de lagere stappen wordt een meer conservatieve benadering gevolgd. In elke hogere stap wordt de beoordeling meer locatie-specifiek, realistischer en derhalve complexer. Als gevolg neemt in elke stap de complexiteit, en dientengevolge de benodigde inspanning en kosten toe. Als in een specifieke stap een onacceptabel risico voor de menselijke gezondheid niet kan worden weerlegd dient de bepaling in de volgende stap te worden uitgevoerd. Het onderliggende principe is: "simpel indien mogelijk en complex indien noodzakelijk".
De stapsgewijze procedure is als volgt gestructureerd: Stap 0 betreft een voorafgaande kwalitatieve evaluatie van de *mogelijkheden* of het consumeren van groenten tot negatieve effecten op de gezondheid kan leiden. In Stap 1 worden de gemeten totale bodemgehalten (gemiddelden of relatief hoge waarden) vergeleken met *Kritische bodemgehalten* (alleen voor cadmium). Deze Kritische bodemgehalten zijn afgeleid op basis van een conservatief blootstellingsscenario. Stap 2 biedt de mogelijkheid voor een *gedetailleerde bepaling van het locatie-specifieke risico op basis van berekening.* Uiteraard verschilt de locatie-specifieke berekening van het contaminant-gehalte in groenten voor metalen, overige anorganische contaminanten en organische contaminanten. Voor metalen zijn Freundlich-achtige plant - bodem relaties (afhankelijk van het totaalgehalte en de belangrijkste bodemeigenschappen) en geometrisch gemiddelden van de BioConcentratieFactoren (gecorrigeerd voor organisch stof- en kleigehalte) gecombineerd. De accumulatie van overige anorganische contaminanten is gebaseerd op passieve opname. De berekening van de concentratie van organische contaminanten in groenten is gebaseerd op een aangepast Trapp en Matthies model. In dit model wordt de partitie van contaminanten tussen het poriewater en de plantenwortels en vervolgens de verplaatsing naar de bovengrondse plantendelen berekend, resulterend in het contaminant-gehalte van de bovengrondse plantendelen. Tenslotte is in Stap 3 *een gestandaardiseerd meetprotocol* beschreven. Met dit protocol kan in het veld een significant aantal representatieve groenten worden bemonsterd, waarvan de eetbare gedeelten worden voorbehandeld in het laboratorium in analogie met de gangbare keukenpraktijk. De gemeten concentratie in groenten kan vervolgens worden ingevoerd in een blootstellingsberekening en indien van toepassing worden getoetst aan toelaatbare concentraties in groenten.
Als belangrijkste onderzoeksaanbevelingen voor de toekomst gelden het uitbreiden van de RIVM plant – bodem dataset voor metalen en het uitvoeren van een uitgebreide validatiestudie.

Nut voor het beleid
De procedure voor de bepaling van de locatie-specifieke humane risico's ten gevolge van consumptie van op verontreinigde bodems geteelde groenten kan dienen als ondersteuning bij

het bodembeheer in geval van de bodemgebruiksvormen “Moestuin/Volkstuin” en “Wonen met tuin”. Bovendien dient het protocol te worden geïntegreerd in de algemene procedure van (stapsgewijze) bepaling van de locatie-specifieke humane risico’s ten gevolge van blootstelling aan verontreinigde bodems. Deze algemene procedure zal onderdeel uit gaan maken van de inhoudelijke basis van de Wet bodembescherming, welke momenteel wordt herzien. Nieuwe toepassingen betreffen het Saneringscriterium en Locale Ambities voor bodemkwaliteit.

Summary

To be able to assess the human health risks of vegetable consumption from contaminated sites in a scientifically-based and efficient way a tiered approach has been developed. Successively, in each tier the degree of conservatism decreases, while site-specificism increases. As a consequence, complexity and hence effort and finances needed also increase in each tier. When in a specific tier an unacceptable human health risk can not be rejected the assessment in the following tier has to be performed. The underlying principle is: simple when possible and complex when necessary.
The tiered approach is laid out as follows: Tier 0 concerns a preliminary qualitative evaluation of the *possibilities* for experiencing adverse human health effects due to vegetable consumption. In Tier 1 the actual total soil concentrations (average or relatively high values) are compared with *Critical soil concentrations* (for cadmium only). These Critical soil concentrations have been derived on the basis of a conservative exposure scenario. Tier 2 offers the possibility for *a detailed assessment of the site-specific risks on the basis of calculation*. Obviously, the site-specific calculation of the contaminant concentration in vegetables differs for metals, other inorganic contaminants and organic contaminants. For metals Freundlich-type plant - soil relations (dependent of the total soil concentration and the major soil properties) and geometric means of the BioConcentrationFactors (corrected for organic matter and clay contents) are combined. The accumulation of other inorganic contaminants is based on passive uptake. The calculation of the concentration of organic contaminants in vegetables is based on an adapted Trapp and Matthies model. In this model the partitioning of contaminants between pore water and roots and subsequently translocation to the upper plant parts is calculated, resulting in the contaminant concentration in the above-ground plant parts. Finally, in Tier 3, *a standardized measurement protocol* has been developed. This protocol allows for sampling of a significant number of representative vegetables in the field, for which the edible parts of the plants are treated in the laboratory in analogy with standard kitchen preparation. Subsequently, the measured concentration can be used in an exposure calculation and, when appropriate, compared to acceptable concentrations in vegetables.
The most important recommendations for future research are extension of the database for metals and the performance of a comprehensive validation study.

Contribution to Dutch soil policy
The procedure to assess the site-specific human health risks assessment for consumption of vegetables from contaminated sites can be used to support planning or soil management in relation to the soil uses “Vegetable garden” and “Residential sites with garden”. Moreover, the protocol should be incorporated in the general procedure on the (tiered) procedure to assess the site-specific human health risks due to exposure to contaminated sites. This general procedure will be included in the technical basis of the Dutch Soil Protection Act, which act

is presently under revision. New applications concern the Remediation criterion and the Local Ambitions for soil quality.

1. Introduction

1.1 Objective

This study is focused on the site-specific human health risk assessment related to consumption of vegetables from contaminated sites. The following *objective* is recognized:

> *Development of the basis for a protocol which allows site-specific assessment of the human health risks for consumption of vegetables from contaminated sites.*

The protocol should be applicable for the following situations:

- vegetable gardens;
- residential sites with gardens that offer the possibility of home-grown vegetables;
- undeveloped or fallow sites that will be taken into development and future land-use enables the possibility for vegetable cultivation.

Besides *the model algorithms for the exposure pathway "exposure due to vegetable consumption" in the CSOIL exposure model must be improved*. These model algorithms are part of the protocol.

1.2 Requirements

The protocol should be applicable for site-specific risk assessment[1] and focuses on metals, other inorganic contaminants and organic contaminants. In the framework of the revision of the Dutch Soil Protection Act (Ministry of VROM, 2003) it must be turned into a user-friendly protocol. Possibly it will be incorporated into a protocol to assess the site-specific risks for human health from a wider perspective. This protocol will replace the present standardized procedure to assess the site-specific risks for human health as included in the procedure to determine the urgency of remediation (Koolenbrander, 1995).

A primary requirement is a sound scientific basis of the protocol and the model algorithms. However, the protocol should also enable a practical, efficient and uniform application.

A crucial factor in relation to the risks of vegetable consumption is the "representative concentration" in vegetables that grow, or could grow, on a contaminated site. It is generally accepted that the assessment of this representative concentration on the basis of a calculation or on the basis of a measurement involves a limited reliability. Therefore the protocol offers the possibilities in higher tiers for calculating as well as measuring this concentration.

[1] For "site-specific risk assessment" often the term "actual risk assessment" is used.

1.3 Tiered approach

With the purpose to optimise the balance between quality and practicability the protocol is based on a tiered approach. This tiered approach includes the possibility for fast screening in case of a clear decision and only compels an intensive effort when necessary ("easy when possible, more complex when necessary"). The following assessment steps will be investigated for inclusion in different tiers:

- a preliminary qualitative evaluation of the *possibilities* for experiencing adverse human health effects due to vegetable consumption;
- comparison of measured soil concentrations (average or relatively high values) with *Critical soil concentrations;*
- a detailed assessment of the actual risk on the basis of a site-specific risk calculation;
- a standardized measurement protocol for sampling and testing of a significant number of representative vegetables in the field.

2. Human health risks due to consumption of home-grown vegetables

The risks for human health depend on exposure and the effects in the human body that follow exposure.

2.1 Human exposure

2.1.1 Site-specific exposure

The exposure to humans largely depends on two factors: the amount of vegetables consumed and the concentration of contaminants in these vegetables. Chapter 4 focuses on the amount of vegetables consumed. The determination of a representative concentration of contaminants in the vegetables is extensively described in chapters 5 (metals), 6 (other inorganic contaminants) and 7 (organic contaminants).

The calculation of exposure is rather straightforward:

$$Exposure_{vegetables} = \frac{\sum Q_{vegetable\,i} \times C_{vegetable\,i} \times f_{home-grown} \times f_{bioavailability}}{W} \qquad \text{Eq. 2.1}$$

in which

$Exposure_{vegetables}$	Exposure due to vegetable consumption	[$mg/kg_{body\ weight}/d$]
$Q_{vegetable\ i}$	Consumption rate of vegetable i	[kg_{dw}/d]
$C_{vegetable\ i}$	Contaminant concentration in vegetable i	[mg/kg_{dw}]
$f_{home-grown}$	Fraction of vegetables that is home-grown	[-]
$f_{bioavailability}$	Correction for relative bioavailability in the human body	[-]
W	Body weight	[$kg_{body\ weight}$]

In almost all European exposure models this pathway is included (*Carlon, in prep.*), like for example in the Dutch CSOIL exposure model (Van den Berg, 1991/ 1994/1995; Otte et al., 2001). This model is also used to derive the human toxicological-based part of the Dutch Intervention Values.

The specification of the fraction of vegetables that is home-grown, $f_{home-grown}$, partly concerns a political decision. Usually, this fraction is not specified for each vegetable separately. In the Netherlands the following default data are used for this fraction, depending on site use:

- "Residential site, with garden": 10%
- "Vegetable garden": 50% for potatoes and 100% for other vegetables.

The differentiation of the fraction of vegetables that is home-grown between potatoes and other vegetables is made, because it is not realistic to use a value of 100% for potatoes. Because consumption rate for potatoes is rather high, an exceptionally large vegetable garden would be needed.

No decent statistics on the amount of the fraction of vegetables that is home-grown in the Netherlands exist. Besides, this fraction will differ between cities and rural areas. The values given above mainly concern a political decision: “the soil quality must offer the possibility to consume at least a specified percentage of vegetables from the own garden”. Since it is a sensitive parameter, it is recommended to further investigate the fraction of vegetables that is home-grown in the Netherlands, in the future.

At this moment no information on the differences between intake and uptake of contaminants via a vegetable matrix exists. Therefore, the correction for bioavailability in the human body, $f_{bioavailability}$, in this protocol is like in any other existing exposure model, 1.0.

To assess the risk due to vegetable consumption it is essential to include exposure due to soil ingestion in most cases, because hand-mouth contact is relatively intensive during gardening.

2.1.2 Reference dose

With the purpose to judge the risk due to exposure, the actual exposure must be compared to a Reference dose (or acceptable or critical exposure; in the Dutch Soil Protection Act: Maximum Permissible Risk for intake; MPR_{human}). To derive values for this Reference dose a distinction has been made between non-threshold contaminants (*genotoxic carcinogens*) and threshold contaminants (*non-carcinogens and non-genotoxic carcinogens*) (Baars et al., 2001). For the threshold contaminants, a Reference dose can be derived for which no adverse effects for humans are likely to occur in cases where this exposure is not exceeded. For the non-threshold contaminants even the lowest exposure rate results in an increased chance of adverse effects for humans.
For non-genotoxic carcinogens and non-carcinogenic contaminants (threshold contaminants), the toxicological Tolerable Daily Intake (TDI) is taken as the Reference dose (Maximum Permissible Risk for intake; MPR_{human}). A TDI is the threshold exposure of a contaminant to which humans can be orally exposed daily on the basis of body weight without experiencing adverse effects on health. For genotoxic carcinogens (non-threshold contaminants), the Reference dose (MPR_{human}) is defined as the dose of a contaminant (based on body weight for oral intake or air volume for inhalative intake) which forms a risk of one additional case of lethal tumour in 10,000 lifelong exposed individuals. This definition is based on a political decision (Ministry of VROM, 1988).

An overview of the most recent values of the Reference dose in the Netherlands is found in Baars et al. (2001).

2.2 Critical soil concentration

A Critical soil concentration can be derived on the basis of exposure and Reference dose. The definition of a Critical soil concentration is: the soil quality resulting in an exposure equal to the Reference dose (see Figure 2.1). In this figure the lifelong-averaged exposure is given for metals and organic contaminants, according to the so-called standardized exposure scenario. Note that only exposure of similar type (i.e. oral or inhalative) should be combined. Soil quality standards, for example the Serious Risk Concentration as one of the two pillars of the Dutch Intervention Values (Swartjes, 1999) are examples of Critical soil concentrations.

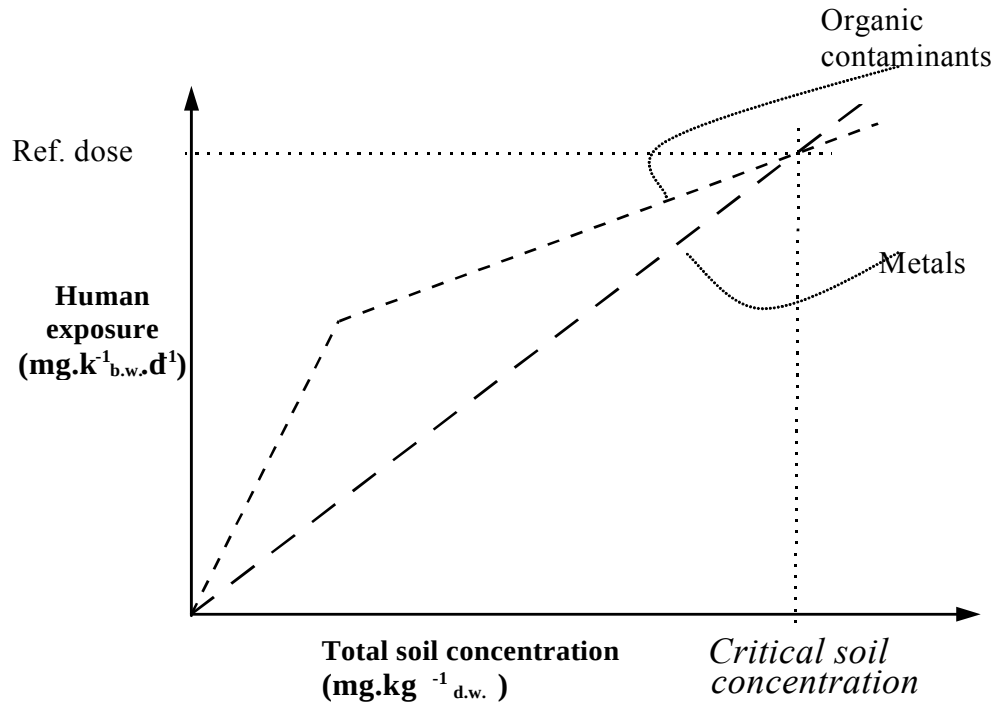

Fig. 2.1: Derivation of a Critical soil concentration.

An exposure scenario must be defined to describe the conditions that are suitable for a specific type of Critical soil concentration. Because in this study Critical soil concentrations are used in an early stage of the tiered approach a rather conservative exposure scenario is appropriate. This conservative exposure is based on:

- higher consumption rates for kitchen gardeners than for the general population (see section 4.2.2);
- a high contribution of vegetable consumption from the contaminated site: 50% of the potatoes and 100% of the other vegetables;

- a BioConcentration Factor (BCF) for conservative soil properties (relatively high plant uptake).

However, in the Netherlands background exposure is not incorporated in most applications (except for the Remediation criterion, i.e. relating to good soil quality after remediation). This is a political decision. Also for the assessment of risks due to vegetable consumption, as proposed in this study, calculated exposure is compared to the Reference dose, not considering background exposure. From a medical viewpoint this is a non-conservative approach.

Since calculation of the concentration in vegetables is relatively unreliable, certainly at low soil concentrations, Critical soil concentrations could also be derived from measured data. When, for example, the concentration in vegetables is plotted against the soil concentration, a Critical soil concentration could graphically be derived from a Critical vegetable concentration.

2.3 Evaluation of contaminant concentrations in vegetables

2.3.1 Toxicological assessment

The human health risks of measured concentrations of contaminants in vegetables could be evaluated in analogy with the derivation of Critical soil concentrations (section 2.2). As a first step a relevant exposure scenario must be defined. As starting point either a standard "Residential site with garden" or "Vegetable garden" scenario can be selected from the CSOIL exposure model. If appropriate, these standard scenarios could be adapted to local circumstances. In the next step the calculated or measured concentration in vegetables must be entered in CSOIL. Finally, the resulting exposure is evaluated against the Maximum Permissable Risk for human exposure (MPR_{human}). To this purpose it is tested whether the sum of the oral (inclusive dermal) and inhalative risk indexes is less than or equal to 1:

$$(\Sigma \text{ oral exposure}/ MPR_{human}oral)+(\Sigma \text{ inhalative exposure}/ MPR_{human}inhalative) \leq 1? \quad \text{Eq. 2.2}$$

In which:

$MPR_{human}oral$	Maximum Permissible Risk for oral intake	[$mg/kg_{body\ weight}$/day]
$MPR_{human}inhalative$	Maximum Permissible Risk for inhalative intake	[$mg/kg_{body\ weight}$/day]

In that case there are no unacceptable risks. When the sum of the oral (including dermal) and inhalatory risk indexes exceeds 1 a next step in the tiered approach must be followed or, when in the final tier, there is an "unacceptable risk" for human health.

Usually exposure to one specific contaminant is considered (no combined exposure). This is a limitation of the protocol described in this report.

2.3.2 Other quality criteria

In case of commercial vegetable production, the contaminant concentrations in crops must be additionally compared to appropriate food quality criteria, according to the Commission Regulation (EC) No 466/2001 (EC, 2004). In Table 2.1 the appropriate food quality criteria are given for cadmium and lead, two important metals in regard to human health effects due to vegetable consumption. Unfortunately, food quality criteria for arsenic, also an important metal in regard to human health effects due to vegetable consumption, are missing in this regulation.

Vegetable group	**Maximal concentration (mg/kg_{fw})**
Cadmium:	
Leafy vegetables, fresh herbs, celeriac, cultivated fungi	0.2
Stem vegetables, root vegetables (excl. Celeriac), (peeled) potatoes	0.1
Other vegetables	0.05
Lead:	
Brassica, leafy vegetabes, cultivated fungi	0.3
Fruit (excl. Berries and small fruits)	0.1
Berries and small fruits	0.2
Other vegetables	0.1

Table 2.1: Food quality criteria for cadmium and lead, according to the Commission Regulation (EC) No 466/2001

2.4 Critical vegetable concentration (cadmium)

When consuming vegetables with an average concentration below a Critical vegetable concentration, human health effects are unlikely. This Critical vegetable concentration can be calculated using the procedure described in section 2.2, with exposure equals the Reference dose (Maximum Permissible Risk for intake, MPR_{human}). Under the assumption that exposure through vegetable consumption is the dominant exposure pathway, which is the case for e.g. cadmium, it follows from Eq. 2.1:

$$C_{vegetable}CR = \frac{MPR_{human} \times W}{\sum Q_{vegetable\ i} \times f_{home-grown} \times f_{bioavailability}} \quad \text{Eq. 2.3}$$

in which

$C_{vegetable}CR$	Critical vegetable concentration	[mg/kg_{dw}]
MPR_{human}	Maximum Permissible Risk for intake	[$mg/kg_{body\ weight}$/day]
$Q_{vegetable\ i}$	Consumption rate of vegetable i	[kg_{dw}/d]
$f_{home-grown}$	Fraction of vegetables that is home-grown	[-]
$f_{bioavailability}$	Correction for relative bioavailability in the human body	[-]
W	Body weight	[$kg_{body\ weight}$]

3. Theory on accumulation of contaminants in plants

The “representative concentration” in plants is the combined result from uptake, transport, accumulation and possibly degradation of contaminants. In this paragraph an overview is given of the characteristics of the major processes.

3.1 Representative concentration in plants

A “representative concentration” refers to the amount of a contaminant (metals, other inorganic contaminants or organic contaminants) that humans are exposed to when consuming these vegetables. This implies that attention is focused on the concentration in or on the edible parts of the vegetables at the moment that these vegetables are harvested and after these vegetables have been prepared (washing, peeling) in a “conventional” way.

3.2 Characteristics of uptake

Figure 3.1 shows the pathways for uptake of contaminants into vegetables.

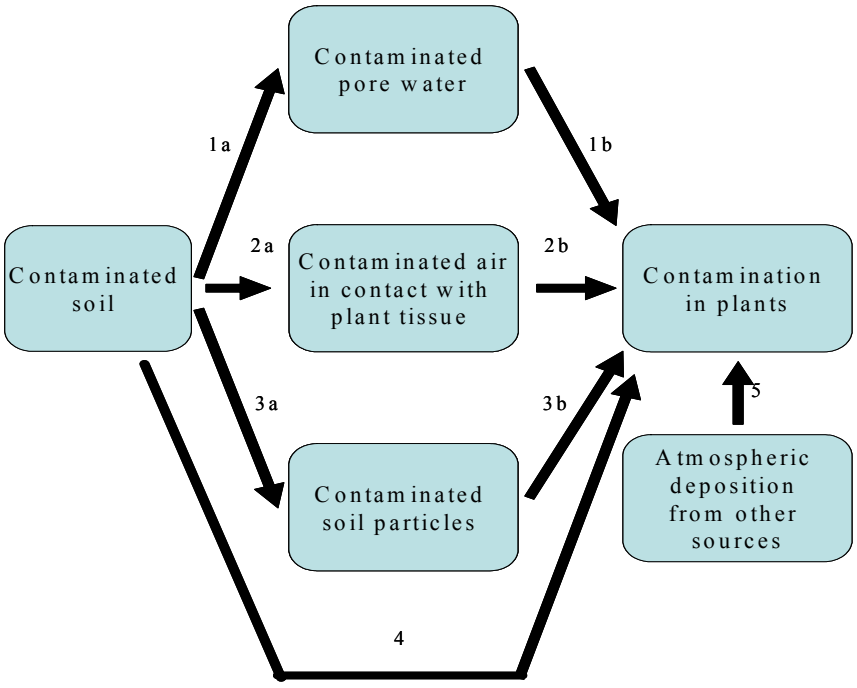

Figure 3.1: Pathways for contaminant uptake in vegetables.
Root uptake via pore water (1a, 1b); foliar uptake via air (2a, 2b); uptake via rain splash (3a, 3b); direct uptake (4); atmospheric deposition from other sources (5).

Uptake through the roots via the pore water (route 1a and 1b) is the dominant pathways. Other pathways are less important (uptake through the leaves, via the air (organic contaminants only, route 2a and 2b; uptake via rain splash: route 3; direct uptake: route 4) or not related to contaminated soil (atmospheric deposition from other sources: route 5).

Roots are primary organs for the contact with ions and uptake from soil. Root uptake can vary with the soil properties, water content of the soil, as well as plant characteristics such as type of root system and lipid content (Paterson and Mackay, 1994). The soil zone surrounding the roots is called the rhizosphere. In the rhizosphere there is a large influence of the plant roots on the soil properties. Plant roots excrete CO_2 and organic substances which can decrease the pH and increase microbial activity.
Contaminants in the pore water may reach the root surface by mass flow, penetrate the root, enter the xylem and be transported in the transpiration stream. Once in the transpiration stream, chemicals may be metabolized (detoxified) by plant enzymes, or escape by gaseous diffusion through stomata in leaves. Climatic conditions as temperature and rainfall determine plant transpiration rates, which in turn control the rates of water movement to the root surface and in the xylem.

Organic contaminants also enter vegetation from the atmosphere by gas-phase and particle - phase deposition onto the *waxy cuticula* or through the stomata of the leaves and are translocated by the phloem. However, from the perspective of soil quality assessment only the contaminants that originate from soil are relevant.

Soil and dust particles can be deposited on the plant by wind or rain (also called rain splash or soil resuspension). Even after washing, a significant fraction of the attached contaminant particles may stick to the leafs, e.g. circa 50% for lead (Meeuwissen, 1989). The contribution to the representative plant concentration is difficult to estimate and depends on many factors (e.g. the geometry of the plant, soil type).

Other factors affecting the differences in uptake of metals, other inorganic contaminants and organic contaminants concern the heterogeneity of contaminants in the soils (e.g. Miller et al. (2004) for cadmium in lettuce) and the heterogeneity of nutrient supply (e.g. Haines (2002) for zinc in *Thlaspi caerulescens*). Also dilution by growth will influence the concentration in the plant.

3.2.1 Metals

Each metal has a different affinity for plant uptake. In general, cadmium or zinc uptake, for example, is higher than mercury or lead uptake. Partitioning processes determine the uptake of metals in the root zone of plants. It is widely recognised that the total soil concentration is an inappropriate parameter for crop uptake (e.g. McLaughlin et al., 2000). The soil properties, which determine the fraction available in the pore water are very important for plant uptake. Most of the metals are more available at lower soil pH. However, some

metalloids (like arsenic) form oxyanions in solution, which show a stronger sorption at lower pH (McLaughlin et al., 1998). Other soil properties that control the availability for metals in the pore water and hence uptake by plants, are the organic matter content, clay content and the presence of (hydr)oxides of manganese, iron and aluminium. Also the availability of soil nutrients plays a role in regard to plant uptake.
The fungi and bacteria in the rhizosphere can also have a strong impact on metal uptake (Kleikamp and Joergensen, 2006). This uptake is also influenced by the lowering of the pH by the excretion of CO_2 and organic acids. Smolders and McLaughlin (1996) investigated the cadmium uptake in Swiss chord [*Beta vulgaris ssp. cicla (L.) Koch, cv. Fordhook Giant*) in the presence of chloro-complexes of cadmium, while the activity of cadmium [2+] in solution was buffered during plant growth. They showed that when solution chloride concentration increased, cadmium concentrations in plant shoots and roots also increased. Another factor influencing sorption and, hence, plant uptake are the redox conditions in the soil affecting the form and reactivity of some soil oxides (e.g. McLaughlin et al., 1998).
There are several active detoxification mechanisms for the plant to defend against excess heavy metal loads in the soil, for example metallothionines synthesis, or enzymatic synthesis of phytochelatines.

3.2.2 Other inorganic contaminants

The group of other inorganic contaminants is a rather heterogeneous group. Not much attention has been focused on the accumulation of other inorganic contaminants in plants, except for compounds that are typically used in agricultural practise like nitrogen and phosphate. In respect to local soil contamination, however, the most relevant representatives of this group concern cyanides and to a lesser extent chloride, bromide and fluorine compounds[2].
Although the group is relatively heterogeneous, a general characteristic of many representatives is that they dissolve easily in pore water and, hence, are well available to for root uptake by plants.

3.2.3 Organic contaminants

Uptake from soil is a passive process for most organic contaminants. Uptake in roots is governed by physical sorption. Water-soluble contaminants that pass the membranes in the roots are transported to the leaf in the transpiration stream in the xylem of the plants.
The foliar uptake via the gas phase is often neglected. In general, air concentrations are too low to result in a significant uptake, because of dilution by the wind. Nevertheless, for closed vegetation close to the ground (e.g. grasses, lettuce) and high soil concentrations, foliar uptake can play a (minor) role. For PAHs the internal concentration measurements give an indication that the concentrations in the foliage of terrestrial plants are mainly the

[2] At present no Intervention Values have been formulated for these compounds (Min. of VROM, 2000). The main reason for this is that these compounds usually have a short residence time in soil.

consequence of uptake of atmospheric PAHs. However, only a limited part of these PAHs originate from soil contamination.
Elimination of an organic contaminant may take place in the leaf tissue by metabolism or photolysis. In plant metabolism organic chemical contaminants are metabolically transformed. Plants have evolved contaminant-specific detoxification (metabolic) pathways and can enzymatically oxidise, reduce, or hydrolyse organic contaminants. The products of these transformation reactions are attached to water-soluble parts, that are removed from the cytoplasm, either by transport into vacuoles or by conversion into insoluble complexes with the cell wall (a process called “lignification”) (Sandermann, 1992). Certain reports in the literature suggest that mineralization of organic contaminants within plant tissues may be possible.
Physicochemical properties that may influence plant uptake of organic contaminants include water solubility, vapour pressure, Henry’s Law constant and hydrophobicity.

3.3 Differences in accumulation between contaminants and plants

3.3.1 Metals

There are plants which can accumulate large amounts of heavy metals in above-ground plants and roots and those which are not effective in the uptake of heavy metals. The former type of plants can sometimes be used for phytoremediation. Plants like *Brassicaceae* (cabbage), *Poaceae* (grasses), *Papilonaceae* (pods) are known as good phytoremediating families (Gawronski, 2000). Some plants are extremely tolerant for soil metals. These plants grow in areas where natural ores of heavy metals occur in the upper layers of the soil. A well-known example of a heavy metal adapted plant is the zinc violet.

