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Organic Contaminants in Sewage Sludge

A Basis for the "Good Sludge and Phosphorus in the Ecocyle"

RAPPORT 5217

Organic Contaminants in Sewage Sludge

Review of studies regarding occurrence and risks in relation to the application of sewage sludge to agricultural soil

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SWEDISH ENVIRONMENTAL PROTECTION AGENCY

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Preface

The Swedish Government has commissioned the Environmental Protection Agency (EPA) to examine the scope for increasing the recycling of phosphorus from sewage systems and to evaluate the need for revised regulations on the use of sewage sludge on agricultural land. The EPA delivered the final report “Aktionsplan för återföring av fosfor ur avlopp” December 16, 2002. This report concerns one of the sub-projects. This report has been financed by the Swedish EPA and the Swedish Water & Waste Water Association.

The report presents a review of a number of different studies regarding the occurrence and risks (for human health and the environment) of different organic contaminants in relation to the use of sewage sludge in agriculture.. The review includes material from Sweden, Norway, Denmark, Germany, the United Kingdom and the USA. In addition, consultant reports in the field compiled for the European Commission are taken into account. The main conclusions of the report are priority-setting of organic substances based on associated risks and an outline for strategies to minimise risks These conclusions form a basis for the identification of strategies and proposals stipulated in the final report.
The author, Lise Samsøe-Petersen of the DHI Water & Environment in Copenhagen, is solely responsible for the content of this report.

Stockholm, February 2003
Swedish Environmental Agency

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Summary

The immediate objective of the project is priority setting of organic substances or groups of substances, based on the risks associated with them, including identification of substances, which could be handled differently at the product level. Furthermore, an outline of principles should be developed for a short-term and a long-term strategy with the aim to minimise the risks, pertaining to organic contaminants in sewage sludge applied to agricultural soil.

The report includes a "state of the art" on the knowledge regarding the occurrence of and risks pertaining to organic contaminants in sewage sludge. This "state of the art" is based on a review of recent reports or studies from the European Union, Sweden, Norway, Denmark, Germany, the United Kingdom and the U.S.A.

The information from the above reports is discussed and the substances of concern, identified in these reports are discussed. Based on their properties and fate in the environment, the substances are grouped. For each group of substances, an assessment as to whether the substances are of concern as contaminants in sewage sludge is made.

For the priority setting, the contribution of substances to agricultural soil from sewage sludge is related to the contribution from other sources; i.e. fertilisers and atmospheric deposition. Furthermore, the fate of organic substances in soil is considered.

Substances, which have not been included in previous risk assessments and/or legislation but which may be of concern, are identified and a few comments on their properties and potential as "candidates for concern" are included. However, detailed assessments of these substances were not part of the project.

Based on the above, a strategy for measures to be taken in order to prevent risks from contamination of agricultural soil with organic substances via sewage sludge is outlined.

Introduction

This project was initiated by the Swedish Environmental Protection Agency (Naturvårdsverket)) based on a need for revision of the Swedish regulation on Sewage sludge.

In 2000 the Swedish parliament passed a series of "Environmental quality objectives" (Miljøkvalitetsmål). These objectives include:

- "Re-circulation": A considerable part of the phosphorous from waste and sewage should be returned to agricultural soil by 2010
- "A poison free environment": The environment must be free from substances and metals, which have been created in or extracted from the society and which can threaten human health or biological diversity

Provided that the phosphorous is not extracted from sewage sludge (and purified) before application, the first objective implies application of sewage sludge to agricultural soil. This in turn implies application of the chemicals, which are present in the sludge and many of these are known to be able to "threaten human health or biological diversity".

Therefore, there is an immediate conflict between these two objectives and the Swedish EPA initiated a series of projects, which will form the basis for the development of a long-term strategy for sustainable use of sewage sludge for agricultural application in Sweden. The outcome of one of these projects is presented in this report.

The immediate objective of the project is priority setting of organic substances or groups of substances, based on the risks associated with them, including identification of substances, which could be handled differently at the product level. Furthermore, an outline of principles should be developed for a short-term and a long-term strategy with the aim to minimise the risks, pertaining to organic contaminants in sewage sludge applied to agricultural soil.

Numerous risk assessments of the application of sewage sludge on agricultural soil have been produced during the latest decade. In order not to copy these, the definition of the task by the Swedish EPA includes that the assessment in this project is based on a review of sludge assessment reports from as many countries as possible.

The report includes a "state of the art" on the knowledge regarding the occurrence of and risks pertaining to organic contaminants in sewage sludge. This "state of the art" is based on a review of recent reports or studies, produced in several industrialised countries.

For the priority setting, the contribution of substances to agricultural soil from sewage sludge is related to the contribution from other sources; i.e.

fertilisers and atmospheric deposition. Furthermore, the fate of organic substances in soil is considered.

Based on their properties and fate in the environment, the substances are grouped. For each group of substances, an assessment as to whether the substances are of concern as contaminants in sewage sludge is made.

For groups of substances, which are identified as being of concern, the possibilities of identification of the sources of their origin are discussed.

Furthermore, substances, which have not been included in previous risk assessments and/or legislation but which may be of concern, are identified and a few comments on their properties and potential as "candidates for concern" are included. However, detailed assessments of these substances were not part of the project.

Based on the above, a strategy for measures to be taken in order to prevent risks from contamination of agricultural soil with organic substances via sewage sludge is outlined.

Overview of reviewed reports and other information

Reports, legislation and other relevant literature from the following countries/authorities were reviewed for the project: Sweden, Norway, Denmark, England, Germany, the European Union and U.S. EPA.

The documents included reports regarding concentrations and sources of organic substances in sewage sludge, other fertilisers and atmospheric deposition as well as risk assessments of sewage sludge use and/or individual substances.

The review of EU reports showed that limit or guidance values have been elaborated in France and Austria for several organic contaminants. The substances include PCBs and PAHs in both countries, and limits for PCDD/Fs and AOX are fixed in Austrian. The French and Austrian background literature has not been available for the project.

Substance names and abbreviations

For the names of most of the substances or groups of substances, which are considered in this report, abbreviations are usually employed in text and tables. Therefore, Table 2.1 gives an overview of the substance names and the abbreviations used in the report.

Table 2.1 Overview of substance names and abbreviations

Name	**Abbreviation**
Polynuclear aromatic hydrocarbon	PAH
Polychlorinated dibenzo-p-dioxins and –furans	PCDD/F
Polybrominated diphenyl ethers	PBDPE
Decabromo diphenyl ether	DecaBDPE
Pentabromo diphenyl ether	PentaBDPE
Octabromo diphenyl ether	OctaBDPE
Polybrominated biphenyl	PBB
Polyclorinated biphenyl	PCB
Linear alkyl benzene sulfonates	LAS
Di-2-(ethylhexyl) phthalate	DEHP
Dibutylphthalate	DBP
Diethylphthalate	DEP
Nonylphenol	NP
Nonylphenol +-ethoxylates	NP(+EO)
Adsorbable organic halogenated compounds	AOX
Volatile organic compounds	VOC

Review of individual documents

The European Union

In the European Union, a revision of the sewage sludge directive (Dir. 86/278/EEC) has been initiated and for this, a number of reports have been prepared, which are all available at a special “sewage sludge web-site” on the EU homepage (EU 2002).

Among these reports, three were identified as being particularly relevant for the present project: “Organic contaminants in sewage sludge for agricultural use” (Erhardt & Prüess 2001), “Pollutants in urban wastewater and sewage sludge (Thornton 2001) and “Disposal and recycling routes for sewage sludge. Scientific and technical sub-component report” (Andersen 2001).

Furthermore, a workshop and a conference have been arranged under EU auspices. The workshop was arranged by the Joint Research Centre of the European Commission in November 1999. The conference, arranged by Environment DG, was held in October 2001. The proceedings from the workshop (Langenkamp & Marmo 2000) and presentations from the conference were available.

Organic contaminants in sewage sludge for agricultural use

The report “Organic contaminants in sewage sludge for agricultural use” (Erhardt & Prüess 2001) is focused on the substances, which are listed in the EU “Working document on sludge” (Environment DG 2000): LAS, DEHP, NP(E), PAH, PCB and PCDD/F. It gives an overview of the occurrence of these organic compounds in sewage sludge, basic toxicological data, a review of persistence of organic contaminants in soil and risk assessments for the various pathways. Furthermore, an attempt is made to identify additional substances or groups of substances, which might cause hazards and should be regulated.

The report is a desk study, which is mainly based on national regulations and risk assessments of sewage sludge from the U.S. and a series of EU countries, including Sweden, Norway, Denmark, Germany, England and Ireland.

The risk assessments are based on assessments of human exposure via each of the routes: Sludge-man, soil-man, soil-plant, soil-(plant)-animal and soil-water. Furthermore, possible effects on microbial activity in soil, soil living animals and plant growth are considered.

The conclusion section is a mixture of conclusions and suggestions for further activities and includes individual recommendations from a series of documents. The only general conclusion in terms of risks, which can be quoted, is that “Persistent compounds such as PCBs, PCDD/Fs and PAHs are generally not transferred from soil to crops, meat and milk although the possible evaporation of PCBs and foliar uptake needs more attention. Little is known about the uptake of phthalates and nonylphenol which are present in relatively high levels in sludge.”

The following (groups of) substances are described and reasons for concern are indicated: Brominated flame retardants (PBB and PBDPE), endocrine disrupting chemicals, chlorobenzenes, “chloroorganic pesticides”, musk ketone, musk xylene, chlorinated paraffins, organotin compounds and volatile organic chemicals.

Disposal and recycling routes for sewage sludge

In the report “Disposal and recycling routes for sewage sludge. Scientific and technical sub-component report” (Andersen 2001), detailed descriptions of all stages of the production and disposal of sewage sludge - including information regarding the contents of organic contaminants in sludge from different countries – are presented. Furthermore, the fate of the substances in soil and uptake routes in plants and livestock are described leading to conclusions regarding human exposure (Section 6). In an appendix (3), individual contaminants are briefly described in terms of their physical-chemical properties, toxicity, origin in sludge, behaviour in soil, transfer to water, uptake by plants and livestock, ecotoxicity and human exposure level. The organic substances in this appendix include PAHs, PCBs, PCDD/Fs, phthalates and LAS.

The conclusions are summarised as follows in the report (Andersen 2001):

“Therefore, considering presently available knowledge on organic compounds, it appears at the present time, that:

- Transfer to water is low, micro-organisms adapt to changing conditions in soil, and numerous organic compounds are rapidly degraded in soil. Attention should therefore mainly be given to compounds with higher half-life time values.
- From the point of view of crop protection, no limit value seems to be necessary as transfers to plant do not occur for most organic compounds.
- Restrictions should focus on bio-accumulative compounds spread on grazing land such as PCBs and PCDD/Fs. In this case deep injection of sludge could reduce the risk of livestock contamination by organic pollutants.
- A survey of organic pollutant levels in sludge should be performed by sludge producers, focusing on the specific organic pollutants identified within the wastewater catchment area of the WWTP.”

Pollutants in urban wastewater and sludge

The primary objective of this report (Thornton 2001) is to determine the sources of pollutants in urban wastewater. Furthermore, the contents of contaminants in wastewater and sewage sludge are evaluated in order to propose measures for reduction of pollution at the source.

The criteria for selection of new organic substances for evaluation are not described, except that the substances could be present in sludge and "maintain their integrity" during treatment and following application to agricultural soil.

The organic substances include chlorinated paraffins, brominated diphenyl ethers, polychlorinated naphthalenes, quintozene (pentachloronitrobenzene), polydimethylsiloxanes (silicone polymers), nitro musks (chloronitrobenzenes), oestrogenic compounds (natural and synthetic estrogens), pharmaceuticals, synthetic fats and polyelectrolytes (i.e. polyacrylamide and cationic copolymers).

As the objective of the report is not risk assessment, only a few comments regarding exposure and effects of some of the substances are included. However, for identification of potential substances of concern, this report introduces several substances, which are not included in any of the other reports.

Sweden

The Swedish EPA has made a series of reports and information material available. The documents can be grouped in three categories:

Informative documents:

- Background information for the sewage sludge handling practices (Naturvårdsverket, LFR & VAV 1995),

Review documents on wastewater and/or sewage sludge:

- Concentrations of substances in sludge (Naturvårdsverket 1992).
- Review regarding human exposure and environmental impact as well as recommendations for sewage sludge handling (Naturvårdsverket 1993).
- Experiences from the first years of the Swedish sewage sludge agreement (Naturvårdsverket 1996).
- Concentrations of substances in sludge and trends of sludge handling (Levlin et al. 2001).

Research projects relevant for agricultural use of sewage sludge:

- Field study with sewage sludge (Andersson & Nilsson 1999).

- Degradation of substances in sludge during composting (Amundsen et al. 2001).
- Accumulation of persistent organic pollutants in earthworms (Matscheko 2001).
- Analyses of field soil in Marks Municipality where sludge from the waste water treament plant of Skene has been applied during the years 1978-1984 (Ek 2002).
- Assessment of possibilities and limitations for chemical risk assessment including chemical analyses of household wastewater (Palmquist 2001).

"Single" substance assessments:

- Microbiological effects of cationic polyacrylamides (Johansson et al. 1998).
- Human exposure assessment of endocrine disrupting substances (Nilsson 1996).

Statistics:

- Sources of contaminants (Naturvårdsverket & Statistics Sweden 1999); included in Levlin et al. 2001.

In summary, these documents include information regarding levels of contaminants in sewage sludge, ecotoxicoloigal properties, human exposure and risk assessments as well as (evaluation of) procedures for sludge handling. Only documents, which include information regarding the Swedish risk assessment process, are reviewed below.

"Sludge - Contents of organic environmentally hazardous substances - Summary and evaluation of analytical results" (Naturvårdsverket 1992). Results of analysis of 27 sludge samples from different WWTP for 70 "priority pollutants" (from an U.S. EPA list) are summarised, showing a series of substances to be present in (almost) all sludge samples.

