A risk-based methodology for deriving quality standards for organic contaminants in sewage sludge for use in agriculture—Conceptual Framework

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Abstract

This paper describes a systematic methodology (Conceptual Framework) to derive quality standards for organic (anthropogenic) contaminants in sewage sludge added to agricultural land, in the context of revision of EU Sludge Directive 86/278/EEC and the broader Soil Thematic Strategy. The overall objective is to ensure, based on a risk assessment approach, a sustainable use of sludge over a long time horizon. ILSI-Europe’s Conceptual Framework is in essence consistent with the EU Technical Guidance Document (TGD) for Environmental Risk Assessment of Chemicals in the soil compartment, or US-EPA’s Sewage Sludge Use and Disposal Regulations, Part 503 Standards. A ‘checklist’ of different exposure pathways and transfer processes for organic contaminants needs to be considered, and the most sensitive relevant toxicological endpoint and its PNEC need to be identified. The additional complexity specific to deriving Sludge Quality Standards (SQS) is that the toxicity results may need—e.g., for (indirect) human toxicity—to be related back to maximum acceptable soil exposure levels (PEC soil). In turn, the latter need to be back-calculated to the maximum acceptable levels in sewage sludge (PEC sludge) at the time of application. Finally, for a sustainable sludge use, the exposure from repeated addition and potential chemical build-up over time (e.g., 100 years) needs to be assessed. The SQS may therefore vary with the (local) sludge application regime, and/or sludge pretreatment processes.

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1. Introduction

This paper is concerned with the development of a systematic and consistent risk-based methodology to

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PEC—predicted environmental concentration)\(^1\) against effects (i.e., PNEC—predicted no effect concentration for the ecosystem). The equivalent in human risk assessment would be the comparison of an acceptable daily intake (ADI—in mg/kg bodyweight day) over exposure (in mg/kg bodyweight day) (ILSI, 2000).

This document does not address other sludge management issues, such as those related to metals, nutrients, pathogens or radionuclides, which have been extensively covered elsewhere (e.g., Andersen, 2001b; Jones, 2003; Langenkamp et al., 2001; Matthews, 1996; Smith, 1996; Vesilind, 2003). Its scope is the specific issue of (anthropogenic) organic chemicals. The existing EC Directive (86/278/EEC) on the use of sewage sludge in agriculture (CEC, 1986), currently in the process of revision, provides limit values for heavy metals in soils and sewage sludges, but new provisions on concentration limits for organic compounds in sludge may be introduced. The latest EC Working Document on Sludge (3rd draft; CEC, 2000c) on the revision of the Directive (86/278/EEC) proposes Limit Values for: AOX (adsorbable organic halogens), LAS (linear alkylbenzene sulphonate), DEHP (diethylhexylphthalate), NPE (nonylphenol ethoxylates), PAH (polynuclear aromatic hydrocarbons), PCB (polychlorinated biphenyls), and dioxins/furans (PCDD/PCDF).

Spreading sewage sludge on farmland of an acceptable quality will remain the best practicable environmental management option for this material. Acceptance by agriculture and society at large will depend on controlling and further reducing pollutant levels, in order to improve the quality and public image of sludge (Andersen, 2002a). The application of sewage sludge to agricultural land, however, implies exposure of soil microbial, plant, and animal life—including humans—to contaminants contained within the sludge. Knowledge of the possible effects of such contaminants upon soil fertility, its biota, the water cycle, the food chain, and human populations are necessary to determine acceptable levels of contaminants within sludge destined for use in agriculture (Andersen, 2002b). Objective safety thresholds can only be determined via the characterization of risks, i.e., the benchmarking of exposure against effects. Risk based standards will apply primarily to soil per se rather than the added sewage sludge. Expression of these standards within sludge necessarily implies consideration of sludge application practices, contaminant fate in sludge and in soil, and a time horizon for the assessment (Fig. 1).

The focus of this paper is on the overarching principles of sludge and soil risk assessment, rather than on technical or methodological details. Some methodological aspects in risk assessment are still under debate, and risk assessment science is progressing continuously (see e.g., ECETOC, 2003). Rather, reference is made to a ‘toolbox’ of state-of-the-art methods and models in the literature and/or the chemical legislation, which should allow the reader to address the different sub-elements of the assessment in scientifically sound way. Also, a number of excellent studies related to sludge contamination statistics, safety and policy issues can be found e.g., at http://europa.eu.int/comm/environment/waste/sludge/index.htm, and will not unduly be repeated here.

The paper also does not intend to deal with the potentially important managerial, logistic and economic consequences of setting Sludge Quality Standards (SQS) in a given country, or for the entire EU. It is essential that a cost/benefit evaluation accompanies the eventual setting of SQS, but this falls within the domain of risk management and policy making. As part of the risk management process the potential drawbacks and environmental impacts associated with alternative fertilizer options, i.e., farm manure or synthetic fertilizer, need to be included for a balanced comparison.

The revision of the Sludge Directive is currently positioned by the EU Commission as part of a broader ‘Soil Thematic Strategy.’ In addition, setting SQS for organic

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**Fig. 1. General principle for deriving risk-based Sludge Quality Standards.**

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\(^1\) Abbreviations used: ADI, acceptable daily intake; AOX, adsorbable organic halogens; BAF, bioaccumulation factor; BCF, bioconcentration factor; BTF, biotransfer factor; CF, conceptual framework; DEHP, diethylhexylphthalate; DM, dry matter; DOC, dissolved organic carbon; EC\(_{50}\), effect concentration 50% (in a toxicity test); EFSA, European Union System for Evaluation of Substances; HPV, high production volume; FAME, fatty acid methyl ester; (log)\(K_{ow}\), octanol/water partitioning coefficient; LAS, linear alkylbenzene sulfonate; LC\(_{50}\), lethal concentration 50% (in a toxicity test); LD\(_{50}\), lethal dose 50% (in a toxicity test); LOEC, lowest observed effect concentration; MOE, margin of exposure; MSW, municipal solid waste; NOEC, no observed effect concentration; NPE, nonyl phenol ethoxylates; NVZ, nitrate vulnerable zone(s); PAH, polynuclear aromatic hydrocarbons; PCB, polychlorinated biphenyls; PCDD, polychlorinated dibenzo-p-dioxins; PCDF, polychlorinated dibenzo-furans; PEC, predicted environmental concentration; PLFA, phospholipid fatty acid; PNEC, predicted no effect concentration; RCF, root concentration factor; R/ID, reference dose; SSD, species sensitivity distribution; SQS, sludge quality standard(s); TGD, EU Technical Guidance Document (on risk assessment of chemical substances); TVC, total viable counts.
compounds only makes sense if other sludge quality issues such as metals, nutrients or pathogens are not the limiting factor for sludge use.

This paper is structured as follows: Section 1 provides general background information on sewage sludge, the practices and the regulatory environment of sludge use in agriculture in Europe. Section 2 offers a short review of typical contaminant levels, and the transfer processes by which they can lead to exposure of the ecosystem, cattle, and humans. Section 3 defines the endpoints that require protection through risk assessment, and some methodological guidance. The whole of the information is then schematized in Section 4 into a Conceptual Framework and a stepwise procedure for a holistic risk assessment of sludge amended soils.

1.1. What is sewage sludge?

The composition of sewage sludge is described, e.g., by Rogers (1996) and Andersen (2001b). It is a by-product of the wastewater treatment process and is essentially settled organic and inorganic solids from the sewage, phosphorus precipitates, as well as the biomass formed during aerobic, anoxic, and/or anaerobic degradation processes. The sludge can also contain inorganic and organic process additives. Organic materials constitute 40–80% by dry weight, depending on the extent and type of sludge treatment. The major organic loading in domestic sewage is from human faecal material, although in industrial catchments, certain industries can add considerably to the organic loading.

The organic fraction of sludge is a mixture of fats, proteins, carbohydrates, lignin, amino acids, sugars, celluloses, humic material, and fatty acids. Live and dead micro-organisms constitute a large proportion of the organic material and provide a large surface area for sorption of lipophilic organic contaminants in the sludge. The properties of sludge are dependent upon its origin and treatment type. It differs in its physical (processability and handleability), chemical (presence of nutrients and contaminants), and biological parameters (microbial activity and presence of pathogens). The characteristics of the sludge in terms of contaminant loading is dependent upon the original pollution loading of the sewage (i.e., domestic, industrial, and mixed) and the type of wastewater treatment the sewage and sludge have received. In many cases some form of post-treatment is applied to the sewage sludge (e.g., anaerobic digestion or aerobic composting), and this can have a major impact on residual contaminant levels in case these are biodegradable.

1.2. Sewage sludge production within the EU

Wastewater treatment within the EU has greatly increased in recent years due to the implementation of the Urban Waste Water Treatment Directive 91/271/EEC and its update 98/15/EC (CEC, 1991b, 1999). Full implementation is required by 2005 and an increase of 69% in treatment capacity in population equivalents has been predicted relative to 1992 (CEC, 1999) and this will have a corresponding impact upon the quantity of sewage sludge produced within the EU, all of which will require disposal (Table 1). The EC predicts that total sewage sludge production within EU15 will reach 9 million tonnes dry solids (tds) per year in 2005, compared with 5.5 million tds in 1992, an increase of 64%. Other sources forecast 10.7 million tds in 2005 (EEA, 1998).

1.3. Current and proposed regulation of sludge use in agriculture

The policy of the European Commission is to encourage the beneficial use of sewage sludge on land, provided that the quality of sludge is compatible with public health and environmental protection requirements, as it represents an apparent long-term sustainable solution to sludge disposal (CEC, 1999).

Within the EU, the use of sewage sludge on agricultural land is controlled under Directive 86/278/EEC. The directive in its current form aims principally at controlling the accumulation of heavy metals in soils following sludge application. Sludge treatment standards are not clearly defined, particularly with regard to pathogen reduction, and organic contaminants are not presently addressed. All EU Member States have implemented the Directive through national legislation. The legislative framework at the EU level, and within individual Member States, was reviewed in Andersen (2001a) and Andersen (2001b).