A BCF (BioConcentrationFactor) is traditionally used to describe the ratio between the concentration of (the edible part of) the plant and the soil concentration (for details see section 5.1). These BCF values, however, are not a constant value for a specific metal in a specific vegetable, but vary with soil properties and type of plants, among others. Therefore, generic BCFs can only be considered as an indicator for the affinity for plant uptake of a specific metal. Details on the limitations of a generic BCF are also given in section 5.1.

In Figure 3.2 the geometric mean of the BCFs for arsenic, cadmium, cupper, lead, mercury, nickel and zinc are given from the RIVM plant –soil database, for several relevant vegetables. From this figure it can be concluded that the affinity for accumulation of different metals differs substantially. Generally the plants exclude metals like arsenic and nickel giving low BCF values. Cadmium however shows a relatively high accumulation for all vegetables.
For the derivation of the human health based risk limits of the revised Intervention Values, exposure due to vegetable consumption is important for all metals (Lijzen et al., 2001). The contribution of exposure due to vegetable consumption to total exposure is very high for

cadmium (93%) and cobalt (96%) and varies between 80% and 90% of total exposure for copper, mercury, molybdenum and zinc. For the other metals this contribution is also substantial: for nickel 55%, for barium 42%, for lead 31% and for arsenic 28%. For most metals, however, ecological effects are far more critical than effects on human health. An exception is lead (ecological and human health risk limits are of the same order of magnitude) and cadmium (ecological risk limit is two times more stringent than human health risk limit), and to a lesser extent arsenic, mercury and molybdenum (ecological risk limits are circa eight times more stringent). For the other metals the ecological risk limits are in between one and two orders of magnitude more stringent than the human health risk limits. When the frequency of occurrence of the different metals is also taken into account, the exposure pathway vegetable consumption, and hence, the determination of a representative vegetable concentration, is mainly important for cadmium and lead and, to a lesser extent, arsenic, mercury, nickel, barium and molybdenum.

Moreover, Figure 3.2 shows that accumulation of metals can vary with one or two orders of magnitude between different types of vegetables. The BCFs are the highest for fast growing leafy vegetables, as spinach and endive. The high BCF values for tomatoes are surprising. Maybe these BCFs have been determined by low metal concentrations and soil properties that stimulate plant uptake.

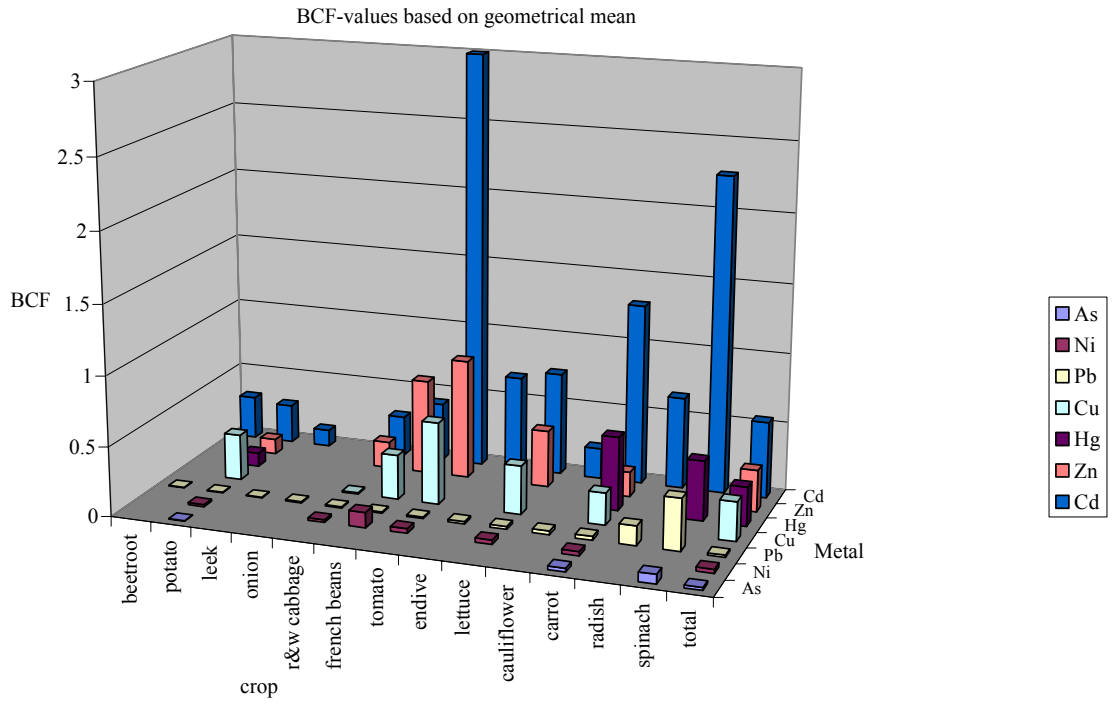

Figure 3.2: Geometrical mean of BCFs (BioConcentrationFactors) from the RIVM plant – soil database, for seven metals and for several relevant vegetables

3.3.2 Organic contaminants

Plant species vary in root lipid content, uptake mechanism (e.g. diffusion or advection) and anatomy, so uptake of organic contaminants is also vegetable-dependent. Moreover, different varieties within certain plant species can also account for large differences. Contaminant concentrations in plants should be normalized to the lipid content of the plants or its leave surface area, especially when directly comparing different species and tissues (Simonich and Hites, 1995). As a consequence the variation between the BCFs of different plants can be large. In zucchini and pumpkin, for example, two orders of magnitude higher concentrations of PCDD/PCDF are found than in other fruits and vegetables (cucumber). As an example the variation between the BCFs for PAHs for different relevant vegetables is illustrated in Figure 3.3.

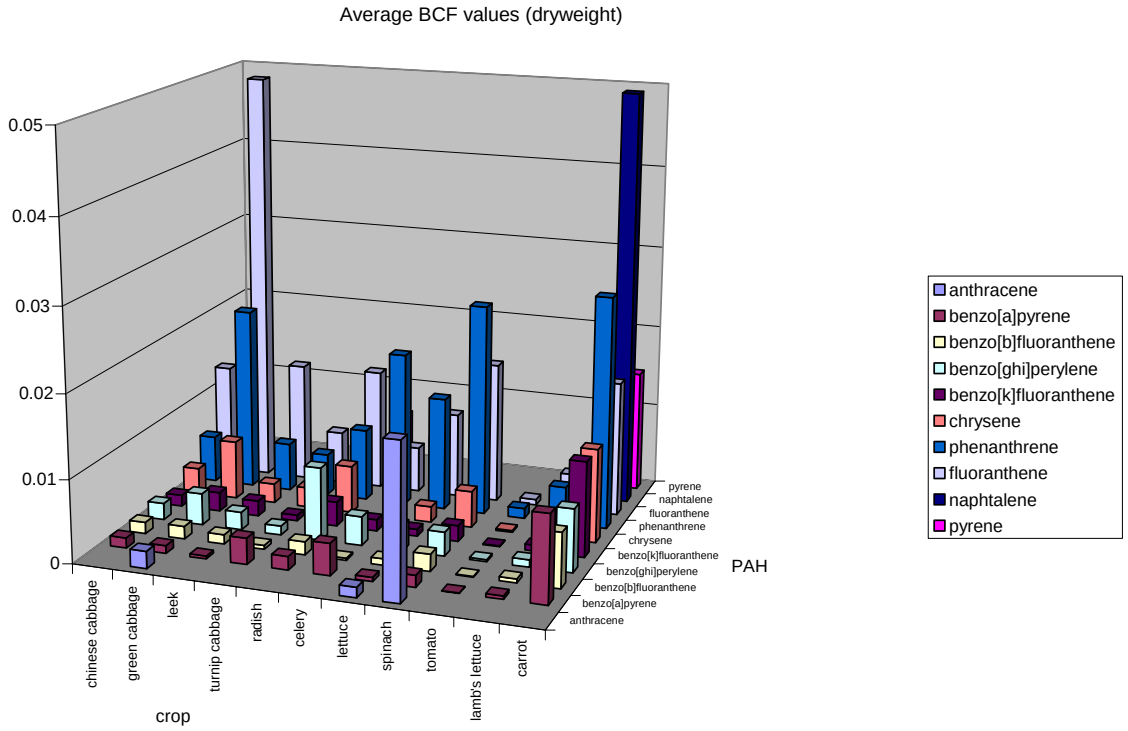

Figure 3.3: BCFs (BioConcentrationFactors) for PAHs, for several relevant vegetables.

Furthermore, Fig. 3.3 shows that accumulation of the different PAHs varies substantially.

For the derivation of the human health based risk limits of the revised Intervention Values contribution of exposure due to vegetable consumption to total exposure is very high for several organic contaminants (Lijzen et al., 2001). The contribution of exposure due to vegetable consumption to total exposure is over 90% for several aromatic compounds (phenols, catechol, resorcinol and hydroquinone), for some chlorinated compounds

(hexachlorobenzene, pentachlorophenol, several PCBs, chlorophenols) and for several pesticides (DDT, DDE, aldrin, carbofuran, propoxur, atrazine, di(2-ethylhexyl)phthalate). For many dioxins this contribution is close to 80%.

3.3.3 Variation between plants

The former sections showed a huge variation in accumulated contaminant concentrations between different plant species (interspecies variation), for both metals and organic contaminants. However, Stenz et al. (1997) show that the metal concentrations in different samples from the same vegetable type (intraspecies variation) from the same plot can differ widely, mainly for cadmium (minimum and maximum values differ often more than one order of magnitude). Alonso et al. (2003) showed large differences for copper and zinc uptake between different samples of the same edible mushrooms species. Also Ruttens (2006a) refers to large intraspecies differences. However, it is generally unknown how this variation relates to the variation in accumulated contaminant concentrations for different plants of the same plant species. This information is important in regard to the number of data needed on soil and plant concentrations for a specific plant species, to be able to derive reliable models that enable calculation of the representative concentration in crops. Moreover, it is important for the field measurement protocol, because it supports the decision on whether to sample many different plants species or (also) many samples for the same plant species. Therefore, it is recommended to further investigate the variation in accumulated concentrations in plants between different plants species versus the variation in accumulated concentrations between plants of the same plant species, in the future.

Besides, the relation between accumulated concentrations in vegetables and in non-edible plants must be further investigated, in the future. When accumulation in non-edible plants represents accumulation in vegetables, data on non-edible plants can be included in the RIVM plant – soil database which forms the basis of the models to calculate accumulated concentrations in vegetables. Besides it would offer more possibilities in field sampling.

4. Vegetable consumption

4.1 Introduction

To be able to determine the "representative concentration" in vegetables, the type of vegetables that are relevant for the development for the protocol must be selected. Because the protocol should be applicable for site-specific risk assessment it is an option to focus on the vegetables that are growing on that specific site at the moment of the assessment. However, this assumption is not always appropriate, for the following reasons:

- These vegetables are not always representative for the long time "representative concentration", because the type of vegetables might change every few years.
- On many contaminated sites no (representative) vegetables are present.

Besides, the policy on soil contamination is often related to "*the possibility* to grow vegetables without experiencing adverse effect on human health". This political basic requirement implies that, independent of the vegetables that are growing on the site, the attention should be focused on a "representative consumption pattern".
Relevant Dutch vegetables are potato, carrot, beet, radish, onion, tomato, cucumber, cauliflower, cabbage, lettuce, spinach, endive, french bean, string bean, nave beans, kidney beans and rhubarb. Other edible vegetables in the Netherlands concern asparagus, leek, celery, brussels sprout, eggplant, okra, green pepper, pod, pea, marrow, lentil, courgette, maize, corn and broccoli.

Note that the *calculation* of the "representative concentration" in plants offers more flexibility in the choice of vegetables than field measurements. In the latter case there is no other option than focusing on the type of plant (vegetable or non-vegetable) that is available, unless a "seeding, growing, harvesting program" is performed. In case no vegetables are present at all, representative vegetable concentrations could be calculated. When measuring is more appropriate, attention could be focused on non-edible plants (when available and assuming that uptake in non-edible plants is related to that in vegetables), or again a "seeding, growing, harvesting program" must be performed.

4.2 Consumption rates

4.2.1 General population

In the Netherlands detailed evaluations of vegetable consumption rates are performed regularly. The latest complete survey is the Dutch National Food Consumption Survey from 1998 (Voedingscentrum, 1998). The average consumption of vegetables in the Netherlands for babies and pre-scholers (1-6 year) and for adults and schoolgoing children (age 7-70 year), for potatoes and other vegetables, is given in Table 4.1.

Table 4.1: The average consumption (g_{fw}/ day) for potatoes and other vegetables in the Netherlands (source: Dutch National Food Consumption Survey; Voedingscentrum, 1998)

Group	**Time span**	**Potatoes**	**Other vegetables**
Babies and pre-scholars	1-6 year	59.5	58.3
Adults and schoolgoing children*	7-70 year	122	139

*About 6% of the consumed amounts of vegetables and about 9% of the consumed amount of potatoes are obtained outdoors. These amounts are not taken into account.

A more recent food consumption survey was performed in 2003. However this survey was focused on the specific age-group of 19 – 30 year and the data were collected in the fourth quarter of 2003 only (Hulshof et al., 2004). Besides, the methodology of the survey was different from earlier surveys. A new baseline survey for all age-groups is foreseen in 2007.

To investigate the development of total vegetable rates in time, several Dutch surveys are compared. National food consumption surveys for all age groups have been performed in 1987-1988 (Ministry of WVC, 1988), 1992 (Voorlichtingsbureau voor de voeding, 1993) and 1997-1998 (Voedingscentrum, 1998). The average lifelong daily consumption rates have been given in Table 4.2. The results of the recent food consumption survey for the specific age-group of 19 – 30 year (Hulshof et al., 2004) are also given in Table 4.2.

Table 4.2: Overview of average lifelong daily consumption rates of potatoes and other vegetables (g_{fw}/day) from Dutch food consumption surveys and for kitchen gardeners (between brackets: data for the specific age-group of 19-30 year)

	General population				**Kitchen gardeners**
	Survey	*Survey*	*Survey*	*Survey*	*Survey*
Year	1988	1992	1998	2003	1988
Potatoes	131	119	114	(96)	147
Other vegetables	150	135	128	(100)	246
Total	**281**	**254**	**242**	**(196)**	**393**

The table indicates that since 1988 there is a decreasing trend in both the total consumption rate for potatoes and other vegetables. This trend seems to continue up to the year of 2003, although these data should be interpreted with care since they reflect a specific age group and the methodology of the survey was different from earlier surveys. However, the average lifelong consumption of the age group of 19-30 year, which is expected to be higher than the lifelong average of all age groups, is low compared with the results of the previous years. The data of 2003 confirm the trend of decrease in consumption rates (pers. comm. K. Hulshof, November 2005).

Preferably, the most recent data must be included. However, the data from 2003 are less representative since they refer to a specific age group (19-30 year) and relate to data collected in the fourth quarter of 2003 only. Thus, at this moment the 1998 data are the most actual for total consumption rates in the Netherlands. Because there is a decreasing trend in total consumption rates for potatoes and other vegetables the 1998 data probably give a slight overestimation of the actual vegetable consumption rates.

4.2.2 Kitchen gardeners

In a study focused on 154 households with kitchen gardens (Hulshof, 1988) the average lifelong consumption of potatoes and other vegetables, both from own vegetable garden and from other sources, was 147 and 246 gram per day, respectively (data also included in Table 4.2). Thus, kitchen gardeners consume more vegetables than the general population. Based on the Dutch National Food Consumption Survey (Voedingscentrum, 1998) and Hulshof (1988) the factor of difference considering the average lifelong consumption of potatoes is approximately 1.1. For the other vegetables this factor is approximately 1.7 for adults and schoolgoing children and 1.2 for babies and pre-scholars.
Although the data for kitchen gardeners refer to a situation in 1988 and the total consumption rates might have changed in time, these factors of difference are considered the most representative for the difference between the consumption rates of kitchen gardeners and of the general population. Applying these factors to the consumption data of the general population results in the average lifelong consumption rates for potatoes and other vegetables for kitchen gardeners as calculated in Table 4.3.

Table 4.3: The (calculation of the) average lifetime consumption rates (g_{fw}/day) for potatoes and other vegetables for kitchen gardeners

Group	Time span	Potatoes	Other vegetables
Babies and pre-scholars	0-6 year	59.5 x 1.1 = 66	58.3 x 1.2 = 70
Adults and schoolgoing children	7-70 year	122 x 1.1 = 134	139 x 1.7 = 236

4.3 Consumption pattern

Besides the total consumption rates also the contribution of different vegetables to the total consumption rate (consumption pattern) is of importance. Dooren-Flipsen et al. (1996) used the data of the Dutch National Food Consumption Survey (Voorlichtingsbureau voor de voeding, 1993) to transform the average consumption rates of foodstuffs into consumption rates of primary agricultural products. They considered 32 vegetables, divided over 9 vegetable groups. Table 4.4 gives the average consumption pattern (average for all age groups and both sexes). The consumption pattern, expressed in gram fresh product per day, is converted to the consumption pattern in gram dry weight per day[3]. Besides the contribution of each vegetable group to total consumption is given.

Note that these data from Dooren-Flipsen et al. (1996) in Table 4.4 and Voorlichtingsbureau voor de voeding (1993) in Table 4.1 show different total rates of consumed potatoes and other vegetables. The reason for this is that the Dutch food consumption survey records in terms of prepared (cooked) actual amounts of foods, which includes composite products. The data in Table 4.4 are based on the fresh product at harvesting (shrink and waste included). The former is applicable for the estimation of the total consumption rates. The latter is applicable for calculation of the generic plant – soil relations (see next chapter). The relative contribution of vegetables to total consumption is expected to be similar in both surveys.

In section 1.3 it was explained that the tiered approach allows for two different steps, i.e. the use of Critical soil concentrations in Tier 1 and a detailed assessment of the actual risk on the basis of a site-specific risk calculation in Tier 2. Both Critical soil concentrations and the detailed assessment of the actual risk on the basis of a site-specific risk calculation should be applicable for the majority of cases. Furthermore, it should be considered that the policy on soil contamination is often related to "*the possibility* to grow vegetables without experiencing adverse effect on human health". This political basic requirement implies that, independent of the vegetables that are growing on the site, the attention should be focused on the "average vegetable pattern", for both calculation steps.
As a consequence, the choice of vegetables does not imply an additional conservative element in the derivation of Critical soil concentrations and the detailed assessment of the actual risk on the basis of a site-specific risk calculation.

[3] Calculation fresh weight - dry weight based upon water content data in EPA Exposure Factor Handbook 1997.

Table 4.4: Average consumption pattern in the Netherlands (Dooren-Flipsen et al., 1996)

no	Group	Vegetable	Average consumption g_{fw}/day	Water content g/100 g product	Average consumption g_{dw}/day	Contribution to total consumption Rate % (vegetable)	% (group)
0	Potatoes	Potatoes	179.7	83.3	30.0	61.6	61.6
1	Roots and tubers	Beetroot	5.2	87.3	0.65	1.3	5.1
		Carrots	13.4	87.8	1.64	3.4	
		Celeriac	0.8	88.0	0.09	0.2	
		Turnip	0.8	91.9	0.07	0.1	
		Radish	0.4	94.8	0.02	0.05	
		Winter carrot	0.2	87.8	0.02	0.04	
2	Bulbous vegetables	Onions	17.0	90.8	1.56	3.2	7.7
		Leek	12.9	83.0	2.19	4.5	
3	Fruiting vegetables	Tomatoes	26.1	94.0	1.56	3.2	5.0
		Cucumber	8.0	96.1	0.31	0.6	
		Melon	2.2	89.7	0.23	0.5	
		Maize	1.4	76.0	0.34	0.7	
4	Cabbages	Cauliflower	16.0	92.3	1.23	2.5	7.6
		Brussels sprouts	4.7	86.0	0.65	1.3	
		White cabbage	7.0	95.3	0.33	1.6	
		Red cabbage	5.1	91.6	0.43		
		Ox heart cabbage	2.0	95.3	0.10	0.2	
		Curly kale	4.9	84.5	0.76	1.6	
		Broccoli	2.0	90.7	0.18	0.4	
5	Leaf vegetables (greens)	Lettuce (head)	8.5	95.4	0.39	0.8	4.4
		Endive	7.4	93.8	0.46	0.9	
		Spinach	10.4	91.6	0.88	1.8	
		Chicory	9.2	95.3	0.43	0.9	
6	Legumes (peas and beans)	Green bean	11.7	90.3	1.13	2.3	6.9
		String/bush bean	3.1	90.3	0.30	0.6	
		Broad/horse/ fava bean	2.5	88.9	0.28	0.6	
		Garden pea	14.8	88.9	1.64	3.4	
7	Beans	Haricot bean	0.9	77.1	0.20	0.4	1.2
		Kidney bean	1.8	77.1	0.40	0.8	
8	Stem and stalk vegetables	Rhubarb	0.7	93.6	0.05	0.1	0.4
		Asparagus	1.7	92.3	0.13	0.3	

5. Accumulation of metals in vegetables

This chapter focuses on the procedure for the calculation of the concentration in vegetables. The last few years many data on metal concentrations in soils and vegetables were published. Unfortunately, evaluation and incorporation of these additional data into the RIVM plant – soil database could not be done within the scope of this study.

5.1 Critical vegetable concentration (cadmium)

In respect to exposure through vegetable consumption, cadmium is an important contaminant. It is a very frequently found in soils, at locally contaminated sites as well as diffusely distributed in agricultural soils. Moreover, human health risks can be crucial for cadmium and the human health risk is dominated by exposure due to vegetable consumption (in section 3.3.1 it was concluded that the contribution of exposure due to vegetable consumption to total exposure is 93% for the standard exposure scenario). For this reasons, attention is focused on the derivation of a Critical vegetable concentration for cadmium, in this separate section.

A Critical vegetable concentration is calculated for cadmium and the Vegetable garden site use, using Eq. 2.3. The consumption rates for potatoes and other vegetables are taken from Table 4.1, corrected for the percentage dry weight given in Table 4.4. As a conservative approach it has been assumed that all other vegetables (i.e. vegetables other than potatoes) concern sensitive crops like lettuce, endive, spinach and broccoli.
In this approach a standard adult has a body weight of 70 kg and consumes 10.2 g_{dw}/d of potatoes (50% of total potatoe consumption of 22.4 g_{dw}/d) and 9.89 g_{dw}/d of the sensitive vegetables (100% of total consumption of other vegetables). A standard child has a body weight of 15 kg and consumes 4.97 g_{dw}/d of potatoes and 4.14 g_{dw}/d of sensitive vegetables. The MTR_{human} for cadmium is 0.5 $\mu g/kg_{body\ weight}$/day (Baars et al., 2001). The water content (see Table 4.4) of potatoes is 83.3% and the average water content for the sensitive vegetables is 92.9%. The correction for relative bioavailability in the human body, $f_{bioavailability}$, is assumed to be 1.0 (i.e. no correction). This results in a Critical vegetable concentration of 1.74 mg/kg_{dw} for adults and 0.82 mg/kg_{dw} for children. As a lifelong-averaged Critical vegetable concentration for cadmium and the Vegetable garden site use a value of 1.66 mg/kg_{dw} results.

In an even more conservative approach the higher consumption rates from kitchen gardeners from Table 4.3 are used. Consumption rates for adults are in that case 11.2 g_{dw}/d of potatoes and 16.8 g_{dw}/d for sensitive vegetables. For children consumption rates are 5.51 g_{dw}/d of potatoes and 4.97 g_{dw}/d for sensitive vegetables. This results in a Critical vegetable concentration of 1.25 mg/kg_{dw} for adults and 0.72 mg/kg_{dw} for children. As a lifelong-

averaged Critical vegetable concentration for cadmium and the Vegetable garden site use (higher consumption rates) a value of 1.20 mg/kg_{dw} results.

5.2 Experimental data in soils and vegetables

Since cadmium is such an important contaminant for human health risk assessment, it is useful to look into more detail at this metal. The RIVM plant - soil database was extended with few novel data and was used subsequently to plot the cadmium concentration in vegetables as a function of the cadmium concentration in soil, for the whole RIVM plant – soil database (Figure 5.1).

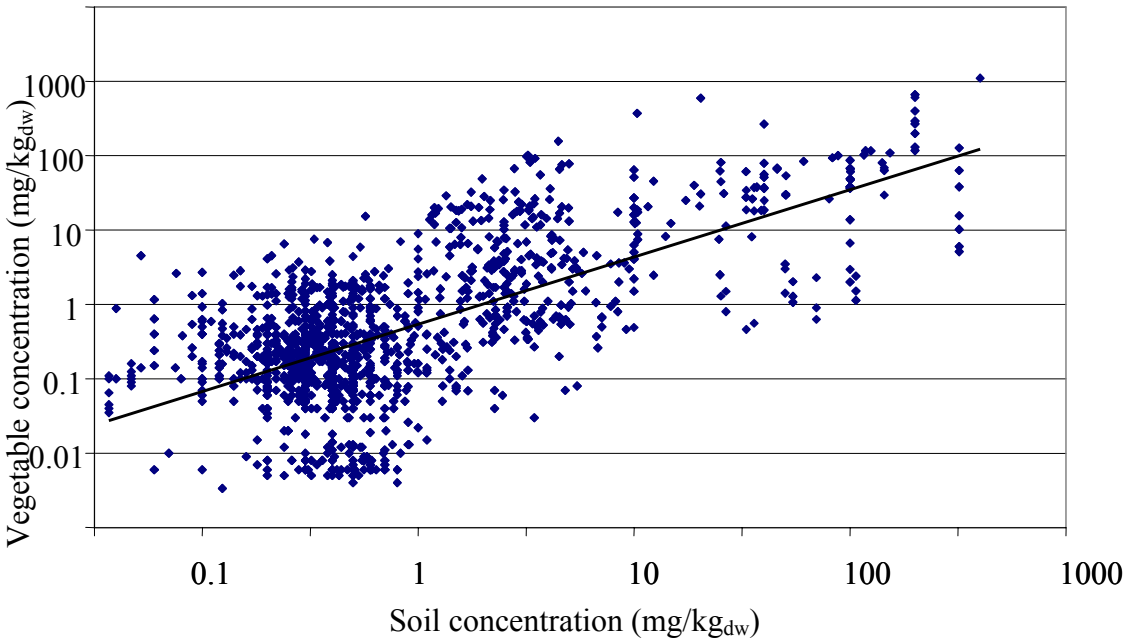

Figure 5.1. The cadmium concentration in vegetables (mg/kg_{dw}) as a function of the cadmium concentration in soil (mg/kg_{dw}), for all available data in the RIVM plant – soil database.

Generally the vegetables have higher cadmium concentrations at higher soil concentrations. However, this figure shows that the relation between the cadmium concentration in vegetables and in soils has a huge variation. At a specific concentration in soil the concentration in different vegetables can vary with three orders of magnitude. The R^2 for this relation is 0.43.

The highest vegetable concentrations are found in the studies were cadmium was added to field plots or in pot experiments. These soils might represent a worst-case situation, because no aging has taken place and, hence, cadmium is readily available for uptake. In most cases,

cadmium present at contaminated sites is the result of historical contamination and is, hence, less available. Therefore a more realistic risk assessment must be focused on studies with historically contaminated soils, in the field.

Figure 5.2 shows the cadmium concentration in potatoes as a function of the cadmium concentration in soil.

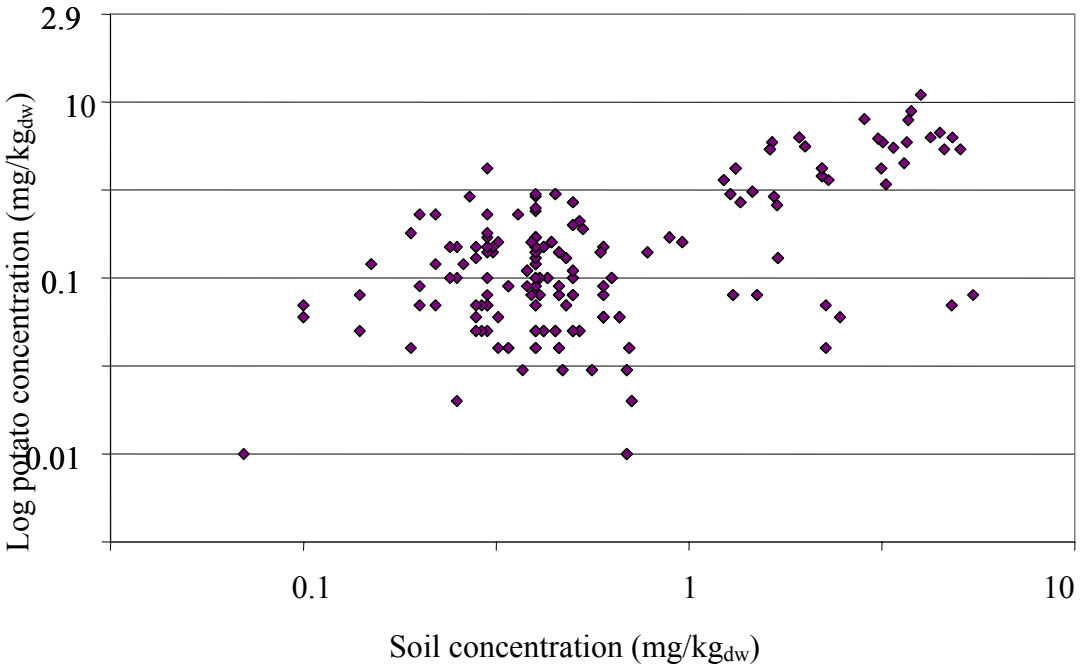

Figure 5.2: The cadmium concentration in potatoes as a function of the cadmium concentration in soils, for all available data in the RIVM plant – soil database.