Furthermore, results of similar analysis of samples of agricultural soil, cattle and pig manure as well as the polyacrylamide used for dewatering of sludge are presented. A few (groups of) substances were detected in the manure samples but except for cresols in both types of manure, the levels of concentration were lower than those in sewage sludge. In the soil samples, only low concentrations of a few organic solvents are inexplicable. The sources of the substances identified in the sewage sludge were examined through analysis of samples of municipal and industrial wastewater.
The fate (degradation, uptake in plants and leaching) of selected substances was investigated by a literature review. Furthermore, atmospheric deposition of these substances was compared to the amounts of substance added by sewage sludge. The substances included PAHs, naphthalenes, phenols, phthalates, toluene, m/p-xylene, PCBs, and dioxins. Most of the substances were considered as being relatively readily biodegradable in soil. The exceptions were PCBs and dioxins. For airborne substances, the contribu-

tion from sludge to the exposure of agricultural soil was considered to be low. Sewage sludge was however, the main contributor of other substances, like NP. With respect to NP, substitution is in progress in Sweden. The concern is effects on aquatic environments of this substance in WWTP discharges and not it's occurrence in sludge. Finally, the limitation of the choice of substances analysed for is pointed out.

"Cleaner sludge - Measures for municipal wastewater treatment plants" (Naturvårdsverket 1993): This report is a follow-up on the previous, as results of analyses for the substances: NP, PCBs (7 congeners), PAHs (6 substances) and toluene (indicator of organic solvents) on a number of sludge samples are presented.

Furthermore, the report includes summaries of a series of investigations and assessments carried out by Naturvårdsverket. These include

- Reviews for identification of potential substances of concern in sewage sludge
- Investigations of the toxicity of sewage sludge and manure to soil microbial organisms/processes
- Field experiments for investigation of possible increases of substance levels in soil and uptake in plants as well as ecotoxicological effects in soil
- Identification of sources of substances in wastewater.

The identification of potential substances of concern is based on comparisons of a database listing "multiproblem-substances" (i.e. substances known to be toxicologically as well as ecologically hazardous) with databases of Swedish high volume production chemicals and of chemicals known to be discharged to public WWTP. Among the resulting substances, those, which were known to be not readily biodegradable and bioaccumulative, were retained for further assessment (34 chemicals). Among the 34 chemicals, 19 were already included in previous analysis (Naturvårdsverket 1992). For the remaining, the existence of reduction measures and/or the relevance for exposure via sewage sludge were assessed.

The conclusion was, that the only potential problem would be persistent organic compounds (PCBs and dioxins and the like) for which it must be assured that no accumulation in soil takes place.

Finally, measures to be taken locally in order to avoid discharge of the substances to wastewater, e.g. identification of sources, are described in detail as well as implements for reduction of discharges to WWTP.

"The agreement on the application of sludge in agriculture between the Federation of Swedish Farmers, the Swedish Water and Wastewater Association and the Swedish EPA - a follow-up on the first years: 1994-

1996" (Naturvårdsverket 1996). In the report, the results of analyses of sludge samples during the first two years of the agreement (1994-1996) are presented. Several measures for reducing the discharge of organic contaminants to public wastewater are listed. In summary, almost two thirds of the sludge samples analysed complied with the limits for organic contaminants set in the agreement as well as with the legally fixed limits for heavy metals. A decreasing trend for most substances is seen. It is further concluded that another substance than toluene should be found to serve as the indicator of organic solvents. At the revision of the sewage sludge agreement in 1999, toluene was removed from the list, partly because it gave analytical problems, partly because it was shown to be formed in the WWTPs, so it was not indicative of sources of contamination.

"Field experiments with municipal sewage sludge from Malmö and Lund during the years 1981-1997" (Andersson, P.-G. & P. Nilsson 1999). In this report, the results of long-term field experiments with application of sewage sludge are presented. The studies included chemical analysis (70 substances) of sludge and soil, monitoring of microbiological parameters and soil living worms as well as degradation of organic contaminants in the soil. As to the organic contaminants in soil and crops, the results show high variation and in most cases, the concentrations of substances, which are detected, can not be correlated to the sludge application. In plants, DEHP is the only substance for which a relationship between sludge application and detection in the crops seems to be present. In soil, no build up of organic contaminants, which can be related to the sludge application, is found. Degradation studies show degradation of PAHs but not of PCBs. Effects on microbial activity and worm populations are stimulation, and Enchytreiid worms are favoured for earthworms.

"Sewage sludge quality and trends of the handling of sludge" (Levlin et al. 2001). Sources of heavy metals and selected organic substances (NP, PAHs, PCBs and phthalates) in wastewater and sewage sludge are reviewed. Results of sludge analyses from existing statistics (Statistics Sweden and Swedish Water and Wastewater Works association) are summarised. The period range from 1969 to 1998. Until 1994, the analyses comprised metals only but from 1995 onwards, organic substances were included. In 1998, most samples had concentrations of NP, PAHs and PCBs below the limit values. Toluene, however was present at levels above the 5 mg/kg dw limit in about 20% if the samples (1997, Tabell 8). The highest concentrations of toluene were found in the smaller WWTPs; i.e. with less than 20.000 pe. The average level of toluene in Swedish sewage sludge doubled between 1995 and 1998 (from 2.3 to 4.5 mg/kg dw), while the level of NP was halved (46.6 to 22.8) but levels of PAHs and PCBs seem to be constant (at 1.8 and 0.1 mg/kg dw, respectively).

Alternative uses of sewage sludge are discussed: Green areas, forests, construction soil, recovery of metals and phosphorous, disposal and incineration. It is recommended that the individual WWTP uses several different ways of marketing the sludge in order to be less sensitive to changes in attitude to the acceptance of sludge for one purpose.

Recent analysis of Swedish sewage sludge

In 2001, the County Administration of Västra Götaland and the Swedish EPA initiated analyses of sewage sludge samples from 19 different Swedish WWTPs for a series of different types of organic contaminants. Furthermore, University of Stockholm has analysed sludge from 50 WWTPs for brominated flame retardants (Nylund et al. 2002). The results are summarised in Table 3.1.

Dependent on the available results, the mean values presented in Table 3.1 are calculated either as the mean of reported sum-values or as the sum of calculated mean values for each substance in a group. Graphs presenting concentrations of individual substances are shown in Annex I.

Table 3.1 Preliminary results of analysis of Swedish sewage sludge 2001, mean values of samples from 19 or 50 (brominated flame retardants) WWTPs. Results from sludge analysis in 1998 (413 WWTPs) and 1995 (420 WWTPs) as well as Swedish guidance values for PAHs,PCBs and NP. All units mg/kg dw.

Group of substances	2001	1998	1995	Guidance
PAHs	1.1	1.6	1.8	3
PCBs	-	0.1	0.1	0.4
4-nonylphenol	3.9	22.8	46.6	100
Brominated flame retardants	0.47	-	-	-
Chlorinated paraffins	< 2.0	-	-	-
Chlorobenzenes	< 0.28	-	-	-
Chlorophenols	< 0.9	-	-	-
DEHP	6.6	-	-	-
EDTA	3.8	-	-	-
EOX	6.1	-	-	-
LAS	252	-	-	-
Organic tin compounds	0.65	-	-	-
PCDD/Fs	0.0013	-	-	-
Perfluorooctansulfonate	0.08	-	-	-
Phthalates	7.3	-	-	-
Triclosan	1.2	-	-	-

-: No information available

Concentrations of PAHs and NP have been decreasing since 1995. The level of PCBs – for which no results are available for 2001 – was constant in 1995 and 1998. The concentrations listed in Table 3.1 are all below the Swedish guidance values (Naturvårdsverket 1996) but it should be remembered that the values are means of results from many WWTPs of which some are higher than the guidance values.

Norway

The available Norwegian reports comprise surveys of sources for organic contaminants in sludge (Midttun & Sorteberg 1997) as well as surveys of concentrations of organic substances in samples of sewage sludge (36 monthly composite samples), compost from source separated household waste (9 samples) and samples of cattle and pig manure (4 samples of each) (Paulsrud et al. 1998).

The focus of the waste surveys (Paulsrud et al. 1998) was prioritisation of possible problematic substances in Norwegian waste products. The measure for assessment was regulations set in other countries and no further assessments were elaborated.

For the selection of substances for analyses, the following criteria were applied:

- Listed as a high priority contaminant by the health and environmental authorities
- Detected in fairly high concentrations in a previous (1989) Norwegian sewage sludge study
- Included in other countries' sewage sludge or compost regulations or guidelines (Denmark, Germany, Sweden)
- Limits to the costs of laboratory analyses

Based on these criteria, six main groups of organic compounds were selected (numbers of substances in parenthesis):
PCDD/Fs (17), PCBs (7), PAHs (16), alkylphenols (NP/-EO, 2- methylphenol, 3,4-methylphenol), phthalates (DEHP, DBP) and LAS.

The reported concentrations were compared to limit/guidance values in the countries mentioned and for most substances, the concentrations in Norwegian samples were generally lower than the values used for comparison. The main exception from this picture was NP(+EO), which was above the Danish and Swedish limit/guidance values for all WWTPs.

There is no document available, describing the Norwegian process of decision but Paulsrud et al. (1998) quote the Norwegian authorities as follows:
"Based upon the results of this survey, the Norwegian authorities have decided not to include limit values for toxic organics in the existing regulations for sewage sludge and compost. In order to reduce the high sewage sludge content of nonylphenol (+ethoxylates), a phasing out of these components in domestic and industrial products has been intensified, and shall be completed within the year 2000." (Paulsrud et al. 1998).

It seems that the Norwegian authorities have simply decided to initiate a phasing out of the major contaminant (NP) at the sources in favour of introducing limit values for organic contaminants in sewage sludge.

Denmark

In Denmark, the rules for sewage sludge application to agricultural soil were based on investigations, which were carried out in 1995 (Tørsløv et al. 1997). Since then, several aspects regarding the fate and effects of organic contaminants in soil as well as effects of different sludge treatment methods have been investigated in a series of individual projects. However, the results of these projects have not (yet) been applied for revision of the rules.

The 1995 project (Tørsløv et al. 1997) aimed at identification of (groups of) substances, which were of concern in Danish sewage sludge as well as development of a basis for regulatory rules for substances of concern. The investigations included chemical characterisation of a total of 20 sludge samples from 19 individual WWTP, one compost sample and slurry from one conventional and one "organic" farm. Furthermore, aqueous extracts of all samples were analysed. The chemical analyses included approx. 110 organic xenobiotics. The possibilities of uptake in plants were assessed by a literature review, including results from experiments with sewage sludge amended or contaminated soil. Based on this, the (groups of) substances, which may be detected with the usual analytical techniques, in (parts of) plants, were identified, and the concentrations in sludge, which may lead to detectable levels in plant tissue, calculated. A screening of the potential for leaching to ground water was based on the concentrations of substances measured in aqueous extracts of the sludge samples.

Based on the results achieved from the above activities and information from other countries, a list was produced showing substances/groups of substances of concern, including PAHs, phthalates (DEHP), LAS and NP (+EO).

Measures for regulation of the levels of these contaminants in sewage sludge were suggested as follows:

- Establishment of limit values for substances found in the waste products
- Limitations in the use of waste products with a critical content of hazardous substances
- Identification and reduction of sources of hazardous substances that, in particular, contribute to the waste products
- Establishment of quality criteria for the maximum permissible content of hazardous substances in agricultural soil in order to ensure the long-term quality of farmlands.

The investigations also included toxicity testing of waste products partly to characterise selected waste products regarding their potential impact on the biological function of soil, partly to provide a basis for the establishment of criteria for the quality of waste products. The laboratory testing comprised organisms relevant to the biological function of agricultural soil (nitrifying soil bacteria, soil dwelling invertebrates (springtail) and germination of lettuce seeds). Furthermore, the applicability of the Microtox test as a screening tool was investigated. The results of the testing were not conclusive in terms of use of ecotoxicological tests for regulatory purposes.

The approach taken by the Danish authorities was to set limit values for PAHs, DEHP, LAS and NP (+EO). In order to encourage substitution of the three latter groups of substances, the subsequent Danish statutory order included limit values for the four groups of substances and declaration of progressive reduction of the limits in years to come.

Germany

In Germany, limit values for AOX, PCBs and PCDD/Fs in sewage sludge are in force. According to Sauerbeck & Leschber (1992), these are not based on assessment of exposure or risks but are simply precautionary.

In stead of a detailed risk assessment of individual substances, the German approach to risk reduction with regard to organic contaminants in sewage sludge has been a priority setting aiming at identification of regulatory and technical measures for reduction of the input of prioritised substances to the sewerage system.

The latest report available (Arbeitsgruppe der Umweltkonferenz 2000) describes the procedure of the priority setting, the prioritised substances and their sources as well as short-term and long-term regulatory and technical measures for reduction of the substances with the highest priority. The appendices (approx. 200 pages) include a detailed description of the prioritisation process and datasheets on 44 (groups of) substances as well as source analyses for each of the prioritised substances. Finally, “sheets of measures” (Massnahmenblatt) for 9 (groups of) substances are included.

The prioritisation was initiated by selection of 44 substances for review. The selection process is not described in detail. It included information from the German Bundesländern regarding levels of contaminants in sludge and a review of investigations and regulatory measures in other countries (the Netherlands, U.S.A., Austria, France, Sweden, Norway and Denmark). Furthermore, 3 EU research projects and 42 German research and monitoring projects regarding organic contaminants in wastewater and sewage sludge were listed. Thus, it seems that the 44 substances were selected as being substances, which are considered as being of concern by some scientific and/or regulatory authority.

The information regarding each (group of) substance(s) included human effects (acute oral, mutagenic, teratogenic and cancerogenic) and limit values for “maximum working place concentrations” (“MAK-Werte”). The ecotoxicological information on the substances included toxicity to soil dwelling organisms (and if not available to aquatic organisms) and bioaccumulative properties. Assessment of the potential for transport between soil and (ground) water as well as uptake in plants was based on experimental evidence and water solubility, log Kow, persistence and possible information regarding the formation of metabolites and/or bound residues in soil. Furthermore, concentrations of the substances measured in sewage sludge and the reliability of this information were considered.

The prioritisation was based on this information and expressed in the placing of each substance in one of four groups of harmful substances. The term “harmful substance” is used here for the German word “Schadstof”. Based on their environmental properties and concentrations in sewage sludge the substances are:

Group 1: Harmful substances, which are considered most relevant. PCDD/Fs, AOX, PCBs, benzo(a)pyrene, NP, LAS, TBT and DEHP.

Group 2: Harmful substances, which are considered relevant. Toluene, 1,4-dichlorobenzene, 1,2,4-trichlorobenzene, hexachlorobenzene, 1,1,1-trichloroethane, tetrachloroethene, DDT and metabolites, lindane, 2,4-dichlorophenol, pentachlorophenol, Ugilec, bromophosethyl, silicone oils and phenol.