The Sludge Directive (CEC, 1986) requires Member States to control the addition of heavy metals to soil,

Table 1

<table>
<thead>
<tr>
<th>Member State</th>
<th>Total sludge production (tds/years × 10³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Austria</td>
<td>195</td>
</tr>
<tr>
<td>Belgium</td>
<td>159</td>
</tr>
<tr>
<td>Denmark</td>
<td>200</td>
</tr>
<tr>
<td>Finland</td>
<td>160</td>
</tr>
<tr>
<td>France</td>
<td>1172</td>
</tr>
<tr>
<td>Germany</td>
<td>2787</td>
</tr>
<tr>
<td>Greece</td>
<td>99</td>
</tr>
<tr>
<td>Ireland</td>
<td>113</td>
</tr>
<tr>
<td>Italy</td>
<td>959</td>
</tr>
<tr>
<td>Luxembourg</td>
<td>14</td>
</tr>
<tr>
<td>Netherlands</td>
<td>401</td>
</tr>
<tr>
<td>Portugal</td>
<td>359</td>
</tr>
<tr>
<td>Spain</td>
<td>1088</td>
</tr>
<tr>
<td>Sweden</td>
<td>323</td>
</tr>
<tr>
<td>UK</td>
<td>1583</td>
</tr>
<tr>
<td>EU15</td>
<td>9611</td>
</tr>
</tbody>
</table>

tds, tonne dry solids.
and sets maximum soil Limit Values. The rate of accumulation may be controlled by a combination of sludge quality standards and limits on sludge dry solids, or by the quantity of heavy metals that may be applied per year (calculated on a 10 year ‘rolling average’). In implementing the Directive, most Member States have set national limits for sludge and soil quality at lower levels than the addition maxima permitted in the Directive. Table 2 summarizes the specific National regulations on sludge addition rate (Andersen, 2001a). Unlisted countries follow the general requirements of the EU Directive 86/278/EEC. In most EU countries the maximum allowable sludge addition rate is around 5 tonnes DM/ha year. In daily practice, however, quantities used on cropland usually do not exceed 2–3 tonnes/ha year (Andersen, 2001a; ECETOC, 2003). Also, application frequency is not necessarily annual.

The EC Working Document on Sludge (CEC, 2000c) proposes to make sludge quality limits mandatory and reduce all of the Limit Values for heavy metals in sludge and soil, with the objective to protect the long-term sustainability of soils in Europe and with the aim to encourage further source reductions in heavy metal discharges to sewer. The EC has commissioned studies to determine appropriate soil quality standards, but the currently proposed values will prevent sludge application to some acid soils due to their natural geochemical composition.

General restrictions on nutrient additions apply in most EU Member States, particularly for nitrogen, because of the potential risk of nitrate leaching and potential impact on surface and groundwater quality. In some states, phosphate additions are also controlled due to a limited adsorption capacity of some soils and risks to water resources. Such controls apply to all sources of nutrients used by the farmer, including sludge. The EC Nitrates Directive 91/676/EEC (CEC, 1991a) imposes a limit on the total nitrogen loadings in designated nitrate vulnerable zones (NVZs). These zones may encompass all of some countries but only limited portions of others, according to whether water quality exceeds, or is at risk of exceeding, the limit value of 50 mg NO₃⁻/L. The maximum annual rate of nitrogen addition within NVZs is 170 kg N/ha.

A principle of the Directive is that nutrient additions from sludge must not exceed crop requirements. Since heavy metal concentrations have progressively reduced in all Member States over the last 20–30 years, these rarely restrict sludge application rates, which are now generally limited by nutrient additions. However, the proposed revision may again make some heavy metals addition rates more restrictive than nutrient additions, which may impact the agronomic value of sludge application as realised by the farmer.

Only a few Member States place additional limit concentrations on some organic contaminants in sludge, principally due to concerns for the potential transfer to the human diet. As a consequence, some Member States have prohibited the application of sludge to grassland due to the potential for grazing animals to directly ingest sludge solids with the possible risk of transfer of organic contaminants into the human food chain through milk and meat. The current EC Working Document (CEC, 2000c) proposes Limit Values for a range of classes of organic contaminants in sludge. It must be remarked that the selection of chemicals and associated Limit Values is essentially based on a compilation of current legislation in a number of EU Member States, and is not the result of a systematic and common assessment procedure.

The requirements for sludge treatment in the current Directive are very general and non-specific, although crop and grazing restrictions are included. The use of untreated sludge is permitted if immediately worked into the soil or is injected into the soil. The EC Draft Working Document proposes sludge treatment conditions for conventional treatment and introduces a new standard of ‘advanced treated sludge’ in which the sludge is effectively disinfected. More stringent crop restrictions would apply where conventional treated sludge is spread, but there would be no crop restrictions on the use of advanced treated sludge products.

In the USA sludge amendment to agricultural land, incineration or sanitary landfill. There are a number of minor outlets, such as land reclamation, forestry, landfill covering and as soil amendment after compo-

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Table 2

<table>
<thead>
<tr>
<th>Country</th>
<th>Sludge application rate in tonnes DM per hectare</th>
</tr>
</thead>
<tbody>
<tr>
<td>Austria</td>
<td>2.5–10 tonnes in 2 years</td>
</tr>
<tr>
<td>Belgium (Flanders)</td>
<td>4 tonnes in 2 years (arable land)</td>
</tr>
<tr>
<td>Belgium (Walloons)</td>
<td>2 tonnes in 2 years (pasture land)</td>
</tr>
<tr>
<td>Belgium (Walloons)</td>
<td>12 tonnes in 3 years (arable land)</td>
</tr>
<tr>
<td></td>
<td>6 tonnes in 3 years (pasture land)</td>
</tr>
<tr>
<td>Denmark</td>
<td>10 tonnes per year</td>
</tr>
<tr>
<td>Germany</td>
<td>5 tonnes in 3 years</td>
</tr>
<tr>
<td>Ireland</td>
<td>2 tonnes per year</td>
</tr>
<tr>
<td>Italy</td>
<td>15 tonnes in 3 years</td>
</tr>
<tr>
<td>Luxembourg</td>
<td>3 tonnes per year</td>
</tr>
<tr>
<td>The Netherlands</td>
<td>2–4 tonnes per year (arable land)</td>
</tr>
<tr>
<td></td>
<td>1–2 tonnes per year (grassland)</td>
</tr>
<tr>
<td>Portugal</td>
<td>6 tonnes per year</td>
</tr>
</tbody>
</table>

Unlisted European countries have no additional limits on application rates, the sludge use is only regulated by conditions and limits according to European Directive 86/278/EEC.

a Depending on the land, the dw content, and the sludge type.

b Depending on the sludge structure (liquid or solid type).
The disposal of sludge to sea is prohibited by the Urban Waste Water Treatment Directive (91/271/EEC) (CEC, 1991b). Methods of sewage sludge treatment and disposal are reviewed by EEA (1998), and practices within individual EU Member States have been reviewed periodically, for example in Hall and Dalimer (1994); Matthews (1996); ADEME (1999); CEC (1999); Andersen (2001b); and Frost and Sullivan (2003).

The proportions of sludge being used or disposed of to different outlets in each Member State are shown in Fig. 2 (ADEME, 1999). There are marked differences between countries in the percentage of sludge used on land compared with that disposed of to landfill and incineration, reflecting differences in conditions and attitudes in different countries.

These disposal patterns are not static and evolve in response to changes in legislative and policy at national and EC levels (see Fig. 3). The most significant of these has resulted in a decline in use of landfill disposal for organic wastes such as sludge, and a corresponding increase in the use incineration. By 2005, incineration is expected to exceed landfilling (CEC, 1999) and may account for around 23% of European sewage sludge disposal by 2009 compared with about 14% in the mid-1990s (Frost and Sullivan, 2003). However, the use of sludge on land, principally in agriculture, will continue to be the most important outlet in Europe, accounting for about 50% of sludge production.

Controlled local use of sludge on agricultural land is generally the preferred option since it can offer the following benefits:

- Improved in soil fertility by adding nutrients (nitrogen and phosphorus) and micronutrients (zinc, copper, etc.), with benefits to crop growth and production.
- Reduced use of fertiliser, saving the farmer money as well as having broader environmental and resource conservation benefits (e.g., limited global reserves of phosphate minerals).
- Sludge application improves the structure of the soil via incorporation of organic material into soil humus. It has positive effects on several other physical properties of the soil such as improvement of water retention capacity and reducing erosion risks, which are particularly important in Mediterranean climates (Albiach et al., 2001; Matthews, 1996);
- Sludge application to land recirculates nutrients for crop production. According to Lansink’s waste hierarchy ladder (VROM, 2001), sludge use in agriculture is therefore ‘recycling’ rather than ‘disposal.’
- Agricultural application is attractive to the wastewater utilities as it is a relatively inexpensive solution (Andersen, 2002a; EEA, 1998). Other options are generally unsustainable (landfill disposal where capacity is scarce), or more expensive (incineration).
Table 3
Overview of sludge pre-treatment processes according to CEC (2000c)

<table>
<thead>
<tr>
<th>Process</th>
<th>Descriptions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Advanced treatments (hygienisation)</td>
<td></td>
</tr>
<tr>
<td>Thermal drying</td>
<td>Ensuring that the temperature of the sludge particles is higher than 80°C with a reduction of water content to less than 10% and maintaining a water activity above 0.90 in the first hour of treatment</td>
</tr>
<tr>
<td>Thermophilic aerobic stabilization</td>
<td>At a temperature of at least 55°C for 20h as a batch, without admixture or withdrawal during the treatment</td>
</tr>
<tr>
<td>Thermophilic anaerobic digestion</td>
<td>At a temperature of at least 53°C for 20h as a batch, without admixture or withdrawal during the treatment</td>
</tr>
<tr>
<td>Thermal treatment of liquid sludge</td>
<td>For a minimum of 30 min at 70°C followed by mesophilic anaerobic digestion at a temperature of 35°C with a mean retention period of 12 days</td>
</tr>
<tr>
<td>Conditioning with lime reaching a pH of 12 or more</td>
<td>And maintaining a temperature of at least 55°C for 2h</td>
</tr>
<tr>
<td>Conditioning with lime reaching and maintaining a pH of 12 or more</td>
<td>For three months</td>
</tr>
</tbody>
</table>

The process shall be initially validated through a $6 \log_{10}$ reduction of a test organism such as *Salmonella senftenberg* W 775. The treated sludge shall not contain *Salmonella* spp. in 50 g (wet weight) and the treatment shall achieve at least a $6 \log_{10}$ reduction in *Escherichia coli* to less than $5 \times 10^2$ CFU/g.