Figure 5.2 shows that the relation between the cadmium concentration in potatoes and in soils has a huge variation. At a specific concentration in soil the concentration in potatoes can vary with more than two orders of magnitude. Despite of the high variation, the cadmium concentration in potatoes is relatively low. Even at the highest tested cadmium concentration in soil of 7 mg/kg_{dw}, the concentration in potatoes is lower than the Critical vegetable concentration for Vegetable gardens of 1.7 mg/kg_{dw} and even than the more conservative Critical vegetable concentration for Vegetable gardens (consumption rates for kitchen gardeners) of 1.2 mg/kg_{dw}.

Figure 5.3 shows the cadmium concentration in different sensitive vegetables as a function of the cadmium concentration is soil. Spinach, endive, lettuce and broccoli are shown, because these vegetables can accumulate significant amounts of cadmium. Besides, abundant data were present for these vegetables in the RIVM plant - soil database.

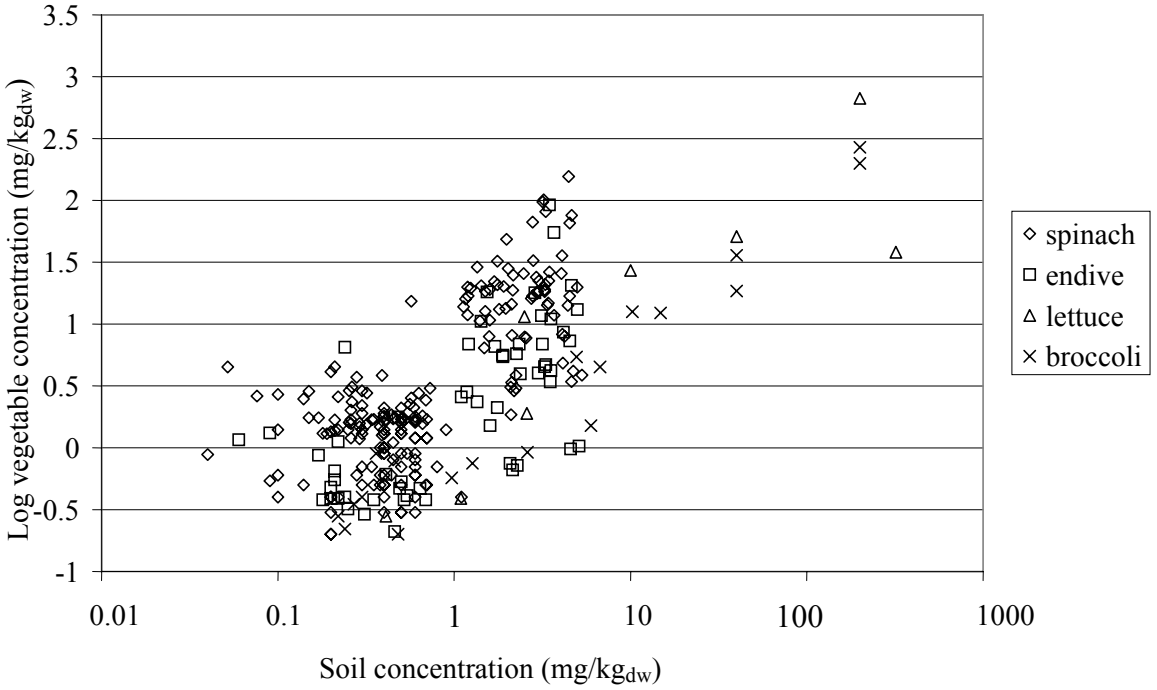

Figure 5.3: The cadmium concentration in sensitive vegetables as a function of the cadmium concentration in soil, for all available data in the RIVM plant – soil database.

The measured cadmium concentrations can be compared with the Critical concentration for cadmium of 1.7 mg/kg$_{dw}$ or 1.2 mg/kg$_{dw}$. It can be concluded that even at cadmium concentrations in soil in between 0.1 and 1.0 mg/kg$_{dw}$ the Critical vegetable concentrations of 1.7 or 1.2 mg/kg$_{dw}$ are exceeded in these sensitive crops, in some cases.

5.2.1 The influence of soil properties

As shown in section 3.1.1 soil properties have a major influence on the uptake of metals by vegetables. A study of Römkens and De Vries (2001) showed that cadmium concentrations in spinach and endive correlated with cadmium concentrations in soil. In addition the percentage of clay, organic carbon and lime were reported in this study. This permits the classification into so-called risk classes. “High-risk” soils have no lime, less than 10% organic matter and less than 5% clay. “Low-risk” soils have more than 5% lime, more than 10% organic matter and more than 25% clay. The soil properties of “intermediate-risk” soils are in between the soil properties of “high-risk” soils and “low-risk” soils. Figure 5.4 shows the cadmium concentration in spinach and endive as a function of the cadmium concentration in soils, for each of the three risk classes of soils.

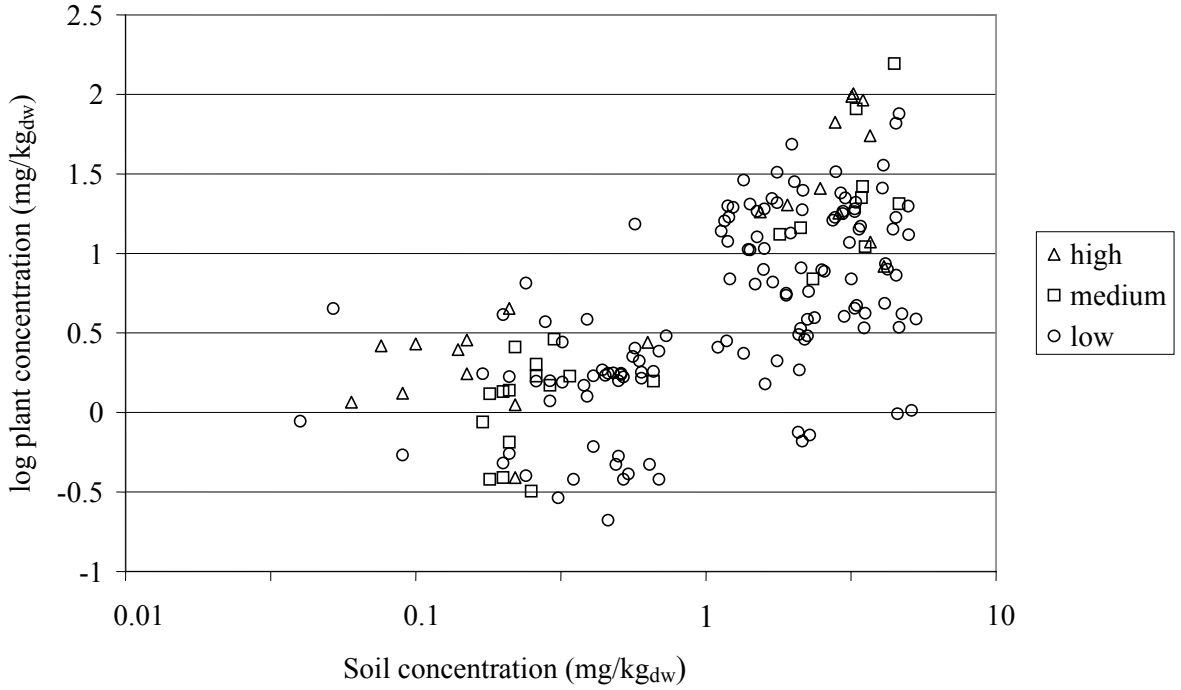

Figure 5.4: The cadmium concentration in spinach and endive as a function of the cadmium concentration in soil for three different "soil risk classes". "High-risk": 0% lime, less than 10% organic matter and 5% clay; "low-risk": more than 5% lime,10% organic matter and 25% clay; intermediate-risk: not "high-risk" or "low-risk"

This figure shows that even the soils in the so-called "low-risk" category can show high cadmium concentrations in spinach and endive. This implies that there is also a considerable human health risk due to consumption of leafy vegetables, even for soils with high amounts of lime, organic matter and clay.

5.3 Calculation procedure

5.3.1 BioConcentrationFactors (BCFs)

A common parameter for estimating the metal concentration in vegetables and subsequently human exposure through consumption of vegetables is the BioConcentrationFactor (BCF), i.e. the ratio between the concentration of (the edible part of) the vegetable and the soil concentration:

$$BCF_{metal} = \frac{Mvegetable}{Msoil} \qquad \text{Eq. 5.1}$$

in which:

BCF_{metal}	BioConcentrationFactor for metals	[(mg/kg$_{dw}$plant)/ (mg/kg$_{dw}$soil)]
Mvegetable	metal concentration in the edible part of the vegetable	[mg/kg$_{dw}$plant]
Msoil	total metal soil concentration	[mg/kg$_{dw}$soil]

Because of their simple application BCFs are widely used (e.g. *Tome et al., 2003*, *Alonso et al., 2003*).

In section 3.3.1 generic BCFs were used as an indicator for the affinity for accumulation of metals in plants. These BCF values, however, are not a constant value for a specific metal in a specific vegetable. Soil properties like the soil pH, clay content, organic matter content, the metal concentration in soil and plant factors like type of plant and growth rate have a strong influence on the actual BCF. The BCF values also differ for different parts of a plant being generally higher for roots and lower for fruits (e.g. Wojciechowska-Mazurek et al. (1995) for metals; Samse-Petersen et al. (2002) for seven trace elements and five PAHs).
The experimental data presented in section 5.2 illustrate the limitation of generic BCF values.

Besides, these generic BCF values are valid within a specific range of soil parameters and extrapolation outside the original range is not appropriate. If for example the calculated Critical vegetable concentrations are much higher than the plant concentrations in the dataset, the calculated Critical soil concentration might be conservative. The use of these BCF values can also lead to Critical soil concentrations which are too high. This is the case when the specific BCF value was derived from a study using a soil with a poor metal bioavailability. A major practical problem is that a *generic* BCF is that does not allow any specifications depending on site specific elements.

Bockting and Van den Berg (1992) derived generic BCF values for the derivation of the Dutch soil quality standards from a wide range of plants. These BCF-values were derived from field data, laboratory experiments and estimations, from which the geometric mean was used. No attention was paid to the vegetable selection related to consumption rates, metal concentration in soil, or to the respective soil properties. Furthermore, the dataset included plants that were irrelevant for consumption.

5.3.2 Plant – soil relations

In the framework of the revision of the Dutch Intervention Values an improved procedure for the assessment of the accumulated concentration in vegetables was developed (Versluijs and Otte, 2001). In this study Freundlich-type plant – soil relations were derived for the calculation of the accumulated concentration in vegetables, as a function of total soil

concentration and the major soil properties. For each vegetable with sufficient and proper data available, the following equation was derived:

$$log[Mvegetable] = a + b\ log[Msoil] + c\ pHsoil + d\ log[\%Clay] + e\ log[\%OC] + f\ [other\ factors] \quad \text{Eq. 5.2}$$

in which:

Mvegetable	metal concentration in the edible part of the vegetable	[mg/kg_{dw}]
Msoil	total metal concentration in the soil in	[mg/kg_{dw}]
pH	pH KCl= -log H^+soil	
%Clay:	clay content of the soil	[%]
%OC:	organic carbon content of the soil	[%]
a, b, c, d, e, f	empirical parameters	[-]

This Freundlich-type plant – soil relations have been used by several other researchers (e.g. Krauss et al. (2001) for accumulation of metals in wheat, *Triticum aestivum*; Hough et al. (2004) for several crops and metals; Römkens et al, 2004 and Römkens et at., 2005 for cadmium and lead in several vegetables the Dutch Kempen region).

In Versluijs and Otte (2001) linear regression was performed to determine the coefficients (a, b, c, d, e and f) of the plant – soil relations, for each vegetable separately. The influence of other factors (e.g. climate, land management, environment) was considered as noise on the data. The resulting plant – soil relations enable site-specific calculation of the accumulated concentration in specific vegetables and the derivation of vegetable-specific BCFs for a specific metal concentration and specific soil properties.

Calculated vegetable concentrations using the plant – soil relations have a relatively large uncertainty when using extreme values (either high or low) for the soil concentration, pH, organic matter content and clay content. Versluijs and Otte (2001) proposed that the application range for the derived plant – soil relations is within the 5th and 95th percentiles of the underlying data for soil concentration, pH, organic matter content and clay content. Outside these boundaries the BCFs were fixed at either the 5th percentile (in case values are lower than this 5th percentile) or at the 95th percentile (in case values are higher than this 95th percentile). This proposal has been followed in this study. The limit values for the concentrations and soil properties are given in Versluijs and Otte (2001), for all relevant metals.

Note that the calculation of Critical soil concentrations in soil with the use of the plant – soil relations concerns an iterative procedure, since the BCF, an important input parameter, is dependent on the soil concentration.

5.3.3 Mixed model

When for a specific vegetable no valid plant – soil relation could be derived, the geometric mean of the BCFs was used in this study, in analogy with Versluijs and Otte, 2001, as schematized in Figure 5.5.

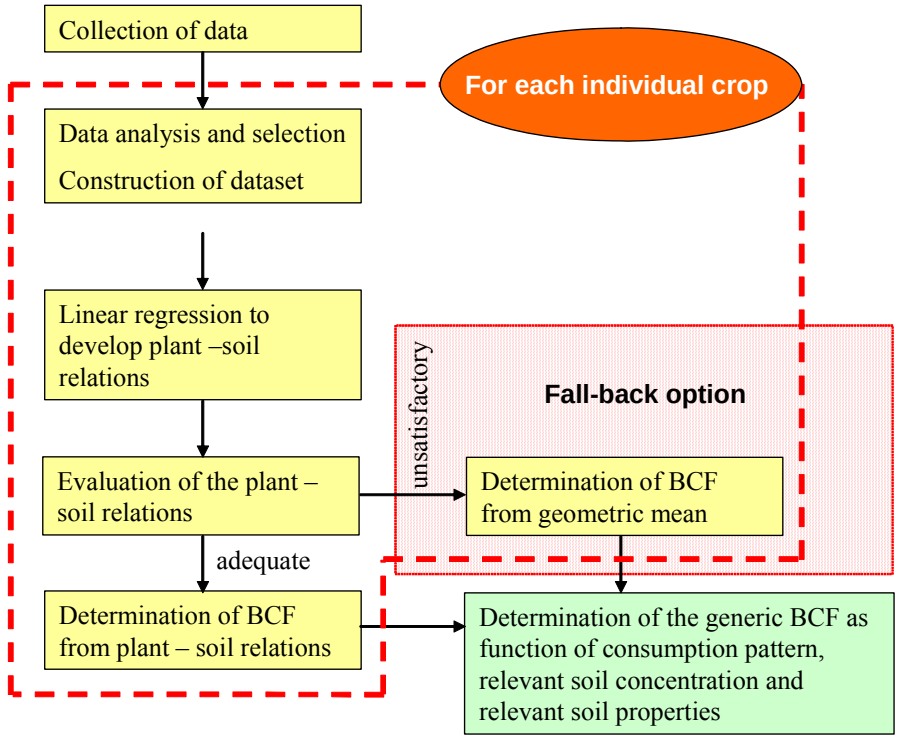

Figure 5.5: Schematisation of the procedure to derive vegetable-specific BCFs ("Mixed model").

5.3.4 Soil type correction

As a consequence, site-specific assessment of risk due to vegetable consumption is only limited, because the geometric means of the BCFs have no relation with soil properties. However, to improve the degree of site-specific assessment in a practical way it is proposed to correct the BCFs from geometric means with the soil type correction factors, which are used to match Target and Intervention values with actual soil properties (Van den Berg et al., 1993). These soil type corrections, which are lacking for antimony, molybdenum, selenium,

tellurium, thallium en silver, are functions of the organic matter and clay content in the soil and are related to the standard soil used to derive the Target and Intervention Values. This standard soil has an organic matter content of 10% and a clay content of 25%. Note that the influence of the pH, although important in relation to plant uptake, is not included in these soil type corrections. The reason for this is that the pH is not a stable parameter, i.e. can change within a relatively short time frame. The correction factor for the Target and Intervention Values, $STcf_{TIV}$, is written as:

$$STcf_{TIV} = \frac{A + (B \; x \; \%Clay) + (C \; x \; \%OM)}{A + (B \; x \; 25) + (C \; x \; 10)} \qquad \text{Eq. 5.3}$$

in which:

%Clay:	clay content of the soil	[%]
%OM:	organic matter content of the soil	[%]
A, B, C:	metal specific variables, see Table 5.1	[-]

Organic matter contents higher than 30% are normalized at 30%; organic matter contents lower than 2% are normalized at 2%. The metal specific variables are given in Table 5.1.

Table 5.1: Metal specific variables for the soil type correction [-], according to Eq. 5.3

Metal	**A**	**B**	**C**
Arsenic	15	0.4	0.4
Barium	30	5	0
Beryllium	8	0.9	0
Cadmium	0.4	0.007	0.021
Chromium	50	2	0
Cobalt	2	0.28	0
Copper	15	0.6	0.6
Mercury	0.2	0.0034	0.0017
Lead	50	1	1
Nickel	10	1	0
Tin	4	0.6	0
Vanadium	12	1.2	0
Zinc	50	3	1.5

To account for the influence of differences in soil type on the BCFs, a soil type correction for BCFs is proposed, related to the medium values of organic matter and clay contents of the RIVM plant - soil database. As a consequence, the soil type correction factor for the BCF, $STcf_{BCF}$, is:

$$STcf_{BCF} = \frac{A + (B\ x\ \%Clay) + (C\ x\ \%OM)}{A + (B\ x\ \%Clay_{average}) + (C\ x\ \%OM_{average})} \qquad \text{Eq. 5.4}$$

in which:

% Clay	actual clay matter content	[%]
%OM	actual organic matter content	[%]
% $Clay_{average}$	average clay matter content of the of the RIVM plant – soil database	[%]
$\%OM_{average}$	average organic matter content of the RIVM plant – soil database	[%]

This correction on the basis of the soil type correction factors is a deviation from Versluijs and Otte (2001) and Otte et al. (2001).

The soil type correction factor for the BCF for cadmium, for example, ranges from 0.61 for a sandy soil with no organic matter up to 1.56 for clay soils with a high organic matter content. To apply this correction the geometric values for the BCF must be divided by the soil type correction factor. This implies for the above given example that the BCF based on geometric means is increased with a factor of 1.64 for a sandy soil with no organic matter (BCF divided by 0.61) and decreased with a factor of 1.56 for clay soils with a high organic matter content.

5.3.5 Vegetable-group-consumption-rate-weighted BCFs

As discussed in section 4.3 “the average vegetable pattern”, including 32 vegetables, is considered a valid basis to calculate the Critical soil concentrations and for site-specific risk assessment. To this purpose the vegetable-specific plant - soil relations of the vegetables that contribute in the consumption pattern has been integrated in *vegetable-consumption-rate-weighted* plant - soil relations. Although the consumption pattern in the Netherlands might have changed since these data were reported, it is believed that Dooren-Flipsen et al. (1996) (Table 4.4) is the best representation of the actual Dutch consumption pattern. The vegetable-consumption-rate-weighted BCF, $BCF_{veg\text{-}cr\text{-}weighted}$, is calculated as follows:

$$BCF_{veg\text{-}cr\text{-}weighted} = \Sigma(w_i\ x\ BCF_{pl\text{-}soil\ relation}) + \Sigma(w_j\ x\ BCF_{geo\ mean}) / STcf_{BCF} \qquad \text{Eq. 5.5}$$

in which

w_i	vegetable-consumption-rate-weighting factor, for vegetables for which a plant – soil relation is available	[-]
$BCF_{pl\text{-}soil\ relation}$	BCF based on plant – soil relation [$(mg/kg_{dw\text{-}plant}) / (mg/kg_{dw\text{-}soil})$]	
w_j	vegetable-consumption-rate-weighting factor, for vegetables for which a geometric mean BCF is used	[-]
$BCF_{geo\ mean}$	BCF based on geometric mean [$(mg/kg_{dw\text{-}plant}) / (mg/kg_{dw\text{-}soil})$]	
$STcf_{BCF}$	soil type correction factor for the BCF	[-]

However, when all individual plant – soil relations would be used, independent of the respective vegetable groups, some vegetable groups for which a relatively large number of different vegetable data are available would have more weight in the overall *vegetable-consumption-rate-weighted* plant – soil relations. With the purpose to prevent that too much stress is given to a specific vegetable group, the overall consumption-rate-weighted plant – soil relation is based on the individual plant – soil relations for the *vegetable groups* instead of on the individual plant – soil relations. As a consequence, the attention is focused on the *vegetable-group-consumption-rate-weighted* BCFs. To this purpose, first a *vegetable-consumption-rate-weighted* average BCF is calculated for each vegetable group on the basis of the BCFs for each vegetable within that group. These BCFs for each vegetable can either result from a plant – soil relation or from a geometric mean with soil type correction. In the following step, the vegetable-group-consumption-rate-weighted BCF, $BCF_{veg\text{-}gr\text{-}cr\text{-}weighted}$, is calculated as follows:

$$BCF_{veg\text{-}gr\text{-}cr\text{-}weighted} = \Sigma (u_i \times BCF_{vegetable\ group}) \qquad \text{Eq. 5.6}$$

in which

u_i	vegetable-group-consumption-rate-weighting factor,	[-]
$BCF_{vegetable\ group}$	vegetable-consumption-rate-weighted average BCF for a specific vegetable group	$[(mg/kg_{dw\text{-}plant}) / (mg/kg_{dw\text{-}soil})]$

In most cases it does not make sense to relate a vegetable(-group)-consumption-rate-weighted BCF to the vegetables that are actually grown at the site. The reason for this is that this vegetable pattern does not represent a sustainable situation. In most cases it is more useful to assess the risks for growing vegetables on the site over a longer time span, represented by an average vegetable package for the Netherlands. An exception could be made when traditionally specific vegetables are grown for example because of regional tradition, on a specific site. In that case a specific BCF could be used on the basis on a “regional” vegetable(-group)-consumption-rate-weighting.

5.3.6 Discussion

The plant - soil relations were evaluated for statistical correctness using an F-test (one-sided exceeding probability) and significance (Versluijs and Otte, 2001). Unfortunately, the statistical significance was insufficient for many of the plant – soil relations. The uncertainty of the coefficients was also quantified by Versluijs and Otte (2001). For some vegetables the standard deviations of the coefficients are large. It is possible, although not necessary, that the “noise” in the datasets will reduce when the volume of the dataset increases by adding other datasets. In this respect, it is highly recommended to extent the RIVM plant – soil

database with other plant and soil data, from other existing datasets and with more recent data from the literature.

In Table 5.2 the characteristics of the Bockting and Van den Berg (1992) approach on generic BCFs and of the Freundlich-type plant – soil relations from Versluijs and Otte (2001) are presented. The table gives a general outline of the underlying data and concepts, the assumptions made and practicability.

Table 5.2: Comparison of two approaches for the determination of metal concentrations in vegetables.

APPROACH	***Generic BCFs (Bockting and Van den Berg, 1992)***	**Freundlich-type plant – soil relations *(Versluijs and Otte, 2001)***
General	Average or median BCF based on measured data	Vegetable-specific plant – soil relations, obtained by linear regression using field data
Dependent on degree of contamination	No	Yes
Soil properties dependent	No	Yes: pH, clay and organic matter content
Consumption pattern considered	No, only the consumption of potatoes and above-ground vegetables	Yes
UNDERLYING DATA		
Datasets	Dataset includes estimations and data from pot experiments	Field data from different sources
Field data	Dominant	Exclusive
Home grown plants	Dominant	Exclusive
Consumable parts	Sometimes	Exclusive
VALIDITY/ USE		
Potential risk assessment	Yes	Yes
Metal concentration range	Relatively low (around Target Value)	Dependent on actual metal concentration
Site-specific risk	No	Yes, within certain ranges for pH, clay and organic matter content

From this comparison it can be concluded that the approach of Versluijs and Otte (2001) has a much better relation with vegetables, instead of plants in general, and with soil concentration and soil properties. The latter implies that, when significant, these relations are more suited for site-specific risk assessment.

In the calculations with the plant – soil relations extreme values for the metal concentration in soil, pH, organic matter content and clay content are fixed at either the 5th percentile (in case values are lower than this 5th percentile) or at the 95th percentile (in case values are higher than this 95th percentile). This has been done to avoid miscalculation due to extrapolation. As a consequence, the calculations of the BCFs outside the 90% confidence interval are not very reliable. That means, for example, that a decent calculation of the cadmium concentration in vegetables is not possible at low concentrations (lower than 0.43 mg/kg$_{dw}$), and for a sensitive soil (pH lower than 5.6, an organic matter content lower than 4.3% and a clay content lower than 10.9%). As another example, no decent calculation of the lead concentration in vegetables is possible at high soil concentrations (higher than 253 mg/kg$_{dw}$) and for non-sensitive soils (pH higher than 7.7, an organic matter content higher than 14.3% and a clay content higher than 25.9%).

The soil type correction, that is performed to correct the geometric means of the BCFs for actual soil properties, rather concerns a practical approach. However, although the scientific foundation is limited, it improves the strength for site-specific application.

In many cases metals are integrated in matrixes, like debris, porcelain, or bullets. It is expected that in these situations metals are less available, at least over a specific time span, than metals adsorbed to the soil solid phase. This phenomenon is not included in this study, which is a shortcoming of the calculation procedure, like in most procedures to calculate concentrations in plants. It is advised to perform further research on the (time span for the) limited availability of metals in these matrixes, in the future.

In some cases complex formation of some metals in the pore water could increase the uptake of other metals, with lower affinity for complex forming (Kalis et al., 2006). This illustrates the important role of competition between metals, which also is not included in this calculation procedure. This is a further serious shortcoming of the calculation procedure in this study, like in most procedures to calculate concentration in plants.

Another improvement of the use of the plant – soil relations would be to test the calculated vegetable concentrations against criteria for phytotoxicity, i.e. levels in vegetables that will damage plant tissue and reduce growth. The reason for this is that when vegetables do not look healthy consumption, and hence exposure, will not take place. As a matter of fact, phytotoxicological limit values in vegetables are the upper limits to what humans can be exposed. In Lijzen et al. (2002) a first approach towards the derivation of these phytotoxicity criteria was made. In this study, effect data on plants were selected from a huge dataset with effect data on organisms and plants. Subsequently a HC5 (Hazardous Concentration for 5% of the plants) and a HC50 (Hazardous Concentration for 50% of the plants) for plants were derived. This procedure could be improved by focussing on vegetables instead of all plants and considering the type of effects in more detail.

5.4 Cadmium

In section 5.1 it was explained that cadmium is a very important contaminant in relation to human health risks. For these reasons, the calculation of the concentration of cadmium in vegetables is discussed in this separate section.

5.4.1 Mixed model

For cadmium the plant – soil relations were statistically significant for a considerable amount of vegetables, i.e. 10 of the 12 investigated vegetables, from 6 different vegetable groups. Table 5.3 gives the coefficients of the plant - soil relations for cadmium for these vegetables, according to Versluijs and Otte (2001).

Table 5.3: Regression coefficients of the plant – soil relations for cadmium, according to Eq. 5.2, for 12 vegetables, according to Versluijs and Otte (2001).

Coefficients / Vegetable	a Const.	b Msoil	c pH	d %OC	e %Clay
Potato	-0.86	0.36	0.06	-0.13	-0.27
Red beet	1.90	0.37	-0.18	-0.30	-0.80
Carrot	0.74	0.45	-0.16	0.20	0.09
Radish	0.00	0.12	-0.32	0.00	0.00
Leek	0.70	0.31	-0.20	-0.29	0.00
Tomatoes*	2	0.1	-0.3	-	-
Cabbage*	12	0.6	-0.6	17	-
Curly kale	1.00	0.39	-0.14	-0.50	-0.40
Lettuce	1.00	0.28	-0.18	-0.19	0.16
Endive	0.00	0.42	-0.10	0.10	0.30
Spinach	1.30	0.28	-0.22	-0.64	0.37
Beans#	11.00	0.13	-0.13	-0.10	-9.00

*Plant – soil relations not significant
#Plant – soil relations uncertain

The available data did not permit the specific consideration of the variable “f” for “other factors”.
For cabbage and tomatoes the F-test proved that the coefficients for the plant – soil relations for cadmium were insignificant (Versluijs and Otte, 2001). Furthermore, Monte Carlo analyses showed a high uncertainty of the plant - soil relations for beans (Versluijs and Otte, 2001). Therefore, the geometric mean was used for cabbage and beans, including the soil type correction (fall-back option Figure 5.5). This is a slight deviation from the procedure from Versluijs and Otte (2001). For tomatoes, the quality of the data was doubted, since other data sources showed significantly lower accumulated concentrations. Therefore, the

geometric mean was not used in the calculations, either. This is a major difference with Versluijs and Otte (2001). For this reason, the vegetable-consumption-group-rate-weighted BCF derived in this study is substantially lower, because these unrealistic high BCF has been excluded.
It is expected that with increasing clay content and increasing organic matter content the plant uptake will decrease, resulting in lower vegetable concentrations. However, for some vegetables the coefficients for clay and organic matter in the Freundlich-type equation show a positive relation, i.e. a higher plant concentration (and, hence, a higher BCF) with higher organic matter and clay contents. This is the case for carrots and endive (positive relation with organic matter and clay content) and for lettuce, spinach (positive relation with clay content). This positive relation cannot be explained with general knowledge on the influence of high molecular organic matter and clay on bioavailability and, hence, on plant uptake. At the other side, low molecular organic matter dissolves easily in the pore water, and hence, might increase plant uptake. For ideal analyses the organic matter fraction must be subdivided in at least two organic matter fractions. However, this would hamper practical application in the field.
Because there is no statistical evidence and insufficient scientific arguments for exclusion, the plant - soil relations with positive relations with organic matter and clay contents have been included in this study.