Group 3: Harmful substances, about which the information available is considered insufficient with respect to their environmental properties and/or concentrations in sewage sludge.
Clofibrin acid (a pharmaceutical) chlorinated paraffins, ethylene diamine tetraacetate, musk-xylene, tri-(chloroethyl)-phosphate, DecaBDPE, PentaBDPE, OctaBDPE, 2,4,6-trichlorophenol, 2,4-dimethylphenol, ethinyl estradiole, polyacrylic acid sodium salt (anionic), polyacrylamide (kationic) and DBP.

Group 4 Mixtures. For this group, effects data were not available due to the lack of definition.
Coplanar polychlorinated biphenyls, EOX, fluorotensides, mineral oil hydrocarbons, PAHs, volatile organic halogenated compounds, “textile auxiliary substances” and glycopeptides.

In addition to these substances, the use of additives for dewatering of sludge is pinpointed as a potential problem. However, in the report (Arbeitsgruppe der Umweltkonferenz 2000), the available information regarding the environmental properties of these substances is considered insufficient.

The German report also mentions that toxicity testing of sludge samples should be considered for the assessment of possible effects on soil living organisms. However, no recommendations are made.

The measures recommended include:

- Monitoring of the priority substances in sewage sludge
- Identification of sources
- Reduction of use or emission of individual substances (including dewatering additives)
- Research to establish relevant information regarding the group 4 substances.

United Kingdom

The only report available from the U.K. is "Organic contaminants in sewage sludges: A survey of UK samples and a consideration of their significance" (Jones & Northcott 2000). This report was prepared for the Department of the Environment, Transport and the Regions (DETR) as a background document for the discussions of the EU "Working Document on Sludge" (Environment DG 2000).

The substances considered in the document include those for which standards have been proposed in the EU (2000) (PAHs, PCBs, PCDD/Fs, phthalates, NP and LAS) as well as organochlorine pesticides, chlorinated paraffins, polychlorinated naphthalenes, polybrominated flame retardants and nitro-musks.

The focus of the study is reported concentrations of these compounds in sewage sludge from WWTP in the U.K. and other European countries as well as the limit values proposed in the EU Working Document (Environment DG 2000). It is stated that the scientific justification for the proposed limit values is insufficient or lacking and that there is a lack of validated analytical procedures.

Part of the discussion in the report is based on the comparisons of reported concentrations of the organic substances in sewage sludge in a series of European countries with the limit values proposed in the EU Working Document. This leads to the conclusion that: "Virtually all sewage sludges in Europe are likely to exceed the proposed limits for PAHs and LAS, for example, even those originating from rural/domestic WWTPs." (Jones & Northcott 2000).

Based on this, it is suggested not to accept the limits proposed but to review each (group of) substance(s) – and in some cases initiate new research – in order to arrive at limit values, which are scientifically motivated.

Therefore, the relevance to the present project of this report lies in the input delivered for identification of possible “new” substances of concern. Based on the properties of the chemicals and concentrations measured in U.K. sludge samples, chlorinated paraffins and nitro musks are identified as being groups of substances, which are potentially of concern.

The UK Water Industry Research (UKWIR) seems to be an important actor in the field of sewage sludge research. However, requests to the UKWIR regarding specific research reports (UKWIR 1996, 2001) have not been answered.

U.S.A.

US-EPA 1996

In the U.S.A., the assessment of sewage sludge for agricultural use is based on risk assessments of individual substances and has been carried out in two rounds. The entire process is described in detail in the “Technical Support Document for the Round Two Sewage Sludge Pollutants” (US-EPA 1996). Only metals (and “total hydrocarbons”) were included in the “Round One Sewage Sludge Regulation”.

Organic contaminants were included for consideration in round two as follows: A national sewage sludge survey involving analyses for 411 individual substances (some of which were inorganic) was carried out in 1988. PCBs (7 Aroclor mixtures) and PCDD/Fs (25 substances) were aggregated into two groups (PCBs and dioxins), thereby reducing the number of “substances” with 30.

The frequency of detection for each substance in sludge samples was considered and if the frequency of detection was less than ten percent, the substance was deleted from the list. 254 substances were not detected (incl. several polybrominated substances) and another 69 substances had a frequency of detection of less than ten percent. These substances are listed in the document (US-EPA 1996) and among those with a frequency of less than ten percent, the following are considered by other countries: Several PAHs, including benzo(a)pyrene and four phthalates.

Of the remaining (55 listed) pollutants, those for which no human health or ecological data were found were deleted from the list.

Based on this procedure, a list of 31 substances was taken as the starting point for a “Comprehensive Hazard Identification study”. Among these, 16 were inorganic, leaving 15 organic (groups of) substances as pollutant candidates for round two. These included DEHP, p-cresol, cyanides, coplanar PCBs and PCDD/Fs.

The risk assessment was based on calculations of human exposure to contaminants in sewage sludge via 15 different pathways. Differences between pathways were whether human exposure took place via e.g. direct ingestion of soil, ingestion of plants or livestock (having ingested soil and/or plants) as well as whether the sludge was applied to the soil surface or mixed with the soil and the percentage of human food intake, which originated from sludge amended soil. Exposure via evaporation and leaching to groundwater were also included. Among these routes of exposure, two pathways were found to comprise the greatest risk from persistent, lipophilic organic compounds. One was adherence of sludge to forage/pasture crops from surface application of fluid sludge, followed by grazing and ingestion of sludge by livestock. The other was direct ingestion of sludge by children (Chaney et al. 1997).

For each pathway, the exposure - and health risks - of "Highly Exposed Individuals" were calculated, using a combination of high-end and average assumptions designed to give a plausible estimate of the individual risk at the upper end of the risk distribution (e.g., above the 90^{th} percentile of the actual distribution). The threshold for carcinogens was an individual risk of $1x10^{-4}$ or higher; for non-carcinogens, a ratio of exposure to the Risk Reference Dose of one or greater; and for ecological risk, a risk quotient of one or greater (US-EPA 1996).

The resulting list of round two sewage sludge pollutants included PCDD/Fs and coplanar PCBs.

In a review and discussion document, Chaney et al. (1997) summarise the pathway analysis of risks from PCBs. They emphasise that there is a need for revision of the exposure assessment in order to include a more realistic estimate of the bioavailability of substances in sludge than what was used for the Round Two background document (US-EPA 1996). They show calculations of risks for PCBs, which conclude that the risk will be far below 10^{-4}.

US-EPA 2001

An exposure analysis for dioxins, dibenzofurans and coplanar PCBs has been made available via the Swedish EPA as a final draft (US-EPA 2001).

From the draft, it appears that a new, national sewage sludge survey, comprising samples form about 100 WWTP, has been conducted in 2001.

The report describes the risk assessment used for determination of the concentrations of PCCD/Fs and coplanar PCBs that can be present in sludge and remain "protective" (below a specific level of risk) of human health.

However, in the available document, there is no summary and the last two sections, which should describe the human health risk characterisation and the analysis of variability and uncertainty, are not included. It seems that the document is still in preparation. Therefore, it is not possible to verify whether the conclusions presented by Chaney et al. (1997) are actually reached in the official U.S. risk assessment.

The latest information available indicates that the National Whistleblower Centre (2002) has initiated a debate regarding the quality of the risk assessments elaborated by the US-EPA. The main objections consider the assessment of the risk caused by pathogens but it is indicated that the calculations of uptake in plants of contaminants from sludge underestimate the availability in soil. Within the limitations of this project, it has not been possible to evaluate this debate in detail.

Overview and discussion of conclusions from individual reports

The objectives and assumptions as well as the conclusions differ in the above assessments of sewage sludge.

For all assessments, except the Swedish, the starting point was the occurrence and concentrations of substances in sewage sludge. Therefore, the possibilities of identifying substances of concern were a priori limited to substances, which were included in the analytical programme. The Swedish assessments, which comprised information regarding sources of potentially problematic chemicals (which may not have been included in the analyses), were still limited by the knowledge regarding environmental and toxicological properties of chemicals at the time.

As the assessments (and subsequent monitoring and/or decision making) were based on chemical analyses of sludge samples from municipal WWTP, the reliability and representativeness of the analyses and samples used are very important. The reliability of the chemical analyses in terms of repeatability (in individual laboratories) and reproducibility (in different laboratories) is discussed in most reports. The discussions are typically based on results of a few repeated analyses and the degree of reliability is variable. The representativeness of the samples taken is a general problem, especially because the levels of contaminants vary with orders of magnitude at different sampling times during the year in many WWTP. E.g. based on five monthly composite samples from each of seven WWTP, Paulsrud et al. (1998) concluded that variations were substantial and that the variations within each plant could be even greater than the variations between different plants.

Table 4.1 gives an overview of the substances, which are considered of concern in individual "countries", the main concerns about them and the parameters, which contributed most to the concern in the assessments. Furthermore, the year of the assessment and/or the main reference cited is included in the table.

Table 4.1 Substances of concern in sewage sludge

Substance	Reasons	Key parameters	Year of assess-ment, reference	Country
LAS	Plant uptake Leaching	Conc. in sludge	1995, Tørsløv et al. 1997	DK
	Properties	and conc. in sludge	AdU 2000	D*
DEHP	Plant uptake Leaching	Conc. in sludge	1995, Tørsløv et al. 1997	DK
	Properties	and conc. in sludge	AdU 2000	D*
NP(+EO)	Plant uptake Leaching	Conc. in sludge	1995, Tørsløv et al. 1997	DK
	Properties	and conc. in sludge	AdU 2000	D*
	-	Conc. in sludge	Paulsrud et al. 1998	N
PAHs	Plant uptake Leaching	Conc. in sludge	1995, Tørsløv et al. 1997	DK
PCBs	Livestock ingestion	P, B, lipophilic	Andersen 2001	EU
	No build up in soil	P, B, lipophilic	Naturvårdsverket 1993	S
	Properties	and conc. in sludge	AdU 2000	D*
	Livestock/ child ingestion	P, B, lipophilic	US-EPA 1996	U.S.
PCDD/Fs	Livestock ingestion	P, B, lipophilic	Andersen 2001	EU
	No build up in soil	P, B, lipophilic	Naturvårdsverket 1993	S
	Properties	and conc. in sludge	AdU 2000	D*
	Livestock/ child ingestion	P, B, lipophilic	US-EPA 1996	U.S.
Benzo(a)pyrene	Plant uptake	Properties and conc. in sludge	AdU 2000	D*
TBT	Properties	and conc. in sludge	AdU 2000	D*
AOX	Properties	and conc. in sludge	AdU 2000	D*

P = persistent, B = bioaccumulative, AdU = Arbeitsgruppe der Umweltkonferenz.
* From the German priority list, only substances from group 1 are included.

In Table 4.2, the substances, which were discussed by Erhardt & Prüess (2001) but for which no conclusions regarding their importance were drawn, are summarised together with the substances listed by Thornton (2001) as having a potential for appearance in sludge. Furthermore, the conclusions drawn by Jones & Northcott (2000) are included.

Table 4.2 Substances, identified as being potentially of concern

Substance	Comments in references	Ref.
Brominated flame retardants	1: P, B, ED, other tox and ecotox effects, 3: P, B [German group 3: Harmful, lack of knowledge]	1: PBB/PBDE 2: PBDEs 3: PBDEs
Chloroben-zenes	Uptake in plants, P [Several in German group 2: "Relevant"]	1
Nitro-musks / polycyclic musks	1: PEC/PNEC, lack of fate knowledge, 2: not P, exposure due to body care and washing >> sludge, 3: High concentrations, not readily biodegradable	1, 2, 3
Chlorinated paraffins	1: P, B, toxic, lack of knowledge, 3: Very high concentrations, P, B	1, 2, 3
Organotins	P, B, toxic, lack of fate knowledge	1
VOCs	Build up in soil	1
Quintozene	Low water solubility, half-life in soil 5-10 months	2
Polychlori-nated naphthalenes	2: Some congeners have dioxin like activity, 3: Similar to PCBs, stable	2, 3
Polydi-methylsilox-anes	Relatively P, not B, no significant environmental toxicity	2
Pharmaceu-ticals	Biological activity, different fate [Clofibrin acid in German group 3: Harmful, lack of knowledge]	2
Synthetic fats	P, not toxic to humans [Danish media: Causes heart failure]	2
Polyelectro-lytes	Degrades relatively slowly in soil, 10%/year. Potential for accumulation in soil	2
Endocrine disrupters	ED, lack of fate knowledge	1
Oestrogenic compounds	Water soluble, major problem in wastewater, not in sludge	2

P = persistent, B = bioaccumulative, ED = endocrine disrupting
1: Erhardt & Prüess 2001, 2: Thornton 2001, 3: Jones & Northcott 2000.

Tables 4.1 and 4.2 reflect the focus and priorities of the assessments, which resulted in identification of concern for individual substances as well as the analytical programmes employed, which again reflect the level of knowledge at the time of the assessments.

In section 8, available knowledge regarding the substances identified as candidates for concern is summarised.

Differences

In all assessments, environmental exposure and effects were included but – except for the German ranking – the reasons for identification of substances as being of concern were the possibilities of build up in soil or human exposure and/or effects .

Differences between the priorities of the countries are obvious. The number of substances is highest in the German priority list and it includes all substances, which are mentioned in the other lists. This reflects the fact that it was not elaborated for the purpose of sludge regulation but for regulation of emission of substances to WWTP. Furthermore, it is evident that the substances included in the Danish list differ from those in the lists from the other countries.

Danish assessments

The Danish investigations in 1995 (Tørsløv et al. 1997) were focused on human exposure assessment in combination with manageable regulatory limits. At the time of the assessments, the lack of knowledge regarding the fate in soil of several substances called for conservative estimates of exposure via groundwater and plants. The relatively rapid degradation of LAS and DEHP in soil under aerobic conditions was taken into consideration. However, the high levels of these substances in sludge raised doubt whether the rate of degradation would be sufficient to exclude the possibilities of leaching and/or plant uptake during the growing season. Because the bioavailability in sludge and soil was included in the assessments, lipophilic substances, which were measured in low concentrations in sludge (like PCDD/Fs and PCBs), were not identified as being of concern. The parameter that turned out to be the most decisive for the identification of concern, was the concentration in sludge.

Since then, a multitude of investigations has been carried out in Denmark. These include identification of sources of contaminants (Kjølholt 1997, Jensen 1997) as well as degradation of organic contaminants in sludge under different storage and/or composting conditions (Dencker-Jensen et al. 1999, Hedeselskabet 2000, Mai & Jungersen 1999).