Conventional treatments

<table>
<thead>
<tr>
<th>Process</th>
<th>Descriptions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermophilic aerobic stabilization</td>
<td>At a temperature of at least 55°C with a mean retention period of 20 days</td>
</tr>
<tr>
<td>Thermophilic anaerobic digestion</td>
<td>At a temperature of at least 53°C with a mean retention period of 20 days</td>
</tr>
<tr>
<td>Conditioning with lime</td>
<td>Ensuring a homogenous mixture of lime and sludge. The mixture shall reach a pH of more than 12 directly after liming and keep a pH of at least 12 for 24h</td>
</tr>
<tr>
<td>Mesophilic anaerobic digestion</td>
<td>At a temperature of 35°C with a mean retention period of 15 days.</td>
</tr>
<tr>
<td>Extended aeration</td>
<td>At ambient temperature as a batch, without admixture or withdrawal during the treatment period (the minimum time length of the treatment shall be laid down by the competent authority taking into consideration the prevailing climatic conditions in the area where the treatment plant is located)</td>
</tr>
<tr>
<td>Simultaneous aerobic stabilisation</td>
<td>At ambient temperature (the minimum time length of the treatment shall be laid down by the competent authority taking into consideration the prevailing climatic conditions in the area where the treatment plant is located)</td>
</tr>
<tr>
<td>Storage in liquid form</td>
<td>At ambient temperature as a batch, without admixture or withdrawal during the storage period (the minimum time length of the treatment shall be laid down by the competent authority taking into consideration the prevailing climatic conditions in the area where the treatment plant is located)</td>
</tr>
</tbody>
</table>

The sludge treatment shall at least achieve a $2 \log_{10}$ reduction in *Escherichia coli*.
Sewage sludge is typically treated prior to use on land by a combination of thickening, anaerobic digestion, composting, lime stabilization, disinfection, dewatering and/or thermal drying. A non-exhaustive compilation of conventional and advanced treatment processes from CEC (2000c) is shown in Table 3. The level of treatment dictates land application practice and restrictions of land use, as prescribed by legislation, in addition to the nutrient and heavy metal constraints, referred to above. The treatment can also significantly affect organic pollutant concentrations in the sludge.

The principal objectives of treatment are to minimise odour potential, avoid microbial health risks, and minimise sludge volume. Depending on the method and intensity of treatment, the physical, chemical, and microbiological characteristics are altered. Most treatments will result in loss of volatile solids, thus increasing the concentration of conservative chemicals in the remaining sludge dry solids. Labile substances such as nitrogen and organic compounds, may be lost by various processes (e.g., mineralisation, volatilisation, etc.).

Sludge is produced initially as a liquid, and may be applied to agricultural land with or without further dewatering. Liquid sludge is usually thickened and digested to 3–5% dry solids, but is typically dewatered to 20–30% to reduce volume and transport costs. The actual dry solids content achieved depends on sludge type, method of dewatering and whether the sludge has received further (advanced) treatment. Advanced treatment of dewatered sludges can produce very dry products, for instance, thermal drying will achieve >90% dry solids.

For liquid sludges, application is most commonly done by tractor-drawn slurry tanker with surface application by splash-plate, although in recent years there have been a number of equipment developments to improve the accuracy of sludge distribution and application rate, and to minimise odour and ammonia emissions and soil damage. Soil injection below the soil surface effectively avoids atmospheric emissions and on grassland and other forage crops, prevents potential contamination of conserved forage and direct ingestion by grazing animals. For dewatered sludge cake, application is usually by traditional muck spreader, although some contractors use specialised large machines to improve efficiency.

The timing for spreading sludge on land depends on cropping patterns, which vary according to season across Europe. The challenge for the sludge manager is to find suitable land for application throughout the year but usually sludge has to be stored, with most sludge being applied during a short period to bare ground prior to cultivation and sowing. Thus the most important operational window is generally in the late summer/early autumn between harvest and autumn sowing of cereals, although in nitrate sensitive areas, applications in the spring is preferred to avoid winter leaching. The potential exposure of sludge on the soil surface is generally short as it is a general requirement to cultivate as soon as practicable (1–2 days maximum) to avoid potential problems of odour, pest attraction, and surface run-off.

Only a small proportion of sludge is applied to growing arable crops due to the practical difficulties involved and the potential for damage to the crop. An exception to this is thermally dried sludge pellets which can be relatively easily applied by conventional fertiliser spreader, and liquid sludge to grassland, although the latter may be subject to constraints, as described above.

2. Contamination aspects

2.1. Organic contaminants within sewage sludge

The occurrence of organic contaminants within sewage sludge has been reviewed by different organizations and for several EU countries, for example by UKWIR (1995); Rogers (1996), Ducray and Huyard (2001), ICON (2001), and Langenkamp et al. (2001). Over 300 substances from a diverse range of classes of compounds have been identified in sewage sludge, and many more can be expected to occur. Concentrations vary from the pg/kg to g/kg range. They include monocyclic aromatics, alkyl and aromatic amines/imines, organotin compounds, aliphatic hydrocarbons, carbonyls, halogenated biphenyls (PCBs), polychlorinated dibenzo-para-dioxins and furans (PCDD/PCDF), pesticides, and polymers of various types and surfactants. Other examples of more recent interests are different types of endocrine disrupting compounds and pharmaceuticals.

Organic pollutants in sludge are subject to a number of fate processes that can dynamically change concentrations over time as well. Factors influencing the fate and behaviour of organic contaminants in sewage sludge are described by Rogers (1996). Accumulation/partitioning into the sludge from wastewater is dependent upon sorption, degradation and volatilisation. Sorption is most often driven by hydrophobicity or lipophilicity and can be described as a simple function of the octanol–water partition coefficient ($K_{ow}$). Volatilisation from sludge is highly dependent upon Henry’s Law constant (H). Degradation processes can be either abiotic (i.e., via hydrolysis or photoysis at the sludge surface) or biotic (i.e., biodgradation/biotransformation). Webber et al. (1996) proposed a qualitative ranking scheme to prioritise high production volume (HPV) contaminants in sewage sludge for further analysis and assessment, based on a consideration of the same physico-chemical properties, i.e., air–water partitioning (Henry’s law constant),
<table>
<thead>
<tr>
<th>Compound group</th>
<th>Physico-chemical properties</th>
<th>Concentration range in sludge (ds)</th>
<th>Degradation</th>
<th>Leaching potential</th>
<th>Plant uptake</th>
<th>Transfer to animals</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polycyclic aromatic hydrocarbons (PAH)</td>
<td>Limited water solubility, volatile to lipophilic</td>
<td>1–10mg/kg</td>
<td>Weeks to 10 years, strongly adsorbed by soil organic matter</td>
<td>Low</td>
<td>Very poor, foliar absorption, and root retention</td>
<td>Possibly but rapidly metabolized and not accumulated</td>
</tr>
<tr>
<td>Phthalate acid esters</td>
<td>Generally lipophilic, hydrophobic, and non-volatile</td>
<td>1–100mg/kg</td>
<td>Rapid, half-life &lt;50 days</td>
<td>Low</td>
<td>Root retention, not translocated</td>
<td>Generally very limited</td>
</tr>
<tr>
<td>Linear alkylbenzene sulphonate (LAS)</td>
<td>Amphiphilic</td>
<td>50–15,000mg/kg</td>
<td>Very rapid</td>
<td>Low</td>
<td>Minimal</td>
<td>Minimal</td>
</tr>
<tr>
<td>Alkylphenols (NPE)</td>
<td>Lipophilic</td>
<td>100–3000mg/kg</td>
<td>Rapid &lt;10 days</td>
<td>Low</td>
<td>Minimal, root retention</td>
<td>Minimal</td>
</tr>
<tr>
<td>Polychlorinated biphenyls (PCB)</td>
<td>Complex, 207 congeners, low water solubility, highly lipophilic, and semi-volatile</td>
<td>1–20mg/kg</td>
<td>Very persistent, half-life several years, strongly adsorbed by soil matter</td>
<td>Low</td>
<td>Root retention, foliar absorption, minimal root uptake, and translocation</td>
<td>Possible into milk/tissues via sludge ingestion, long half-life</td>
</tr>
<tr>
<td>Polychlorinated dibenzo-p-dioxins and furans (PCDD/F)</td>
<td>Complex, 75 PCDD congeners, 135 PCDF congeners, low water solubility, highly lipophilic, and semi-volatile</td>
<td>&lt;few µg/kg</td>
<td>Very persistent, half-life several years, strongly adsorbed by soil organic matter</td>
<td>Low</td>
<td>Root retention, foliar absorption, minimal root uptake, and translocation</td>
<td>Possible into milk/tissues via sludge ingestion, long half-life</td>
</tr>
<tr>
<td>Organochlorine pesticides</td>
<td>Varied, lipophilic to hydrophilic (e.g., 2,4-D), some volatile</td>
<td>&lt;few mg/kg</td>
<td>Slow, half-life &gt;1 year, loss by volatilization</td>
<td>Low to moderate</td>
<td>Root retention, translocation not important, foliar absorption</td>
<td>Via sludge ingestion, persistent in tissues</td>
</tr>
<tr>
<td>Monocyclic aromatics</td>
<td>Water soluble and volatile</td>
<td>&lt;1–20mg/kg</td>
<td>Rapid</td>
<td>Moderate</td>
<td>Limited owing to lower persistency, rapidly metabolized</td>
<td>Rapidly metabolized</td>
</tr>
<tr>
<td>Chlorobenzenes</td>
<td>Water soluble/volatile to lipophilic</td>
<td>&lt;0.1–50mg/kg</td>
<td>Lower MW compounds lost by volatilization, higher MW compounds persistent</td>
<td>Low to high</td>
<td>Possible via roots and foliage, may be metabolized</td>
<td>Important for persistent compounds</td>
</tr>
<tr>
<td>Short-chained halogenated aliphatics</td>
<td>Water soluble and volatile</td>
<td>&lt;5mg/kg</td>
<td>Generally rapid loss by volatilization</td>
<td>Moderate</td>
<td>Foliar absorption, possible translocation</td>
<td>Low</td>
</tr>
<tr>
<td>Aromatic and alkyl amines</td>
<td>Water soluble and low volatility</td>
<td>&lt;1 mg/kg</td>
<td>Slow</td>
<td>Moderate to high</td>
<td>Possible</td>
<td>Low</td>
</tr>
<tr>
<td>Phenols</td>
<td>Varied, lipophilic, and high water solubility</td>
<td>&lt;5 mg/kg</td>
<td>Rapid</td>
<td>Low to moderate</td>
<td>Possible via roots and foliage</td>
<td>Generally very limited</td>
</tr>
<tr>
<td>Organic compound</td>
<td>Member States</td>
<td>Congeners</td>
<td>Threshold limits in sludge</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>------------------</td>
<td>---------------</td>
<td>-----------</td>
<td>---------------------------</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>AOX Halogenated organic compounds</td>
<td>Germany and some Lander in Austria</td>
<td></td>
<td>500 mg/kgdm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LAS Linear alkylbenzene sulphonates</td>
<td>Denmark</td>
<td></td>
<td>2600 mg/kgdm (until 30/6/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1300 mg/kgdm (from 1/7/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>100 mg/kgdm (until 30/6/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>50 mg/kgdm (from 1/7/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>DEHP Di(2-ethylhexyl) phthalate</td>
<td>Denmark</td>
<td></td>
<td>50 mg/kgdm (until 30/6/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>10 mg/kgdm (from 1/7/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>NPE Nonylphenol and nonylphenol ethoxylates with 1 or 2 ethoxy groups</td>
<td>Denmark</td>
<td></td>
<td>50 mg/kgdm (until 30/6/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>10 mg/kgdm (from 1/7/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Sweden*</td>
<td></td>
<td>50 mg/kgdm (until 30/6/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PAH Polycyclic aromatic hydrocarbons</td>
<td>Denmark</td>
<td>Acenaphthene, phenanthrene, fluorene, fluoranthene, pyrene, benzo(b+j+k) fluoranthene, benzo(a)pyrene, benzo(g)perylene, indeno(1,2,3-cd)pyrene</td>
<td>6 mg/kg dm (until 30/6/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>France</td>
<td>Fluoranthene, Benzo(b)fluoranthene Benzo(a)pyrene</td>
<td>3 mg/kgdm (from 1/7/00)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Sweden*</td>
<td>Sum of six</td>
<td>5 mg/kgdm (4 on pastureland)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.5 mg/kgdm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2 mg/kgdm (1.5 on pastureland)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>3 mg/kgdm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PCB Polychlorinated biphenyls</td>
<td>Germany</td>
<td>28, 52, 101, 138, 153, and 180</td>
<td>0.2 mg/kg dm for each congener</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>France</td>
<td>28, 52, 101, 118, 138, 153, and 180</td>
<td>0.8 mg/kgdm for the sum</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Sweden*</td>
<td>28, 52, 101, 118, 138, 153, and 180</td>
<td>0.4 mg/kgdm for the sum</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PCDD/F Polychlorinated dibenzo-p-dioxins/dibenzofurans</td>
<td>Denmark and some Lander in Austria</td>
<td></td>
<td>100 ng TEQ/kg dm</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Bold numbers in the last column indicate the Limit Values as currently proposed in CEC, 2000c.