The medium values for the clay and organic matter content for cadmium in the RIVM plant – soil database, respectively, 16% and 7%. As a consequence, the soil type correction factor for the BCF of cadmium, $STcf_{BCF}Cd$, becomes according to Eq. 5.4:

$$STcf_{BCF}Cd = \frac{0.4 + (0.007\ x\ \%Clay) + (0.021\ x\ \%OM)}{0.659} \qquad \text{Eq. 5.7}$$

5.4.2 Vegetable-group-consumption-rate-weighted BCF

The vegetable-consumption-group-rate-weighted BCF for cadmium is calculated according to Eq. 5.6. The BCFs based on the plant – soil relations and geometric means are given in Table 5.4, for several vegetables, for a hypothetical but common soil. The characteristics of this soil are: pH = 7, organic matter content = 2% (organic carbon content = 1.18%), clay content = 5% and the total cadmium concentration in soil = 3 mg/kg_{dw}. The BCF values that are used in the derivation of the overall vegetable-group-consumption-rate-weighted plant – soil relations are given **in bold**.

The vegetable-consumption-rate-weighted average BCF for each vegetable group has also been given in Table 5.4. Also the consumption-group-rate-weighted BCFs for potatoes and other vegetables are given separately. The reason for this is that this enables a differentiation

of the fraction of consumption of home-grown potatoes to total consumption of potatoes in comparison with this fraction for other vegetables. Note that the overall Vegetable-group-consumption-rate weighted BCF is very sensitive to the BCF for potatoes due to the high contribution of potato consumption rate from 62%.

*Table 5.4: BCFs ([(mg/kg$_{dw\text{-}plant}$) / (mg/kg$_{dw\text{-}soil}$)] for cadmium based on the plant – soil relations[*] or geometric means, for several vegetables and vegetable groups and vegetable-group-consumption-rate-weighted BCFs (**in bold**: the values that are used for the calculation of the overall vegetable-group-consumption-rate-weighted BCF).*

Vegetable type	**BCF from Geomean**	**BCF from plant – soil relation**	**Vegetable group**	**Vegetable-consumption-rate-weighted BCF**
Potatoes	0.275	**0.156**	Potatoes	**0.156**
Red beet	0.309	**0.194**	Root vegetables	**0.260**
Carrot	1.318	**0.288**		
Radish	0.661	**0.002**		
Leek	0.120	**0.074**	Bulbous cops	**0.074**
Tomatoes	Not reliable	insignificant	Fruit vegetables	-
White and red Cabbage	**0.326**[#]	insignificant	Cabbages	**0.364**
Curly kale	0.219	**0.368**		
Lettuce	0.741	**0.303**	Leafy vegetables	**0.362**
Endive	0.676	**0.286**		
Spinach	2.291	**0.427**		
White and brown beans	**0.525**[^]	uncertain	Beans	**0.534**
Overall Vegetable-group-consumption-rate-weighted BCF:				
			Potatoes	**0.156**
			Other vegetables	**0.260**
			All vegetables	**0.187**

[*] pH = 7, organic matter content = 2% (organic carbon content = 1.18%), clay content = 5% and the total cadmium concentration in soil = 3 mg/kg$_{dw}$.
[#] Average soil type correction factor of 0.88 (BCF based on geometric mean increased with a factor of 1.14)
[^] Average soil type correction factor of 0.81 (BCF based on geometric mean increased with a factor of 1.23)

A more detailed analysis is given in Table 5.5. In this table the following four sections are presented:

- Information on the crops, i.e. type of crop, weighting factor in regard to total consumption rate, moisture content (columns 2 - 4).
- BCFs for each separate vegetable (dry weight and fresh weight), the basis of the BCF (plant – soil relation or geometric mean), inclusion of the BCF in the calculation of the Vegetable-group-consumption-rate-weighted BCF (yes or no) (columns 5 - 8).

- BCFs for each separate vegetable group (dry weight and fresh weight), vegetable group weighting factor in regard to consumption rate, inclusion of the BCF (yes or no) (columns 9 -12).
- Soil properties, i.e. soil content, pH, percentage organic matter, percentage clay, soil type correction factor (columns 13 - 19). In the first row the selected soil properties are given. In the following rows the actual values are given after correction for the range of application (in between 5^{th} and 95^{th} percentiles).

In the final rows the average vegetable-group-consumption-rate-weighted-BCFs for cadmium are given for aboveground vegetables, root crops and potatoes. Moreover, the average vegetable-group-consumption-rate-weighted BCF for cadmium is given for all vegetables and for all vegetables, exclusive potatoes.

Table 5.5: Table including all information for the calculation of the average vegetable-group-consumption-rate-weighted-BCFs for cadmium, for a hypothetical soil (pH = 7, organic matter content = 2% (organic carbon content = 1.18%), clay content = 5% and the total cadmium concentration in soil = 3 mg/kg$_{dw}$) (page 52).

	Crop information				BCFs vegetables				BCFs vegetable groups				Soil properties						
	Crop group	Crop	Weighting factor crop	Moisture content %weight basis	BCF dry weight	BCF wet weight	Model (1) or geom. mean (0)	Participation yes(1)/ no (0)	BCFcrop group dry weight	BCFcrop group wet weight	Weighting factor group	Participation group yes(1)/ no (0)		soil content	pH	OM%	OC%	Clay%	soil type corr
													selected	3.00	7.00	2.00	1.18	5.00	
0	potatoes	potatoes	61.6	83.30	0.156	0.026	1	1	0.156	0.026	61.6	1	adapted:	1.70	7.00	2.00	1.18	6.00	0.73
						0.000													
1	root vegetbales	beetroot	1.3	87.30	0.194	0.025	1	1	0.260	0.032	5.09	1	adapted:	3.00	7.00	5.78	3.40	13.00	0.92
		carrots	3.4	87.80	0.288	0.035	1	1					adapted:	2.70	7.00	2.00	1.18	5.00	0.72
		celeriac	0.2	88.00															
		turnip	0.1	91.90															
		radish	0.05	94.80	0.002	0.000	1	1					adapted:	3.00	7.00	6.97	4.10	24.00	1.07
		winter carrot	0.04	87.80															
2	bulbous crops	onions	3.2	90.80					0.074	0.013	7.7	1							
		leek	4.5	83.00	0.074	0.013	1	1					adapted:	2.50	7.00	5.78	3.40	12.00	0.91
3	fruit vegetables	tomato	3.2	94.00	3.416	0.205	0	1	3.416	0.205	5.0	0	adapted:	2.40	7.00	4.93	2.90	12.00	0.88
		cucumber	0.6	96.10															
		melon	0.5	89.70															
		maize	0.7	76.00															
4	cabbages	cauliflower	2.5	92.30					0.347	0.036	7.6	1							
		brussels sprout	1.3	86.00															
		white cabbage	0.7	95.30	0.326	0.015	0	1					adapted:	2.40	7.00	4.93	2.90	12.00	0.88
		red cabbage	0.9	95.30	0.326	0.015	0	1					adapted:	0.40	7.00	5.78	3.40	12.00	0.88
		oxheart cabbag	0.2	95.30															
		curlt kale	1.6	84.50	0.368	0.057	1	1					adapted:	0.40	7.00	5.78	3.40	12.00	0.91
		brocolli	0.4	90.70															
5	leafy vegetables	lettuce	0.8	95.40	0.303	0.014	1	1	0.362	0.026	4.4	1	adapted:	3.00	7.00	4.93	2.90	12.00	0.88
		endive	0.9	93.80	0.286	0.018	1	1					adapted:	2.40	7.00	5.78	3.40	12.00	0.91
		spinach	1.8	91.60	0.427	0.036	1	1					adapted:	3.00	7.00	2.00	1.18	5.00	0.72
		chicory	0.9	95.30															
6	legumes	green beans	2.3	90.30					0.000	0.000	6.9	0							
		string/bush bee	0.6	90.30															
		broad/horse/fav	0.6	88.90															
		gaden peas	3.4	88.90															
7	beans	haricot bean	0.4	77.10	0.525	0.120	0	1	0.525	0.120	1.2	1	adapted:	3.00	7.00	2.55	1.50	12.00	0.81
		kidney beans	0.8	77.10	0.525	0.120	0	1					adapted:	3.00	7.00	2.55	1.50	12.00	0.81
8	stem and stalk vege	rhubarb	0.1	93.60					0.000	0.000	0.4	0							
		asparagus	0.3	92.30															
	All vegetables		99.89								99.89		average:	2.35	7.00	4.50	2.65	11.83	0.86
	not significant						BCFabove-ground vegetables: group 2 t/m 8		BCFroot crops: group 0 and 1			BCFpatatoes: group 0		BCFall vegetables, but potato group 1 t/m 8				BCF overall:	
	uncertain																		
						dry weight	0.260		0.164			0.156		0.260				0.187	
	Cadmium					wet weight	0.030		0.027			0.026		0.031				0.027	

5.4.3 Comparison

In Table 5.6 the BCF values for cadmium from this study, from Bockting and Van den Berg (1992) and several other sources have been listed. To this purpose the BCF for the same hypothetical soil as in section 5.4.2 is included (integrated BCF = 0.19).

Table 5.6: Comparison of BioConcentrationFactors (BCFs) from seven sources.

This study	***Bockting and Van den Berg, 1992***	***Van Driel et al., 1988***	***Baes at al., 1984***	***Bechtel Jacobs, 1998***
Consumption - averaged	Geometric mean	Consumption-averaged	Median	Median
0.19	0.37	0.26	0.55	0.51

From this table it can be concluded that the vegetable-group-consumption-weighted BCF from this study is relatively low. The Bockting and Van den Berg (1992) BCF and the Van Driel et al., 1988 BCFs are respectively 2 and 1.5 times higher than this BCF. The two other measured BCFs are almost a factor of three higher. The main reason for the relatively low BCF from this study is that in the plant - soil relations a high weighting is given to the BCF of potatoes due to the high contribution to total consumption (62%). And this BCF for potatoes is relatively low. Another explanation could be that in many uptake experiments conditions are created that stimulate uptake (low pH, plants that show a relatively high uptake) and, hence, results in higher BCFs. At the other side, the vegetable-group-consumption-weighted BCF from this study includes more data from slightly contaminated soils than data from higher cadmium levels. For example, the median soil content of cadmium in the RIVM plant –soil database is only 0.45 mg/kg_{dw} (10^{th} percentile is 0.12 and the 90^{th} percentile is 4.2). For these slightly contaminated soils, plant concentrations and, hence, BCFs are higher than BCF values from soils with higher cadmium levels.

5.5 Critical soil concentrations for cadmium

In several studies Critical soil concentrations for cadmium have been derived. When this value is not exceeded it is assumed that vegetables can be cultivated and consumed without unacceptable risks for humans. Although the conditions to which these Critical soil concentrations apply are slightly different, the variation in these Critical soil concentrations is remarkable. A value of 0.15 mg/kg_{dw} was proposed for the regulation of food crops in agricultural practice in Poland. For the Belgium Kempen region[4] a Critical soil concentration

[4]The Kempen region is a region on the border of the Netherlands and Belgium (Flanders), contaminated with cadmium and other metals due to long-time emissions from the zinc industry.

of 3 mg/kg_{dw} is used for soils with a pH(H_2O) of at least 7.5 and "sufficient" organic matter (Ondersteuningscel logo's, 2006). Under these conditions every vegetable could be grown on the site. For the Dutch Kempen region a Critical soil concentration of 2 mg/kg_{dw} is proposed for a soil with a pH(H_2O) of at least 6.5 and an organic matter content of at least 5% (Projectbureau Actief Bodembeheer de Kempen, 2007). Under these conditions it is justified to grow vegetables, except leafy vegetables. Below a Critical soil concentration of 0.5 mg/kg_{dw} no limitations on vegetbale type or soil conditions are applied.
Römkens and De Vries (2001) derived a table with Critical soil concentrations for cadmium as a function of pH, organic matter and clay contents. In this study Critical soil concentrations varied from 0.26 for a soil with pH = 4, organic matter and clay contents = 2% up to 19.9 for a soil with pH = 7, organic matter content = 10% and clay content = 20%. Dudka et al. (1996) propose a value as high as 30 mg/kg_{dw} as a safe concentration for soils with a neutral pH to grow crops, for Polish farm land. The reason for this high value is probably the fact that the metals originate from smelter flue-dust, with low availability.
In this section the possibility for the derivation of Critical soil concentrations for the present purpose, i.e. a lower tier assessment of human health risks in regard to cadmium uptake due to vegetable consumption, is investigated. The Critical soil concentrations can be calculated with the CSOIL exposure model (Van den Berg, 1991/1994/1995; Otte et al., 2001), as described in section 2.2. Moreover, because it concerns a lower tier assessment, the Critical soil concentrations must be conservative. Because Critical soil concentrations must be conservative the vegetable garden exposure scenario must be selected. The main characteristic of this scenario in relation to vegetable consumption is the assumption that 50% of the potatoes and 100% of the other vegetables are taken from the contaminated site. The Critical soil concentrations should be *generic* (applicable for the majority of cases) and must preferably be focused on the "average vegetable pattern" (see section 4.3).

To balance between generic application and practicability the derivation of Critical soil concentrations for several combinations of organic matter and clay contents has been investigated. The reason for this is that these are sustainable parameters, i.e. stable in time. It is, for example, not realistic to derive conservative Critical soil concentrations for vulnerable soils and to apply these to soils with a high organic matter and clay content. The pH, however, might change in a shorter time frame and was fixed at the rather conservative value of 5. Therefore, the derivation of Critical soil concentrations, on the basis of vegetable-group-rate-weighted BCFs, has been investigated for the following combinations of organic matter and clay contents: organic matter = 0, 2%, 5% and 10%, clay content = 0, 5%, 10% and 25%. However, the resulting differentiation in BCFs and hence in Critical soil concentrations proved to be limited. The reason for this is that the organic matter clay contents for the calculation of the plant – soil relations are limited within the 5^{th} and 95^{th} percentiles of the data in the RIVM plant – soil database. The average 5^{th} percentile for percentage organic matter = 4.1. This implies that no variation in the BCFs due to a difference in organic matter of 0% and 2% can be assessed for most vegetables. Besides, the average 5^{th} percentile for percentage clay = 10.3. This implies that no variation in the BCFs due to a difference in clay

content of 0%, 5% and 10% can be assessed for most vegetables. Because of this limitation no Critical soil concentrations for several combinations of organic matter and clay contents has been calculated in this study.

Instead of using this detailed differentiation in soil properties, the procedure that was implemented for the information brochure on the possibilities for private vegetable cultivation in the Kempen region was followed (Projectbureau Actief Bodembeheer de Kempen (2007). As a consequence the following contaminant classes are proposed for Tier 1:

- Cadmium concentration is lower than 0.5 mg/kg_{dw}: every crop can be cultivated and consumed without unacceptable risks to human health.
- Cadmium concentration is in between 0.5 and 2.0 mg/kg_{dw}: the pH(H_2O) of the soil must be at least 5.5 and the percentage organic matter at least 5%: every crop can be cultivated and consumed without unacceptable risks to human health.

Note that the 0.5 mg/kg_{dw} is in the same order of magnitude as the 95th percentile of the background concentrations for cadmium in Dutch soils (Lamé and Nieuwenhuis, 2006). This value is 0.6 mg/kg_{dw} for the Dutch standard soil, which implies a value in between 0.35 and 0.5 mg/kg_{dw} for more vulnerable soils. As a consequence, the criterion "every crop can be cultivated and consumed without unacceptable risks to human health" is not often met, even in relatively unpolluted areas (nature reserves and agricultural land).

The value of 2.0 mg/kg_{dw} is compatible with the Critical soil concentration that is used as basis for the derivation of Reference Value for good soil quality in the Netherlands, for the land-use "Vegetable garden" of 1.2 mg/kg_{dw}, for "a standard soil" (organic matter content = 5%; clay content = 25%) and a pH of 5 (Dirven - Van Breemen et al., 2007). In the derivation of this value a background exposure of 40% has been assumed. Without this background exposure the value would be around 2.0 mg/kg_{dw}.

5.6 Lead

In section 3.3.1 it was concluded that the exposure pathway vegetable consumption is also important in regard to lead. Although the contribution of exposure due to vegetable consumption to total exposure is "only" 30% for the standard residential scenario (for cadmium 93%), lead is important because it is a very frequently found contaminant. Moreover, human health risks (more specific risks to children; mainly due to soil ingestion, but second also to crop consumption) are often crucial for lead. For these reasons, this contaminant is discussed in this separate section.

For lead plant – soil relations were significant for a considerable amount of vegetables, i.e. 10 of the 13 investigated vegetables, from 6 different vegetable groups. Table 5.7 gives the

coefficients of the derived plant - soil relations for lead for 13 vegetables, according to Versluijs and Otte (2001).

Table 5.7: Regression coefficients of the plant – soil relations for lead for 13 vegetables according to Versluijs and Otte (2001).

Coefficients / Vegetable	a Const.	B Msoil	c pH	d %OC	e %Clay
Potato	-2.0	0.67	0.12	-0.02	-0.50
Red beet	0.5	0.75	-0.08	-0.64	-1.2
Carrot	-0.64	0.56	-0.04	-0.16	-0.03
Radish*	1.8	0.25	-0.27	-0.3	0.22
Onions*	-1.0	0.4	0.1	-	-
Leek	0.80	0.5	-0.12	-0.61	-0.57
Tomatoes*	-2	-1.2	0.4	-	-
Red an white cabbage	7.0	0.6	-0.30	-3.1	-5.0
Curly kale	2.00	0.29	-0.11	-0.62	-0.65
Lettuce	-0.60	0.90	-0.07	-0.34	-0.19
Endive	1.9	0.52	-0.17	-0.68	-.8
Spinach	-0.12	0.36	-0.03	0.25	-0.23
Beans	3.2	-0.10	-0.2	-5.3	-

* Plant – soil relations not significant

For radish, onions and tomatoes the relations were not significant. For these vegetables the geometric means of the BCFs have been used, including the soil type correction. The medium values for the clay and organic matter content in the RIVM plant - soil database for lead are, respectively, 16% and 7% (coincidently the same as for cadmium). As a consequence, the soil type correction factor for the BCF of lead, $STcf_{BCF}Pb$, becomes according to Eq. 5.4:

$$STcf_{BCF}Pb = \frac{50 + \%Clay + \%OM}{73} \qquad \text{Eq. 5.8}$$

The vegetable-consumption-group-rate-weighted BCF for lead is calculated according to Eq. 5.6. Table 5.8 shows an example for the calculation of the vegetable-consumption-group-rate-weighted BCF for lead for the same soil as described in section 5.4.2, for the illustration of the calculation of the vegetable-consumption-group-rate-weighted BCF for cadmium. However, a more common soil concentration for with lead contaminated soils is chosen, i.e. 100 mg/kg_{dw}. The explanation of the table is the same as given in section 5.4.2. In the final rows the average vegetable-group-consumption-rate-weighted-BCFs for lead are given for aboveground vegetables, root crops and potatoes. Moreover, the average vegetable-group-

consumption-rate-weighted BCF for lead is given for all vegetables and for all vegetables, exclusive potatoes.

Table 5.8: Table including all information for the calculation of average vegetable-group-consumption-rate-weighted-BCFs for lead, for a hypothetical soil (pH = 7, organic matter content = 2% (organic carbon content = 1.18%), clay content = 5% and the total lead concentration in soil = 100 mg/kg$_{dw}$) (page 58).

	Crop information				BCFs vegetables				BCFs vegetable groups				Soil properties						
	Crop group	Crop	Weighting factor crop	Moisture content %weight basis	BCF dry weight	BCF wet weight	Model (1) or geom. mean (0)	Participation yes(1)/ no (0)	BCFcrop group dry weight	BCFcrop group wet weight	Weighting factor group	Participation group yes(1)/ no (0)		soil content	pH	OM%	OC%	Clay%	soil type corr
													selected	100.00	7.00	2.00	1.18	5.00	
0	potatoes	potatoes	61.6	83.30	0.006	0.001	1	1	0.006	0.001	61.6	1	*adapted:*	100.00	7.00	2.00	1.18	6.00	0.85
						0.000													
1	root vegetbales	beetroot	1.3	87.30	0.009	0.001	1	1	0.015	0.002	5.09	1	*adapted:*	100.00	7.00	3.23	1.90	12.00	0.96
		carrots	3.4	87.80	0.015	0.002	1	1					*adapted:*	100.00	7.00	2.00	1.18	5.00	0.84
		celeriac	0.2	88.00															
		turnip	0.1	91.90															
		radish	0.05	94.80	0.167	0.009	0	1					*adapted:*	100.00	7.00	2.00	1.18	5.00	0.84
		winter carrot	0.04	87.80															
2	bulbous crops	onions	3.2	90.80	0.009	0.001	0	1	0.010	0.001	7.7	1							0.96
		leek	4.5	83.00	0.010	0.002	1	1					*adapted:*	100.00	7.00	5.78	3.40	12.00	1.00
3	fruit vegetables	tomato	3.2	94.00	0.010	0.001	0	1	0.010	0.001	5.0	1	*adapted:*	100.00	7.00	4.93	2.90	12.00	0.98
		cucumber	0.6	96.10															
		melon	0.5	89.70															
		maize	0.7	76.00															
4	cabbages	cauliflower	2.5	92.30					0.034	0.005	7.6	1							
		brussels sprout	1.3	86.00															
		white cabbage	0.7	95.30	0.007	0.000	1	1					*adapted:*	100.00	7.00	3.23	1.90	12.00	0.96
		red cabbage	0.9	95.30	0.007	0.000	1	1					*adapted:*	100.00	7.00	5.78	3.40	12.00	0.96
		oxheart cabbag	0.2	95.30															
		curlt kale	1.6	84.50	0.060	0.009	1	1					*adapted:*	100.00	7.00	5.78	3.40	12.00	1.00
		brocolli	0.4	90.70															
5	leafy vegetables	lettuce	0.8	95.40	0.030	0.001	1	1	0.026	0.002	4.4	1	*adapted:*	100.00	7.00	3.40	2.00	5.00	0.86
		endive	0.9	93.80	0.035	0.002	1	1					*adapted:*	100.00	7.00	5.44	3.20	12.00	0.99
		spinach	1.8	91.60	0.020	0.002	1	1					*adapted:*	80.00	7.00	2.00	1.18	5.00	0.84
		chicory	0.9	95.30															
6	legumes	green beans	2.3	90.30					0.000	0.000	6.9	0							
		string/bush bee	0.6	90.30															
		broad/horse/fav	0.6	88.90															
		gaden peas	3.4	88.90															
7	beans	haricot bean	0.4	77.10	0.013	0.003	1	1	0.013	0.003	1.2	1	*adapted:*	100.00	7.00	3.23	1.90	12.00	0.96
		kidney beans	0.8	77.10	0.013	0.003	1	1					*adapted:*	100.00	7.00	3.23	1.90	12.00	0.96
8	stem and stalk vege	rhubarb	0.1	93.60					0.000	0.000	0.4	0							
		asparagus	0.3	92.30															
	All vegetables		99.89								99.89		*average:*	98.33	7.00	3.90	2.29	9.67	0.93
	not significant						BCFabove-ground vegetables: group 2 t/m 8		BCFroot crops: group 0 and 1		BCFpatatoes: group 0		BCFall vegetables, but potatoes group 1 t/m 8					**BCF overall:**	
	uncertain																		
						dry weight	0.020		0.007		0.006		0.019					**0.010**	
	Lead					wet weight	0.002		0.001		0.001		0.002					**0.001**	

5.7 Arsenic

Arsenic concerns the third important metal in regard to human health effects due to vegetable consumption. However, for arsenic at least two problems arise:

- The phytoavailable fraction is strongly related to other soil properties then organic matter and clay contents. The major soil properties concern (hydr)oxide and phosphate contents and these soil properties are not measured in standard soil quality risk assessment.
- For arsenic there hardly is a relation between the concentration in vegetables and the concentration in soil.

Versluijs and Otte (2001) only found one significant plant – soil relation for arsenic (on the basis of concentration in soil and organic matter and clay contents) out of the three investigated vegetables, i.e. for carrots. The relations for potatoes and spinach were not significant. This mean that the present calculation of the vegetable-consumption-group-rate-weighted BCF for arsenic, according to Eq. 5.6, is mainly based on geometric means of the BCFs for many vegetables, including the soil type correction. The medium values for the clay and organic matter content in the RIVM plant - soil database for arsenic are, respectively, 11% and 2.7%. As a consequence, the soil type correction factor for the BCF of arsenic, $STcf_{BCF}\,As$, becomes according to Eq. 5.4:

$$STcf_{BCF}\,As = \frac{15 + (0.4\,x\,\%Clay + 0.4\,x\,\%OM)}{21} \qquad \text{Eq. 5.8}$$

Cornelis and Swartjes (in progress) concluded, after a telephone conference with plant uptake experts and the investigation of several measured data of arsenic in vegetables and soils, that the use of a fixed value for the concentration of arsenic in vegetables, that is independent of the concentration of arsenic in soil and the soil properties, is the best available option. This would imply that the use of a BCF overestimates the arsenic concentration in vegetables, at higher soil concentrations.
It is strongly recommended to investigate the relation between arsenic concentration in vegetables and soils in more detail, in the future. This investigation must focus on the (lack of) influence of the arsenic concentration in soils on the arsenic concentration in vegetables and the possibilities for use of less common soil properties, like the (hydr)oxide and phosphate contents.

5.8 Other metals

Unfortunately, for the other metals the statistical significance for the majority of the relations in Versluijs and Otte (2001) was insufficient: for arsenic 1 out of the 3 of the investigated vegetables; for cupper 2 out of the 6 of the investigated vegetables, for mercury none of the 3 investigated vegetables; for nickel 2 out of the 6 of the investigated vegetables.

For zinc, only 1 out of 6 vegetables resulted in a significant relation. However, zinc is an essential metal for human beings and is not very toxic. The concentrations in plants that match exceedance of the Reference dose, when the standard exposure scenario which forms the basis of the Dutch Intervention Values is used, must be higher than 8300 mg/kg_{dw}. The corresponding Critical soil concentration is 46,000 mg/kg_{dw}. Lijzen et al. (2002) show that at a soil concentration of 97 mg/kg_{dw} (and 215 mg/kg_{dw} as 97.5% upper confidence limit) a phytoxicological level of 50% affected fraction of plants prevails. From this it can be concluded that plants will not survive at concentration levels in soils needed to have a significant exposure, i.e. 46,000 mg/kg_{dw}. Also Ruttens (2006b) states that almost no risk for toxicity of zinc, but also copper, exists, because the accumulation of these elements is lethal to the plants well before concentrations become harmful to man. Therefore, human health risks for zinc due to vegetable consumption are precluded.
It is recommended to investigate the possible preclusion of human health risks for copper due to vegetable consumption.

For the other metals the only option is to combine the BCFs from the significant relations with geometric mean values for the BCFs, in analogy with the procedure schematized in Figure 5.5.

For metals that are not included in the RIVM plant – soil database a BCF can be derived from the distribution coefficient soil -water, according to Baes et al. (1984) as follows:

$$\ln BCF_{generic} = 2.67 - 1.12 \; x \; \ln Kd \qquad [-] \qquad \text{Eq. 5.9}$$

in which:

$BCF_{generic}$	generic BCF	[(mg/$kg_{wet\ weigt\text{-}plant}$) / (mg/$kg_{dw\text{-}soil}$)]
K_d	distribution coefficient soil-water	[kg/kg_{dw}: kg/m^3 water]

Again it is proposed to divide the BCF by the soil type correction factor to account for actual soil properties:

$$BCF_{site\text{-}specific} = BCF_{generic} / STcf_{BCF} \quad \text{Eq. 5.10}$$

in which:

$BCF_{site\text{-}specific}$	site-specific BCF	[(mg/kg$_{wet\ weigt\text{-}plant}$) / (mg/kg$_{dw\text{-}soil}$)]
$STcf_{BCF}$	soil type correction factor for the BCF	[-]

The reliability of BCFs derived from distribution coefficients soil –water is limited.