Furthermore, a 4-year research programme, focusing on sustainable use of waste products (especially sludge) in agriculture, is being finalised at present. It has included laboratory and field investigations of fertilising properties as well as fate and effects of selected contaminants (LAS, NP(+EO), DEHP, pyrene (PAH), LAE and antibiotics). The activities are in the process of being published, mainly in separate papers, describing investigations of single parameters.

Results from several investigations were summarised in a newsletter from the Danish Environmental Research Programme (DERP 2001).

The results of the individual sub-projects are combined for an overall risk assessment of the model substances, when applied to agricultural soil (Petersen & Rasmussen 2001, Rasmussen et al. 2002). The risk assessment is based on mathematical modelling of the fate of each contaminant, including existing models (MACRO amended with DOC) and using research results as input for parameters like half-life (aerobic and anaerobic) in soil and sorption. Maximum and "typical" values are used for concentrations in sludge and sludge application rates. The result is estimates of concentrations (PEC) in the soil as a function of depth and time, concentrations in percolating water at any predefined depth as well as degree of accumulation in the soil and degree of leaching. PNEC values for environmental effects assessment are based on results of laboratory tests with soil living organisms (from the project and literature) and for human risk assessment, the limits for drinking water are used as “PNEC-values”. Uptake in plants is not yet included in the model. Repeated applications of sewage over a period of 30 years are modelled. The conclusions of the resulting risk assessment are:

- Probably no risk to soil organisms if the concentrations in sludge are below the Danish cut-off values
- The predicted degree of leaching is low for all compounds
- The predicted concentration in percolate is below the limit value for drinking water
- The accumulation in soil is negligible for NP, LAS and LAE
- DEHP and pyrene may accumulate in soil.

These conclusions imply, that the reason for concern with respect to these substances, would be the possibility of accumulation in soil – in spite of the degradability of DEHP and NP in soil.

Results of the field experiments, which were conducted as part of the research programme, are not yet fully published. However, a preliminary evaluation of the results of the chemical analyses indicates that accumulation of the substances in soil cannot be measured.

Laboratory experiments and field trials with plants (barley, carrot, rapeseed and oats) have shown that the substances LAS, NPEO, pyrene, nonylphenol and DEHP can be adsorbed to/taken up by plant roots from aqueous solutions in the laboratory. However, the uptake from soil is negligible, and under field conditions, possible uptake is not measurable (Mortensen et al. 2001).

Sweden, Germany and U.S.A.

The other assessments focused on exposure and risk assessment and the substances included in the investigations were as numerous as in the Danish. However, in these assessments, the degradability under aerobic conditions and availability for leaching and plant uptake of substances in soil were key parameters and the importance of other sources of the contaminants was considered.

Therefore, substances, which were known to be readily biodegradable (under aerobic conditions in water) and/or undergo rapid degradation in soil, were sorted out during the assessments. Consequently, LAS and DEHP were not included in these lists of substances of concern. Information regarding the degradation of NP in soil was not conclusive but in the Swedish assessment, the lack of demonstration of plant uptake in field experiments was given priority. NP is on the list of substances, for which Swedish guidance values have been set, mainly because of the aquatic toxicity. It is anticipated that the monitoring of NP in sludge can be used as a measure of the development of discharges of NP to WWTP.

The consideration of other routes of exposure (to agricultural soil or man) has lead to the conclusion that possible human exposure to PAHs via sewage sludge is very much lower than via atmospheric deposition (to soil) or human intake of processed food with high levels of PAHs (e.g. barbecued or smoked). However, PAHs are on the list of substances, for which guidance values have been set in Sweden, in order to give an indication of the degree of discharges of contaminated water from run-off.

In conclusion, the group of substances, which is considered as being of concern in most assessments, is the persistent, lipophilic organic compounds (PCDD/Fs and PCBs). The motivation differs but the decisive parameters are the properties of the substances.

The above discussion shows that the basic differences in the focus and principles used for prioritisation in different countries were more decisive for the differences in the resulting lists than the actually measured concentrations of the substances in sludge samples. Therefore, comparisons of the concentrations in sludge from different countries at the time of the assessments will not be made. (Available information regarding concentrations of substances of potential concern in sewage sludge is included in section 8.)

Groups of substances in sludge

The parameters, which were decisive for the outcome of the individual assessments, can be used as a basis for a grouping of the substances for further discussion. The grouping is summarised in Table 4.3, based on qualitative indications of the parameters.

Table 4.3 Grouping of contaminants

Parameter	**Quality**	**Concentration in sludge**	
		High	Low
Degradation in aerobic soil	Rapid-fair	LAS, DEHP, NP, low weight PAHs	None
	Slow-none	None	PCBs, PCDD/Fs, benzo(a)pyrene a.o. high weight PAHs
Sorption (lipophilicity)	Low-medium	LAS, NP, low weight PAHs	None
	High	DEHP	PCBs, PCDD/Fs, benzo(a)pyrene a.o. high weight PAHs
Bioaccumulation	Low	LAS, DEHP, NP, low weight PAHs	None
	High	None	PCBs, PCDD/Fs, benzo(a)pyrene a.o. high weight PAHs

The substances identified as being of concern by one or more assessments belong to one of two groups:

1. Biodegradable, not bioaccumulative substances, which are found in high concentrations in sludge
2. Persistent, bioaccumulative substances, which are generally found in low concentrations in sludge.

The exception from this, is the PAHs for which their properties vary with the size of the molecule. The identification of PAHs as being of concern in the Danish assessment was mainly based on low weight PAHs, which would fit into group one above. In other countries, the lack of biodegradability of the high weight PAHs led to the inclusion of PAHs on the lists of substances of concern.

The Swedish identification of PCBs and PCDD/Fs as being of concern was based on the potential for build up of these substances in soil after repeated applications of sewage sludge. This concern would not be dependent on the application rates of these substances to soil.

Fate of organic contaminants in sewage sludge amended soil

For the substances, which are considered of concern due to high concentrations in sludge, and which may be degraded in soil, parameters affecting the concentration in soil may be important for an assessment of whether these substances should be considered of concern in Sweden.

In this section, the parameters, which are decisive for the concentrations of organic contaminants in soil, resulting from the application of sewage sludge, are summarised. They comprise application rate and "dilution" of the sludge in soil during application followed by metabolisation and other routes of dissipation, e.g. volatilisation and leaching.

Application rate and dilution

The degree of dilution is determined by the rate of application of sludge (tonnes dry weight/ha) and the possible incorporation into the soil through injection, harrowing or ploughing.

Calculations of dilution are based on the assumption that the sewage sludge is evenly distributed in soil after application and ploughing/harrowing. However, in practise, the sludge will typically be present as lumps in the soil and therefore, the concentrations of individual substances will be high in the lumps and low (or zero) in between them. After some time, bioturbation due to soil living animals and plant roots in combination with microbial degradation will result in a more even distribution. Investigations of the importance of the heterogeneous distribution of sludge in agricultural soil were included in the Danish Environmental Research Project (DERP 2001). Transient effects on soil living organisms were recorded in field experiments in areas of high sludge concentration (Krogh 2001) but uptake in plants could not be observed in experiments with heterogeneous distribution of sludge (Mortensen et al. 2001).

The maximum rate of application (of sludge complying with contaminant limits) is typically defined in national regulation. The limits may be based on sludge dry matter or nutrient content of the sludge. Furthermore, application rate limits may be related to the frequency of application, so that at yearly applications, the maximum will typically be a proportional fraction of the maximum allowed at longer intervals between applications. In some countries, different rates are fixed for different crops.

In Denmark, the maximum amount-based application rate for sludge is 2 tonnes/ha/year, which may be applied annually or as 6 tonnes/ha once every 3 years. This may be overruled by limits set for addition of phosphorous to soil, which may also be applied as yearly doses or as a total once every three years.

In the Swedish legislation, limits are based entirely on additions of both phosphorous and nitrogen. As in Denmark, the dosage of phosphorous for several years (up to seven) may be added at one application, while the addition of nitrogen (maximum of 150 kg/year) can be applied only as yearly doses.

In practice, the maximum amount of sewage sludge added would vary with the contents of nutrients in the sludge, and based on this, no fixed dosage can be used for calculations. Therefore, for risk assessments (and the planning of field studies), it is necessary to define a "maximum, realistic" application rate.

In Denmark, the maximum, realistic rate used for calculations is 2 tonnes/ha/year or 6 tonnes/ha once every three years (based on sludge dry weight). The calculated maximum concentration in soil, immediately after application is therefore based on a rate of 6 tonnes/ha/year.

In Sweden, the corresponding rates are a maximum of 1.25 ton/ha/year or 5 tonnes/ha once every four years (Naturvårdsverket, pers. comm.) Therefore, expected, maximum concentrations in soil in Sweden are 5/6 (or 0.83) of those in Denmark.

The other parameter, decisive for dilution, is the depth of incorporation in the soil.

In Denmark, the soil is typically harrowed (10 cm depth) after sludge application but ploughing (20-30 cm depth) is also applied. A conservative, realistic estimate of the dilution is mixing with the upper 10 cm of the soil. Using an application rate of 6 tonnes/ha and a density of soil of 1.5 kg/L, the resulting dilution factor becomes 250.

Swedish agricultural practices also include harrowing. Assuming the same depth (10 cm) as used for the Danish assessments and an application rate of 5 tonnes/ha, the dilution factor for calculations of concentrations in soil under Swedish conditions becomes 300.

Dissipation

Following application to soil, organic substances may dissipate via volatilisation, leaching and/or degradation.

For NP, DEHP and LAS, degradation is considered the most important dissipation process. Degradation rates reported in literature are often based on laboratory tests, which are conducted under optimum conditions (e.g. with respect to oxygen) and at relatively high temperatures (typically 20°C). Therefore, results should be interpreted with care – especially for sludge amended soil, where anaerobic microhabitats are frequent and in countries, where the soil temperature is considerably lower during most of the year.

In the Danish assessments (Tørsløv et al. 1997), the reported relatively high degradation rates for the substances were considered but were not included in any quantitative way. However, the reduction of biodegradability under anaerobic conditions was considered seriously, because of the occurrence of anaerobic microhabitats especially in sludge amended soil.

In the recent Danish research project (DERP 2001), the degradation of LAS, NP and DEHP was studied in detail in laboratory experiments, which were simulating conditions in sludge amended soil (Henriksen et al. 2001). They demonstrated, that biodegradation in soil and sludge amended soil of DEHP and NP decreased with time. As described above, the modelling results indicate, that NP and DEHP may accumulate in soil at repeated applications, while increasing concentrations in soil do not seem to be measurable in field experiments (Petersen & Rasmussen 2001). At present, the possibility of build up of DEHP and pyrene in soil cannot be excluded.

Relevance to Swedish conditions

The relevance of revising the Swedish conclusions regarding LAS, DEHP and NP (i.e. that they are not of potential concern in sewage sludge in Sweden) can be summarised as follows:

LAS concentrations in Swedish sewage sludge are very low due to consumer's avoidance of LAS containing products (detergents). Thereby, the major reason of concern is omitted, and LAS is not expected to be a substance of concern.

NP concentrations in Swedish sewage sludge are declining due to phasing out of NP from the Swedish market. The results in Table 3.1 show that the level in 2001 (mean: 3.9 mg/kg dw) is well below the Danish limit value, which was set in 1996 (50 mg/kg). The lower application rates of sludge in Sweden imply that the "margin of safety" is relatively high.

DEHP concentrations were measured in two samples of Swedish sludge from 1997 at levels of 70-160 mg/kg dw. Samples from preceding years from the same WWTPs do not give any indication of trends. However, the results from the samples from 19 WWTPs collected in 2001 (mean of sum of phthalates: 7.5 mg/kg dw) indicate a decrease. Compared to the original Danish limit value of 100 mg/kg, the reasons for concern in Denmark do not seem to be relevant in Sweden.

Input from sewage sludge, other fertilisers and air

For decisions regarding Swedish regulatory measures with respect to individual substances, the input to agricultural soil related to input from other routes of exposure is requested. This section comprises an overview of available information regarding the contents of organic contaminants in different fertilisers and includes estimates of contributions to concentrations in agricultural soil from dry or wet deposition. The estimates are expressed on a yearly basis.

For sewage sludge, the estimated “maximum application rate” (e.g. kg/ha/year) is multiplied with the concentrations of each substance (e.g. mg/kg) in sludge in order to achieve a measure of the “load” (e.g. mg/ha/year). 2001-2002 values were made available from Naturvårdsverket (2002 and Nylund et al. 2002).

For Swedish manure, only results of analyses in 1989 were available. None of the substances analysed for were detected (Naturvårdsverket 1992).

The only information available regarding concentrations of organic contaminants in Swedish compost was on PCBs. In recent analyses of compost samples, Nilsson (2002) measured concentrations of PCBs in the range 0.022-0.048 mg/kg dw.

For atmospheric deposition, information on PAHs and PCBs was available IVL (2002).

For inorganic fertilisers no analyses seem to have been conducted. According to KEMIRA in Denmark (Jens Jacob Larsen, pers.comm.), the production of synthetic fertilisers is based on inorganic raw materials and no organic substances are added during the production. The product is dried with air, which is heated by natural gas, and it seems likely that possible contamination with combustion products is negligible. Therefore, the possible contribution of organic contaminants from synthetic fertilisers is considered to be negligible. The available information is summarised in Table 6.1.

Table 6.1 Concentrations of organic substances in Swedish sewage sludge, maximum application rate and loads of organic contaminants to soil in Sweden - unit: mg/m²/year

Substance	Conc. in sludge[1]	Sludge application rate	Sludge load	Deposition[2]
	mg/kg dw	kg/ha/y	mg/m²/y	mg/m²/y
NP	3.9	1250	0.49	-
PAHs	1.1		0.14	0.056-0.083
PCBs	0.1		0.01*	0.00029-0.00057
PCDD/F	0.0013		0.00016	-
DEHP	6.6		0.83	-

*: 1998 value. 1: Naturvårdsverket 2002, 2: IVL 2002, range representing city area and remote area, respectively.

Table 6.1 shows that the average yearly input of PAHs from atmospheric deposition is in the same order of magnitude as the input from application of sewage sludge at the maximum rate. For PCBs, the load from sludge (based on 1998-value) is up to two orders of magnitude the load from atmospheric deposition (2001). However, as PCBs are continuously being phased out, the atmospheric deposition as well as the expected concentration in sludge are decreasing. Therefore, the load from sludge is probably overestimated.