* In non-binding guidelines agreed among farmers, Swedish EPA and water companies.
organic carbon-water partition coefficient, and biodegradation and hydrolysis rates.

For most compounds, their presence in sludge will be influenced primarily by the presence of potential sources within the catchment of the wastewater treatment plant. Some compounds, however, may actually be formed de novo during wastewater treatment (e.g., alkylphenols from alkylphenolethoxylates). Details of the concentration range of the principal organic contaminants found in sewage sludge are presented in Table 4 (Smith, 2000).

New provisions on Limit Values are considered for introduction into the revised Sludge Directive for a few specific organic compounds (or classes of compounds), i.e., AOX, LAS, DEHP, NPE, PAH, PCB, and PCDD/PCDF. Existing standards within individual European countries for these organic contaminants in sewage sludge applied to agricultural land, and the proposed new European values, are summarized in Table 5 (Smith, 2000). This paper is concerned with the questions how the Limit Values as proposed by the EU can be justified by risk assessment science, and what is their environmental relevance.

The potential impact of these new Limit Values on disposal options for sludge has been studied, e.g., by Ducray and Huyard (2001) for France. The need to include certain compound groups and establish Limit Values is still the subject of ongoing debate. Several authors have for example expressed their doubts regarding the justification of the inclusion of a sum-parameter like AOX (e.g., Mueller, 2003). Organohalogens are a heterogeneous group of compounds that can be highly toxic or totally harmless, easily degradable of very persistent. In addition, there is a large natural production of organohalogens in the environment, and AOX is naturally occurring in soils (Asplund and Grimwall, 1991; Gribble, 1994). More than 80 plant species are known to produce halometabolites (Engvild, 1986). Also in the specific case of PCDD/PCDF, the US EPA in 2003 made the final decision not to regulate dioxins in land-applied sludge based on extensive studies and a risk assessment procedure for human health and the environment (www.epa.gov/waterscience/biosolids).

2.2. Exposure pathways and transfer processes for organic chemicals

Following application to land the contaminants are subject to various chemical or biological transformation processes, as well as physical transfer processes (i.e., leaching, run-off, erosion, and volatilization) that will influence their concentration in various environmental media and along the food chain. These processes are described in more detail, e.g., in van Leeuwen and Hermens (1995) or CEC (2000b) and have potential implications for environmental and human health. Soil exposure levels for chemicals (PECs) can be calculated as shown below:

1. The concentration in soil at any time is given by:
   \[ PEC_{\text{soil}}(t) = \frac{F_{\text{air}}}{k} - \left[ (F_{\text{air}}/k) - PEC_{\text{soil}}(0) \right] e^{-kt}. \]

2. The average PEC_{\text{soil}} over a given period (T) is:
   \[ PEC_{\text{soil-average}}(T) = \frac{F_{\text{air}}}{k} + 1/(kT) \times PEC_{\text{soil}}(0) - (F_{\text{air}}/k) \times [1 - e^{-kt}], \]

3. The steady state PEC_{\text{soil}} after multiple additions is:
   \[ PEC_{\text{soil-steady state}} = \frac{F_{\text{air}} + F_{\text{sludge}}}{k} \times H \times R, \]

where
- \( F_{\text{air}} \) = chemical flux from air (in mg/(kg.day) in Eqs. (1) and (2), and in kg/(m².day) in Eq. (3).
- \( F_{\text{sludge}} \) = chemical flux from sludge application (kg/(m².day)).
- \( k \) = total removal rate (1/d) = sum of first order biodegradation, leaching and evaporation rate).
- \( PEC_{\text{soil}}(0) \) = initial concentration (mg/kg).
- \( H \) = soil depth (m).
- \( R \) = soil density (kg/m³).

Although human health is central in a risk assessment, other organisms, which also can be affected by organic pollutants applied through sewage sludge, should be included. Sludge application to arable land means an exposure of a range of organisms to organic contaminants, either directly or indirectly. The soil fauna, soil microflora, and plants growing in sludge-amended soil are directly exposed. Domestic and wildlife animals can be affected consuming plants or predating soil biota from sludge-amended soil. Leaching, runoff, and erosion of organic contaminants to natural waters can affect aquatic organisms. The intake of drinking water from wells, if contaminated by sludge-amended soil, may affect humans and domestic animals. Soil ingestion by children’s mouthing or intake through unwashed vegetables, soil breathing of air-born dust, and of course dietary intake of plant or animal products and drinking water can affect humans.

To structure the assessment, a variety of exposure pathways are being considered in the EU TGD (CEC, 1996, 2003) by which a chemical can reach a target organism. Similarly, according to the US-EPA (1993), 14 pathways of exposure to contaminants in sewage sludge can be identified, which have been slightly simplified to 13 pathways in this review:

*Paths to the environment;*
- Sludge–soil–soil biota (including soil microorganisms).
- Sludge–soil–plant.
Pathways to livestock and humans via direct exposure, or indirectly through food chain transfer:

- Sludge–soil–child.
- Sludge–soil–animal.
- Sludge–soil–animal–human (incl. via aquatic animals).

Pathways to humans (and also animals) through dispersion to other environmental media:


Sludge application to agricultural soil means, first of all, that the soil-plant system is exposed. Thus primarily, the living components of this system, plants, soil microflora and soil fauna (biota) can be affected. In the case of soil microflora and plants, the uptake of contaminants takes place through the soil water phase. The transfer of chemicals from the soil water phase to the plant roots or microorganisms can be described by a root transfer factor (RTF) or bioconcentration factor (BCF), respectively. In addition, volatilized compounds may be taken up through the plant canopy. The soil fauna can both ingest soil particles and take up dissolved compounds from the pore water.

The uptake of contaminants by wildlife via predation of the soil fauna, by domestic animals via fodder, by aquatic organisms through natural water and by humans through food products and drinking water takes place in a diluted form. These processes can be described by a bioconcentration (BCF—uptake via medium) or bioaccumulation factor (BAF—if uptake via the food chain is involved) (Trapp and McFarlane, 1995; Van de Meent et al., 1995). However, the contaminant concentration is generally lower in the organism or crops than in soil to which a contaminant was applied via sludge (see e.g., ICON, 2001; Kirchmann and Tengsved, 1991; Smith, 1996).

The scientific case for extending SQS to include organic contaminants with potential effects via the food chain (i.e., indirect exposure) was critically examined in Smith (2000) and Jones and Stevens (2002). A strong policy-driven motivation for the adoption of such controls is evident in several EU countries. Several authors have assessed and questioned the relative toxicological and ecotoxicological significance of organic contaminant transfer to crops and subsequently up the food chain. Indirect exposure risk assessment for selected compounds have been published, e.g., for PCDDs/PCDFs by Wild et al. (1994) and Greenwood et al. (2004).

Dean and Suess (1985) have concluded that total human intake of organic pollutants from sludge application to agricultural land is limited as compared to other sources, and unlikely to cause adverse health effects. According to Stark and Hall (1992) actual risk under normal sewage sludge management practices is likely to be minimal. Several subsequent reviews support this stance (Jones and Stevens, 2002; Smith, 2000; UKWIR, 1995). This line of thinking is also reflected in the decision of the US EPA to delete criteria for organic pollutants from the 503 Rule for sewage sludge (US-EPA, 1993).

Surface run-off and erosion do occur, mainly driven by meteorological events and influenced by soil management practices and terrain conditions. Contaminants that are transported to surface waters as diffuse pollution can either be in the dissolved or sorbed state.

The relative importance and relevance of these transfer pathways can, however, be very different according to the chemical considered. Some general rules of thumb are provided below (see also Andersen, 2002b):

- Most organic chemicals are not significantly taken up and transported by plants via the roots. Organic chemicals with the highest potential for uptake and transportation in the plant are those with a logKow of 0–3, i.e., neither hydrophilic nor hydrophobic (Briggs et al., 1982; Trapp et al., 1994).
- Bioavailability to plants via root uptake will also be limited as a consequence of adsorption to soil particles (UKWIR, 1995). Related to soil sorption is the aspect of aging and humification. It has been shown for a number of compounds (e.g., pesticides) that their extractability and bioavailability decreases significantly over time (Hatzinger and Alexander, 1995). This suggests that some irreversible binding, diffusion into microbially inaccessible micropores, or incorporation into the humus fraction can take place over time (Alexander, 1995; Bollag et al., 1992).
- Leaching to groundwater that may be used for drinking water preparation can occur for water-soluble compounds or metabolites, but is unlikely to occur for lipophilic organic contaminants chemicals with a high affinity for organic matter, or for positively charged compounds with affinity for clay minerals.
- Run-off, when it occurs, may play an important role in the transfer to surface water.
- Direct ingestion by livestock of sludge particles or soil associated with grass constitutes an exposure scenario with some potential for transfer into the foodchain, and is theoretically the main route of human expo-
sure to organic contaminants from sewage sludge used in agriculture. It is a more important transfer route than the soil–plant pathway.