Since, many data on metal concentrations in soils and vegetables were published the last few years, it is highly recommended to extent the plant – soil database, specifically for arsenic, mercury, nickel, barium and molybdenum, with the purpose to derive more plant – soil relations for these metals, in the future.

6. Accumulation of other inorganic contaminants in vegetables

6.1 Introduction

The category of other inorganic contaminants includes a wide scope of contaminants. One of the most important contaminants is this category is cyanide. Cyanide is often found at contaminated sites and the calculation of the accumulated concentration in plants is not very accurate. Furthermore, several inorganic forms of the anions bromide, chlorides, fluorine and sulphates and nutrients like nitrate and phosphorous are found in soils. For several reasons these contaminants have not been included in the Dutch Soil Protection Act (Swartjes, 1997), which means that at this moment no formal Intervention Values exist for these elements (Ministry of VROM, 2000). Nevertheless, the anions of elements bromide (Br) and fluorine (F) are incorporated in this study. The reason for this is that the *uptake behaviour* of these contaminants might be relevant for other contaminants in the group of other inorganic contaminants.

6.2 Present procedure

In the present procedure a fixed BCF (BioConcentrationFactor) is inferred when no detailed information on bioconcentration of inorganic contaminants is available (Van den Berg, 1991/1994/1995). Assuming that plants consist of 80% of water and that the concentration of contaminants within the plant is equal to the soil pore water concentration, than the bioconcentration based on fresh weight will be 0.8. Based on dry weight the BCF will be 4.

Since not enough information about uptake of inorganic contaminants is available to validate or refine the above assumptions, an exploratory study was performed to gain insight in the uptake of two inorganic contaminants: fluorine and bromide. For both contaminants a Target Value has been derived. For each contaminant plant - soil relations were looked up in literature and BCF values will be assessed where possible. With these BCF values the validity of the present BCF value of 4 will be evaluated. If there is reason to believe that the value of 4 leads to underestimation of risks to human health, then reconsidering of the value is needed. When the value of 4 leads to overestimation of risks, then the value can be regarded as conservative.

6.3 Cyanide

Cyanide, or CN, occurs in three major forms in the environment, as free CN, as complex CN and as thiocyianate (SCN). Köster (2001) accomplished an extended study of historical cyanide contamination, mainly gas manufacturing plants. From his work the following summary is derived, completed with work of other authors.
Free CN has a limited lifetime in soils, either at low or in high concentrations. At high concentrations leaching will be the predominant way of CN removal if it is not included into metal hexacyanide complexes. Iron hexacyanide complexes are the major source of free CN, although the solubility of such complexes is low and determined by kinetics rather than by equilibrium (Meeussen, 1992). However, the dissociation of ferri ferrohexycyanides decreases with decreasing pH (Kjeldsen, 1999). The data on plant uptake mechanisms of free CN are limited. Usually a reference is made to Wallace et al. (1971) and Wallace et al. (1981) and Boening and Chew (1999)), who showed that CN is usually taken up as a chelate linked to a metal.
The cyanogenic plants, like the *Brassica* species and cassava, produce naturally, endogenously non-free CN. Free CN is generally produced in all plants. This free CN is rapidly conjugated by enzymes and finally metabolized to particular amino acids within the plant. This conversion of free CN appears to be common for plants and this mechanism is responsible for keeping the free cyanide levels at a non-toxic level. Köster (2001) found no reports on toxic effects for humans and domesticated animals of free CN (and SCN) in food from cultivated non-cyanogenic plants. He concluded that it will probably not be induced by CN contamination, as long as the exogenous CN levels are below phytotoxic levels. Larsen et al. (2004) and Yu et al. (2005) found also that free CN levels are reduced by conversion in plants and they both suggest to use plants as a bioreactor for cyanide removal of gold mining wastewaters or other sources. It can be assumed that if CN levels exceed the phytotoxic level, the unhealthy and hampered plants will not be used for food consumption.
Köster concluded that CN uptake below phytotoxic levels results in the conversion of CN to amino acids, that free CN exposure through plant consumption can be regarded as negligible. No references to Critical vegetable concentrations, which can be related to a TDI, were found in this study and, considering the conversion, will probably be difficult to derive. Therefore the plant uptake route will not be considered in this study.
Plant uptake of metals is enhanced using CN as chelate. Anderson et al. (2005) showed that during a demonstration of phytoextraction of gold using CN as chelate that phytotoxic effects which finally destroyed the plants was caused by an excess uptake of copper instead of phytotoxic levels of cyanide. However, this side effect of CN uptake is not considered in this study.

A result, human health risks for free CN due to vegetable consumption are precluded in the protocol to assess human health risks due to vegetable consumption.

6.4 Fluorine

Within the field of medical geochemistry fluorine is well known as a natural hazard to human health in areas with elevated natural fluorine concentrations, mostly related the a certain geological setting (Plant et al., 2001; Lin et al., 2004). Anthropogenic sources of fluorine are the aluminium melting industry, burning fluorine rich coal and mining of fluorine bearing ore deposits.
Several studies on fluorine uptake by plants have been published. Singh et al. (1995) studied plant uptake in irrigation water by ladyfinger (*Abelmorchus esculentus*). They found that fluorine uptake in artificial quartz sand was higher than fluorine uptake in cultural soil. In both cases the fluorine uptake was more ore less linearly related with the fluorine concentration in the irrigation solution. The accumulation in the plants grown on the quartz sand did not show any regular pattern, while the plants grown on the cultural soil had decreasing concentrations in respectively the roots, leaves, fruit and shoot. Sing et al. (1995) concluded that up to 120 µg fluorine/litre irrigation water did not harm the plants. The ingestion of fluorine by humans from plants irrigated with 10 µg/litre would be 0.20 mg per 100 g ladyfinger.
Arnessen (1997) showed that fluorine concentrations nearby a contamination source in white clover (*Trifolium repens*) and ryegrass (*Lolium multiflorium*) were highly correlated with the concentration H_2O and 0.01M $CaCl_2$ extractable fluorine in soils from a pot experiment. The author added natriumfluorine (NaF) to uncontaminated sand and loamy sand and assessed the fluorine concentration in the plants. Fluorine concentrations larger than 200 mg/kg_{dw} soil resulted in phytotoxic effects for the clover and concentrations larger than 400 mg/kg_{dw} showed toxic effects in all the plants. The correlation between H_20 extractable fluorine and plant tissue for NaF values in soil below 200 mg/kg_{dw} showed strong linear (curvilinear for ryegrass on sand) relations with the fluorine concentration in the shoot. Fluorine concentrations in clover exceeded 30 mg/kg_{dw}, even in moderately contaminated soils. The values from the pot experiment did not compare with the results from experiments with common bent (*Agrostis capillaris*) grown in 12 Cambic Arenosols from areas around aluminium smelters. Concentrations in the common bent did not show a relation between extractable fluorine and fluorine concentrations in plant tissue.
In solution culture experiments Stevens et al. (1998) exposed tomatoes (*Lycopersicon esculentum*) and oats (*Avena sativa*) to fluorine. They showed that growth of tomatoes was limited at fluorine activities higher than 1473 µM in solution, while oats showed no effect at activities up to 5130 µM. The conclusion of this study was that at low activity of fluorine in solution (<1684 µM), a positive linear relation existed with fluorine concentrations in shoots. But when the fluorine activity reached a certain threshold (larger than 1476 µM) the fluorine concentration increased rapidly towards an upper asymptote, resulting in an S-shaped curve.
Fung et al. (1999) studied the relation between soil fluorine and fluorine concentration in tea. Their data show that the fluorine concentration in tea plants (*Camellia sinensis*) is linearly related to the fluorine concentration in soil. In general the authors mentioned above note that

the, mostly linear, relation between soil fluorine concentrations and fluorine activity and fluorine uptake is dependent of phytotoxicity, type of plant, type of soil, type of ionic fluorine species and soil pH. Therefore a common relations between extractable soil fluorine concentration and plant uptake is difficult to derive. However, based on the data of above mentioned authors, accumulation of fluorine in plant tissue seems to be limited. An exception is white clover (Arnesen, 1997). Based on the present BCF factor of 4, it can be concluded that fluorine accumulation in general will not exceed this value.

6.5 Bromide

Major anthropogenic sources of bromide in the environment are the use as a tracer for hydrological experiments, fuel scavenger, waste product of potassium mining, and as a residue for bromomethane, a soil fumigant to control nematodes and other pests. Natural bromide origins from the sea and sea spray, resulting in higher natural bromide concentrations nearby marine environments.
Flury and Papritz (1993) compiled information about the occurrence of bromide in the environment and summarized ecotoxicological data. They outlined the low acute and chronic toxicity of Br. Regarding plant uptake they reported that the percentage of relative plant uptake, calculated as the ratio of uptake and bromide application, had a mean around 0.3, ranging from 0.09 to >0.5.
The relation between bromide concentration in the pore water and plant uptake is reported to be linear (Magarian et al., 1998) or to follow Michealis-Menten kinetics (Xu et al., 2004). The latter described the uptake in wetland flora, while the former author looked at alfalfa herbage.
In the FAO evaluations on the bromide ion (FAO/WHO, 1984) a study is summarized on bromide concentrations in lettuce after application of bromomethane. Soil concentrations of the treated soils were a factor of 1 to 4 higher than untreated soils. Concentrations in lettuce were a factor 0.4 to 6.2 (with a mean of 2.3) higher than the soil concentration. The soil was not treated with water after the bromomethane application, as is common in agricultural practice.

Based on the data of bromide there is no reason to assume that the current method, using a BCF factor of 4, will lead to underestimating of the risk. However, there are not enough data for a more specific approach for estimating the risk. Based on the low toxicity of bromide and the low uptake and accumulation found in the limited data above it might be assumed that the current method may lead to overestimation of plant uptake.

6.6 Résumé

The discussed example of bromide and fluorine uptake showed that the relation between soil concentrations, uptake and plant concentration is plant specific. The studied literature does not indicate a general relation, nor general BCF values. However, the data suggest that uptake of these two anions is limited.
Considering the choice to set the concentration in plants equal to the concentration in the pore water, the validity of this assumption has not been shown nor has it been falsified. No better alternative could be derived from the studied literature. Regarding that the current approach is conservative, easy to use, and probably does not lead to overestimation of risks, there is no reason to disregard or modify the approach.

7. Accumulation of organic contaminants in vegetables

7.1 Introduction

Several authors evaluated the concepts for the uptake of organic contaminants by plants (Jager and Hamers, 1997; Jager, 1998; Schwartz et al., 2000; Trapp and Schwartz, 2000; Rikken et al., 2001; Rikken and Lijzen, 2004). Fryer and Collins (2003) compared nine models for predicting the uptake, translocation, and elimination of organic contaminants by plants, ranging from simple screening tools to complex mechanistical models. They concluded that the selection of the appropriate model depends on the requirement of the assessment, the nature of the environmental media and the duration of the source term.

A description of the present method for the calculation of the accumulated concentration of organic contaminants, as incorporated in the CSOIL exposure model (CSOIL 2000), is given in this chapter. However, also some improvements are discussed. Besides, an evaluation of the model is carried out by comparing the model estimations with limited literature data.

The processes and parameterisation depend on the type of plant. Models have been developed describing specific processes for each type of plant (Figure 7.1). A distinction can be made in root vegetables (e.g. carrot), leafy vegetables (e.g. lettuce), tuberous vegetables (e.g. potato) and fruits (e.g. apple), according to Kulhánek et al. (2005).

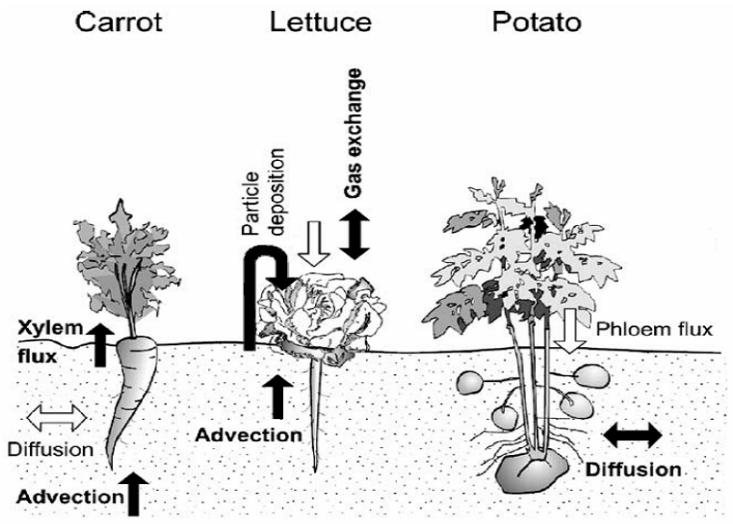

Figure 7.1: Principles of the plant models used for the uptake of organic contaminants (Kulhánek et al. 2005).

The models for root vegetables and leafy vegetables are described further in the next section of this report. A tuber is a storage organ of a plant that is not a root, but is morphologically a modified stem (Kulhánek et al., 2005). Tubers are connected to the phloem with a flow from the leaves to all other parts of the plants. Unlike roots, an advective transport of soil contaminants into the potato is unlikely. For the time being the root model is also used for tuberous vegetables, because a validated tuber or potato model is currently not available. Trapp et al. (2003) developed a fruit tree model for the uptake of neutral organic chemicals from contaminated soils into fruits. This model considers an influx into fruit via phloem and xylem from the woody stem. Since uptake in fruits in relatively limited, human health risks due to fruit consumption are not considered in this study.

7.2 Calculation procedure

7.2.1 Model description

A description of the method for the uptake of organic contaminants in plants as implemented in the CSOIL 2000 is given in this section. This method is mainly based on Trapp and Matthies (1995) and includes also the contribution of deposition or rain splash on plants (Rikken et al., 2001). The uptake routes and fate processes of the plant model are schematized in Figure 7.2. More details on the boundary conditions and restrictions of the Trapp and Matthies plant model can be found in Rikken et al. (2001).

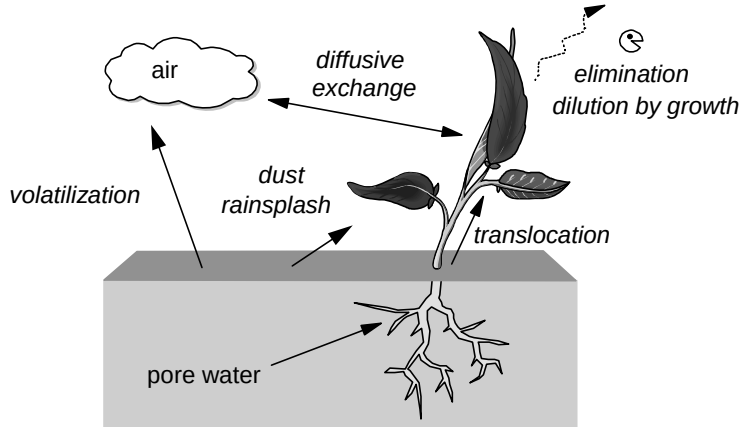

Figure 7.2: Uptake routes and fate processes accounted for in the plant model.

To determine the concentration in root vegetables the method of Trapp (2002) is proposed, which is currently not implemented in CSOIL 2000. Roots that are consumed (e.g. carrots) are considered to be thick roots and are specified in this report as root vegetables. Fine roots are considered not to be consumed (e.g. roots of lettuce). In CSOIL 2000 a difference has been made between uptake in subterranean plant parts (roots) and uptake in above-ground plant parts (stem or leaf).

Root vegetables (Trapp)
The model of Trapp (2002) is a dynamic flux model for the uptake of neutral lipophilic contaminants into root vegetables. This model is developed to estimate the concentration in thick roots were, unlike fine roots, a chemical equilibrium can hardly be reached. The contaminant input from soil into the root is with the transpiration water and the sink is the sum of the exponential growth rate, loss with the transpiration stream and metabolism. The sink term is described by one parameter (k).

Leaf vegetables (Trapp and Matthies)
In the model of Trapp and Matthies (1995) the uptake and translocation of organic contaminants via pore water and the uptake/elimination via air is included. Deposition from contaminants by rain and particles from air can also be important. However, this route is not taken into account in this study, because these deposition sources are not related to local soil contamination.

Uptake routes and fate processes
Pore water → root → leaf.
Uptake from soil is a passive process for most organic contaminants, directed by the transpiration stream in the xylem of the plant. Water-soluble contaminants that pass the membranes in the root are transported to the leaf with the transpiration stream. In the leaf water will evaporate and contaminants can accumulate. For most (neutral) contaminants the phloem flow back to the root is negligible (Bromilow and Chamberlain, 1995).

Soil → air (gas phase) → plant.
This route is often neglected but can play an important role in heavily contaminated soils. For example PCDD/Fs are poorly transported with the transpiration stream but can be taken up via this route in substantial amounts (Trapp and Matthies, 1994). Because dilution by wind is an important factor, air concentrations stay low. Nevertheless, for closed vegetation close to the ground (e.g. grasses, lettuce) this route can have a relevant contribution. However, the route soil to air to plant is not included in this study.

Soil → air (particle-bound) → plant.
Soil and dust parts can be deposited on the plant by wind or rain (also called rain splash or soil resuspension). The contribution is difficult to estimate and depends on many factors (e.g. the geometry of the plant, soil type). Nevertheless this route can be important in case of contaminated soils. Soil particles that are directly deposited on the leaves can expose plants. It is difficult to estimate to what extent the internal concentration is influenced.

Elimination processes.
Elimination of contaminants from the leaf can be important for volatile contaminants. Photo-degradation and metabolism of organic contaminants can occur in the plant, although

prediction of these processes using physical-chemical properties of the contaminant is difficult. Also dilution by growth will influence the concentration in the plant.

Deposition on leaves (soil resuspension / rain splash)

Root uptake and dry gaseous deposition (concentration in air) are taken into account in the concept of Trapp and Matthies. Other exposure routes of plants are dry particle deposition, wet particle deposition and soil particle resuspension (rain splash) (Smith and Jones, 2000; McLachlan, 1999). Dry and wet particle deposition, originating from air pollution, must be excluded from the risk assessment of human vegetable consumption at (potentially) contaminated soils. Estimation of the soil particle resuspension can be an important source of contaminants in plants for some contaminants, especially for contaminants with a high octanol-air partition coefficient (log K_{oa})[5], a low $K_{air\text{-}water}$ and a high log K_{ow} (McLachlan, 1999). The contribution of this route is set at a provisional value of 1% dry soil per dry plant, based on an evaluation of Rikken et al. (2001).

7.2.2 Model equations

Roots

The partitioning between water and plant tissue is based on sorption to plant fats. Trapp and Matthies (1995) give the following relation for plant tissue.

$$K_{plant-water} = F_{water} + F_{fat} \cdot K_{ow}^{\ b} \qquad \text{Eq. 7.1}$$

in which:

$K_{plant\text{-}water}$:	partition coefficient between plant and water	[kg/m^3 plant: kg /m^3 water]
F_{water} :	volume fraction of water	[m^3 water/m^3 plant]
F_{fat}:	volume fraction of plant fat	[m^3 fat/m^3 plant]
K_{ow}:	octanol water partition coefficient	[m^3 water/m^3 octanol]
b:	correction exponent for differences between plant fat and octanol	[-]

A separate partition coefficient between plant and water is calculated for root vegetables and leafy vegetables, based on the root or leaf parameters in Table 7.1.

For the concentration in fine roots (C_{root}) a relation can be given with the concentration in pore water:

[5] There is a relation between the octanol-air partition coefficient, logK_{oa} (=logK_{ow}/log $K_{air\text{-}water}$) and the $K_{leaf\text{-}air}$ (McLachlan, 1999) and therefore the concentration in the plant strongly relates to the K_{oa}. In the model of Trapp and Matthies the $K_{leaf\text{-}air}$ is estimated from K_{ow} and $K_{air\text{-}water}$ (dimensionless Henry coefficient).

$$C_{root} = \frac{K_{plant-water} \cdot C_{water}}{RHO_{plant}} \quad [\text{kg/kg}_{ww}] \qquad \text{Eq. 7.2}$$

in which:

$K_{plant\text{-}water}$:	partition coefficient between root and water see also calculation of $K_{plant\text{-}water}$ via Eq. 7.1	[kg/m^3 plant to kg/m^3 water]
RHO_{plant} :	plant density	[kg/m^3]
C_{water} :	concentration in the pore water	[kg/m^3]

Above-ground plant parts

Mass transport to above-ground plant tissue

The mass transport in the xylem (N_{xy}) from the pore water is described as:

$$N_{xy} = C_{water} \cdot TSCF \cdot Q \quad [\text{kg/d}] \qquad \text{Eq. 7.3}$$

in which:

C_{water}:	concentration in the pore water (calculated with C_{soil}/K_d)	[kg/m^3]
TSCF:	Transpiration Stream Concentration Factor	[-]
Q:	transpiration stream	[m^3/d]

The TSCF is defined as the concentration ratio between xylem sap and external solution (pore water) and can be calculated in two ways:

$$\text{TSCF} = 0.784 \cdot \exp\left[\frac{-(\log K_{ow} - 1.78)^2}{2.44}\right] \qquad \text{Eq. 7.4}$$

with range for log K_{ow}: -0.5 - 4.5 (Briggs et al., 1982)

$$\text{TSCF} = 0.7 \cdot \exp\left[\frac{-(\log K_{ow} - 3.07)^2}{2.78}\right] \qquad \text{Eq. 7.5}$$

with range for log K_{ow}: 0.96 - 5.3 (Hsu et al., 1990)

Exchange with air

The partition coefficient between leaf and air determines the exchange with air:

$$K_{leaf-air} = \frac{K_{plant-water}}{K_{air-water}} + F_{air} \quad [m^3/m^3] \qquad \text{Eq. 7.6}$$

where:

$$K_{air-water} = \frac{Vp \cdot MOLW}{SOL \cdot R \cdot T} \quad [m^3/m^3] \qquad \text{Eq. 7.7}$$

in which:

$K_{plant-water}$:	partition coefficient plant vs. water	[-]
$K_{air-water}$:	partition coefficient between air and water (=dimensionless Henry)	[m^3/m^3]
Vp:	vapour pressure	[Pa]
MOLW:	molecular weight	[kg/mol]
SOL:	water solubility	[kg/m^3]
R:	gas constant (=8.314 Pa.m3/mol.K)	[$Pa.m^3/mol.K$]
T:	environmental temperature	[K]
F_{air} :	volume fraction air in the plant	[m^3/m^3]

F_{air} can be left out, because it hardly makes a difference in calculations (pers. comm. Trapp, 2000).

The net flux between leaf and air, N_A, is:

$$N_A = A \cdot g \cdot \left(\frac{C_{air} - C_{leaf}}{K_{leaf-air}} \right) \quad [kg/s] \qquad \text{Eq. 7.8}$$

in which:

A:	leaf surface	[m^2]
g:	conductance	[m/s]
C_{air}:	concentration in air	[kg/m^3]
C_{leaf}:	concentration in leaves	[kg/m^3]
$K_{leaf-air}$:	partition coefficient between leaf and air	[m^3/m^3]

Mass balance

The actual model for calculating the concentration in leafs can be described with a simple differential equation: $d(C_L V_{leaf})/dt$. The source term (beta) and sink term (alpha) are:

Source: flux from soil via the xylem to the stems, N_{xy}
Source/sink: flux from/to air, N_A
Sink: photo-degradation, k_{photo}
Sink: metabolism, k_{metab}

$$alpha = \frac{A \cdot g}{K_{leaf-air} \cdot V_{leaf}} + k_{\text{elim}} + k_{growth} \quad \text{(sink)} \qquad \text{Eq. 7.9}$$

$$\text{beta} = C_{water}.TSCF \cdot \left(Q_{transp}/V_{leaf}\right) + C_{air} \cdot g \cdot \left(A/V_{leaf}\right) \quad \text{(source)} \qquad \text{Eq. 7.10}$$

in which:

g:	conductance	[m/d]
A:	leaf surface area	[m^2]
V_{leaf}:	leaf volume	[m^3]
k_{elim}:	rate constant for elimination (metabolism, photo-degradation)	[d^{-1}]
K_{growth}:	rate constant for dilution by growth	[d^{-1}]
C_{air}:	concentration in air	[kg/m^3]

The steady-state concentration in leaf is calculated as the source term divided by the sink term:

$$C_{leaf} = \frac{beta}{alpha \cdot RHO_{plant}} \quad [\text{kg/kg}_{ww}] \qquad \text{Eq. 7.11}$$

Deposition on leafs (soil resuspension / rain splash)

The contribution of this route is set at a provisional value of 1% dry soil per dry plant, based on an evaluation of Rikken et al. (2001). The dry weight fraction of leafy vegetables (F_{crops}) is used to convert the concentration in leafs from dry weight to fresh weight.

$$C_{leaf\ dep} = DP_{const} \cdot C_{soil} \cdot F_{crops} \quad [\text{kg/kg}_{ww}] \qquad \text{Eq. 7.12}$$

in which:

DP_{const}:	deposition constant (=0.01)	[-]
C_{soil}:	concentration in soil	[kg/kg_{dw}]
F_{crops}:	dry weight fraction of leafy vegetables (=0.098)	[-]

Concentration in leafs
The resulting concentration in leafs is calculated as follows:

$$C_{leaf\ total} = C_{leaf} + C_{leaf\ dep} \quad [\mathrm{kg/kg_{ww}}] \qquad \text{Eq. 7.13}$$

7.3 Evaluation of important plant parameters and model concepts

7.3.1 Plant parameters

To implement new model concepts into the plant module, input parameters have to be evaluated and selected. The parameters in Table 7.1 have to be known to calculate the concentration in root or leafy vegetables, based on the total concentration in soil and in groundwater.

The plant parameters are mainly based on the reports of Jager and Hamers (1997), Rikken et al. (2001) and Rikken and Lijzen (2004), in which plant uptake models were evaluated. These evaluations showed that some parameters are much more critical than others, also depending on the contaminant. The resulting parameters are presented in Table 7.1. Two parameters, i.e. the volume fraction water (F_{water}) in roots and the empirical factor b, were further evaluated, because there were inconsistencies between the different references (see Table 7.1.

*Table 7.1: Plant parameters according to various model evaluations and the proposed values for a default scenario (selected parameters **in bold**).*

	Symbol	Unit	Trapp and Matthies (1995)	Jager and Hamers (1997)	Rikken et al. (2001)	CSOIL 2000	Proposed
Plant parameters							
Transpiration stream	Q	$m^3.d^{-1}$	**0.001**	-	0.001	0.001	0.001
Rate constant growth dilution	k_{growth}	d^{-1}	**0.035**	-	0.035	0.035	0.035
Rate constant for elimination	k_{elim}	d^{-1}	**0**	-	0	0	0
Deposition constant (soil resuspension)	DP_{const}	-	-	-	**0.01**	0.01	0.01
Leaf parameters							
Volume fraction fat	F_{fat}	$m^3.m^{-3}$	-	**0.01**	0.01	0.01	0.01
Volume fraction water	F_{water}	$m^3.m^{-3}$	-	**0.65**	0.65	0.65	0.65
Volume fraction air	F_{air}	$m^3.m^{-3}$	-	**0.30**	0.30	0.30	0.30
Bulk density tissue (wwt)	RHO_{leaf}	$kg.m^{-3}$	-	**800**	800	800	800
Leaf surface area	A	m^2	**5**	-	5	5	5
Leaf volume	V	m^3	**0.002**	-	0.002	0.002	0.002
Conductance	G	$m.d^{-1}$	86.4	-	**80**	80	80
Empirical factor [1)]	B	-	**0.95**	-	0.95	0.95	0.95
Root parameters							
Volume fraction fat	F_{fat}	$m^3.m^{-3}$	-	**0.005**	0.005	0.005	0.005
Volume fraction water	F_{water}	$m^3.m^{-3}$	-	0.93	0.93	0.833[2)]	**0.89**[3)]
Bulk density tissue (wwt)	RHO_{root}	$kg.m^{-3}$	-	**1000**	1000	1000	1000
Empirical factor [1)]	B	-	-	-	**0.77**	0.8[4)]	0.77
Sum of exponential growth rate and loss by upward flux and metabolism	K	d^{-1}	-	-	-	-	**0.01**[5)]

[1)] Empirical correction factor for differences between plant lipids and octanol.
[2)] Based on dry weight fraction for potatoes (0.167).
[3)] Water content of carrot (*Trapp, 2002*).
[4)] The value from *Rikken et al. (2001)* was adopted; however 0.77 was rounded off to one decimal.
[5)] This report proposed to use a loss rate of 0.01 d-1 for the root model of *Trapp (2002).*

7.3.2 Root parameters

Volume fraction water in roots

The volume fraction of water in roots of 0.93, proposed by Jager and Hames (1997), corresponds to fine roots, which is not relevant for root vegetables. Therefore, in CSOIL 2000 the volume fraction of water was adapted to be more relevant for root vegetables. In CSOIL 2000 a volume fraction of water of 0.833 is used that is based on potatoes. Trapp (2002) used a volume fraction of water of 0.89, which is based on carrots. In Figure 7.3 the minimum and maximum fractions are used to estimate the BCF for root vegetables according to Trapp (2002). This figure shows that the water fraction of roots is not a sensitive parameter for the calculation of the BCF. The estimated BCF differs only slightly for contaminants with a log K_{ow} lower than about 2. From this it can be concluded that the volume fraction of water for roots is not a critical parameter. It is proposed to use a fraction of water in roots that is based on root vegetables other than potatoes, because the model of Trapp (2002) that is proposed to use for root vegetables is not applicable for potatoes. The selected value can be in the range between of about 0.65 and 0.90. In this study the value of 0.89 from Trapp (2002) is selected, because this is the volume content of water for carrots, because Trapp (2002) based his model on this figure for carrots.