Assessment of substances of concern

For the two groups of substances of concern, the information gathered in the previous sections, can be summarised as follows:

1. Biodegradable, not very bioaccumulative substances, which are found in high concentrations in sludge in some countries (LAS, NP, DEHP)

- LAS: Low concern, due to low concentrations in Swedish sludge
- NP: Low concern. The phasing out of NP in Sweden seems to be effective and concentrations in Swedish sewage sludge (2001) are one order of magnitude lower than the Danish limit value
- DEHP: Low concern. Concentrations in Swedish sewage sludge (2001) are almost two orders of magnitude lower than the Danish limit value

2. Persistent, bioaccumulative substances, which are generally found in low concentrations in sludge.

- PAHs: Levels in Swedish sludge seem to be decreasing. The contribution from sludge (2001) is about twice the magnitude of atmospheric deposition. Based on concern regarding possible accumulation in soil, a limit could be considered, the present guidance value is 3 mg/kg.
- PCBs: Not measured in 2001. Based on concentrations in 1995 and 1998 (equal, at 0.1 mg/kg dw), the contribution from sludge is one order of magnitude above the contribution from atmospheric deposition. Based on concern regarding possible accumulation in soil, a limit could be considered. The present guidance value is 0.4 mg/kg dw.
- PCDD/Fs: No atmospheric deposition reference available for Swedish conditions. Based on concern regarding possible accumulation in soil, a limit could be considered. For comparison, the risk based American limit value (US-EPA 1999) is 0.0003 TEQ[1] mg/kg. The equivalent concentration reported from Swedish sewage sludge analysis (Naturvårdsverket 2002) is 0.000019 TEQ[1] mg/kg.

Information regarding contributions to agricultural soil from other sources than sewage sludge in Sweden is scarce and the above conclusions may well be modified if more information is gathered.

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[1] TEQ: TCDD-EQuivalents is a unit, which is expressing the toxic potential of the substances. There are several sets of toxicity factors (TEFs) for making these calculations and it has not been possible to identify, whether the same set of factors was used for the Swedish and American calculations. The numbers may therefore not be immediately comparable.

In summary, for the substances, which are presently considered as being of concern by one or more countries, the levels of concentrations in Swedish sewage sludge do not give rise to concern with respect to risks of toxic effects on man or the environment. However, for substances, which are persistent or not readily biodegradable (PAHs, PCBs and PCDD/Fs), there is reason for concern because accumulation in soil is possible.

Substances of potential concern

In section 4, substances for which the possibility of concern has been raised, were identified (Table 4.2). In this section a preliminary assessment of the potential for concern is made for some of these substances. A detailed assessment of whether each group of substances should be considered as being of concern is outside the scope of this project.

Except from the endocrine disrupting chemicals, the common denominator of these substances is persistence – or low degradability. For several, lack of knowledge regarding their fate in soil and/or toxicological and environmental effects, is indicated in Table 4.2.

Therefore, based on available information, each group of substances is briefly characterised with respect to its environmental and toxicological properties as well as sources and levels reported in sewage sludge.

For convenience, Table 4.2 is repeated here as Table 8.1.

*Table 8.1 (=4.2)		Substances, identified as being potentially of concern*

Substance	Comments in references	Ref.
Brominated flame retardants	1: P, B, ED, other tox and ecotox effects, 3: P, B [German group 3: Harmful, lack of knowledge]	1: PBB/PBDE 2: PBDEs 3: PBDEs
Chloroben-zenes	Uptake in plants, P [Several in German group 2: “Relevant”]	1
Nitro-musks / polycyclic musks	1: PEC/PNEC, lack of fate knowledge, 2: not P, exposure due to body care and washing >> sludge, 3: high concentrations, not readily biodegradable	1, 2, 3
Chlorinated paraffins	1: P, B, toxic, lack of knowledge, 3: Very high concentrations, P, B	1, 2, 3
Organotins	P, B, toxic, lack of fate knowledge	1
VOCs	Build up in soil	1
Quintozene	Low water solubility, half-life in soil 5-10 months	2
Polychlori-nated naphthalenes	2: Some congeners have dioxin like activity, 3: Similar to PCBs, stable	2, 3
Polydi-methylsilox-anes	Relatively P, not B, no significant environmental toxicity	2
Pharmaceu-ticals	Biological activity, different fate [Clofibrin acid in German group 3: Harmful, lack of knowledge]	2
Synthetic fats	P, not toxic to humans [Danish media: causes heart failure]	2
Polyelectro-lytes	Degrades relatively slowly in soil, 10%/year. Potential for accumulation in soil	2
Endocrine disrupters	ED, lack of fate knowledge	1
Oestrogenic compounds	Water soluble, major problem in wastewater, not in sludge	2

P = persistent, B = bioaccumulative, ED = endocrine disrupting
1: Erhardt & Prüess 2001, 2: Thornton 2001, 3: Jones & Northcott 2000.

Brominated flame retardants

This group of substances was pinpointed in all of the documents cited in Table 4.2 and is included in the German priority list under group 3 (harmfull substances, lack of knowledge). It comprises polybrominated biphenyl ethers (PBDPEs), tetrabromobisphenol A (TBBPA) and hexabromocyclo-dodecane (HBCD).

Focus has recently been set on the substances in this group and they are being investigated very thoroughly at present; e.g. Chemosphere vol. 46 2000, is a special issue devoted to brominated flame retardants. (In this issue de Witt (2000) gives a thorough overview of the current state of the knowledge and areas for further research are recommended.) Furthermore, these substances have been included in the EU-programme for existing substances and risk assessment documents have been elaborated (Andersson 2002).

- Chemistry: Very close to PCBs (bromide instead of chlorine). Physical-chemical properties and (lack of) degradability similar to PCBs. Volatile.

- Toxicology, ecotoxicology: May affect the thyroid gland, the liver, and cause neurological disorders. Toxic to aquatic organisms and may cause long-term adverse effects.

- Use and sources: Furniture (polystyrene, polyurethane foam), textiles, electronic equipment (wire and cable insulation and electrical connectors). Washing of textiles (→WWTP), volatilisation from products during use and after disposal → atmospheric deposition (not quantifiable (Andersson 2002)).

- Levels in wastewater: Palmquist (2001) made analyses of different fractions of household wastewater (blackwater, i.e. toilet water and greywater, i.e. other household water). Specific chemical analyses of 91 organic substances included a.o. 13 brominated flame retardants, and DEHP. Discharges per person per day and per year were calculated for both types of wastewater. Tetra- and hexaBPDEs, were discharged with greywater at rates of 3.0 and 0.09 mg/person/year, respectively. For comparison, the most frequent substance in greywater, DEHP, amounted to almost 1400 mg/person/year.

- Levels in sludge: Available data regarding measurements of PBDPE are summarised in Figure 5.1 (Andersson 2002). Several data are from Swedish sewage sludge samples. The concentrations vary depending on the level of bromination. DecaBDPE seems to be present in the highest concentrations (with maximum concentrations of 350 (Sweden) and 9160 (U.S.A.) µg/kg sludge dry weight). Labels on the diagram indicate these values. In analyses of pentaBDPE, different congeners were identified, so the total level of pentaBDPE congeners is exceeding the impression on the figure.

- Other sources to contamination of agricultural soil: Atmospheric deposition is described as a very (the most?) important route of dispersal of these substances (Andersson 2002), but no quantitative information is available.

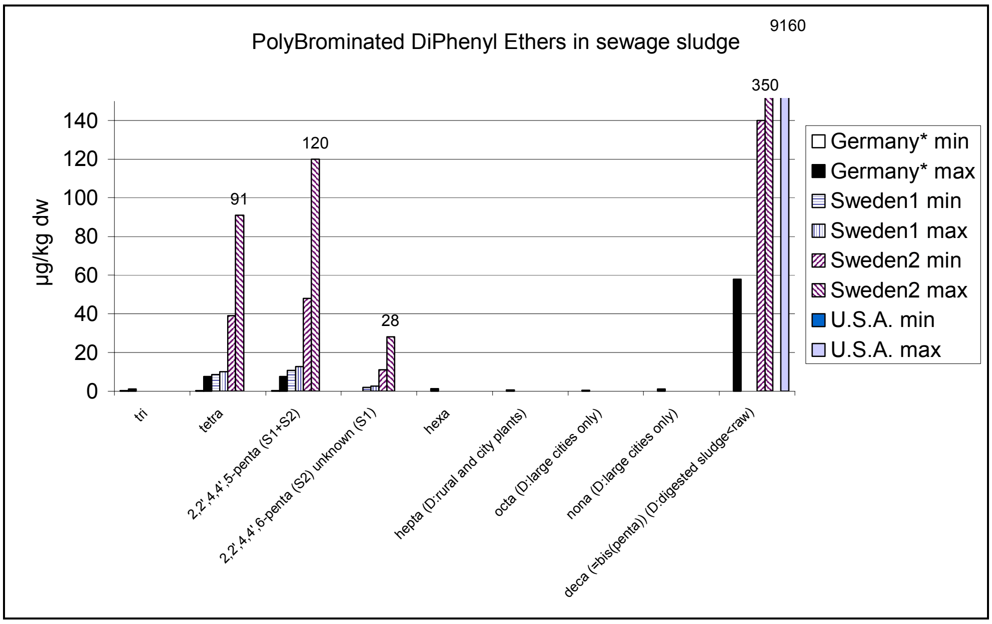

Figure 5.1 Concentrations of polybromonated diphenyl ethers in sludge, [µg/kg dw]. Source: EU risk assessments (Andersson 2002).

- Additional information: Matscheko (2001) investigated concentrations of PCBs, PCDD/Fs and polybrominated diphenylethers (PBDPEs) in agricultural soils, which had been amended with sewage sludge (2-18 years before sampling) and in control soils. Concentrations of PBDPEs showed elevated levels in sludge amended soils, and the ratio of concentrations in sludge-treated soil to untreated soil was significantly greater than one. In one field, which had received large amounts of sewage sludge with high levels of PBDPEs in the years 1978-82, the ratios were 2000-12000, indicating that the PBDPEs had not been removed from the sol during the latest 18 years. Accumulation in earthworms was investigated in field collected worms. Biota to soil accumulation factors (BSAFs, soil organic matter/worm lipids) varied between <1 and 34 for PBDPEs, which were considered almost as bioaccumulative as PCBs with an average BSAF for ortho-PCBs of five.

Subsequently, the local municipality (Marks kommun) analysed samples from the same field as Matscheko (Ek 2002) and from seven other fields in the area, which had received sewage sludge at the same time. Furthermore, samples from three fields, which had not received sewage sludge, were analysed. The results showed low levels of brominated substances in all fields except one, which was different from the one in which Matscheko had recorded high levels and there was no correlation between sewage sludge application and levels of brominated flame retardants (or PCDD/Fs and nonyl phenol) in the 11 sampled fields. It is suggested that single samples showing high levels of organic contaminants from sludge treated fields could be indicative of the place in the field, in which the sewage sludge was originally deposited (Ek 2002). The results of the two investigations are conflicting regarding the concentrations of flame retardants in the field, Matscheko (2001) sampled. The evidence from several fields collected by

Marks Kommun (Ek 2002) demonstrate that for the assessment of levels of contamination in individual fields, results of analysis of samples taken at random are not sufficient.

The investigations carried out by Marks Kommun included samples of cow milk and cereal grains from the farm, in which Matscheko had measured high levels of PBDPEs in one field. These samples did not show elevated levels of flame retardants (Ek 2002).

Several other documents refer measurements of PBDPEs in human tissue and breast milk.

In summary, PBDPEs are frequently occurring in sludge samples – some substances primarily in urban wastewater. The database is however too limited to get a picture of the concentration levels. The substances are persistent and bioaccumulative and bioaccumulation has been demonstrated in earthworms and man. Based on this information, the brominated flame retardants should be considered as being of concern in sewage sludge in terms of the possibility of accumulation in agricultural soil. However, the toxicity of these substances are generally lower than that of e.g. PCBs and PCDD/Fs and a recent, Swedish risk assessment concludes that there is no risk of toxic effects of these substances as a result of application of sewage sludge (Hellström 2000). In order to assess the relative importance of sewage sludge for the exposure of agricultural soil, information regarding concentrations in other fertilisers (manure, compost) and atmospheric deposition is needed.

Nitro musks / polycyclic musks

Jones & Northcott (2000) make a distinction between nitro musks (substituted trinitro- and dinitrobenzenes) and polycyclic musks, stating that the latter are replacing the nitro musks on the market.

- Chemistry: Mono- or polycyclic organic compounds of which the monocyclic compounds are nitrosubstituted. Not readily biodegradable, but some degradation will take place in WWTP and some in soil (Thornton 2001). Degradation rate in soil half-life = 12 days (Thornton 2001), which indicates biodegradability in soil. However, Jones and Northcott (2000) describe the musks as “not particularly biodegradable”. Many are volatile (Jones & Northcott 2000), which would increase their potential of long range atmospheric transport.
- Toxicology, ecotoxicology: Very toxic to aquatic and terrestrial organisms (Tas et al. 1997).
- Use and sources: Perfume and body care products, washing agents, fabric softeners, air freshners etc. Release dominated by domestic discharges to sewer.
- Levels in wastewater: No information available.

- Levels in sludge: High, order of magnitude milligrams (mg/kg sludge dry weight): UK: Σ11: 2-86, The two dominant musk substances from the UK results have been measured in German and Dutch sewage sludge as well: D: 0.1-8.9, and 0.1-5.2, NL: 0-34 and 0-63 mg/kg, respectively. (Jones & Northcott 2000).
- Other sources to contamination of agricultural soil: No information.
- Additional information: None available. (EU risk assessment in Tas et al. 1997).

In summary, information regarding occurrence, effects and fate of these substances is insufficient for conclusions to be drawn with respect to risks to man and the environment. The fact that they are less persistent than other substances of concern indicates a lower priority, while the high concentrations measured in sludge points to prioritisation.

Chlorinated paraffins

This group of substances is identified as being of concern in all three documents (Erhardt & Preüss 2001, Thornton 2001, Jones & Northcott 2000).