- Lipophilic compounds can be bioconcentrated in predator organisms and livestock due to continuous direct intake of contaminated material. Bioconcentration in fat and milk of domestic animals is possible for lipophilic compounds via this route (UKWIR, 1995). Such compounds therefore deserve extra attention.

- Some direct contamination of the foodchain may potentially occur from the surface of plants which are consumed raw (e.g., fodder, salad crops), although sludge application to such crops is currently not permitted as a hygiene precaution.

More specifically, Table 4 details the potential for leaching, plant uptake and transfer to animals of the principal organic contaminant classes in sewage sludge. Smith (2000) addressed the question of why there is limited evidence for transfer to the foodchain of these organic contaminants within sewage sludge. The intrinsic physico-chemical properties of organic contaminants appear to provide barriers to their transfer to the foodchain. Organic compounds can typically be grouped into three categories on the basis of their behaviour: (i) readily volatilized compounds that are lost to the atmosphere from sludge and sludge amended soil, (ii) compounds that are rapidly biodegraded by micro-organism and therefore do not persist long enough for any significant uptake, and (iii) persistent compounds that are often strongly adsorbed onto the organic components of sludge and soil. Potential effects from such contaminants are consequently minimized when sewage sludge is used in agriculture.

A compilation of the medium transferring pollutants in Table 6 shows that, based on the relative exposure concentration, a highly exposed group (either continuous or intermittent) and a low-to medium exposed group of organisms can be distinguished. The low-medium exposure is associated with secondary exposure scenarios. In many earlier soil studies only plant species were considered for inhibition and toxicity tests when applying pollutants. The soil microflora (Van Beelen and Doelman, 1997) and recently also the soil-living organisms (Jensen, 1999) have been recognized as important groups for toxicity tests, which is in accordance with the above reasoning. Thus, in future risk assessment of contaminants applied to soil, the soil microflora, the soil fauna, plants and certain target groups of humans should be considered as relatively more exposed.

The need for SQS is the subject of debate for several years now, and was taken up as a challenge by ILSI’s Environment and Health Task Force and its Expert Group on Risk Assessment of Sewage Sludge, to arrive at a consistent approach to dimension the risks on a scientific and practical basis.

3. Risk assessment endpoints

There are a variety of important endpoints that need to be assessed in order to provide a comprehensive risk assessment of the agricultural use of sewage sludge. The soil environment is vital to sustain terrestrial life, and consequently soil protection has become an important global concern. The greatest source of soil contamination is chemical pollution, and therefore there is increasing public and regulatory pressures to assess, understand and minimise the impact through risk analysis and risk management. Concerns about the possible risk to public health from the application of sludge was first recognised in 1970, when guidelines were published in the UK on advisory levels of zinc, copper, nickel, and boron. As the use of sewage sludge grew, various guidelines were introduced by EU Member States. The aim of this assessment is therefore to protect soil microbial pro-

### Table 6

Relative exposure concentration of organisms to organic pollutants if applied with sewage sludge to soil

<table>
<thead>
<tr>
<th>Affected organisms</th>
<th>Medium transferring the organic pollutants</th>
<th>Relative contaminant concentration in the medium</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Primary continuous exposure</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Soil microflora</td>
<td>Soil solution</td>
<td>High</td>
</tr>
<tr>
<td>Soil fauna</td>
<td>Soil particles and soil solution</td>
<td>High</td>
</tr>
<tr>
<td>Crops</td>
<td>Soil solution and gaseous flux in canopy</td>
<td>High</td>
</tr>
<tr>
<td><strong>Primary intermittent exposure</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Certain human populations</td>
<td>Soil particles, air-borne dust</td>
<td>High</td>
</tr>
<tr>
<td>Domestic animals</td>
<td>Soil particles, air-borne dust</td>
<td>High</td>
</tr>
<tr>
<td><strong>Secondary exposure</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Wildlife</td>
<td>Soil fauna</td>
<td>Low–medium</td>
</tr>
<tr>
<td>Aquatic organisms</td>
<td>Run-off and leached soil solution</td>
<td>Low–high</td>
</tr>
<tr>
<td>Domestic animals</td>
<td>Crops</td>
<td>Low</td>
</tr>
<tr>
<td>Humans</td>
<td>Crop and animal products and drinking water</td>
<td>Low</td>
</tr>
</tbody>
</table>

* Primarily farmers and children (mouthing).
cesses (i.e., ‘function’), soil flora and fauna, and limit the
direct and direct transfer of organic contaminants to
animals and humans via food and water, to avoid detri-
mental health effects.

Decomposers such as micro-organisms are important
for the mineralisation of the organic matter and conse-
quently nutrient cycling in the ecosystem. Symbiotic
microorganisms such as Rhizobium species and Mycorh-
izae are important for nutrient and water supply, and
plant health in general. Soil invertebrates are important
for creating and maintaining a good soil structure and
are a vital link in the food chain. Finally, plants are the
main primary producers for the ecosystem and provide
a source of food for all heterotrophic organisms. To date,
environmental risk assessment has focused on the aquatic
environment, and at present there are few standardised
tests for the terrestrial compartment. However, there is
still a general understanding that the effects of sewage
sludge application on microbial activity, soil fauna and
flora are difficult to study, because they are influenced
by a multitude of interdependent factors (e.g., fertilisa-
tion, water capacity, mixture toxicity, heavy metal con-
tent, method of sludge application, etc.).

As a common principle of regulation, the hazard and
risk assessment of chemicals is based on data obtained
by testing. It is important to note that ‘hazard’ refers to a
potential source of harm (e.g., a chemical-based on
inherent properties), and ‘risk’ is the combination of
the probability of the occurrence of harm and the sever-
ity of that harm (ISO, 1999). The assessment of risks
from chemical compounds can be based on the risk ra-
tio, i.e., PEC/PNEC, which expresses the relationship
between a predicted environmental concentration
(PEC) and a predicted no-effect concentration (PNEC).

It is important to consider the type of soil used in
testing, in relation to soil characteristics like organic
matter and clay content, soil pH and soil moisture con-
tent, which affect the bioavailability of the test com-
 pound. In addition, factors such as ageing of organic
soil contaminants should also be considered. In the con-
text of sludge risk assessment, the most realistic and
therefore preferred procedure to account for bioavail-
ability effects in biodegradation and ecotoxicity tests,
is to dose chemicals to soil in a sludge matrix rather than
as aqueous solutions.

The calculation of a PECsoil is not laid out in detail
in this document since adequate guidance can be found
for example in van Leeuwen and Hermens (1995), CEC
(2003) or ECETOC (2003). The PNECsoil for a chemi-
cal and the soil ecosystem is determined by the use of
ecotoxicological test methods for effects on endpoints
such as survival, growth, reproduction, or behaviour.
As soil is such a complex and heterogenous environ-
ment, it is important to use soil organisms that represent
different and significant ecological functions in the eco-
system. On the other hand, risk assessment for sludge
in agricultural soil applies to a man-made production
environment; therefore the selection of plant test species
should preferably cover typical crop species. Sustained
crop productivity will be a useful overall indicator of
the health of the soil ecosystem.

The basis for the derivation of safe thresholds for or-
ganic contaminants within soil is given in the TGD
(CEC, 1996, 2003). This typically involves the use of
deterministic assessment factors to extrapolate from sin-
gle species laboratory based studies to the ecosystem. A
factor of 1,000 is employed to the lowest endpoint (LC50
or EC50) from short-term (acute) tests on plants, inver-
tebrates or micro-organisms. Factors of 100, 50 or 10
are applied to endpoints (NOECs) from long-term
(chronic) tests on one, two or three species from different
trophic levels, respectively. An alternative methodology
for data-rich compounds is statistical extrapolation
from a theoretical distribution function based on
chronic endpoints from single species tests. A prescribed
percentile of the single species distribution is used as the
criterion. Several distribution functions are available,
such as log-triangular (OWRS, 1985), log-logistic (Koo-
These extrapolation methods aim at deriving a 95% pro-
tection level, which is the hazardous concentration for
5% of the species (HC5). For a critical review of extrap-
olation methods the reader can refer to Suter (1993);
CEC (2000b) or Posthuma et al. (2002).

3.1. Effects on microbial systems and soil fertility

Exposure occurs via the sludge–soil–(micro)biota
pathway. An active and balanced soil microbial activity
is essential to ensure well functioning nutrient cycles and
plant health. Key soil functions are decomposition of or-
ganic matter, mineralisation and cycling of nutrients,
and synthesis of humus. Elements of the carbon, nitro-
gen, phosphorus and sulphur cycles are also associated
with the soil. Some soil micro-organisms also play a role
in nitrogen fixation and plant pathogen control.

Micro-organisms are particularly suitable indicators
of effects on soil function as they are ubiquitous with
population densities as high as 10^6–10^8 bacteria per
gram of soil (Doelman and Vonk, 1994). Soil microbes
also live in direct contact with the soil microenvi-
ronment and experience a high level of exposure due to their
small size and therefore high surface area to volume ra-
tio. In addition, soil micro-organisms are vital in the
transformation of synthetic organic compounds and
natural waste materials into inorganic forms that are
environmentally acceptable (Torstensson, 1996).

One of the main problems in studying the microbial
community in sludge amended soils over a period of
time is the sheer diversity of the initial population and
the problems of comparing amended and non-amended
sites. Most of the research carried out on community
structure using the methods described above have found that it is the long term accumulation of heavy metals in sewage sludge that may limit the diversity of microbial communities. However, the increased carbon and energy sources in the sludge appear not to cause a decrease in total microbial numbers. Usually the opposite is true, i.e., the total microbial biomass is larger but with a different bacterial population structure than in the non-amended soil (Seselitsch et al., 2001). It is therefore difficult to obtain data on the effects of organic contaminants alone. Evans et al. (1997) report the absence of stress from sludge addition in the soil microbial biomass.

Since microbial communities are highly adaptive, in constant evolution (e.g., via frequent exchange of genetic information) and their composition and ‘niche’ difficult to describe, it is recommended that functional rather than structural microbial endpoints should be used in a sludge risk assessment. Also, the addition of chemicals in a sludge matrix—which is mainly composed of microbes—will induce significant changes to the composition of the soil microbial community structure, at least in the short term.

The best approach for evaluating the effects of chemical substances on microbial populations is to use a battery of different tests to assess the condition of the microbial population and ecosystem in soils. A number of these have been described below.