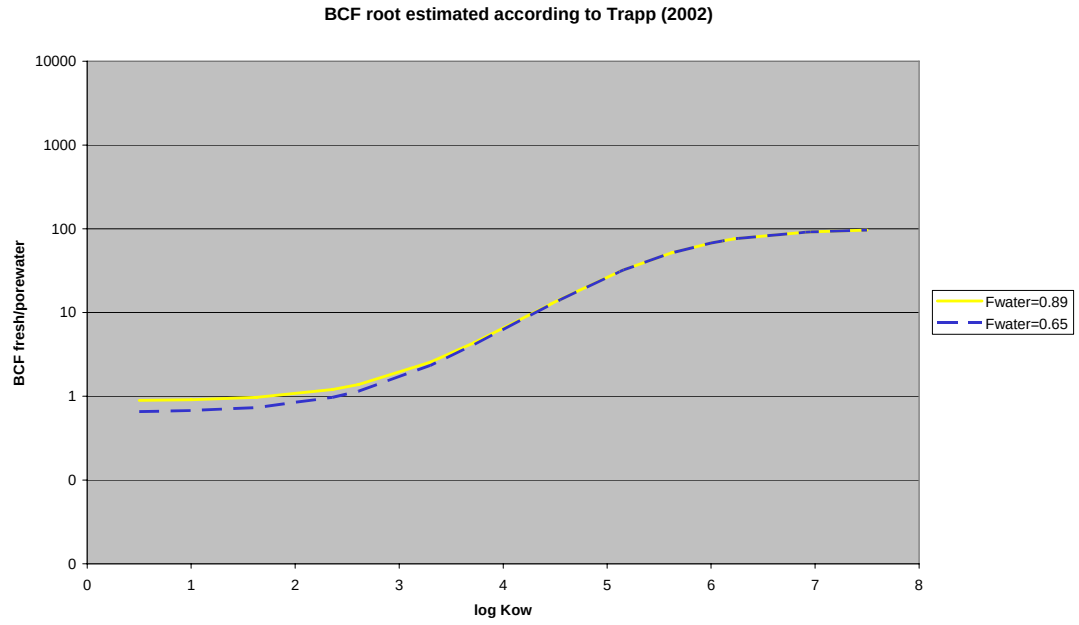

Figure 7.3: BCF for roots as a function of K_{ow} calculated according to Trapp (2002), using volume fractions of water for roots.

Empirical factor for roots

The empirical factor b is used to correct for differences between plant lipids and octanol. This empirical factor can vary from 0.77 for thick roots to 0.95 for fine roots of barley shoots (Rikken et al., 2001). The effect of this empirical factor on the BCF for roots is presented in

Figure 7.4. From this graph it can be concluded that the empirical factor b is a critical parameter for a wide range of K_{ow} values. The value of 0.77 proved to be most appropriate for root vegetables, based on the evaluation of Rikken et al. (2001). In CSOIL 2000 this value was rounded off to 0.8, because it was assumed that 0.77 suggests too much accuracy. Nevertheless, it is proposed in this study to use the value of 0.77, because it is the most appropriate value and it has an unbiased scientific basis.

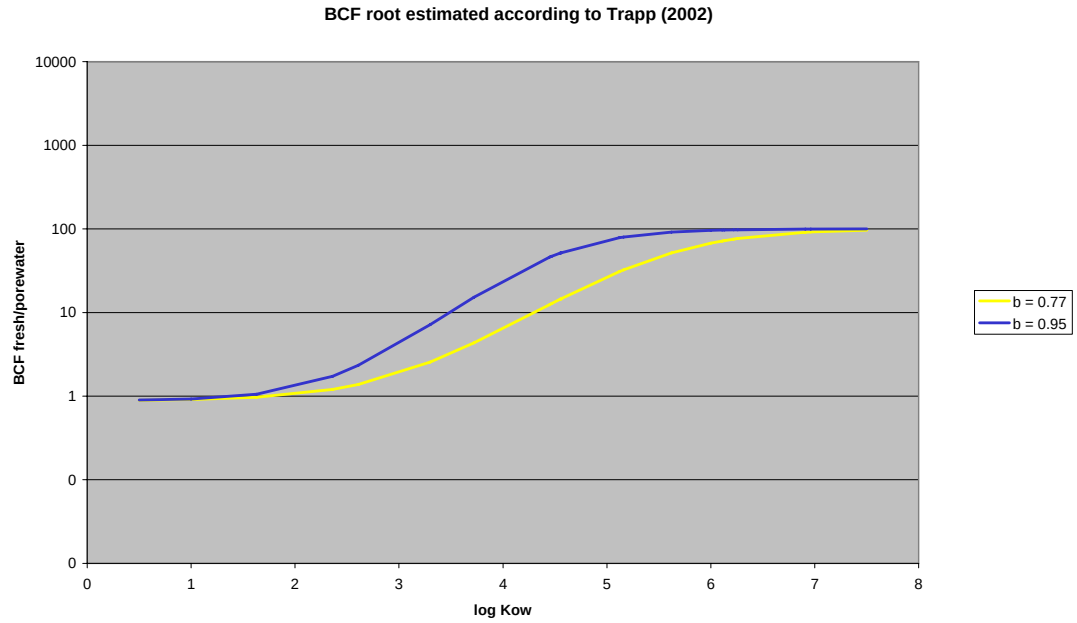

Figure 7.4: BCF for root as a function of K_{ow} calculated according to Trapp (2002), using different values for the empirical factor b.

Model concept for roots

A feasibility study of Versluijs et al. (1998) concludes that the calculation of the concentration in the roots is satisfactory. However, in Rikken et al. (2001) it was observed that the calculation of Briggs was conservative. Trapp (2002) showed that the concentration of lipophilic organic chemicals in thick roots of root vegetables (e.g. carrot) is not accurately predicted with this equilibrium approach. For the concentration in thick roots (C_{root}) Trapp derived the following relation:

$$C_{root} = \frac{Q}{Q \Big/ K_{plant-water} + k \cdot V_{root}} \cdot C_{water} \quad [\text{kg/kg}_{ww}] \qquad \text{Eq. 7.14}$$

in which:

Q:	transpiration stream	[m^3/d]
k:	sum of exponential growth rate and loss by upward flux and metabolism	[d^{-1}]
V_{root}	root volume	[m^3]
C_{water}:	concentration in the pore water (calculated with C_s/K_d)	[in kg/m^3]

In this report a comparison is made between the BCF values for fine and thick roots that are calculated with both the equations and measured data. The results are presented in section 7.5.1.

7.3.3 Above-ground plant parts parameters

Model concept for above-ground plant parts: TSCF

The Transpiration Stream Concentration Factor (TSCF) is defined as the concentration ratio between xylem sap and external solution (pore water) and can be calculated according to Briggs et al. (1982) and Hsu et al. (1990). The TSCF relation according to Briggs et al. is presented in Figure 7.5. The TSCF equation of Hsu et al. gives a higher log K_{ow} optimum than the TSCF relation of Briggs et al. (see Figure 7.6). This could be explained by the fact that Hsu et al. used a pressure chamber technique that gives faster xylem fluxes and less time for equilibration. In the European Union risk assessment (EC, 2003) only the relation of Briggs is used. From a comparison of both TSCF equations Trapp and Matthies (1998) found that the TSCF is an uncertain parameter with measured values that have a large variance. They proposed to use the highest value for the TSCF from both equations for further calculations (Trapp and Matthies, 1995, 1998). In this report a comparison is made between BCF values that are calculated with both TSCF equations and measured data. The results are presented in section 7.4.4, for which it was proposed in Rikken et al. (2001) to use the highest result of both equations in CSOIL 2000.

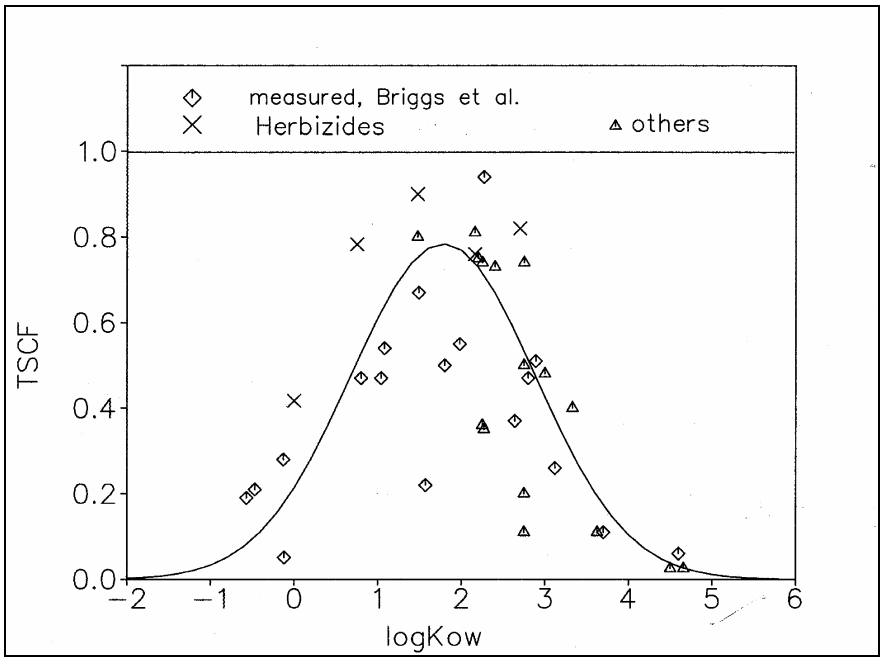

Figure 7.5: TSCF as a function of log K_{ow}; adapted from Briggs (Trapp and Matthies, 1998).

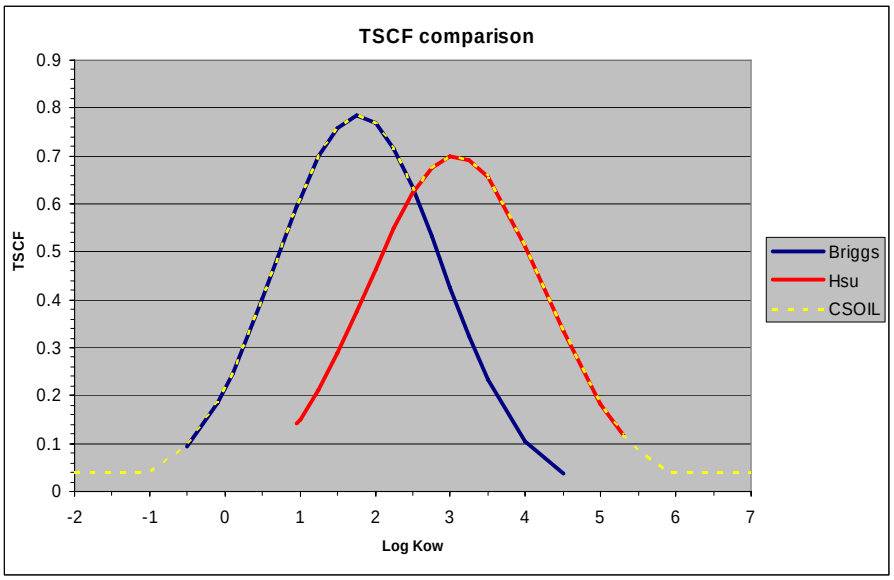

Figure 7.6: Transpiration Stream Concentration Factor (TSCF) as a function of log K_{ow}, according to Briggs et al. (1982), Hsu at al. (1990) and as implemented in CSOIL 2000.

7.4 Comparison of BCFs for roots with above-ground plant parts

In this chapter the BCFs for roots and above-ground plant parts are compared. Separate BCF values for roots and leafs have been calculated and presented in Figure 7.7. The BCF values for roots are calculated according to Trapp (2002), using the empirical factor b of 0.77 as explained in section 7.3.2. The BCF values for leafs are calculated according to Trapp and Matthies (1995) and include resuspension. The resuspension part is based on measured data of section 7.5.2. It is possible that more BCF values for leaf are calculated, that are based on more than one value for resuspension, for the same substance with one log K_{ow} value. Figure *7.7* shows that in almost all cases the BCF for roots is larger than for above-ground plant parts. The BCF for above-ground plant parts of only two substances, with a low log K_{ow}, exceeds the BCF for roots. The difference between the BCF for root and leaf is within 2 orders of magnitude in almost all cases, but can rise to 3 orders of magnitude.

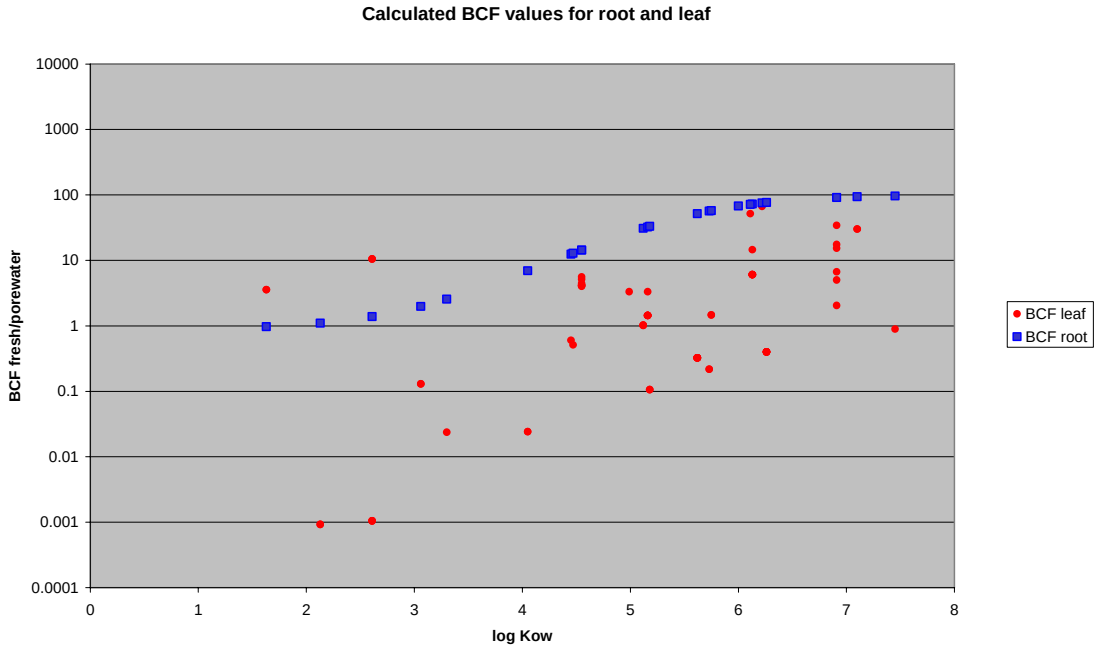

Figure 7.7: BCFs for roots and leafs from pore water as a function of log K_{ow}.

7.5 Comparison of model estimations with measured data

Rikken et al. (2001) compared the results of various model concepts for plants with measured data from literature to get an impression of the extent in which the calculations fit measurements. This section includes the results of that evaluation. Additionally, critical parameters and new model concepts have been evaluated. The literature data are mainly

based on a limited search that was performed to quantify the accumulation of organic contaminants in plants (Cornelese and Lijzen, 2000). The references used for this comparison are presented in Table 7.2.

Table 7.2: References used for comparison of measured BCFs with estimated BCFs.

Contaminant	Number of references	References
PAHs	4	Wild and Jones, 1992; Delschen, 1996; Ye et al., 1991; Trapp et al., 1990; O'Connor et al., 1990; Bellin et al., 1990; Webber et al., 1994; Delschen, 1996
PCBs	6	
Pesticides	12	Harris and Sans, 1969; Heinrich and Schulz, 1996; Voerman and Beseme., 1975; Beall and Nash, 1971; Pylypiw et al., 1993; Shone and Wood, 1974; Burken and Schnoor, 1996; Rouchaud et al., 1991; Trapp et al., 1990; Aplada-Sarlis et al., 1994; Nair et al., 1993
Dioxins	1	
Chlorofenols	2	
Chlorobenzenes	1	Isensee and Jones, 1971
Trichloroethene	1	Bellin and O'Connor, 1990; Casterline et al., 1985
Benzene	1	Topp et al., 1989; Tam et al., 1996.
Phthalates	1	Schnabel et al., 1997 Topp et al., 1989 O'Connor et al., 1990

7.5.1 Roots

Several studies concluded that the calculation of the concentration in the roots according to the approach of Trapp and Matthies (1995) is satisfactory. Nevertheless, a recent study of Trapp (2002) showed that the concentration of lipophilic organic chemicals in thick roots of root vegetables (e.g. carrot) is not accurately predicted with this equilibrium approach. Therefore, the estimated BCF for roots, according to Trapp and Matthies (1995) and Trapp (2002) are compared with BCF data from literature.

BCF for roots according to Trapp and to Trapp and Matthies compared to data of all kinds of roots

In Figure 7.8 the relation between log K_{ow} and calculated BCFs for roots is given for both concepts and the data from the literature search. The literature data include all kind of roots, from fine root of e.g. lettuce, to thick roots of e.g. carrots. In his study, Trapp mentioned two loss rates (k), 0.1 d^{-1} and 0.01 d^{-1} (corresponding to a half time of 6.9 d and 69 d), describing the sum of the exponential growth rate and the loss by upward flux and metabolism. For the root model of Trapp, calculated BCFs based on both loss rates are presented in Figure 7.8.

The BCF values, estimated with Trapp, do not increase at log K_{ow} values higher than 7. The values found in literature fit relatively good with the values of Trapp and Trapp and Matthies with the proposed b=0.77. This comparison makes also clear that the BCF values for roots, calculated with a loss rate of 0.1 d^{-1}, are underestimated. Therefore it is proposed to use the loss rate of 0.01 d^{-1} for estimations according to the Trapp model.

BCF-root according to Trapp and to Trapp and Matthies compared to root vegetables
To be able to create a standard scenario to calculate the risk of vegetable consumption at contaminated soils, all literature data representing fine roots that are not consumed (of e.g. spinach) are eliminated from the database. Figure 7.9 presents the relation between log K_{ow} and the BCF for root vegetables only. Consumption plants, like potato and carrot, appear to have lower bioconcentration factors. For these root vegetables most literature data are lower than the calculated BCFs. Here, the Trapp and Matthies approach can be seen as the upper boundary of the data found in literature and is therefore a conservative estimate for root vegetables. The model of Trapp performs better, mainly because the model concept represents no equilibrium of thick roots with the pore water. It assumes equilibrium with the peel only. Using this concept leads to a more realistic estimate of the bioconcentration factors for root vegetables, although the feasibility could not be checked for contaminants with a log K_{ow} higher than 7, because there was no literature data available. Nevertheless, it is proposed to use the concept of Trapp, mainly because of the improved concept for root vegetables and the better fit with literature data.

BCF-root according to Trapp compared to tuberous vegetables
In section 7.1 it was stated that the root model is used to estimate the concentration in tuberous vegetables (e.g. potato), although a tuber is not a root but a storage organ of a plant. In Figure 7.10 the limited literature data for tubers are compared to the BCF values estimated according to Trapp. The results show that the BCF values for tuberous plants do not deviate much from those for root plants. Therefore, it is proposed to use the Trapp concept also for tuberous plants.

Résumé
It can be concluded that for the estimation of the concentration in roots the model of Trapp (2002) with a loss rate of 0.01 d^{-1} is preferred above the model of Trapp and Matthies (1995).

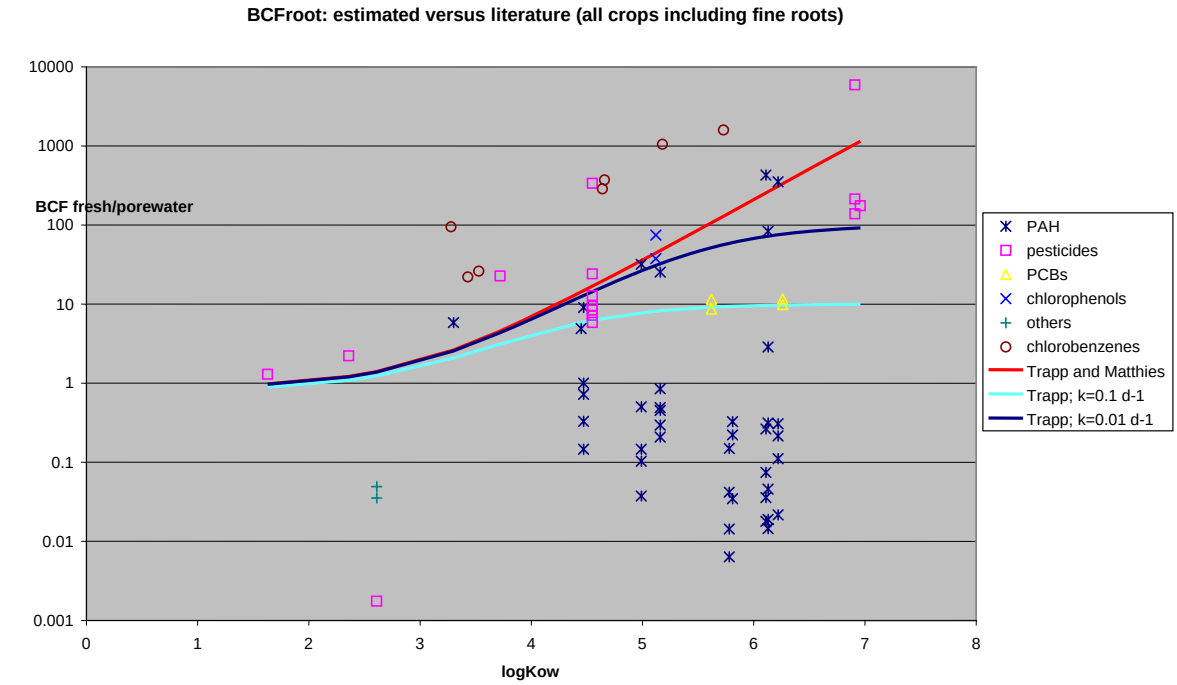

Figure 7.8: BCF from pore water for roots of all plants (incl. fine roots) as a function of log K_{ow}, calculated according to Trapp (2002) and Trapp and Matthies (1995) versus literature data, using various values for the loss rate k.

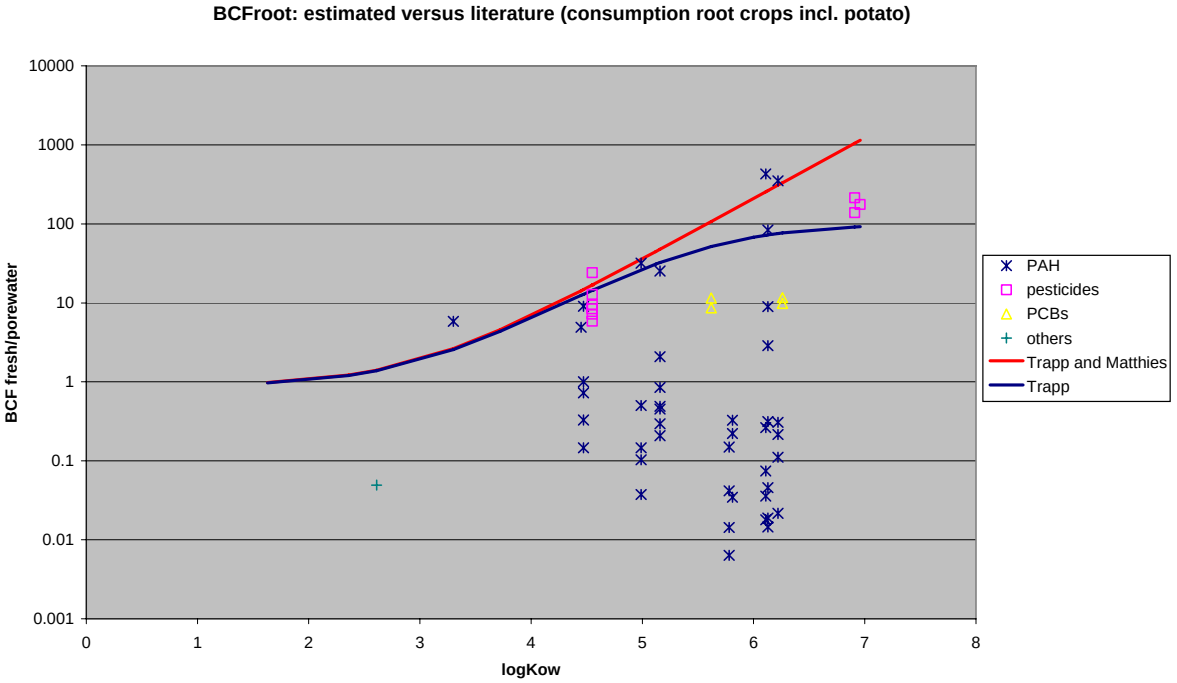

Figure 7.9: BCF from pore water for root plants only (incl. potato), calculated according to Trapp (2002) and Trapp and Matthies (1995) versus literature data.

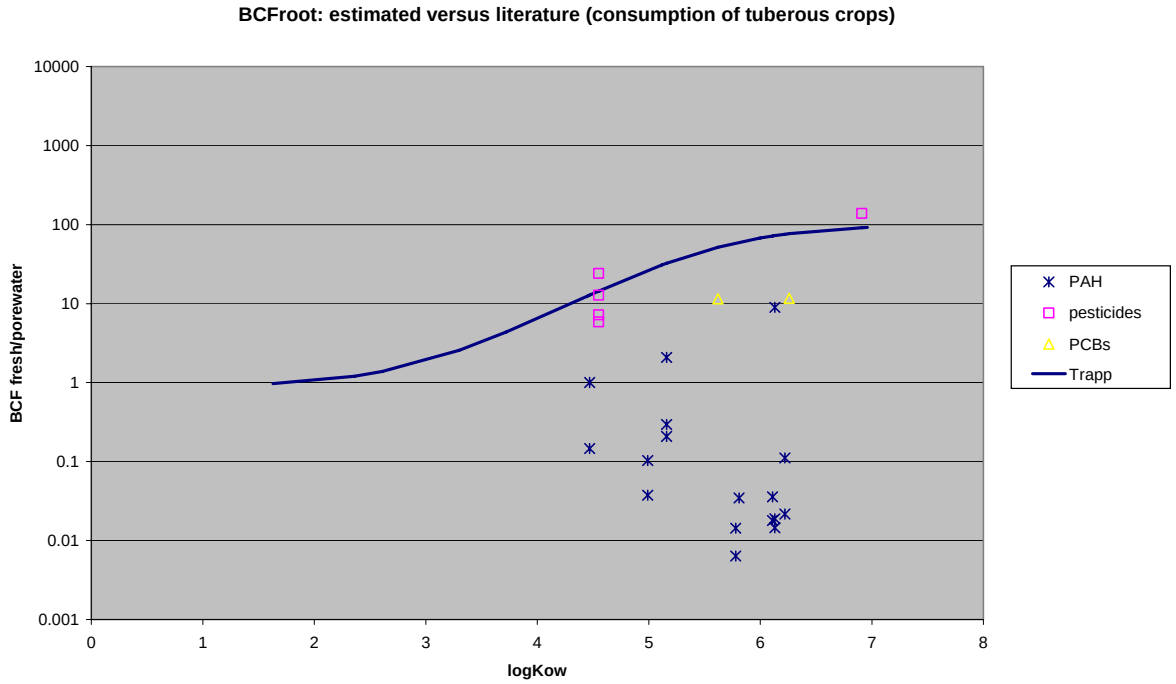

Figure 7.10 BCF from pore water for tuberous plants (e.g. potato) calculated according to Trapp (2002) versus literature data.

7.5.2 Above-ground plant parts

The Trapp and Matthies (1995) model proved to be suitable to calculate the concentration in above-ground plant parts (Rikken et al. 2001; Rikken and Lijzen, 2004). Therefore, the Trapp and Matthies model concept has not been compared to other model concepts and measured data. An important part of the Trapp and Matthies approach is the use of the Transpiration Stream Concentration Factor (TSCF) as is described in the sections 7.2.2. and 7.3.3. The publications of Briggs et al. (1982) and Hsu et al. (1990) are available to estimate the TSCF. Currently, in CSOIL 2000 also the highest result of both TSCF estimation methods is used, in analogy with Trapp and Matthies (1998). However, foundations were lacking to take the highest result of both TSCF methods. Therefore, a comparison has been made between BCF values that are calculated with both TSCF methods and measured data. In this way it could be checked if the calculated BCF values are possibly conservative. The need for a comparison was further supported by the fact that the TSCF is an uncertain parameter that can have a substantial variance and that in the European Union risk assessment (EC, 2003) only the relation of Briggs is used. The estimated BCF values, according to Trapp and Matthies (1995), for the above-ground plant parts are compared with BCF data from literature.