- Chemistry: Chlorinated paraffins or polychlorinated n-alkanes, chlorinated straight chain hydrocarbons. Mixtures of chain length: short: C10-C13, medium: C14-C17, long: C18-C30. Persistent, bioaccumulative and toxic. Possibly volatile.
- Use and sources: Plasticisers in PVC and other plastics, extreme pressure lubricant additives, flame retardants, paint and sealant additives. Illicit disposal of oils to wastewater may be an important source. "Approximately 50% of the lubricating oils used by industry in Sweden may be released into the air or wastewater" (Thornton 2001).
- Levels in wastewater: Not available.
- Levels in sludge: Order of magnitude mg/kg. UK (1980): 4-10 mg/kg, 1995-data from industrial area: short chain: 1-65 mg/kg. UK (2000): Σshort chain: 7-200 and Σlong chain: 37-9700 mg/kg sludge dry weight (Jones & Northcott 2000).
- Other sources to contamination of agricultural soil: No information available.
- Additional information: None available.

In summary, these substances, which have properties similar to POPs (Jones & Northcott 2000) and are measured in levels as high as those of LAS and DEHP in sludge, are high ranging candidates for concern.

Polyelectrolytes

Polyelectrolytes based on polyacrylamide and cationic copolymers are identified as potential substances of concern by Thornton (2001) and listed in the German Group 3, indicating concern but lack of knowledge.

- Chemistry: Linear polymers. Degradation slow in soil, 10% per year. Sorbs strongly to soil. Acrylamide monomer residuals are rapidly biodegraded in soil.
- Toxicology, ecotoxicology: Acrylamide is potentially toxic to humans and is a reported carcinogen.
- Use and sources: Dewatering agent used for sewage sludge.
- Levels in wastewater: No information.
- Levels in sludge: 2500-5000 mg/kg sludge dry weight.
- Other sources to contamination of agricultural soil: None known.
- Additional information: In short-term tests with or without addition of sludge, no adverse effects of cationic polyacrylamides on soil respiration, biomass or nitrification were found (Johansson et al. 1998).
- Investigations of biodegradation of polyacrylamide are at present being carried out, including identification of degradation pathways (DHI, pers. comm.).

In summary, the low rate of biodegradation in soil and the high concentrations of polyelectrolytes in sewage sludge qualify this group of substances to being considered as of concern. However, more information is needed regarding their fate and possibilities of accumulation in soil for conclusions to be drawn.

Endocrine disrupting chemicals

An array of substances has been identified as having endocrine disrupting effects. Among these are phthalates, NP, PCDD/Fs and PCBs and still new substances are added to the list. The effects may be similar or antagonistic to natural hormones, and several hormonal systems can be affected. Therefore, this group of substances cannot be assessed as a whole.

Furthermore, the scientific literature regarding investigations of endocrine disrupting effects of single substances as well as risk assessments is voluminous and still increasing.

Therefore, no assessment of these substances is made but a Swedish exposure study will be reviewed briefly.

Nilsson (1996) elaborated human exposure assessments of four groups of possible endocrine disrupting substances with (anti)estrogenic effects. The substances included phthalates, NP, PCDD/Fs and PCBs. For each (group of) substance(s), exposure of soil via sewage sludge, synthetic fertilisers and manure was evaluated as well as uptake in plants and livestock. Furthermore, the persistence (and possibilities of accumulation) in soil were considered as well as the presence of the substances in drinking water. Estimates of human exposure to the substances via other routes of exposure were used to estimate the relative importance of the exposure caused by sludge amendment of soil.

The conclusions were that human exposure caused by sewage sludge application to

- Phthalates is probably very low and insignificant compared to total exposure. However information is insufficient regarding all routes of exposure. They are not expected to accumulate in soil or leach to groundwater
- NP is probably very low but total exposure is unknown. It is not expected to accumulate in soil or leach to groundwater
- PCDD/Fs is probably low compared to other sources. However, PCDD/Fs may accumulate in soil but are not expected to leach to groundwater
- PCBs is probably low, because PCB levels in sludge are low at present. However, PCBs may accumulate in soil but are not expected to leach to groundwater.

Summary of substances of concern

The above assessments, of which those for substances of potential concern are preliminary, are summarised in Table 9.1.

Table 9.1 Overview of assessments of substances of concern and potential substances of concern

Substance	**Assessment in summary**
LAS	Low concern
NP	Low concern
DEHP	Low concern
PAHs	Concern: Accumulation in soil
PCBs	Concern: Accumulation in soil
PCDD/Fs	Concern: Accumulation in soil
Brominated flame retardants	Concern: Accumulation in soil
Chlorobenzenes	Not assessed here
Nitro-musks / polycyclic musks	Lack of information regarding degradability in soil, possible concern, no conclusion
Chlorinated paraffins	Concern: Persistent, bioaccumulative, toxic, high conc. in sludge
Organotins	Not assessed here
VOCs	Not assessed here
Quintozene	Not assessed here
Polychlorinated naphthalenes	Not assessed here
Polydimethylsiloxanes	Not assessed here
Pharmaceuticals	Not assessed here
Synthetic fats	Not assessed here
Polyelectrolytes	Concern: Persistent and bioaccumulative, lack of information
Endocrine disrupters, incl. oestrogenic	Different properties and occurrence, cannot be assessed as a group, no conclusion

Strategies for risk reduction

The long-term objective is reduction of concentrations of critical organic contaminants in sewage sludge by elimination of the sources of emission.

The most efficient way of reducing the contamination of sewage sludge with organic chemicals would be to separate toilet outlets from all other sewage and use only sludge from toilet sewage for agricultural soil. An evaluation of the practical and economic implications of this measure is outside the scope of this report. In this section, only strategies for risk reduction in connection with the present construction of most Swedish sewerage systems is considered.

The legislative framework of the application of sewage sludge to agricultural soil in Sweden forms the basis of the development of strategies for reduction. As mentioned in the introduction, this framework includes the specific sewage sludge legislation as well as the Environmental Quality Objectives as introduced by the Swedish parliament. The most relevant paragraphs of these are summarised below.

Background: Swedish legislation

The national Swedish sewage sludge directive (SNFS 1998:4) includes requirements for the application of sewage sludge, which imply risk reduction by means of exposure reduction through limitations in areas of application (§7: grazing pasture - harvest within 10 months).

The Environmental Quality Objectives in SOU 2000:52 contain two objectives, which are relevant to sewage sludge.

The first prescribes that re-circulation of a considerable part of the phosphorous from waste and sewage should be returned to agricultural soil 2010. This implies one of two options. It is possible to extract P from sewage sludge (mentioned in some the EU reports) but this option is not considered in this report. The alternative option is application of sludge to agricultural soil.

The other Environmental Quality Objective requires “a poison-free environment: The environment must be free from substances and metals, which have been created in or extracted from the society and which can threaten human health or biological diversity.” A prerequisite for fulfilling this objective is that substances, which may threaten human health or biological diversity, can be identified.

Identification of substances of concern

"New" substances are continuously being identified as potential substances of concern and these have not been measured prior to the identification (e.g. the brominated flame retardants). No matter how many substances are included in the analytical programmes, there is never a guarantee that there will not be other substances present in sludge, which may be of concern.

All the substances used in production and households and led to sewage systems, as well as substances emitted to the atmosphere and subsequently deposited on consolidated areas will reach the WWTPs. Among these substances, those with a relatively high ability to adsorb to sludge particles and which are not fully mineralised during the retention time in the treatment plant will end up in the sludge.

The reason for concern is basically the toxic properties of substances that causes danger to humans or the environment combined with the ability to persist in the environment and/or to bioaccumulate.

In general, the risk assessments (in terms of effects on human health or the environment, which have been carried out by several authorities (e.g. US-EPA 1996 (PCBs, TDCC/Fs), Hellström 2000 (PBDPEs)) indicate very low immediate risks from the application of sewage sludge to soil - especially with the restrictions in the application practice (reducing human exposure), which are imposed by SNFS 1998:4.

Therefore, the main problem is whether a long-term build-up of concentrations of persistent contaminants in agricultural soil is acceptable, from a human health and environmental point of view, even if the immediate risk is negligible. This should be seen in the light of the continuous identification of "new" substances of concern but also the continuous awareness in society and authorities of persistent organic contaminants and the subsequent measures taken to reduce the use and/or formation of these.

In other words: In a long-term perspective, a sustainable use of sewage sludge in agriculture requires control and reduction of persistent pollutants in sludge. The short-term goal should include measures that can ensure a safe use of sludge in agriculture. Taking PCBs and NP as examples, it seems likely that due to the phasing out of these substances, the concentrations in sludge will keep falling and sooner or later there will be no need for limits for those. In the meantime, new substances will be identified (e.g. the brominated flame retardants) - and monitoring of these will be relevant. If phasing out is initiated, the monitoring could be seen as a monitoring of the progress of this process, e.g. this has hitherto been the reason for monitoring the concentrations of NP in sludge in Sweden.

Because of the limited range of substances included in the analytical programmes, this will not ensure that there is no risk from application of sludge if limit/guidance values are kept - and therefore, monitoring of sewage sludge quality, based on the current substances of concern, may seem to give a false security. However, if the sources of the substances of concern are known, monitoring may indicate the level of contamination of the sludge from these sources - also with respect to substances, which have not yet been identified as being of concern.

In order for monitoring programmes to be effective, the list of substances included in a programme should be updated continuously based on the most recent knowledge on dangerous substances, e.g. based on the REACH strategy as described below.

Limits to concentrations in sewage sludge could also be used as a measure for motivating local authorities for identification of sources for contamination and subsequently taking measures for reduction of these sources - e.g. by putting taxes on the discharge of the substance(s).

Measures

The measures available for reduction of risks caused by substances in sewage sludge are directed at different steps in the pathways of the chemicals from discharge to human and/or environmental exposure.

One option is treatment of sludge for reduction of contents of xenobiotics before agricultural application. Results of several research programmes indicate that composting is promising as a mean of reduction of organic contaminants. E.g., the Danish research programme included investigations of the degradation of organic contaminants in sewage sludge during composting. The results showed that DEHP, LAS, NPE and 9 PAHs were degraded during 25-90 days of composting as up to 91% of the DEHP and up to 97% of NP were degraded. Degradation of NPE was dependent on temperature (Mogensen et al. 2001). Norwegian investigations support these findings (Amundsen et al. 2001). However, evaluation of the applicability of such measures is not included in this project.

Another option includes measures aiming at avoiding exposure of crops/livestock to chemicals present in sludge. The regulation of crops and/or methods of application are such measures.

The option most suitable for a sustainable agricultural use of sewage sludge is the introduction of measures directed at reducing and eliminating sources of critical chemical substances.

Short-term measures

Short-term measures could include:

- Regulation of application

- Limit values for contaminants in sludge
- Treatment (e.g. composting) of sludge, not discussed here

The avoidance of exposure can be handled by the sewage sludge legislation, partly by the setting of limit values, partly by regulation of crops for which the sludge can be used and/or rules for methods of application. Furthermore, limit values for specific substances in sewage sludge and industrial discharges to public sewage systems can be fixed. The latter has been proven to be an important tool for the management of sludge quality.

These measures can be applied immediately and could be considered as the corner stones of short-term measures for risk reduction.

Medium-term measures

Source tracking of critical substances and reduction of specific local point sources is the main item, which could be regarded as a "medium-term measure".

At a local scale, the possibility of tracking sources of critical substances and initiating reduction of the specific discharges may be a very efficient way of reducing emissions to waste water. In principle, this can be initiated immediately but the effects will most often appear with a certain delay.

Reduction of discharges of chemicals from industries to waste water can be obtained by technical measures. Dependent on the source of the discharge, the instruments could include:

- Substitution of specific substances
- Recirculation inside plant
- Collection of dangerous fractions of waste water for special treatment
- Pre-treatment of waste water before discharge to public sewerage system

Long-term measures

General reduction of emissions to waste water will require other measures, of which some may be (and are) applied immediately while the effects can only be expected on a long-term scale. They would include national measures for reduction of local emissions as well as international measures for reduction of especially airborne dangerous chemicals. Several measures of this type have already been applied as described below.

The discharge of chemicals to waste water can be efficiently reduced by avoiding the use of chemicals (in products and/or production), i.e. by substitution. The measures for reduction of emission of chemicals to waste water will partly be dependent on the source of the chemicals and are summarised below.

Waste water from industry

- Regulatory: Ban on individual chemicals
- Voluntary: Agreements on phasing out / reduction
- Consumers avoidance of products containing the substances (public awareness campaigns, e.g. eco-labelling)
- Economic incentives:
- “Negative”: Taxing discharge of specific substances
- “Positive”: Economic support for development of cleaner technology (substitution)

Waste water from households

- Separation of toilet outlets from all other sewage and agricultural use of sludge from this source only
- Avoidance of certain chemicals in consumers products - see “industrial”
- Avoidance of release of used chemicals to sewer
- Public awareness campaigns combined with good facilities for collection of chemical wastes

Regulation of diffuse sources leading to chemicals in run-off from streets etc.

Reduction of contamination from vehicles, roofs, gutters, road surfaces and combustion would all result in reduction of the amounts of chemicals reaching the WWTPs and consequently the sewage sludge. The measures could include:

- General: Encourage exchange of old cars for new with better technology for reduction of spill as well as exhaust treatment
- Low taxes on new cars
- Pollution dependent tax on vehicles
- Extra “road tax” on old cars
- Specific:
- Pre-treatment or separate treatment (low technology methodologies like sand filters etc.)
- Regulations including technical standards
    - E.g. demands for catalytic exhaust purifiers and/or filters
    - E.g. obligatory, regular control of car quality

- Choice of materials for roofs, gutters and road surfaces (e.g. PVC gutters, asphalt roads)
- Demands for combustion processes and/or smoke cleaning
- Public awareness campaigns regarding fuels for private heating, e.g. fireplaces, wood burning stoves
- Improved combustion plants for public incineration, heating and electricity
- Planning of heating, e.g. district heating
- Long-term: International agreements on reduction of/ban on chemicals etc. (e.g. the Stockholm Convention includes PCDD/Fs)

Not identified substances of concern

For substances, which have been identified as being of concern in sewage sludge, the above measures can be applied. However, the preceding chapters of this report point to the fact that even the most qualified list of such substances may not be exhaustive, simply because there may be substances that possess POP-properties but to which no attention has yet been paid due to lack of knowledge.

For possible identification of such "new" substances of concern, regular reviews and comparison of knowledge (databases) regarding the use and properties of substances, which are used for the production of and/or present in manufactured products, could be a measure. This has already been practised in connection with the former Swedish assessment of chemicals in sludge (Naturvårdsverket 1993) as described in Chapter 3 of this report.