- **Microbial numbers**: Various workers have evaluated simple microbial methods involving enumeration for assessing the effect of a sewage sludge addition on microbial processes in soil. Examples by Lawlor et al. (2000) included a direct microscopic count using acridine orange in fluorescence microscopy; a more direct plating method determining the Total Viable Count (TVC); and the Biolog Ecoplate method (Garland and Mills, 1991). However, the new insight that non-culturable microbial species often outnumber the culturable ones, limits the relevance of microbial counts.

- **Microbial biomass**: An alternative to enumeration is an evaluation of microbial biomass. One way in which the microbial biomass is estimated is via determination of microbial biomass carbon (Khan and Scullion, 1999). The same technique was used by Albiach et al. (2000) to assess the changes in a soil population over a four year cycle of sludge amendment.

- **Microbial metabolic activity**: Monrozier et al. (1993) used the method of Tate and Jenkinson (1982) to measure ATP in soil and used this as an indicator of microbial activity in unsaturated soils containing xenobiotics. Fatty Acid Methyl Ester analyses (FAME) has also been used to detect diversity in microbial populations (White, 1995) and as such lends itself to looking at differences in sludge and non sludge amended soils (Lawlor et al., 2000). A more specific method of analysis is PLFA (phospholipid fatty acid). This is the analysis of fatty acids specifically linked to the polar lipid fraction. The resulting data leads to an increase in sensitivity (over FAME analysis) and an increased specificity for detecting microbial populations (Baird and White, 1985). Another method looking at the effect of toxicity on gene expression is the luminescence bioassay.

- **Respirometer based methods**: Changes in the respiration rate of soils with and without sludge amendment have been used as a basis of monitoring changes in the microbial population (Khan and Scullion, 1999; Saviozzi et al., 1999).

- **Molecular biology based techniques**: The use of nucleic acid based techniques for examining the diversity of micro-organisms in a matrix is rapidly gaining in popularity. Methods such as rRNA extraction using for example a hot phenol extraction protocol (Moran et al., 1993) allow the evolutionary relationships between species to be identified. However, the application of molecular techniques to study sewage sludge amended soils has been limited so far.

A number of field studies have been carried out in sludge-amended soils using the methods described above. For further details the reader can refer, e.g., to Khan and Scullion (1999), Saviozzi et al. (1999), Sandaa et al. (1999), and Lawlor et al. (2000). In summary, all of the above studies have found that the main problem for microbial communities in soil amended with sewage sludge seems to be the effect metal ions can have on species diversity. Few studies have been carried out on the effect of specific chemicals on soil communities. The methodology to carry out such a study does exist, but the effects of metal ions need to be excluded or separated in the study design.

### 3.2. Effects on soil flora

Exposure occurs via the sludge–soil–plant pathway. Plants are essential primary producers in the terrestrial ecosystem, providing a food source for all heterotrophic organisms, and forming the basis of the food chain. In addition, the crop yield and quality are important success criteria in agriculture. Therefore it is important to identify potential phytotoxins and understand the magnitude of their impact on different terrestrial ecosystems. Although many data have been published over the last 50 years on the toxicity of organic chemicals to vascular plants, most of these have been generated by the pesticide industry. Consequently there is a considerable amount of data on plant hormones and herbicides, but relatively little data on the
phytoxicity of other organic contaminants in sewage sludge (Fletcher et al., 1988).

To improve our understanding, O’Connor, 1996) evaluated the data on the uptake of organic compounds by crops grown in sludge-amended soils. He came to the conclusion that contamination of crop plants with known toxic organic contaminants is negligible. However it should also be noted that when Engwall and Hjelm (2000) used a sensitive bioassay, they were able to detect low concentrations of dioxin-like compounds in all carrot, zucchini, and cucumber samples grown in soil that had been amended in sludge. In addition, Smith et al. (2001) concluded that persistent organic contaminants may also be introduced into the grazing animal food chain if sewage sludge is applied to pasture land.

There are a variety of different protocols available to evaluate potential phytotoxins, and these involve different combinations of plant species, plant parts, exposure conditions, and measured endpoints. Chemicals can be taken up actively or passively via the seed, embryo, the seedling or adult plant. The choice of test will depend on a variety of criteria such as expense, safe use of hazardous chemicals, reproducibility of controls, and most importantly the reliability and usefulness of the test. Phytotoxicity tests may be conducted during different stages in the life cycle of a plant and a variety of endpoints can be measured. The most commonly used and recognised standardised tests available are tests for monocotyledon and dicotyledon plants (ISO, 1993a,b; OECD, 2000a).

Plant tests have also been criticised because often the number of plant species per test is too low and wild plants, in particular, have not been studied (Roembke and Moltman, 1996). This is a significant problem, as an analysis of tests conducted with 151 different species indicates that the sensitivity within vascular plants alone varies on average by a factor of 10.5 and in individual cases by more than 100 (Fletcher et al., 1990). In addition, a taxonomic evaluation of the phytotoxicity literature through the PHYTOTOX database (Fletcher et al., 1988) revealed that most published data is heavily biased towards north-temperate agricultural species and information on tropical plants is almost non-existent. In the context of a sludge risk assessment in agricultural soil, information on crop plants is obviously the most informative.

3.3. Effects on soil fauna

Exposure occurs via the sludge–soil–biota–predator pathway. There are an incredible variety of life forms and life histories of soil invertebrates living either permanently or temporarily in or on the soil. Hence, ecotoxicologists have a very complex and difficult task of attempting to assess effects on these species and the interactions between them. The selection of species for a battery of tests with soil invertebrates should take into account the most important trophic levels in the soil compartment (e.g., decomposers, parasites, and predators), the different routes of exposure to the chemical substrates and the different sensitivities to the toxicants (Laskowski et al., 1998).

Ecotoxicological tests for the terrestrial compartment are summarised in van Leeuwen and Hermens (1995) or in Løkke and Van Gestel (1998). Overall there is a lack of data available on the effects of industrial chemicals on soil fauna, especially in sludge amended soils. However in the last few years, a number of high quality studies have been carried out at the NERI Institute in Denmark, including, e.g., a study to evaluate the effects of LAS, soil type and sewage sludge amendment on the Collembolan Folsomia fimetaria and the earthworm Aporrectodea caliginosa (Holmstrup et al., 2001).

A comprehensive listing of test species and protocols for both invertebrate and plant crops is provided in the EU TGD (CEC, 1996, 2003). They include standardised tests with invertebrates such the compost worm Eisenia fetididae (ISO, 1996a,b; OECD, 1984), the oligochaete Enchytraeus albidus (OECD, 2000b; Roembke, 1989) and the springtail Folsomia candida (ISO, 1993c). Additional tests are mainly from Løkke and Van Gestel (1998). Tests are conducted using an exposure system (or combination) as expected in the field i.e.: (i) ingestion/oral uptake of food/soil, (ii) dermal uptake from soil, and (iii) respiration via tracheae.

3.4. Biomagnification and secondary poisoning of top predators

Exposure occurs in this case via the sludge–soil–biota–predator pathway. Bioaccumulation/biomagnification will be most pronounced in birds and mammals that prey on lower organisms which are already highly contaminated. In risk assessment, potential exposure in the predator bird or mammal is determined using a bioconcentration factor (BCF) derived from experimental data for earthworms (Connell and Markwell, 1990). Exposure concentrations in prey species correspond to the soil concentrations multiplied by a BCF: PEC preceded by sludge (PECsludge) * BCF.

A means of establishing a predicted no-effect concentration (PNEC) for worm-eating birds and mammals from subchronic data for laboratory mammals is detailed in Romijn et al. (1994). Toxicity endpoints (mg/kg body weight) from the laboratory studies are converted to maximum concentrations in the prey diet (mg/kg diet; PECprey), using conversion factors based on food consumption rates and assessment factors to extrapolate from the laboratory to the environment. If the predicted concentration in food (PECprey) exceeds the PNEC, secondary poisoning can be a critical pathway. To back-calculate safe thresholds in sludge...
amended soil, the secondary poisoning PNEC in the prey species is divided by the BCF.

Evaluation of secondary poisoning in risk assessment is more fully dealt with in van Leeuwen and Hermens (1995), and in the TGD (CEC, 1996, 2003). Duarte-Davidson and Jones (1996) proposed a simple screening model to assess the likelihood of organic contaminants from sewage sludge accumulating within the food chain. The model aims to predict root surface uptake, translocation potential, foliar uptake and animal intake based on a small number of physico-chemical parameters and fate properties. Over 300 compounds were screened for potential transfer from soils to plants via retention on root surfaces, root uptake and translocation, foliar uptake and animal intake via soil and herbage ingestion. The classes of compounds prioritized as having the potential for accumulation in plants and animals were: heavy (> tri-) chlorobenzenes, heavy (> tri-) chlorophenols, PCBs and dioxins/furans. A recent plant uptake model intercomparison was published by Collins and Fryer (2003).

3.5. Leaching and groundwater quality

Understanding the potential for chemicals to leach from the surface to ground water is important to protect the groundwater layers. These are often used as a source of drinking water, requiring normally only minimal treatment. The potential for organic contaminants in sewage sludge to leach to groundwater following application to agricultural land was assessed by Wilson et al. (1996). Various models were employed to predict compound mobility in soil on the basis of physico-chemical properties (water solubility, vapour pressure, octanol-water partition coefficient, organic carbon-water partition coefficient, Henry’s law constant and loss half-life). Different criteria were used to define high leachability, depending upon which model was under consideration. The same models and criteria may similarly be applied to assess the likelihood of groundwater contamination from compounds under regulatory scrutiny vis-à-vis concentration limits in sewage sludge. Wilson et al. (1996) also provided a means of predicting soil aqueous phase concentrations in sludge amended soils. This would allow the back-calculation of safe concentrations in sewage sludge. The relevant benchmarks to start from might be groundwater limits or drinking water standards.

Gustafson (1989) has formulated a model and index to estimate the leaching behaviour of a substance using simple properties (i.e., sorption behaviour and half-life). A specific risk assessment system for the groundwater module has, for example, been developed by Swartjes et al. (1993) based on the PESTLA model. The SESOIL screening-level model (Bonazountas and Wagner, 1984; http://www.scisoftware.com) was designed to simultaneously predict one-dimensional vertical water and sediment movement, as well as pollutant fate. A similar functionality is provided by the MACRO model (http://www.mv.slu.se/bgf/macro.htm). Leaching columns in the laboratory or lysimeters in the field are often used as experimental systems to assess leachability. According to Andersen (2001b), however, leaching of organic compounds from sludge to groundwater may be relevant in a few particular cases, but seems in general to be a negligible phenomenon.