Consequences of the range of TSCF
The TSCF can be calculated, while accounting for the minimum and maximum K_{ow} of the dataset mentioned in the publications (see Eq. 7.4 and 7.5). Figure 7.11 presents the resulting TSCF values when the K_{ow} is outside the range of the minimum or maximum K_{ow}. This method is applied in the European Union risk assessment and in CSOIL 2000 (see Figure 7.11). In Figure 7.12 and in Figure 7.13 the resulting BCF values are plotted against the octanol-air partition coefficient (log K_{oa}), with and without soil resuspension.
The TSCF can also be calculated assuming that the relations of Briggs and Hsu are also valid outside the K_{ow} range. These TSCF relations can be seen in Figure 7.14, which is more in agreement with the relation according to Briggs in Figure 7.5 (Trapp and Matthies, 1998). The resulting BCF values are shown in Figure 7.15 and Figure 7.16.

Comparison with measured data excluding soil resuspension
Figure 7.12 and Figure 7.15 show that the BCF values from the literature are higher than calculated with Trapp and Matthies, in most cases. In general, the BCF values from literature increase with higher K_{oa}, when the range of the K_{ow} is taken into account (Figure 7.12). This picture is less clear in Figure 7.15 for data points with a logK_{oa} between 10 and 12, when the valid range is not taken into account. The high concentrations found in literature could originate from deposition (dry and wet), from soil resuspension (rain splash) or from air born contaminants. In these model calculations deposition or resuspension is not included.

Comparison with measured data including soil resuspension
Soil resuspension can be included in the model calculations when 1% of the soil concentration on a dry weight bases is added to the estimated BCF with Trapp and Matthies (Rikken et al., 2001). In Figure 7.13 and Figure 7.16 these model calculations are plotted against the literature data. These figures show that not only the differences between the estimated BCF and the measured BCF are smaller than in Figure 7.12 and Figure 7.14, but also the differences between the BCF based on the two TSCF methods (Briggs and Hsu) are smaller. Further, there are almost no differences between the methods that account or do not account for the range of the K_{ow} to calculate the TSCF.

Résumé
It can be concluded that the choice to use the highest options of both TSCF estimation methods leads to BCF values that are more often lower than higher than the literature data, even when soil resuspension is taken into account. However, the BCF values would be even lower when only one of the two TSCF methods is applied. This conclusion supports the proposed use of the highest result of both TSCF methods in this study. The consequences of the omission or use of valid range of the TSCF are not noticeable when the soil resuspension is taken into account.

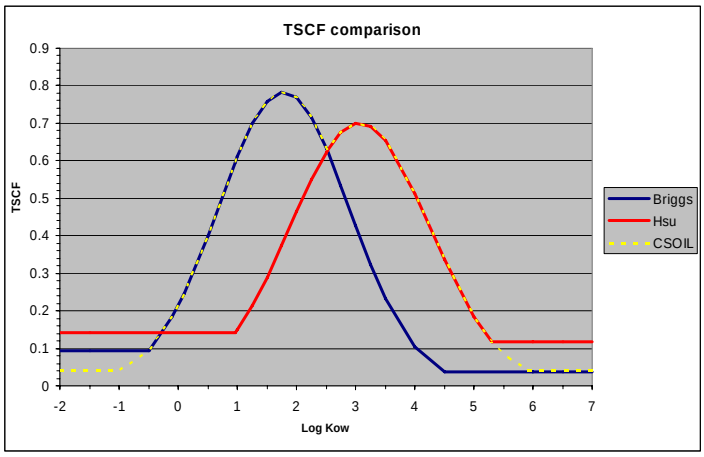

Figure 7.11: Transpiration Stream Concentration Factor (TSCF) according to Briggs, Hsu and the implementation in CSOIL 2000, including the range borders, used to calculate Figure 7.12 and Figure 7.13.

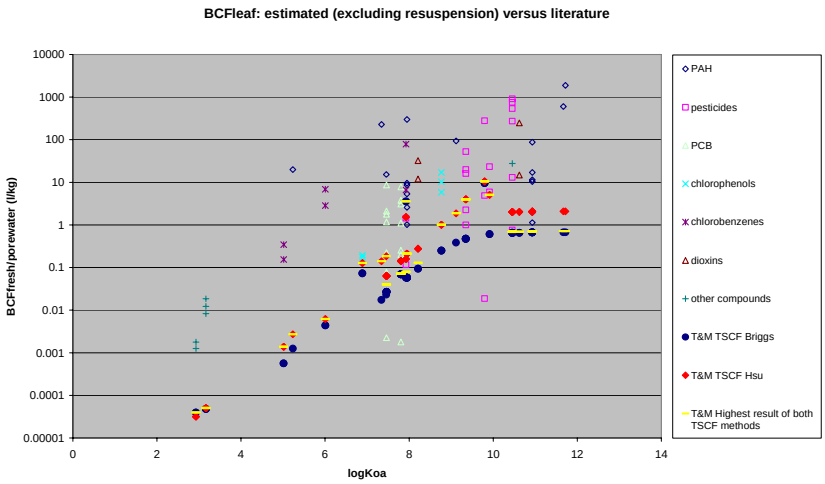

Figure 7.12 BCF for leaf from pore water calculated according to Trapp and Matthies (1995) using various models for the TSCF, versus literature data.

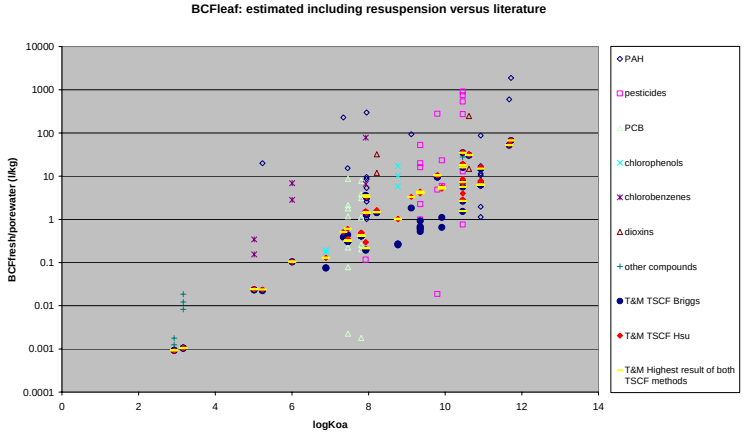

Figure 7.13: BCF for leaf from pore water calculated according to Trapp and Matthies, including resuspension, using various models for the TSCF, versus literature data.

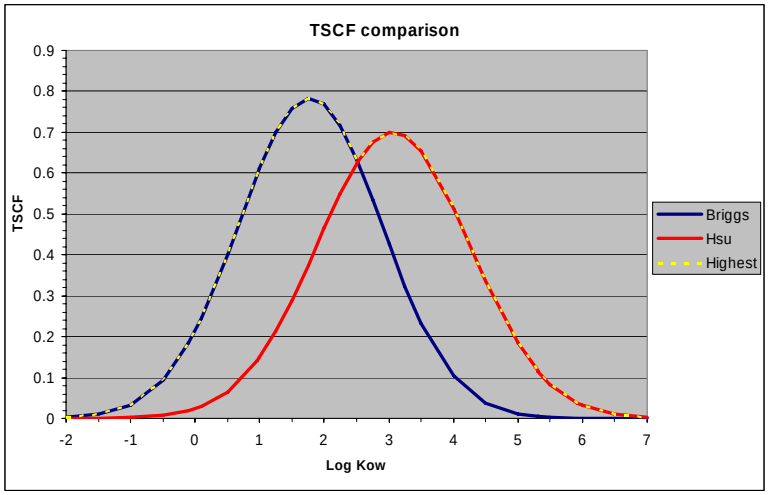

Figure 7.14: TSCF according to Briggs, Hsu and the highest result of both references, used to calculate Figure 7.15 and Figure 7.16.

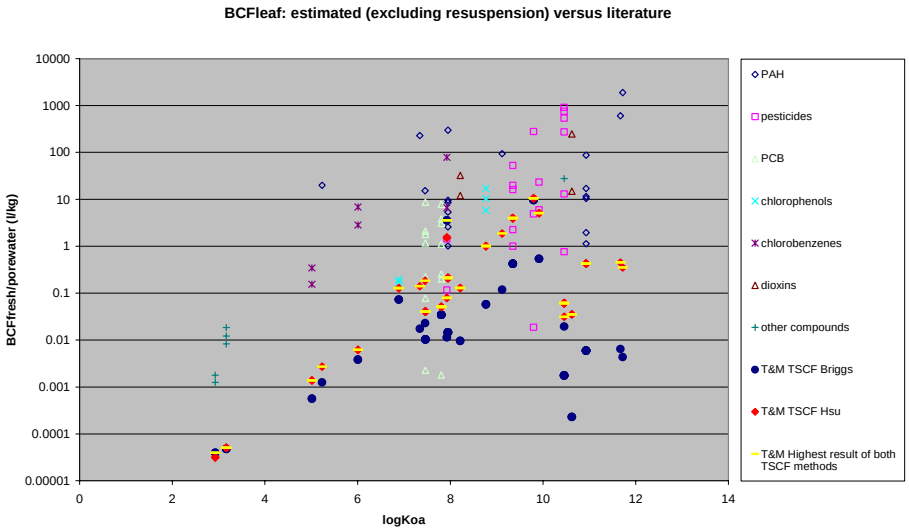

Figure 7.15: BCF for leaf from pore water calculated according to Trapp and Matthies (1995) using various models for the TSCF, versus literature data.

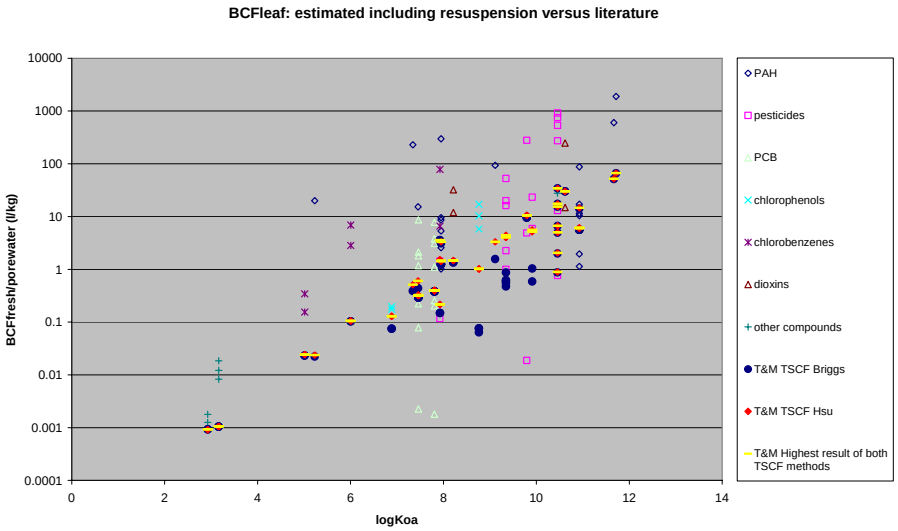

Figure 7.16: BCF for leaf from pore water calculated according to Trapp and Matthies, including resuspension, using various models for the TSCF, versus literature data.

7.6 Résumé

The relative contribution of accumulation of organic contaminants is larger for roots than for above-ground plant parts. The BCF for above-ground plant parts of only two substances, with a low log K_{ow}, exceeds the BCF for roots.

7.6.1 Roots

The Trapp (2002) concept leads to a more realistic estimate of the bioconcentration factors for root vegetables, mainly because the model concept represents no equilibrium of thick roots with the pore water and the proper fit with measured data. Nevertheless, the measured dataset was very limited. It is strongly recommended to check the feasibility of this concept further with measured data for root vegetables of more contaminants, of which preferably also some contaminants with a log K_{ow} higher than 7. A loss rate of 0.01 d^{-1} is proposed to estimate the concentration in roots with the Trapp model, but this value should be considered in more detail in the future.

A large part of the average consumption basket is taken up by potatoes. A potato is a tuberous vegetable that is not a root, but is morphologically a modified stem. Unlike roots or root vegetables, an advective transport of soil contaminants into tuberous vegetables is unlikely. Currently, the root model of Trapp is proposed to use for tuberous vegetables, but it is preferred to estimate concentrations in tuberous vegetables according to a diffusive uptake model. For future revisions it is proposed to consider the diffusive approach for tuberous plants in more detail. A starting point could be the planned validation study of the Samsøe-Peterson et al. (2002) diffusive model concept (Kulhánek et al., 2005).

7.6.2 Above-ground plant parts

The Trapp and Matthies (1995) approach is used to calculate the concentration (and BCF values) for above-ground plant parts. Apparently, the Trapp and Matthies approach underestimates BCF values when they are compared with BCF values based on literature data. It is unknown if this underestimation is caused by the deposition of contaminants by rain and particles from air, that could be included in the measured data. Deposition from air is not accounted for in the Trapp and Matthies model. Therefore, a thorough investigation of the validity of this approach compared to more appropriate experimental data is recommended.

The soil resuspension concept (rain splash deposition on leaves) plays an important role in the determination of the BCF for leaves. This can be concluded from the evaluation of TSCF estimation methods, where large differences between the estimated BCF values were not noticeable anymore when the soil resuspension data were added. The contribution of this soil resuspension route was set at a value of 1% dry soil per dry plant (Rikken et al., 2001). This figure should be evaluated in more detail.

Fruits are not considered in the average consumption basket. Therefore, the uptake of contaminants in fruits is not evaluated in this report. Nevertheless, Trapp et al. (2003) developed a fruit tree model for the uptake of neutral organic chemicals from contaminated soils into fruits. This model must certainly be considered if it is decided to add fruits to the consumption basket in future revisions.

8. Measuring the available fraction in soil

8.1 Introduction

It is well recognized that only part of the contaminant pool in soils is available to plants. For that reason several researchers assume that the pore water concentration has a better relation with concentration in plants than the total soil concentration. In fact, not only the total soil concentration in the pore water is relevant for plant uptake, but the uptake rate also strongly depends on the speciation in the pore water. Besides, the pore water concentration is not always easy to measure and is not stable in time. It might fluctuate, for example with, redox conditions and, hence, change in between rainy period and periods of drought. Several methods exist to extract a fraction of contaminants from soil that represent the bioavailable fraction, under standardized conditions in the laboratory. Although plants are less divers than soil organisms in regard to uptake mechanisms, there is debate on the most appropriate measurement technique or extraction method for assessing the contaminant concentration in soils that relates to the accumulated concentration in plants. Most research on these techniques and extraction methods has been performed for metals.

It is investigated in this chapter if measurement of the available fraction in soil has potential to be included in Tier 3, i.e. the measurement protocol. Either as an alternative for or an addition to measurements in crops.

8.1.1 Metals

Many researchers find a good relation between free metal ion activity and plant uptake (e.g. Datta and Young, 2005). Several methods exist to measure the free metal activity, among them the Donnan Membrane Technique (Temminghoff et al., 2000) and the Permeation Liquid Membrane (Senn et al., 2004). Other methods also include measurement of the labile fraction in soil, e.g. Diffuse Gradients in Thin Films (Davison and Zhang, 1994). However, these procedures are relatively complex and time consuming and not suitable for routine soil investigation. For this reason, often the pore water concentration is taken as representative for the fraction that is taken up by plants. For metals several extraction methods exist that are supposed to mimic a representative pore water concentration, e.g. 0.025 M EDTA with a pH of 4.6 and shake for 90 minutes at soil: solution ratio of 1:25 (Sims and Johnson, 1991). Stenz et al. (1997) use a $NaNO_3$ extraction, while Novozamsky et al. (1993) refer to a 0.01 M $CaCl_2$ extract. However, many other options exist, differing in “power” to extract metals from the sorption sites or even from the soil particles skeleton. In fact extractions methods differ from mild extractions methods like shaking with a mild EDTA solution to complete destruction, for example with a concentrated solution with HF, HCl and HNO_3.

Chojnacka et al. (2005) give an overview of several extraction methods:

- Deionized water;
- $MgCl_2$ (1 ml/l);
- $NaNO_3$ (0.1 ml/l);
- CH_3COOH (2%);
- HCOOH (2%);
- NA_2EDTA (0.05 mol/l);
- EDTA(0.1 ml/l);
- KNO_3 (0.5 ml/l);
- Ammonium citrate (2%);
- Citric acid (2%);
- $K_2P_2O_7$ (0.1 ml/l);
- $HCl + HNO_3$ (0.1 ml/l).

The authors conclude that the 2% ammonium citrate extraction shows statistically significant relations with plant concentrations for all the nine metals investigated (relatively low metal concentrations), with the exception of nickel. The correlation of plant concentrations with the ammonium citrate extracted fraction shows an important increase in correlation significance compared to the relation between plant and soil concentrations.

Because no method is recognized as the ideal method, the use of a procedure to extract metals form contaminated soils is not further explored in this study. However, identification of such an ideal method could improve the field measurement, possibly in combination with the calculation of the uptake from the pore water into the plants, in the future. Therefore, it is recommended to perform research with the purpose to identify the potential of and the most appropriate extraction method for assessing the metal concentration in soils that relates to the accumulated metal concentration in plants.

8.1.2 Organic contaminants

For organic contaminants also several extraction methods exist that result in a representative pore water concentration. Most methods use solid phase or membrane-based extractions. A very popular solid phase-based procedure is an extraction with tenax (e.g. Cornelissen et al., 1997; Ten Hulscher et al., 2003). Alternatively, an extraction with SPME (Solid Phase MicroExtraction) is often used (e.g. Leslie et al., 2002; Van der Wal et al., 2004). Several authors compared these two methods. Stephen et al. (1997), for example, compared these two methods for volatile compounds. The latter concluded that tenax shows higher extraction rates than SPME. In a recent article You et al. (2006) compared these two methods for accumulation of hydrophobic compounds in the oligochaete *Lumbriculus variegatus* (an organisms resembling tubifex worms, although from a different Order). They concluded that both procedures provide good estimations of bioaccumulation.

In analogy with extraction procedures for metals, no method is recognized as the ideal method. Therefore, the use of a procedure to extract organic contaminants from contaminated soils is not further explored in this study. However, identification of such an ideal method could improve the field measurement, possibly in combination with the calculation of the uptake from the pore water into the plants, in the future. Therefore, it is recommended to perform research with the purpose to identify the potential of and the most appropriate extraction method for assessing the organic contaminant concentration in soils that relates to the accumulated organic contaminant concentration in plants.

9. Measuring of contaminants in vegetables

9.1 Introduction

Measuring the representative contaminant concentration in vegetables is complicated, because of the many choices that must be made (e.g. location of the samples, type of vegetable, number of samples) and the huge heterogeneity of contaminant concentrations in soil and, hence, crops. Nevertheless, a measured concentration is considered as the most realistic value for the representative concentration in vegetables, in the highest tier. Wegener Sleeswijk and Kleijn (1993) also recommend a measurement procedure in case when it can not be concluded from site specific factors that an unacceptable risk for vegetable consumption is unlikely.
This chapter aims to provide guidance on the determination of representative contaminant concentrations in vegetables, by measurement in the field. The presented procedures relate to local lots of cultivated land with variable size and a different degree of homogeneity and are applicable for a variety of home-grown vegetables. Guidance presented in this document covers the following aspects:

- Field sampling procedures (plant sampling in general, properties and size of the sampling location, types of plants and quantity required for analyses, quality restrictions): section 9.2.
- Sample pre-treatment in the laboratory (cleaning, fragmentation, homogenization, storage, drying and grinding): section 9.3.
- Proxy plant: section 9.4
- A seeding, growing harvesting program: section 9.5.

The methods in this guideline are revised versions of a compilation of existing guidelines used by WU, Alterra (Römkens et al., 2004), IB-DLO en RIVM. The regional Health inspectorate (GGD) in the Netherlands also provides guidance on sampling food products (Van Brederode, 2002).

No formal guidelines for the chemical analysis of plants exist, i.e. no Dutch NEN standards, neither ISO standards. Therefore, it is advised to perform analysis in analogy with soil samples. Laboratories that perform the chemical analysis must comply with ISO-17025 for good laboratory practice.

9.2 Field sampling procedures

9.2.1 Preliminary investigation

Before sampling the owner or tenant of the parcel of land under investigation must give permission to perform the sampling. It may also be essential to ask the owner or tenant for information on soil type, manuring or fertilising practice, type of tillage, sowing and harvest season and the type of cultivated vegetables.

Additionally, the stage of growth and the health condition of the plants must be registered. By taking a series of (digital) photographs, the conditions of the site and the plants must be visualized and documented. Aspects of importance in the documentation process are:

- condition of the garden;
- lay-out of the site (size and localization of the allotments);
- micro relief (ridges and furrows);
- type and variety of the plants;
- health condition of the plants.

Preferably, date and time stamped copies of these photographs (jpg files) are stored on a separate CD-ROM, together with additional written information in a text file format, for each separate allotment.

9.2.2 Properties and size of the sampling location

The sampling procedure for vegetables is dependent on the size and the lay-out of the site, micro relief, the spatial distribution of the contaminants and soil properties.

For a relatively small site, such as a private kitchen garden in a residential area, the results should reflect the contaminant concentration in vegetables for the entire site. The same holds for larger areas where there is low spatial variability in regard to contaminant concentration, soil properties, tillage and (micro)relief, or when this variability is unknown. For every homogeneous site a composite sample is taken for all individual type of vegetables that grow on the site. If a particular vegetable grows at several locations within one parcel, the composite sample should contain sub-samples from every location.

For a larger site where the variation in regard to contaminant concentration, soil properties, tillage and (micro)relief is considerable and known, several contaminant concentrations, representative for several sub-sites within the site, could be determined. Therefore, these sub-sites must be sampled separately. Alternatively, a worst-case sampling could be performed at a sub-site where relatively high risks may be expected. Sub-sites with relatively high risk may be indicated by comparatively high levels of contaminants in combination with vulnerable soil properties (sand, low organic matter content, low pH). In case the worst-case risk assessment does not exclude an unacceptable human health risk, extension of the

sampling program to other sub-sites may be considered. On a site with a number of separated garden allotments, it may also be necessary to evaluate each allotment separately.

In the design stage of a sampling program, the purpose of sampling must be specified in relation to available data on the distribution of differences in the contaminant levels, soil properties and tillage practice. Common purposes are:

- determination of the representative vegetable concentration for the entire location;
- determination of the representative vegetable concentrations for several sub-sites;
- determination of vegetable concentrations under worst-case conditions, to guide a possible extension to the determination of representative crop concentrations at one or more sub-sites;
- determination of the variation in vegetable concentrations for a larger area.

Moreover, for research purposes it is strongly recommended to extent crop sampling with an appropriate soil sampling program. However, although the additional costs are relatively limited, this is not suitable for routine risk assessment.

9.2.3 Types of plants and the quantity required for analysis

Obviously, only plants that are present on the site can be sampled. In case several plants are present, the selection of the type of plants to be sampled is subject to the following criteria:

1. Vegetables versus non-edible plants; preferably vegetables must be sampled.
2. Frequency of occurrence in kitchen gardens, e.g. lettuce, beans, potatoes are frequently found in kitchen gardens.
3. Representativeness of different kitchen garden vegetables; preferably plants of different plant groups must be sampled (see section 4.3: potatoes, root and tubers, bulbous vegetables, fruit vegetables, cabbages, leafy vegetables, legumes, beans and stem and stalk vegetables.
4. Contribution to the total consumption rate (see section 4.3).
5. Affinity for accumulation of contaminants; preferably (also) high accumulating vegetables like spinach, lettuce and endive must be sampled.

It is essential to take a representative number of samples of all the available types of edible plants at the time of sampling. Only the edible parts of the plants must be sampled.

The vegetable groups that are preferred for sampling are given in Table 9.1. Based on accumulation potential as well as consumption preference and frequency of cultivation, Table 9.1 also presents the vegetables that are most appropriate to be sampled individually (**in bold**). In view of the criteria given above, a minimum requirement is to sample potatoes, root and tuber vegetables and leafy vegetables. In Table 9.1 also the required amount of sample per vegetable is quantified, both in numbers and in weight (optimum and minimum value).

If the vegetables available are not sufficient for sampling, or there are no vegetables present at all, there is an option for sowing additional plants solely for the purpose of human health risk assessment (see section 9.5).

Table 9.1: Vegetable types and the amount of vegetable material needed for sampling. ***Bold print*** *indicates the preferred vegetables for sampling.*

Group of vegetables	**Vegetable**	**Amount of fresh material (g)** (optimum / minimum)	**Number of plants or plant parts** (optimum / minimum)
Potatoes	**Potato**	1000/ 500	10 (from 5 plants)/ 5 (from 3 plants)
Root and tuberous vegetables	**Carrot**	500/ 200	10 piece/ 5 piece
	Beetroot	500/ 200	10 piece/ 5 piece
	Black salsify	500/ 200	10 piece/ 5 piece
	Celeriac	500/ 200	10 piece/ 5 piece
	Turnip	500/ 200	10 piece/ 5 piece
	Radish	500/ 200	25 piece/15 piece
	Winter carrot	500/ 200	10 piece/ 5 piece
Bulbous vegetables	**Leek**	500/ 200	10 piece/ 5 piece
	Onion, Eschalot	500/ 200	25 piece/15 piece
	Garlic		25 piece/15 piece
Fruits	**Tomato**	500/ 200	10 (from 5 plants)/ 5 (from 3 plants)
	Courgette	500/ 200	10 (from 5 plants)/ 5 (from 3 plants)
	Strawberry	500/ 200	25 (from 5 plants)/15 (of 3 plants)
	Cucumber	500/ 200	10 (from 5 plants)/ 5 (from 3 plants)
	Melon, Pumpkin	1000/ 500	10 (from 5 plants)/ 5 (from 3 plants)
	Gherkin	500/ 200	10 (from 5 plants)/ 5 (from 3 plants)
	Capsicum, Chilli	500/ 200	10 (from 5 plants)/ 5 (from 3 plants)
	Eggplant	500/ 200	10 (from 5 plants)/ 5 (from 3 plants)
	Maize	500/ 200	10 piece/ 5 piece

Cabbage	Conical or red cabbage	1000/ 500	5 piece/ 3 piece
	Kale	1000/ 500	5 piece/ 3 piece
	Sprout cabbage	1000/ 500	5 piece/ 3 piece
	White cabbage	1000/ 500	5 piece/ 3 piece
	Green and Savoy cabbage	1000/ 500	5 piece/ 3 piece
	Broccoli	1000/ 500	5 piece/ 3 piece
Leafy vegetables	**Lettuce**	1000/ 500	5 piece/ 3 piece
	Endive	1000/ 500	5 piece/ 3 piece
	Spinach	500/ 200	Number of leaves to reach weight
	Chicory	1000/ 500	10 piece/ 5 piece
	Chervil, Parsley and other herbs	500/ 200	Number of leaves to reach weight
Fresh pod vegetables	Green bean	500/ 200	50 pieces/20 pieces
	Garden pea	500/ 200	50 pieces/20 pieces
	French bean	500/ 200	50 pieces/20 pieces
Beans	Garden pea	1000/500	100 pieces/ 50 pieces
	Navy bean	1000/500	100 pieces/ 50 pieces
	Brown bean	1000/500	100 pieces/ 50 pieces
	Broad bean		100 pieces/ 50 pieces
Stem and stalk vegetables	Rhubarb		10 stems/5 stems
	Asparagus		10 stems/5 stems
	Blanched celery		10 stems/5 stems

Individual plants of a single species should be cut at the same distance from the ground, in order to have a comparable level of contamination. Preferably, the plant material should be sampled under dry weather conditions. The plant parts of interest should preferentially be cut with a titanium scissors, or be harvested by hand. Avoid contamination of the plant sample with soil material. Soil particles adhering to the sampled plant parts should as far as possible be removed by shaking. Collect the plants in bags that can be properly closed. Use paper bags for samples that may contain organic contaminants. Perform, if possible, an analysis of fresh weight of the sampled plant material while still at the sampling location, since the dry matter content can change during transport and storage.
Transport and storage of the sample must take place under refrigerated conditions (no more than 4 °C).

9.2.4 Quality restrictions

For the purpose of plant sampling, there are no existing guidelines that specify the quality of the plant sample. Therefore, it is possible to adhere to the sampling guidelines specified by the Food and Drug Act (*Warenwet*) or by the EU on the analysis of pesticide residues in primary products (EC, 2002).

In case of edible plants, i.e. vegetables, the sampled plant parts should be in the stage of growth where they are actually harvested for consumption. When edible parts of vegetables "do not look good", do not sample them. Follow the same selection as is normal household practice. A specific remark for sampling potatoes must be made: damaged specimen should not be sampled. The minimum size of the potatoes is 2 cm.