Substances, for which information regarding their environmental properties is available, could be identified this way. However, for many existing substances, no such information is available. Properties like biodegradability and toxicity of organic substances may be estimated on the basis of their chemical identity by means of Quantitative Structure-Activity Relationship (QSAR). This approach is being used by the Danish EPA and has hitherto led to the publication of a Guidance list for self-classification of dangerous substances (DK-EPA 2001a), containing 20,624 substances.

Measures already adopted for reduction of use

In the Swedish chemicals policy, several measures for reduction of the use of hazardous chemicals have already been adopted. Some are initiated at the national level, some are consequences of international agreements. The Swedish National Chemicals Inspectorate has made most of the following information available.

General national regulation

The take-off for the national regulation is the Environmental Quality Objective requiring that “the levels of xenobiotics in the environment are close to zero”. With respect to this quality objective, the Swedish parliament has adopted six sub-objectives among which the third concerns newly manufactured products, specifying that they should be free from:

- Carcinogenic, mutagenic and reproductive disturbing substances by 2007, if the products are intended to be used in such a way that they are released to the eco-cycle
- New, organic substances that are persistent and bioaccumulative as soon as possible, but no later than 2005. Additional organic substances that are very persistent and very bioaccumulative, by 2010.
- Other organic substances that are persistent and bioaccumulative, by 2015.

Furthermore, these substances may not be used in production processes unless the company can prove that human health and the environment will not be harmed. The sub-objective applies to man-made or extracted substances and includes substances giving rise to substances with the above properties, including those formed unintentionally.

EU regulation on new and existing chemical substances

At the international level, the most important general effort to reduce substances with undesirable properties is the EU White Paper “Strategy for a future Chemicals Policy”, in which proposals for a strategy are presented, and which has as its overriding goal: Sustainable development. The central part of the proposal is the REACH system, which introduces requirements for Registration, Evaluation and Authorisation of CHemical substances. Essentially, this would be a system of hazard identification, risk assessment and risk management. An authorisation system is proposed for the risk management of Substances of Very High Concern (SVHC). According to the White Paper, these are specifically categories 1 and 2: Carcinogens, mutagens and reproductive toxicants (CMRs) and substances with “POPs characteristics”. The Council has invited the Commission to add also PBT (Persistent, Bioaccumulative and Toxic chemicals) and vPvB substances (very Persistent, very Bioaccumulative chemicals) to the group of SVHC. A key feature of the authorisation proposal is that it reverses the burden of proof by requiring that, for each single use of a SVHC substance, industry must demonstrate that the risk is negligible. The Commission is expected to present a proposal for amended legislation this autumn.

The EU Water Framework Directive (Annex X) includes priority lists of dangerous substances, which will be updated regularly. The lists are derived by a systematic selection of substances based on their potential risk to the water environment. WFD includes provisions for reduction/elimination of emission of these substances to the water environment.

National measures for reduction of individual substances of concern

Nonylphenol/-etoxylates (NP/Es) and *Chlorinated paraffins (CPs)* have been subject to voluntary phase-out efforts in Sweden since the beginning of the 1990s, in line with goals set up in Government Bills. The use of NP/Es has decreased by over 90% and the use of short-chained CPs by approx. 80% (KemI, 2001a).

Efforts aimed at *brominated flame retardants* were also initiated in the beginning of the 1990s. The National Chemicals Inspectorate has proposed a strategy for the phasing out of PBDEs and PBBs (KemI, 1999). It has been noted that these two groups of substances are indeed subject to on-going phase-out while it is difficult to verify any changes with respect to other brominated flame retardants (KemI 2001a).

Phase-out goals for *DEHP and other harmful plasticisers* in PVC were introduced in Government Bill Prop. 1997/98.145. A progress report on the achievements was presented by the National Chemicals Inspectorate in December 2000 (KemI, 2001b). According to the report, the rapid phase-out of outdoor products, indicated in the Government's Bill, will not be fully implemented. In the case of other product groups, the phase-out will depend on the acceptance by foreign suppliers of a replacement of DEHP.

EU measures regarding individual substances

Many of the industrial chemical substances/groups of substances high-lighted in this report are, or have already been, subject to comprehensive risk assessments at the EU level within the Existing Substances Programme. Where a concern for human health or the environment has been concluded, discussions and negotiations on restrictions on marketing and use have followed. Below, a brief summary is given of the status for some sub-stances/groups of substances.

Nonylphenol/-ethoxylates (NP/Es)
The risk assessment concluded that there is a concern for effects in the terrestrial and aquatic compartments of the environment. A strategy for reducing the risks, including a set of measures, has been agreed and a proposal to restrict the use for several purposes is currently under discussion at the Commission level.

DEHP and other phthalates
Although some details remain to be discussed, the risk assessment of DEHP for human health will conclude that there is a need for limiting the risks. A risk reduction strategy is currently being worked out by Sweden. Other phthalates subject to risk assessment are DINP, DIDP, DBP and BBP.

Chlorinated paraffins (CPs)
The uses of *short-chained* CPs for metal working fluids and leather goods are about to be banned. The remaining uses will be subject to a review. A risk assessment of the *medium-chained* CPs is on-going.

PBDEs
The use of PentaBDE will soon be banned. Risk assessments for Octa- and DecaBDE are about to be finalised and a discussion on risk reduction measures has been initiated.

Other international restrictions on the use of specific chemicals

The Stockholm Convention on Persistent Organic Pollutants was acknowledged by the UN Intergovernmental Negotiating Committee in December 2000 and is a legally binding treaty that requires governments to minimise and eliminate some of the most toxic chemicals ever created. The 12 initial POPs include *eight pesticides* (aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, mirex and toxaphene), two industrial chemicals (*PCBs and hexachlorobenzene*, which is also a pesticide), and two unwanted by-products of combustion and industrial processes (*dioxins and furans*). Further POPs will be included in future revisions of the list. As this is an international effort, involving more countries than e.g. the EU, it is expected that addition of new substances to the POP list will be less rapid than the results of the REACH system. The importance of the Stockholm Convention mainly lies in its agreement of minimising the use/formation of some of the substances known to be subject to long range (intercontinental) atmospheric transport.

The international convention on the control of harmful antifouling systems on ships was agreed in October 2001. According to the convention, *tributyltin* (TBT) may not be applied or re-applied on ships from 1 January 2003. Additional restrictions for marketing will be introduced within the EU.

Musk xylene appears on the OSPAR List of Chemicals for Priority Action. The Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR) and the Convention on the Protection of the Marine Environment of the Baltic Sea Area (Helsinki convention, by the HELsinki COMmission (HELCOM)) both include lists of substances, which should be reduced. Switzerland is responsible for presenting a background document on musk xylene and other musk compounds. The document will include recommendations to reduce environmental exposure.

Outline of a strategy for risk reduction of organic chemicals in sewage sludge

In summary, a strategy for risk reduction of organic chemicals in sewage sludge should consist of measures, which can be initiated immediately and would be effective in a short-medium term perspective, and measures aiming at long-term elimination of emissions of such substances to waste water.

Short-medium term measures

- Treatment of sludge before use, e.g. composting (rules, which will support increased composting of sewage sludge are in the process of being implemented in Swedish legislation)
- General limitations on sludge application (are already in force)
- Identification of possible “new” substances of concern followed by risk assessments and possible limit values
- Limit values for specific substances in sludge
- Limit values for discharge of such substances with industrial waste water
- Source tracking of critical substances and reduction of specific point sources
- Establishment of a system for monitoring of sewage sludge and waste water in combination with continuous updating on the latest scientific development for identification of potential “new” substances of concern

As a starting point for decisions regarding identification of substances of concern and their inclusion in a monitoring programme as well as the setting of limit values, conclusions regarding the substances, which have been assessed in the preceding chapters, are summarised in Table 11.1.

Table 11.1 Overview of conclusions regarding substances assessed above

Substance	Conclusion	Monitoring	Limit value
LAS	Low concern	No	No
NP	Low concern	No	No
DEHP	Low concern	No	No
PCBs	Concern: Accumulation in soil – concentrations expected to be very low and decreasing in sludge from all WWTPs	No	No
PAHs	Concern: Accumulation in soil – information regarding occurrence in sludge from individual WWTPs needs to be analysed	Dependent on result of analysis of information*	Dependent on result of analysis of information*
PCDD/Fs	Concern: Accumulation in soil - information regarding occurrence in sludge from individual WWTPs needs to be analysed	Dependent on result of analysis of information*	Dependent on result of analysis of information*
Brominated flame retardants	Concern: Accumulation in soil	Yes	?
Nitro-musks / polycyclic musks	Possibly of concern, more information needed	?	?
Chlorinated paraffins	Candidates for concern, more information needed	Yes	?
Polyelectro-lytes	Possibly of concern, more information needed	Yes	?
Endocrine disrupters	Cannot be assessed as a group, more information on specific substances needed	?	?

*: The variation in concentrations measured in sludge from different WWTPs must be considered in order to evaluate whether some WWTPs give rise to sludge with alarmingly high concentrations. In that case, monitoring may be relevant.

Table 11.1 shows that most of the substances, which have been considered as being of concern and monitored, are no longer causing concern; mainly due to low and decreasing levels in sludge (LAS, NP, DEHP and PCBs). The concern regarding PAHs and PCDD/Fs is also expected to be low but further analysis of the variation between samples is needed before a conclusion can be drawn.

The remaining substances in Table 11.1 are substances, which have been identified as potential "new" substances of concern during the literature study. Regarding these substances, the main reason of concern is not the risk of toxic effects on man or the environment. The main reason of concern is that the substances may accumulate in soil and thereby violate the Swedish Environmental Quality Objective regarding a poison-free environment.

Long-term measures

- Phase-out of critical chemicals as initiated by national Swedish and international (UN, EU, OSPAR/HELCOM) strategies for reduction

- Continuous focus on critical chemicals in a follow-up on the above monitoring system (of sewage sludge and waste water in combination with continuous updating on the latest scientific development for identification of potential "new" substances of concern)
- Development of source separation of waste water in order to restrict use of sewage sludge for agricultural purposes to that originating from toilets

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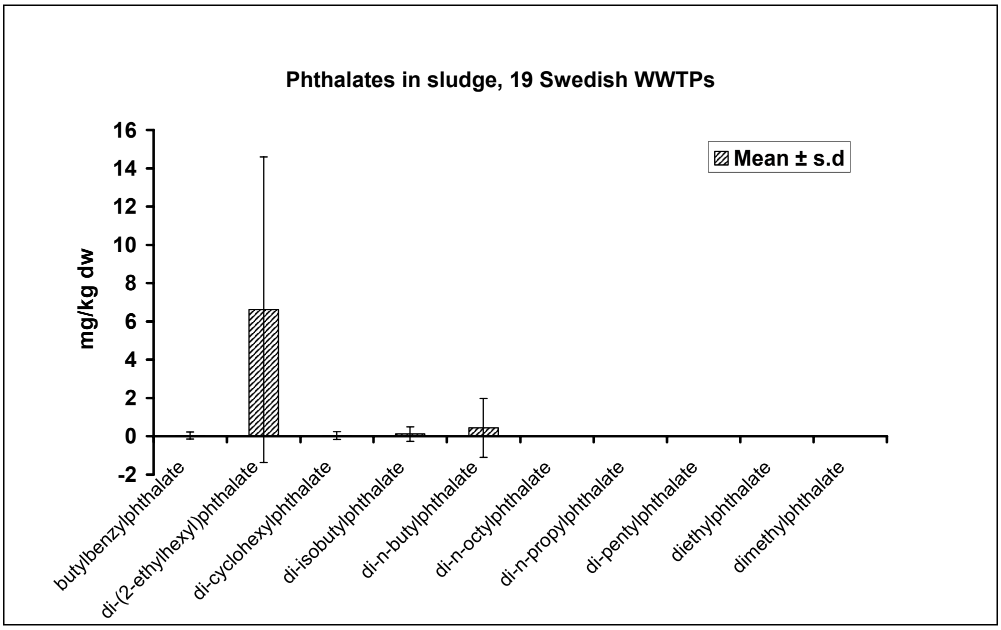
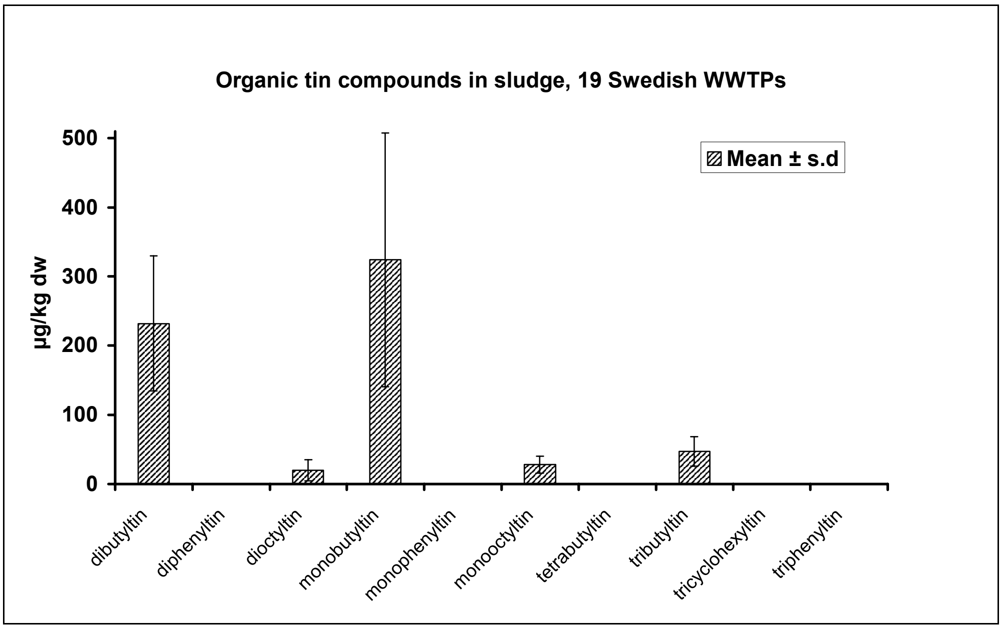
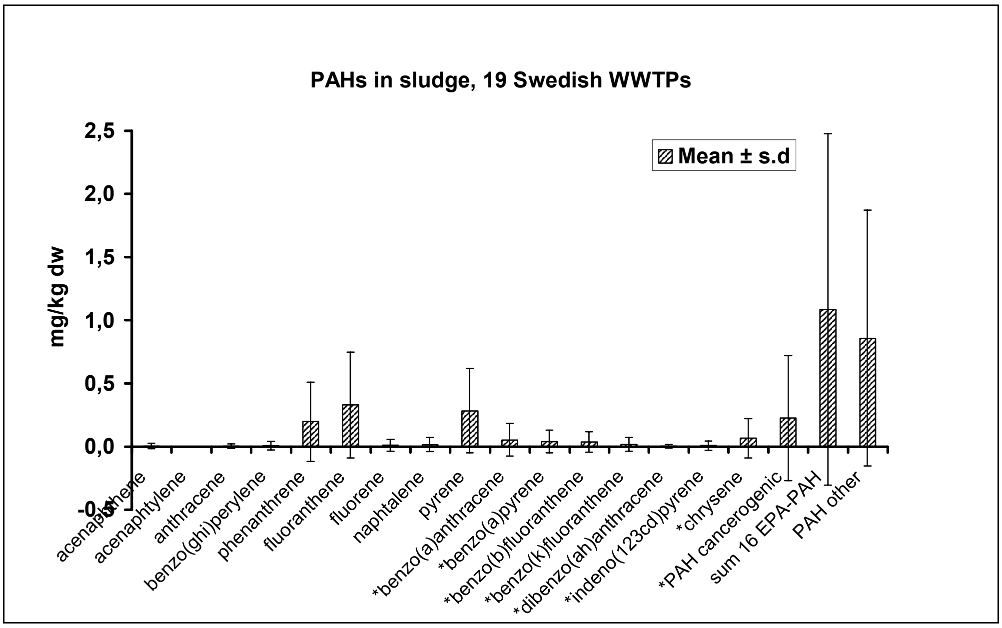
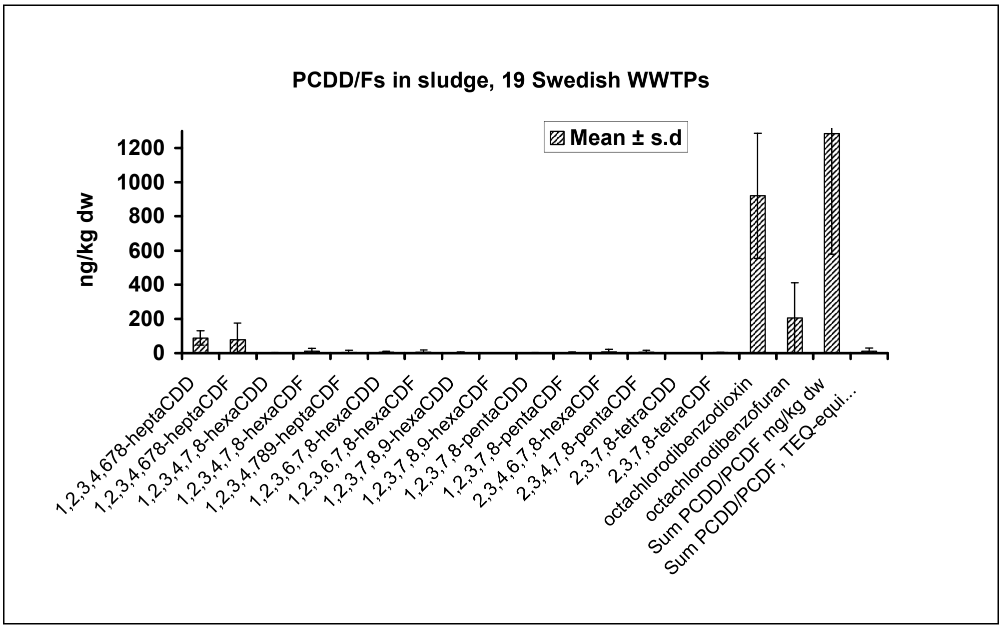
Annex I - Swedish sludge 2001/2002