3.6. Run-off, erosion, and surface water quality

Chemicals can be transported from agricultural land to surface water due to run-off caused by meteorological events (rain, wind). This is also referred to as ‘diffuse pollution,’ and when occurring, it is usually a major process (Andersen, 2001b). The chemicals can be in a dissolved state, but most often a large fraction is bound to soil particles and other suspended matter. A risk assessment for sludge added to agricultural land should assess the potential for diffuse pollution and the impact on surface- and drinking water quality (Greenwood et al., 2004). Inputs from diffuse sources should be added to those of point sources. Good Agricultural Practice recommends against the use of sewage sludge and manure in conditions with a high likelihood of run-off.

For chemicals that rapidly break down after their addition to soil, it is not envisaged that diffuse-source transfers will regularly make a significant contribution to the total surface water load. Highest fluxes are most likely to occur during extreme (i.e., high magnitude, low frequency) storm events, which generally coincide with high discharge in the receiving waters and, therefore, a high degree of dilution.

An overview of state-of-the-art models in run-off and contaminant transport from land is provided e.g., in White et al. (2001). These are EPIC, GLEAMS, OPUS, PRZM, ANSWERS-2000, SWAT-2000, SWATCATCH, etc. None of these models explicitly includes the capability to model transport of organic compounds in sewage sludge. The goal of the CEFIC-LRI ‘TERRACE’ project (http://www.cefic.org/lri), is to develop and validate SWAT-2000 for such purpose. The concentration of chemicals in surface waters originating from point sources can for example be modelled with GREAT-ER (Schowanek et al., 2001).

3.7. Indirect human exposure

Indirect human exposure can potentially occur via different pathways, i.e., via food crops, animal uptake to meat and milk, or drinking water (following leaching to groundwater or surface run-off). Chemicals can reach plants via the roots or via deposition on upper parts (O’Connor, 1996). The organics that will be of the great-
est concern to animal and human health are those that (Chang et al., 2002):

- can be absorbed by plants,
- appear frequently at high enough concentrations to be analytically detectable, and,
- for which there is evidence of food chain transfer.

Priority for consideration of indirect human exposure is given to organics that are reproductive toxicants (including endocrine disruptors and some pharmaceuticals), carcinogenic substances or chemicals that bioconcentrate in the food chain.

The modelling of indirect human exposure to environmental contaminants in food is described, together with some examples, in Van de Meent et al. (1995). This approach has been implemented in EU chemical risk assessment procedures (TGD) and in the software EUSES (CEC, 1996, 2003). Dietary intake from drinking water, fish, food crops, milk, and meat are combined in order to calculate a total daily uptake via indirect exposure. It is often difficult to quantify the actual exposures to various chemicals that might be contained in the sludge. Therefore models that predict the transfer of toxics from one environmental compartment to another along the exposure route (e.g., from the soil to the crop, from the soil into the water, etc.) are used. They rely on linear transfer coefficients based on lipophilicity and other properties. An example is the pollutant partition factor between plant and soil $K_{sp}$, (also known as Root Concentration Factor, RCF) which is estimated based on the $K_{ow}$ as measure of lipophilicity (Travis and Arms, 1988). Similar linear transfer coefficients, called bio-transfer factors (BTFs) and also based on lipophilicity, can be used to estimate steady state chemical concentrations in meat and milk based on concentration in the food source.

For example: $\text{BTF}_{\text{meat}} \text{ (day/kg)} = \text{concentration in the meat (mg/kg)/uptake via fodder (mg/day)}$.

Determining specific human exposures requires the use of validated assumptions for model parameterization regarding daily food and drink intake, percentage of crops eaten that have been grown on the sludge amended soil, average body weights, chemical absorption by the body through the different exposure routes, amounts of air respired, etc. Once these quantities have been estimated the total dietary (and other) intake can be derived by adding the exposures from drinking water, fish, food crops, milk, and meat and through inhalation.

The estimated exposure to any one chemical (including the total exposures expected from other non-related sources, e.g., food grown outside the area, air pollution, drinking water, etc.) can then be compared to an assessment factor such as an ADI (acceptable daily intake—ILSI, 2000) or RfD (Reference Dose). ADIs and RfDs are concentrations of a chemical to which an average adult can be exposed to on a regular basis (daily) for a lifetime (70 years) that will not cause adverse effects, or in the case of carcinogens the risk of cancer will not exceed one excess case in 100,000 exposed individuals (WHO, 1996). The ADI or RfD can be used to establish guidelines for safe chemical threshold values in the soil based on the expected exposure to a chemical resulting from the application of sludge to agricultural land (Chang et al., 2002).

Based on computational modelling of the environmental exposure pathways described above, the organic chemicals that are likely to pose the greatest risk to human health are those that have a combination of low or very low acceptable daily intakes (i.e., are highly toxic, e.g., dioxin) and are readily absorbed by plants (see Table 3). Often the primary determinant of whether an organic chemical found in the sludge will pose a health threat or not is based on its potential for absorption by the plant. Relatively toxic chemicals that are not absorbed by plants (e.g., PAHs) are less likely to result in a threat to human health through food chain transfer as a result of the application of sewage sludge to agricultural land (Chang et al., 2002). According to Andersen (2002b), however, soil and sludge ingestion on land used for grazing of cattle, and not the plant route, is the main route for animal contamination.

### 3.8. Direct human exposure

There is a need to differentiate between the general population, which is rarely exposed to sewage sludge and sludge-amended soil in a direct way, and some specific subgroups like farmers or sludge workers, for which exposure is more realistic and may require risk assessment.

#### 3.8.1. Soil ingestion

Humans can be exposed to sludge-related organic chemicals in soils by directly consuming the contaminated soil. For example, ingestion of soil can occur directly when contaminated hands transfer soil to the mouth, or through inhalation and swallowing (Gerba et al., 2002; Abrahams, 2002). People that exhibit soil-pica\(^2\) behaviour or infants and small children up to 18 months old are at particular risk from this type of exposure (Lacey, 1990). Direct exposure to sludge-related organic chemicals in soils could occur in several situations, the most relevant of which is in the field to farm workers and possibly their families. Exceptional situations would be in home gardens where soil conditioners with sewage sludge are used, and in residential areas converted from

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\(^2\) Pica is an eating disorder that results in cravings for nonfood items (e.g., soil, paint chips, clay, hair, soap, and plant leaves) or for unusual amounts of food items (Lacey, 1990).
farmland where sewage sludge was previously applied (a scenario mainly relevant for heavy metals).

Risk assessment identifies soil ingestion by children as being amongst the most sensitive environmental exposure pathways to a range of inorganic and organic substances, if this vulnerable group is exposed to contaminated soil. The ingestion of soil by children is not usually a significant route of exposure from the conventional use of sewage sludge as an agricultural fertiliser. Land use restrictions provide an effective barrier against the transmission of enteric disease. A small proportion of children exhibit soil-pica behaviour and can ingest significantly larger amounts of soil than a typical child in the general population. Some children have been reported to ingest as much as 25–60 g in a single day and ingestion rates in the range >1–13 g/day are also reported for pica children (Calabrese et al., 1991; Calabrese and Stanek, 1993). LaGoy (1987) considered a value of 5 g day\(^{-1}\) to be a reasonable estimate of a maximum single-day exposure for a child with habitual pica. Calabrese et al. (1991) suggested that an assumption that one in 200 children ingests about 1 g of soil 4 days per week for 4 years during the 1–6 years age span would be appropriate to account for soil-pica behaviour in risk assessment analyses. An overview of potential soil ingestion rates is provided in Table 7.

The level of exposure to chemicals from this route can be quantified by using the following information: concentration of chemicals in the soil, the amount of soil consumed, the body weight of the consumer, the percentage of the chemical absorbed by the body, and the duration of the exposure. This estimated exposure level then can be compared to an ADI or RfD as described in Section 3.7.

Children that exhibit soil pica may ingest large amounts of soil resulting in a risk of acute toxicity due to contaminants ingested in soil or sludge. Calabrese et al. (1999) showed that contaminant intakes during soil pica episodes could exceed acute toxicity dosages causing illness in humans. For certain compound types, apparently conservative soil/sludge criteria based on chronic lifetime exposures may not be protective of children during acute soil pica events.

3.8.2. Exposure to airborne dust and via volatilization

The exposure of an individual inhaling vapours of any volatile pollutant that may be in the sludge should be assessed. Defining exposure from volatilization can be quite complex. Expert judgement or a (multimedia) model calculation with a sensitivity analysis based on chemical properties related to volatilization and sorption can help to define when this would be needed.

Volatilization models depend on parameters such as Henry’s law coefficient, sorption, surface, diffusivity, etc., in combination with assumptions about the air volume passing over the site. A simple screening approach for calculating volatilization rate is provided in Andersen (2001b) or in ECETOC (2002).

Existing occupational and consumer exposure models can be used or reformulated to perform a risk assessment of volatile and particle bound chemicals respired from air. US-EPA (1992) provides a source of technical guidance to develop occupational exposure scenarios for chemicals in sludge.

4. Conceptual framework for sludge risk assessment

4.1. Formulating the conceptual framework

In this section, the principal elements related to a risk assessment of organic contaminants applied with sewage sludge to soil are schematized in a Conceptual Framework (Fig. 4). Conceptual models serve to summarize the results of the problem formulation and provide a roadmap to the actual assessment. The goal of any risk assessment is to find threshold levels—here called Sludge Quality Standards—for critical effects below which no significant adverse risk is expected. The ultimate aim is to broadly protect public health and the environment, while maintaining the possibility for sludge recycling in agriculture.

The proposed CF emphasizes the need for a holistic and systematic approach to risk assessment for sludge in agriculture. It is equally applicable to any persistent and/or toxic metabolite that may be formed from a parent compound. In order to keep a sustainability perspective, the assessment should consider both immediate effects of both parent compounds and metabolites, as well as long term application and potential accumulation of chemicals in the soil.