9.3 Sample pre-treatment in the laboratory

9.3.1 Cleaning

In general, the concentration of contaminants in soil is much higher than in the plant tissue, certainly for hydrophobic contaminants. Therefore, the sample needs to be cleaned from adhering soil particles prior to analysis. Moreover, hydrophobic contaminants (mainly PAHs) are known to deposit on plant material from the air. These deposits also have to be removed prior to analysis. Cleaning may consist of washing, peeling, fragmenting and other plant specific methods, sometimes with additional washing in between.
The cleaning process to be applied should as much as possible resemble the common household practice of preparing the vegetables for consumption. Because in real life humans do ingest a fraction of the soil particles attached to the vegetables, exposure from contaminants in not-easy-to-remove particles must be part of the risk assessment. Therefore, a more intense cleaning procedure than the common household practice is dissuaded.
In the process of peeling and cutting, materials made of titanium or other inert materials must be used, just as in the process of sampling in the field. Vegetable specific domestic preparation procedures are summarized in Table 9.2 for the most common vegetable garden vegetables.

Table 9.2: Summary of domestic preparation procedures, wash and peel descriptions for the most common vegetable garden vegetables.

Vegetable	Peeling and cutting	Washing
Potato	Peel and cut to cubes of approx. 2 cm^3	Wash with tap water before peeling and rinse once peeled
Carrot	Scrub with a hard brush and water and cut in small slices	Wash with tap water before scrubbing and rinse after scrubbing
Beetroot	Peel and cut in small slices (0.5 cm)	Wash with tap water before peeling and rinse once peeled
Black calcify	Peel and cut in small slices (0.5 cm)	Wash with tap water before peeling and rinse once peeled
Celeriac	Peel and cut to cubes of approx. 2 cm^3	Wash with tap water before peeling and rinse once peeled
Turnip	Peel and cut to cubes of approx. 1 to 2 cm^3	Wash with tap water before peeling and rinse once peeled
Radish	Remove root	Rinse with tap water
Winter carrot	Scrub with a hard brush and water and cut in small slices	Wash with tap water before scrubbing and rinse after scrubbing
Leek	Remove outer leave and root base, cut in 1 cm slices	Rinse the cut leek with tap water
Echalot	Remove outer peel and cut in 5 mm cubes	None
Onion	Remove outer dry peels and cut in 5 mm cubes	None
Garlic	Remove outer dry peels and cut in 5 mm cubes	None
Tomato	Remove sepals and cut in parts of approx 3 cm	Wash with tap water before cutting in parts
Courgette	Cut in pieces of approx. 1 cm.	Wash with tap water before cutting in parts
Strawberry	Remove sepals	Wash with tap water before removing the sepals
Cucumber	Peel and cut in small slices	Wash with tap water before peeling and cutting
Melon, Pumpkin	Peel and cut in parts	None
	Peel and cut in parts	Wash with tap water before peeling and cutting
Capsicum, Chilli	Remove stem and seeds	Wash with tap water before removing stem and seeds
Egg plant	Remove stem and cut in parts	Wash with tap water before removing stem and cutting in parts
Maize	Remove leaves and collects seeds.	Wash with tap water before collecting the seeds
Conical or red cabbage	Remove outer leaves and stem and cut in slices of 1 cm	Rinse the cut cabbage with tap water

Kale	Collect the leaves and remove hard veins. The leaves are cut in small pieces of approx 1 cm^2	Rinse the cut cabbage with tap water
Sprout cabbage	Remove outer leaves	Wash with tap water after removal of outer leaves
White cabbage	Remove outer leaves and stem and cut in slices of 1 cm	Rinse the cut cabbage with tap water
Green or Savy cabbage	Remove outer leaves and stem and cut in slices of 1 cm	Rinse the cut cabbage with tap water
Broccoli	Remove leaves and divide the cabbage in sizable florets	Rinse the cut cabbage with tap water
Lettuce (maximum of 2 dominant varieties)	Cut or tear the leaves (depending on variety)	Wash whole leaves with tap water. Iceberg lettuce should be cut first
Endive	Remove outer leaves and the plant base, cut in small pieces	Rinse the cut leaves with tap water
Spinach	Remove thick stems	Rinse leaves with tap water until soil particles are removed
Chicory	Remove outer leaves and plant base, cut in small pieces	Wash cut leaves with tap water
Chervil and Parsley	Cut in small pieces	Wash herbs prior to cutting.
Herbs	Cut in small pieces	Wash herbs prior to cutting.
Green bean	None	Rinse with tap water
Garden pea	Remove stalk	Rinse with tap water
Broad bean	Remove the beans from the pod	Rinse unshelled beans with tap water
Peas	Remove the beans from the pod	Rinse unshelled beans with tap water
Navy bean	Remove the beans from the pod	Rinse unshelled beans with tap water
Brown bean	Remove the beans from the pod	Rinse unshelled beans with tap water
Rhubarb	Remove leaves and cut stalks in pieces of 2 cm	Wash with tap water before cutting in pieces
Asparagus	Peel and remove base of the stem	Wash with tap water after peeling
Blanched celery	Cut the stems loose	Wash stems with tap water

9.3.2 Fragmentation and homogenization of sub samples

The plant sample must be homogenized, before chemical analysis. Depending on the analytical detection limit a quantity of homogenized plant material in between 10 and 100 grams is needed. For all plant samples a fragmenting homogenizer must be used that does not emit heavy metals. Examples are plastic or glass household blenders with titanium or zirconium knives, onion cutters with carbon steel knives and a plastic housing, or by hand with titanium or teflon cutlery.

9.3.3 Storage of plant samples

Fresh plant samples are best kept under refrigeration (<4 °C) for a limited time, also during transport to the laboratory. For a prolonged period of time, dried plant samples can be stored in a dark and dry place. Homogenized samples can be stored for a prolonged period of time in the freezer at a minimum temperature of -30 °C.

9.3.4 Drying of plant samples

Drying of the plant samples must be done for two reasons, i.e. as preparation of the chemical analysis and for the determination of the dry matter content of the plants. The plant-specific dry matter content may be highly variable.

Chemical analysis

Samples that are analyzed for heavy metals must be dried before analysis. Drying is done in an oven at a temperature of 70 °C, until no further weight loss occurs. To avoid leafy plants like lettuce, spinach, chicory and endive to get slimy, these vegetables must be pre-dried at a temperature of 35 °C. The plant material is dried in anodized aluminium trays that are lined with a clean inlay of paper. Sugar holding material is dried in glass petri dishes, disposable aluminium trays or in packed in heat resistant plastic foil, to avoid sticking and the loss of plant material. Samples that only contain metals can also be freeze-dried. Tomatoes should not be dried, because they will loose some of their cell contamination concentration. Samples with organic contaminants also should not be dried.

Determination of the dry matter content

Usually, the concentration in plants is expressed on the basis of dry weight. Human exposure in the CSOIL 2000 model is expressed on the basis of fresh weight consumption rates. Therefore it is essential to determine the dry matter content from both the dry weight and humidity of a fresh plant sample. There are no generally accepted ways to perform this type of analysis. Different laboratories have their own methodology. In this protocol the fresh sample must be dried at a temperature of 70 °C, in order to avoid loss of organic material. The fresh weight of plant material must be determined by weighing a sub-sample on an analytical balance and registering the fresh weight. Subsequently, the sub-sample must be put in a paper bag and dried in an oven for 24 hours at a temperature of 70 °C. After 24 hours, the bag must be cooled in desiccators for at least 30 minutes. Then the dried plant material must be transferred from the paper bag to the analytical balance and the dry weight must be determined. The dry matter content in terms of percentage (%DMC) with respect to fresh weight is calculated straightforward:

$$\%DMC = \frac{Mvegetable_{wet\ weight} - Mvegetable_{dry\ weight}}{Mvegetable_{wet\ weight}} x\,100\% \quad [-] \qquad \text{Eq. 9.1}$$

in which:

Mvegetable	concentration in the vegetable	[mg/kg] (either dry weight or fresh weight)

In most cases drying can serve both purposes, i.e. the preparation of the chemical analysis and for the determination of the dry matter content of the plants. For tomatoes and samples with organic contaminants, for which drying for analytical purposes is dissuaded, separate samples must be dried to determinate the dry matter content.

9.3.5 Grinding of dried samples

For grinding of dried samples, a grinder or blender is used that does not introduce (additional) heavy metals. Small samples are grinded in a blender with a glass beaker, using titanium or zirconium knives. For some sugar holding products (e.g. fruits) the grinded material may stick to the blender beaker. It may help to freeze these products in liquid nitrogen immediately, prior to grinding with a mortar and pestle.

9.4 Proxy plant

For convenience, in an ideal situation measurement activities should focus on a “proxy” plant, for which accumulation represents the accumulation for a “representative consumption pattern”. Measuring the accumulated contaminants in such a plant, which can be a vegetable or non-edible plant, could improve the simplicity and the quality of the measurement of the accumulated concentration in vegetables. Requirements for a suited proxy plant are:

- Representing the appropriate accumulation rate (e.g. for a “representative consumption pattern”).
- Present during the whole year (e.g. grasses).
- Easy to grow and manage.

At this moment no such plant is known. It is recommended to focus research on a proxy plant, in the future.

9.5 A seeding, growing harvesting program

9.5.1 A field program

In situations were no suitable crops are growing, or no crops are growing at all, vegetables could be cultivated in a seeding, growing, harvesting program. The disadvantage of this procedure is that the program is time consuming and the results will only be available at the end of the growing season. It is a case by case decision if the effort is worth the value of measured data. No detailed protocol is described in this section. However, guidance is given with the purpose to optimize the seeding, growing, harvesting program.

9.5.2 Vegetable selection

The big advantage of a seeding, growing, harvesting program is that the type of vegetables can be chosen. A representative vegetable package could be seeded, but this is a rather extended activity. Alternatively one or two vulnerable vegetables could be grown, i.e. vegetables that show a high affinity for uptake. In analogy with Wegener-Sleeswijk and Kleijn (1993) attention could be focused on spinach and curly kale for cadmium, endive and curly kale for lead and spinach and endive for mercury. In any situation is it recommended to grow crop species of regional importance.

Preferably, the seeding, growing, harvesting program is performed at the site under investigation. However, this is only possible within a specific time of the year. In general it is possible to sow plant between March and October (see Table 9.3 for a sowing time calendar; *http://www.devolkstuin.nl/tuin*). Besides, row distance and distance between crops within a row have been specified in this table.

Table 9.3: Sowing time calendar, row distance and distance between crops within a row

Sowing time calendar	J	F	M	A	M	J	J	A	S	O	N	D	Row distance [cm]	Distance within rows [cm]
Strawberry													45cm	45cm
Endive													30cm	30cm
Artichoke													100cm	80cm
Egg Plant													45cm	45cm
Gherkin													100cm	35cm
Beet root													35cm	10cm
Chicory													30cm	10cm
Large-leaved chicory													30cm	10cm
Chervil													25cm	
Cucumber													100cm	15cm
Cauliflower													60cm	60cm
Broccoli													45cm	45cm
Curly kale													45cm	45cm
Chinese cabbage													30cm	15cm
Red Cabbage													60cm	60cm
Savoy cabbage													60cm	60cm
White cabbage													60cm	60cm
Sprout cabbage													60cm	60cm
Turnip													45cm	45cm
Rutabaga													35cm	25cm
Maize													50cm	30cm
Melon														
Capsicum (Paprika)													50cm	35cm
Chili													50cm	35cm
Parsley													30cm	
Pumpkin													100cm	15cm
Purslane														
Leek													30cm	10cm
Turnip tops														
Radish													25cm	
Winter radish													30cm	10cm
Field mustard													30cm	10cm
Black salsify													30cm	15cm

Sowing time calendar	J	F	M	A	M	J	J	A	S	O	N	D	Row distance [cm]	Distance within rows [cm]
Celeriac													30cm	30cm
Blanched celery													30cm	30cm
Celery													25cm	
Romaine lettuce													30cm	30cm
Head lettuce													30cm	30cm
Loose-leaf lettuce													30cm	
Cut lettuce													30cm	
Lamb's lettuce													30cm	
Beet greens													30cm	
Spinach													25cm	
Tomato													45cm	45cm
Garden cress														
Onion													30cm	10cm
Carrot													35cm	
Sorrel													25cm	
Slicing beans														
Green beans													40cm	10cm
Broad bean													60cm	15cm
Garden peas													40/60cm	7cm
		Sowing in cold frame												
		Sowing outside												

For most crops several sub-species and varieties are commercially available (e.g., www.vreeken.nl). For the natural flora, the selection can be based on the seed list of the KNNV (www.knnv.nl). Select a regionally preferred variety for which the sowing period matches the time of year. A sowing time calendar (e.g., www.devolkstuin.nl/tuin) may help in the selection of a proper variety of crop species.

If needed, the growing season can be lengthened by cultivation in a cold frame. When the seeding, growing, harvesting program needs to be initiated outside the growing season, the cultivation can take place in the laboratory on sampled soil. The cultivated crops should be harvested and analyzed in the same manner as crops grown on the field.

9.5.3 Soil management

Soil fit for sowing is well drained and loose. Surplus deposition should be able to drain quickly. In impervious soil it is not possible to grow any crop. The soil should be turned over one spade deep to get a loose texture. During soil turn over, the soil conditions can be altered. If the soil organic matter content is very low (< 2%), clay soils should be mixed with some garden peat, while sandy soils should be amended with organic fertilizer.

Depending on the type of crop, there are two ways to transfer the seeds to the soil. Broad-cast sowing, where the seeds are evenly spread. Direct seeding in lines, where the seeds are put on seeding lines with some distance between them. With the latter method, weeds are easier to remove. Fine seeds are sown very shallow or pressed on the soil the surface. Courser seeds can be sown to a depth of maximal 1 – 2 cm. At the stage where the seedlings shed their seed leaves, they need to be thinned out. The proper distance between the seedlings is always printed on the seed package. The remaining seedlings are firmed in the soil and individually watered. Thinning is not needed for all seedlings. This is also stated on the seed package. Use wind screens to protect the crop against the wind. Cold frames or plastic cloches can also be used to lengthen the growing season by sowing early in the year. A cold frame looses less heat during winter and can be kept frost free by applying insulation materials.

10. Basis of the protocol

10.1 Layout

The proposed tiered approach is laid out as follows (see Figure 10.1): Tier 0 concerns a qualitative evaluation of the possibilities for vegetable consumption, the so-called preliminary qualitative evaluation. In Tier 1, concentrations in soil (average or relatively high values) are compared with Critical soil concentrations. Critical soil concentrations only are incorporated for cadmium. Tier 2 offers the possibility for site-specific calculation. This calculation is supposed to be “realistic worst case”. The procedure differs for the site-specific calculation of accumulation of metals, other inorganic contaminants or organic contaminants in vegetables. Finally, in Tier 3, a standardized measurement protocol has been developed. In this protocol representative vegetables are sampled, for which the edible parts of the plants are treated in the laboratory in analogy with standard cooking preparation.

The scientific basis for the assessments in each tier have been described in more detail in chapter 4 to 9.

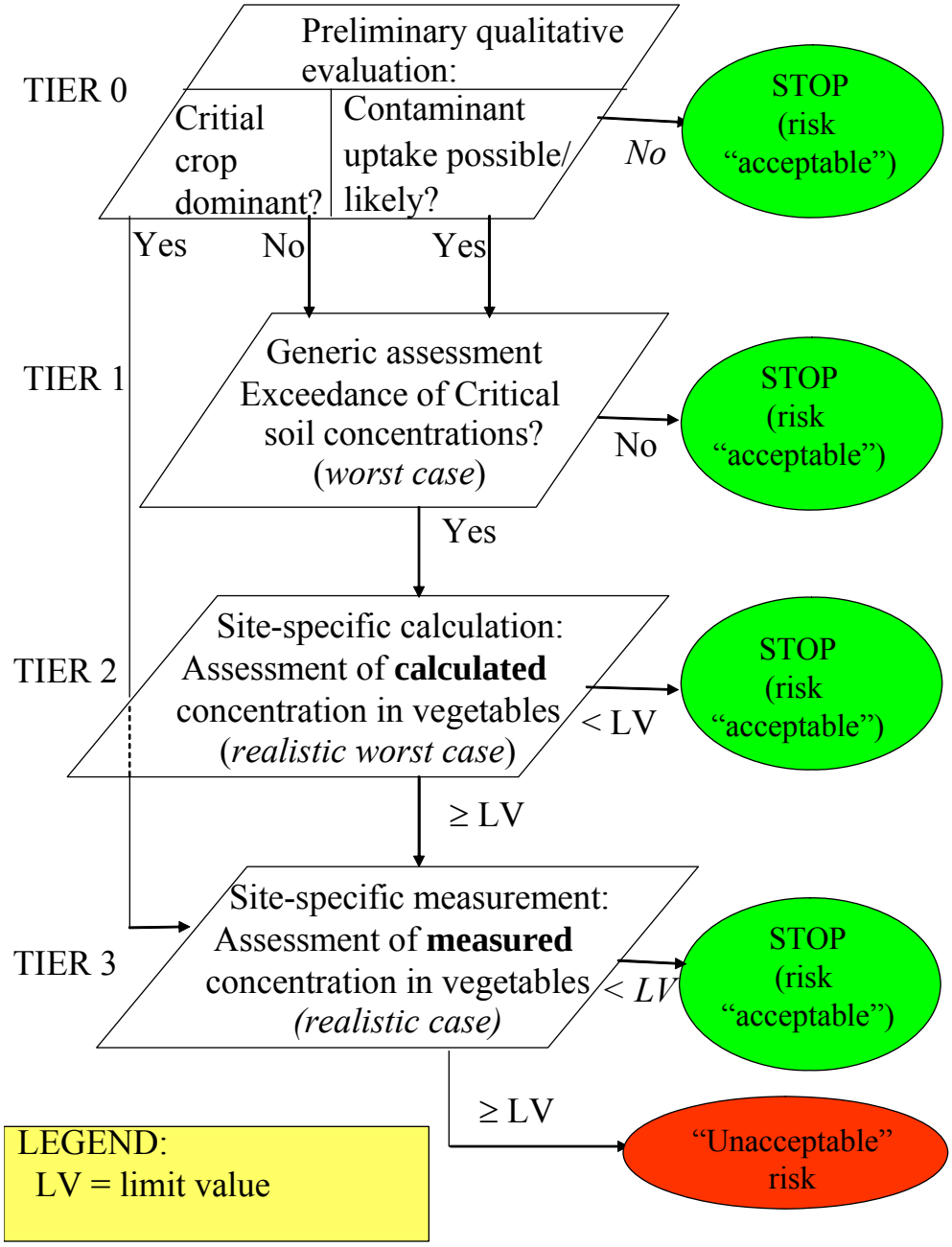

Figure 10.1: The layout of the proposed tiered approach to assess the risks of vegetable consumption from contaminated sites.

10.2 Preliminary qualitative evaluation (Tier 0)

The following aspects determine the need for a more detailed assessment in higher tiers:

- situation of the site;
- type of contaminant;
- sensitivity of the vegetables.

There are two situations in which it is unlikely that vegetable consumption from contaminated sites results in adverse health effects, independent of the soil concentration:

- The site or the type of soil use does not allow the cultivation of vegetables. This could be the case, for example, when the contaminated site is situated in an industrial area or concerns a residential garden next to a traffic intersection.
- The contaminant concerns an essential metal, which humans need for a proper physical functioning and for which it is unlikely, even in the case of a vegetable garden, that intake via vegetable consumption contributes to risks to human health. Besides, the concentrations in vegetables that are critical for humans would be so high that the vegetables would not survive. At this moment *zinc* is considered to meet these criteria.
  Also for *free CN* it is highly unlikely that human health risks occur due to vegetable consumption. This contaminant is also excluded from risks to human health.

When either of these two situations applies it can be concluded that no unacceptable human health risks exists. In any other case an unacceptable human health risk can not be excluded and Tier 1 has to be performed.

Leafy vegetables like spinach, endive or broccoli show relatively high metal uptake rates. When it can not be excluded that these vegetables are the dominant vegetables on a contaminated site (for example when it can not be excluded that more than half of the crops concern one or more of these vegetables) risk of vegetable consumption is relatively high, even at low contaminant levels. In that case a Critical soil concentration (Tier 1) is not applicable and calculation of actual concentrations in vegetables (Tier 2) not reliable. As a consequence Tier 1 and 2 must be skipped and Tier 3 (measuring the concentration of contaminants in vegetables in the field) is appropriate when these vegetables are present on the site.

10.3 Critical soil concentrations (Tier 1)

The procedure for derivation of a Critical soil concentration has been described in section 2.2. However, no Critical soil concentrations have been derived on the basis of an exposure calculation, since the calculated BCFs for cadmium are not reliable for sensitive soil (low organic matter and clay contents). Instead, the Critical soil concentrations for the Dutch Kempen region have been adopted, as explained in section 5.6. The following Critical soil concentrations resulted:

- 0.5 mg/kg_{dw}: when this concentration is not exceeded cultivation of vegetables is possible, independent of soil properties, without unacceptable risks to human health.

- between 0.5 and 2.0 mg/kg_{dw}: when the pH(H_2O) of the soil is at least 5.6 and the percentage organic matter at least 5%: in this concentration range cultivation of crops is possible, without unacceptable risks to human health.

When these conditions are not met, Tier 2 has to be performed.

10.4 Site specific calculation (Tier 2)

Tier 2 offers the possibility for site-specific calculation. This calculation is supposed to be "realistic worst case". Obviously, the procedure differs for the site-specific calculation of accumulation of metals, other inorganic contaminants or organic contaminants in vegetables. The assessment in Tier 2 is based on comparison of the realistic worst case exposure with the reference dose (MPR_{human}). To this purpose, the resulting concentrations in vegetables must be included in a CSOIL calculation. It is essential to include exposure due to soil ingestion in most cases, because hand-mouth contact is relatively intensive during gardening. When exposure does not exceed the MPR_{human} it can be concluded that no unacceptable risk exists. In any other case Tier 3 has to be performed.

In case of commercial vegetable production, the calculated contaminant concentrations in individual vegetables are additionally compared to appropriate food quality criteria.

The exposure scenarios are related to actual soil-use, i.e. "Residential with garden" or "Vegetable garden". This means that either the consumption rates of the general population in combination with a 10% contribution of vegetable consumption from home-grown vegetables is used ("Residential with garden"), or consumption rates of gardeners in combination with a 50% (potatoes), or 100% (other vegetables) contribution ("Vegetable garden") of home-grown vegetables to total consumption. In specific situations the risk assessor can use deviant data for contribution of vegetable consumption from home-grown vegetables. This has to be motivated. The consumption rate and pattern is based on an average consumption pattern (see section 4.3).

The calculation of the metal concentration in vegetables is based on the plant – soil relations as described in chapter 5. The calculation of the concentration of other inorganic contaminants in vegetables is similar to the present approach and is described in chapter 6. The calculation of the concentration of other organic contaminants in vegetables is based on the adapted Trapp and Matthies model, as described in chapter 7.

Besides, these calculation procedures are suited to improve the model algorithms for the exposure pathway "exposure due to vegetable consumption" in the CSOIL exposure model must be improved (the additional objective as formulated in section 1.1)

10.5 Measurement (Tier 3)

The measurement protocol in chapter 9 must be followed. The resulting concentrations in vegetables must be included in a CSOIL calculation. It is advised to replace each calculated BCF for a vegetable group by the geometric mean of the measured BCF in the vegetables of that vegetable group. As a consequence, the BCF is a combination of measured and calculated values, unless vegetables of all vegetable groups are measured.
Again, exposure due to soil ingestion must also be included in the exposure calculation. Subsequently, exposure must be compared to the Reference dose (MPR_{human}). When the MPR_{human} is exceeded there is an unacceptable risk for human health.

Again, in case of commercial vegetable production, the measured contaminant concentrations are additionally compared to appropriate food quality criteria.

11. Conclusions and recommendations

11.1 Conclusions

To be able to assess the human health risks of vegetable consumption from contaminated sites in a scientifically-based and efficient way a tiered approach has been developed. Ultimately, the protocol should be applicable for vegetable gardens, residential situations with garden that offer the possibility of home-grown vegetables and undeveloped or fallow sites that will be taken into development. Besides, this report offers the possibility to improve the model algorithms for the exposure pathway “exposure due to vegetable consumption” in CSOIL 2000. The protocol focuses on metals, other inorganic contaminants and organic contaminants.

Successively, in each tier the degree of conservatism decreases, while site-specificism increases. As a consequence, complexity and hence effort and finances needed also increase in each tier. When in a specific tier an unacceptable human health risk can not be rejected the assessment in the following tier has to be performed. The underlying principle is: simple when possible and complex when necessary.
The tiered approach is laid out as follows: Tier 0 concerns a preliminary qualitative evaluation of the *possibilities* for experiencing adverse human health effects due to vegetable consumption. In Tier 1 the actual total soil concentrations (average or relatively high values) are compared with *Critical soil concentrations* (for cadmium only). These Critical soil concentrations have been derived on the basis of a conservative exposure scenario. Tier 2 offers the possibility for *a detailed assessment of the site-specific risks on the basis of calculation*. Obviously, the site-specific calculation of the contaminant concentration in vegetables differs for metals, other inorganic contaminants and organic contaminants. For metals Freundlich-type plant - soil relations (dependent of the total soil concentration and the major soil properties) and geometric means of the BioConcentrationFactors (corrected for the actual organic matter and clay contents) are combined. This procedure does not account for the presence of specific matrixes (like debris, porcelain, or bullets), or phytotoxicological limit values in vegetables as the upper limits to what humans can be exposed. The accumulation of other inorganic contaminants is based on passive uptake. The calculation of the concentration of organic contaminants in vegetables is based on an adapted Trapp and Matthies model. In this model the partitioning of contaminants between pore water and roots and subsequently translocation to the upper plant parts is calculated, resulting in the contaminant concentration in the above-ground plant parts. Finally, in Tier 3, *a standardized measurement protocol* has been developed. This protocol allows for sampling of a significant number of representative vegetables in the field, for which the edible parts of the plants are treated in the laboratory in analogy with standard kitchen preparation. Subsequently, the

measured concentration can be used in an exposure calculation and, when appropriate, compared to acceptable concentrations in vegetables.

The most important recommendations for future research are extension of the dataset for metals (specifically for cadmium, lead, arsenic, mercury, nickel, barium and molybdenum) and the performance of a comprehensive validation study.

Contribution to Dutch soil policy

The procedure to assess the site-specific human health risk assessment for consumption of vegetables from contaminated sites can be used to support planning or soil management in relation to the soil uses “Vegetable garden” and “Residential sites with garden”. Moreover, the protocol should be incorporated in the general procedure on the (tiered) procedure to assess the site-specific human health risks due to exposure to contaminated sites. This general procedure will be included in the technical basis of the Dutch Soil Protection Act, which act is presently under revision. New applications in the Netherlands concern the Remediation criterion and the Local Ambitions for soil quality.

11.2 Recommendations

A distinction has been made in recommendations for the short term and the long term.

11.2.1 Short term

In the short term this basis for a protocol must be turned into a user-friendly protocol, in the framework of the revision of the Dutch Soil Protection Act. Probably it will be incorporated into a protocol to assess the site-specific risk for human health from a wider perspective. This protocol will replace the present standardized procedure to assess the site-specific risk for human health as included in the procedure to determine the urgency of remediation. New applications concern the Remediation criterion and the Local Ambitions for soil quality.

11.2.2 Long term

In the longer term research should focus on the following topics:

Inter-plant variation:

- The interspecies variation (variation in accumulated contaminant concentrations in plants of the different plant types) versus the intraspecies variation (variation in accumulated contaminant concentrations in different plants of the same plant types).
- The variation in accumulated concentrations in non-edible plants versus the variation in accumulated contaminant concentrations in vegetables.

Calculation of accumulation concentrations in vegetables:

- Improvement of the plant – soil relations for cadmium, lead and arsenic, by extending the dataset by combining the RIVM plant – soil database with other existing datasets and with more recent data from the literature.
- Investigation of the relation between the arsenic concentration in vegetables and soils, focusing on the (lack of) influence of the arsenic concentration in soils on the arsenic concentration in vegetables.
- Extension of the plant – soil database, specifically for cobalt, barium, mercury, molybdenum and nickel, with the purpose to derive more plant – soil relations for these metals.
- The (time span for the) limited availability of metals in matrixes like debris, porcelain, or bullets (metals) or tar (organic contaminants).
- A more extended investigation on the calculation of the accumulated concentration of other inorganic contaminants in plants.
- The evaluation of the feasibility of the concept for calculating the concentration of organic contaminants in root vegetables for more contaminants, also including contaminants with a log K_{ow} higher than 7.
  The evaluation of the feasibility of the concept for calculating the concentration of organic contaminants in above-ground plant parts, which is known to often underestimate BCF-values.
- Implementation of phytotoxicological limit values in vegetables, as the upper limits to what humans can be exposed.

Measuring contaminant concentration in plants:

- The use of a “proxy” plant, which can be a vegetable or non-edible plant, which represents the ideal accumulation pattern. Such plant could improve the simplicity and improve the quality of the measurement of the accumulated concentration in vegetables.
- The potential for and the identification of the most appropriate extraction methods for assessing the “bioavailable” contaminant concentration in soils that relates to the accumulated concentration in plants.

Exposure to contaminants due to vegetable consumption:

- The fraction of vegetables that is home-grown in the Netherlands.
- The possible preclusion of human health risks for copper due to vegetable consumption.

Last but not least an extended validation study is recommended for metals, other inorganic contaminants and organic contaminants, in which the results from different tiers must be compared with measured concentrations in vegetables.

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