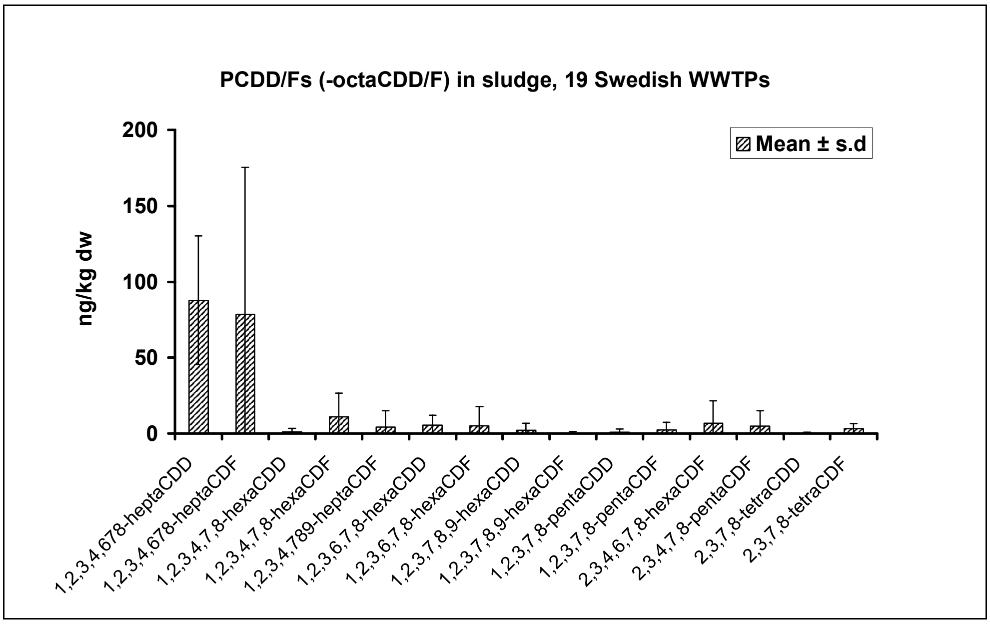
The Swedish EPA has analysed samples of sewage sludge from 19 different Swedish WWTPs for a series of different types of organic contaminants. Furthermore, University of Stockholm has analysed sludge from 50 WWTPs for brominated flame retardants (Nylund et al. 2002). The results are summarised in the figures below, in which the units are different as indicated next to the Y-axis.




Organic tin compounds in sludge, 19 Swedish WWTPs
Mean ± s.d
µg/kg dw
500
400
300
200
100
0
dibutyltin
diphenyltin
dioctyltin
monobutyltin
monophenyltin
monooctyltin
tetrabutyltin
tributyltin
tricyclohexyltin
triphenyltin


PAHs in sludge, 19 Swedish WWTPs
Mean ± s.d
mg/kg dw
2,5
2,0
1,5
1,0
0,5
0,0
-0,5
acenaphthene
acenaphtylene
anthracene
benzo(ghi)perylene
phenanthrene
fluoranthene
fluorene
naphtalene
pyrene
*benzo(a)anthracene
*benzo(a)pyrene
*benzo(b)fluoranthene
*benzo(k)fluoranthene
*dibenzo(ah)anthracene
*indeno(123cd)pyrene
*chrysene
*PAH cancerogenic
sum 16 EPA-PAH
PAH other


PCDD/Fs in sludge, 19 Swedish WWTPs
Mean ± s.d
ng/kg dw
0
200
400
600
800
1000
1200
1,2,3,4,678-heptaCDD
1,2,3,4,678-heptaCDF
1,2,3,4,7,8-hexaCDD
1,2,3,4,7,8-hexaCDF
1,2,3,4,789-heptaCDF
1,2,3,6,7,8-hexaCDD
1,2,3,6,7,8-hexaCDF
1,2,3,7,8,9-hexaCDD
1,2,3,7,8,9-hexaCDF
1,2,3,7,8-pentaCDD
1,2,3,7,8-pentaCDF
2,3,4,6,7,8-hexaCDF
2,3,4,7,8-pentaCDF
2,3,7,8-tetraCDD
2,3,7,8-tetraCDF
octachlorodibenzodioxin
octachlorodibenzofuran
Sum PCDD/PCDF mg/kg dw
Sum PCDD/PCDF, TEQ-equi...


PCDD/Fs (-octaCDD/F) in sludge, 19 Swedish WWTPs
Mean ± s.d
ng/kg dw
0
50
100
150
200
1,2,3,4,678-heptaCDD
1,2,3,4,678-heptaCDF
1,2,3,4,7,8-hexaCDD
1,2,3,4,7,8-hexaCDF
1,2,3,4,789-heptaCDF
1,2,3,6,7,8-hexaCDD
1,2,3,6,7,8-hexaCDF
1,2,3,7,8,9-hexaCDD
1,2,3,7,8,9-hexaCDF
1,2,3,7,8-pentaCDD
1,2,3,7,8-pentaCDF
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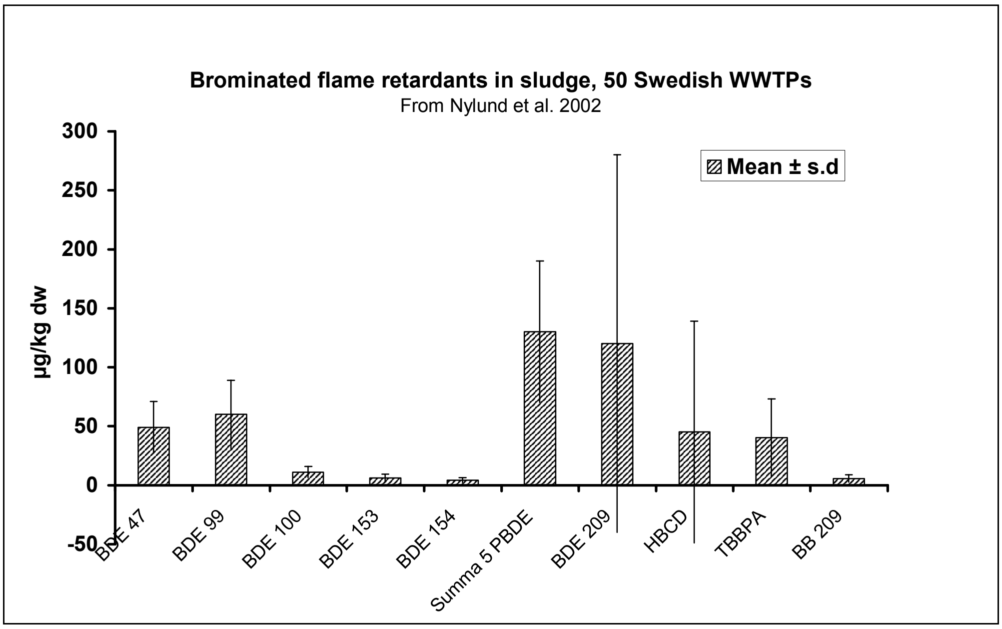
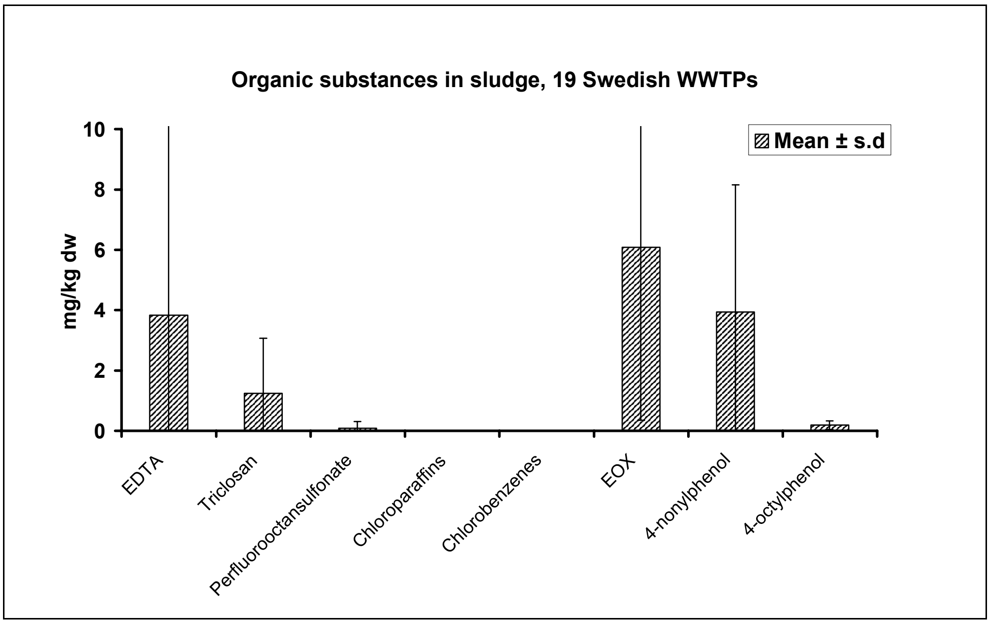
2,3,4,7,8-pentaCDF
2,3,7,8-tetraCDD
2,3,7,8-tetraCDF


Brominated flame retardants in sludge, 50 Swedish WWTPs
From Nylund et al. 2002
µg/kg dw
300
250
200
150
100
50
0
-50
Mean ± s.d
BDE 47
BDE 99
BDE 100
BDE 153
BDE 154
Summa 5 PBDE
BDE 209
HBCD
TBBPA
BB 209


Organic substances in sludge, 19 Swedish WWTPs
mg/kg dw
10
8
6
4
2
0
Mean ± s.d
EDTA
Triclosan
Perfluorooctansulfonate
Chloroparaffins
Chlorobenzenes
EOX
4-nonylphenol
4-octylphenol

REPORT 5217

Organic Contaminants in Sewage Sludge

The Swedish Environmental Protection Agency (EPA) was commissioned to examine, in consultation with the relevant authorities and stakeholders, the issues concerning the environmental and health protection requirements for sewage sludge and its use as well as possible restoration of phosphorus from wastewater to arable land. This assignment is reported in the form of the "Aktionsplan för återföring av fosfor ur avlopp", report 5214 from EPA. The present report is a sub-report to the main report mentioned above and presents a review on studies regarding the occurrence and risks of different organic contaminants in relation to the use of sewage sludge in agriculture. Material from Sweden, Norway, Denmark, Germany, the United Kingdom and the USA is considered as well as consultant reports compiled for the European Commission. As a result, priority setting of organic compounds and strategies to minimise risks are proposed.

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