Starting from a risk assessment in the soil, in makes the link to the technical practice of sludge treatment (Table 3) and use (Good Agricultural Practice). The CF is aimed to serve as a ‘roadmap’ to perform a scientifically consistent and holistic sludge risk assessment or organic chemicals. As such, the CF can be used for priority setting of chemicals for further assessment, and where needed, for setting regulatory SQS and sludge management strategies.
<table>
<thead>
<tr>
<th>Comment</th>
<th>Mean (mgday⁻¹)</th>
<th>50 percentile (mgday⁻¹)</th>
<th>95 percentile (mgday⁻¹)</th>
<th>Max (gday⁻¹)</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fifty nine children, Al and Si tracers</td>
<td>181–184</td>
<td></td>
<td></td>
<td>0.1–0.5</td>
<td>Binder et al. (1986)</td>
</tr>
<tr>
<td>Normal child</td>
<td>25–100</td>
<td></td>
<td></td>
<td>5</td>
<td>LaGoy (1987)</td>
</tr>
<tr>
<td>Pica child</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Children in day care</td>
<td>0–90 (geometric)</td>
<td>Up to 190 (90 percentile)</td>
<td></td>
<td></td>
<td>Van Wijnen et al. (1990)</td>
</tr>
<tr>
<td>Children in camping groups</td>
<td>30–200</td>
<td>Up to 300 (90 percentile)</td>
<td></td>
<td></td>
<td>Van Wijnen et al. (1990)</td>
</tr>
<tr>
<td>Pica child (1 in 200) aged 1–6 years</td>
<td></td>
<td></td>
<td></td>
<td>1g 4 day week⁻¹ for 4 years</td>
<td>Calabrese et al. (1991)</td>
</tr>
<tr>
<td>Sixty children</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Calabrese and Stanek (1992)</td>
</tr>
<tr>
<td>Six children, average aged 3.1 years</td>
<td>10 (Zr), 19 (Al), and 36 (Ti)</td>
<td>200</td>
<td></td>
<td>&gt;1</td>
<td>Calabrese and Stanek (1992)</td>
</tr>
<tr>
<td>Soil-pica child aged 3.5 years</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Calabrese et al. (1991);</td>
</tr>
<tr>
<td>Sixty four children</td>
<td>45</td>
<td>13</td>
<td>138</td>
<td></td>
<td>Calabrese and Stanek (1992)</td>
</tr>
<tr>
<td>Sixty four children aged 1–4 years</td>
<td></td>
<td>&lt;1</td>
<td>160</td>
<td>0.5</td>
<td>Calabrese and Stanek (1995b)</td>
</tr>
<tr>
<td>Twelve children aged 1–3 years</td>
<td></td>
<td></td>
<td></td>
<td>0.5–3.05</td>
<td>Calabrese et al. (1997a)</td>
</tr>
<tr>
<td>Children</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Cohen et al. (1998)</td>
</tr>
<tr>
<td>Soil-pica child</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Calabrese et al. (1999)</td>
</tr>
<tr>
<td>Young children</td>
<td>110 (central estimate)</td>
<td></td>
<td></td>
<td></td>
<td>Stanek et al. (2001)</td>
</tr>
<tr>
<td>Children</td>
<td>24 ± 4 (median)</td>
<td>91 ± 16.6</td>
<td></td>
<td></td>
<td>Stanek et al. (2001)</td>
</tr>
<tr>
<td>Adult</td>
<td></td>
<td></td>
<td></td>
<td>&lt;0.48</td>
<td>Kissel et al. (1998)</td>
</tr>
<tr>
<td>Ten adults</td>
<td>10 ± 94</td>
<td>1</td>
<td>331</td>
<td></td>
<td>Stanek et al. (1997)</td>
</tr>
<tr>
<td>Pet dog</td>
<td></td>
<td></td>
<td></td>
<td>20</td>
<td>Calabrese and Stanek (1995a)</td>
</tr>
</tbody>
</table>
4.2. Data availability, quality assurance, and validation aspects

A sludge risk assessment according to the CF, with derivation of an SQS, and can in principle be based solely on the data required by the so called ‘Base Set’ for the notification of new chemicals in the EU (CEC, 1992). It is quite likely that such a ‘tier 1’ assessment will come with considerable uncertainty associated with the end result. It is good scientific practice to define the areas of greatest uncertainty—possibly via a mathematical uncertainty and sensitivity analysis of the models used (e.g., by a Monte Carlo tool)—to define where additional data generation can be most relevant and efficient. Uncertainty analysis is mostly feasible on distinct and smaller parts of the fate and/or exposure models, but rapidly becomes very complex for large and comprehensive risk assessment models, such as for example EUSES. The CF follows the basic principle of tiered risk assessment, where the identification of risk and significant data gaps should lead to further testing, and a new round of iteration of the risk assessment.

It can be expected in practice that SQS setting will only be considered for (classes of) chemicals with known hazard and elevated sludge levels. The CF is therefore conceived primarily for the more data-rich chemicals, for which reliable fate, partitioning, bioaccumulation and (eco)toxicity data are already available. But even then, specific data (like for example measured soil-crop transfer coefficients) are often lacking, and not all parts of the exposure assessment will be equally accurate.

The ILSI-Europe Expert Group also recommends the selection of realistic test methods, exposure scenarios, and lab-to-field validation. For example, bioavailability effects of sorptive compounds can be incorporated in the laboratory test design by dosing via sludge rather than pure aqueous solutions. Field experiments at realistic temperatures and root penetration depths should be preferred over pot trials in the greenhouse. Functional rather than structural endpoints should be used in microbial tests, and the soil fauna—and plant species selected should be—where possible—typical for agricultural fields in the climate zone of interest.

Finally, it should be mentioned that the conclusions from a risk assessment should be verified for their plausibility against other sources of data. Such information can come, for example, from epidemiological studies for health risk, biodiversity studies, or sustained agronomic performance demonstrated in large scale field studies. If sludge of a certain, known, origin has been used in agriculture for a prolonged time period, the farmer’s assessment of crop yield and appreciation of plant health can provide a way to (in)validate the phytoxicity assessment based on laboratory experiments.

4.3. Stepwise procedure to derive sludge quality standards

The recommended risk assessment procedure based on the Conceptual Framework in Fig. 4. is as follows:

Step 1—Effects assessment. Define, based on the evaluation of the different relevant endpoints and risk assessment the lowest relevant PNEC value for a chemical. This may be a direct PNECsoil number, or a back-

Fig. 4. Conceptual framework, processes, endpoints, and boundaries for agricultural soil risk assessment, and risk-based derivation of SQS. Transport and transfer processes by which organic pollutants applied to agricultural land through sewage sludge can reach soil biota, plants, aquatic organisms, humans, domestic animals, and wildlife are given in italics. Numbers in the boxes refer to the respective sub-sections where this endpoint is discussed.
calculated PNECsoil from, e.g., predator- or human health effects.

Step 2—Exposure Assessment and derivation of SQS. The PNECsoil will define the maximum acceptable exposure level (PECsoil) at all times. A Sludge Quality Standard (or set of Standards) can be derived from the PECSoil based on typical application regimes. This SQS relates to the concentration in the sludge (PECsludge) at the time of application. It can be further extrapolated to fresh sludge concentrations, in order to reflect eventual known treatment and conditioning of the sludge prior to application. It is known that sludge treatment processes like anaerobic digestion, aerobic stabilization or composting (see Table 2) will often alter the PECsludge. This, of course, requires insight in the fate processes that may occur during sludge treatment.

An alternative way of presenting and communicating the SQS can be to express it as mass flux (kg chemical/ha.year) rather than as a sludge concentration.

Step 3—Validation. Because of the usually repeated application of sludge to land, it is also important to define the time horizon for which the assessment should be valid (typically 20, 50 or 100 years). For a sustainable application regime it is essential that there is no build-up in the soil over time above the soil PNEC value. Simple soil mass balance models have been proposed for metals and organics e.g., by Andersen (2001b) and Jones and Stevens (2002) to address the risk of accumulation over time. The State Institute for Environmental Protection Baden-Wuertemberg (2003) observed that as a consequence of many years of sludge application persistent organic compounds such as PCDD/PCDF, polycyclic musks and organotins were building up in the soils. By contrast, soil build-up for readily biodegradable chemicals has not been observed.

In case there is a risk of accumulation, either the sludge application rates or the SQS should be adjusted downward. When the actual or predicted concentration of a chemical in sludge is below the SQS, there should be no further concern or basis for specific regulations.

4.4. Concluding remarks

In the chain from source to sink—i.e., from factory or household to agricultural soil—the control of certain pollutant groups in sewage sludge can be a pragmatic approach to secure continued sludge use in agriculture, to protect soil quality and the food chain, and to stimulate emission reductions at the source. This paper aims to provide a holistic and systematic methodology to derive quality standards for organic anthropogenic contaminants with potential environmental or human health risk. The outcome of such a risk assessment could be used, in combination with analytical, economical, and management considerations, to decide whether there is a need to legislate a certain class of compounds.

The Conceptual Framework can propose appropriate Sludge Quality Standards that would allow a safe use for a selected sludge application regime and time horizon.

It must be acknowledged at the same time that the accuracy of the risk assessment calculation depends heavily on quality and field relevance of the input data. Only few organic pollutants come with a sufficiently complete test data set allowing to apply the Conceptual Framework, and with robust and affordable analytical methods in sludge and soil matrices. In other cases, such as for the analytical sum parameter AOX, the Conceptual Framework cannot be applied for methodology reasons.

This raises the question how to deal with chemicals with limited data availability, chemical mixtures and variability in field conditions. These issues also surface in other domains of risk assessment. While methodology and test data gaps are being filled over time, the regulator has the challenging task to define whether or not interim (precautionary) limits for sludge are to be set. This situation could eventually support an approach where regulatory ‘Limit Values’ are used next to ‘Guide Values.’ Limit Values are to be implemented only for those chemicals in sludge for which a robust assessment suggests a significant long term risk. To deal with data-poor and uncertain cases, indicative ‘Guide Values’ could be considered. These would be based on a preliminary and conservative risk assessment calculation, or direct extrapolation from other chemical groups. Guide Values aim to provide an indication of an acceptable sludge concentration, and could be monitored flexibly depending on regional relevance and legislative priorities. As more data come available, a ‘Guide Value’ could either become a ‘Limit Value,’ or the chemical may be taken off the list.

4.5. Preview to the LAS case study

A methodological example will be presented in a separate paper by the ILSI-Europe Expert Group on Risk Assessment of Sludge how (a set of) SQS can be derived based on the approach outlined in the Conceptual Framework. The compound chosen is LAS (linear alkylbenzene sulphonate, CAS 68411-30-3), for which a Limit Value of 2600mg/kg sludge is currently proposed in the third Draft of the Working Document on Sludge on Sludge (CEC, 2000c), and a value of 1300mg/kg sludge in the Danish National legislation). The anionic surfactant LAS is the most important surfactant worldwide, and 350,000—400,000 tonnes/year are used in Western Europe, mainly in laundry detergents.

LAS comes with a very considerable set of test data. The ILSI-Europe risk assessment for LAS is based on robust summaries of the most recent and
quality-assured fate and (eco)toxicity data, and updates previous soil and sludge assessments in the literature. However, also the ILSI assessment and the derived SQS may need to be revised in an iterative way in the future for more, or more realistic data come available.

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