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FINAL REPORT FOR DISCUSSION

Centre for Environmental Control and Waste Management
Department of Civil and Environmental Engineering

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The Implications for Human Health and the Environment of Recycling Biosolids on Agricultural Land

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FEBRUARY 2008

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EXECUTIVE SUMMARY

Recycling sewage sludge to agricultural land, to gain benefit from the essential plant nutrients and organic matter it contains, is regarded by most scientific and regulatory authorities as the most pragmatic and environmentally sustainable approach to managing the sludge generated from urban wastewater treatment. The opinion of the European Commission is that the use of sewage sludge on agricultural soils as a fertiliser is the best environmental option provided that it does not pose any threat to the environment as well as to animal and human health. The European Parliament reports that there are no cases of human, animal or crop contamination due to the use of sludge on agricultural soils following the provisions of Directive 86/278/EEC. Furthermore, it considers that the provisions of the Directive have prevented the spreading of pollution from the use of sludge and the Commission considers that a monitored and well-regulated land spreading of sludge should be encouraged and sustained. The flexible provisions of the Directive have enabled effective management of sludge use in agriculture whilst protecting human health and the environment.

Despite the international support for recycling sludge to land, the acceptance of this practice amongst different Member States of the EU varies considerably, and recycling has declined markedly in some countries. Under these circumstances, a decrease in the use of sludge in agriculture has resulted in the necessary expansion of incineration as the only viable alternative outlet for sludge. However, the Commission considers that this development contravenes the Waste Hierarchy, which favours recycling above incineration.

In the light of the increased production of sewage sludge across the Community in response to the implementation of the Urban Wastewater Treatment Directive, and recognising the need to ensure stakeholder confidence in the reuse of sludge in agricultural soils, the Commission plans to undertake a comprehensive review of the provisions contained in the Directive. In particular, this is to ensure they take into account potential long-term effects on soil quality.

An extensive body of scientific evidence has been generated internationally relating to health, environmental and soil quality aspects of recycling sewage sludge to agricultural soil. This evidence has also been synthesised into a number of reports and risk assessments, designed to draw together the comprehensive amount of data available to provide a consensus opinion on the measures required to manage sewage sludge on land in a safe and sustainable manner. The purposeof this report, therefore, is to give a holistic overview of the scientific knowledge and provide an objective summary of the key evidence to assist in the assessment of suitable, technically based, management practices and controls that ensure human health and the environment are protected when sewage sludge is recycled on farmland.

The overall conclusion of this review is that recycling sewage sludge on farmland as a soil conditioner and alternative fertiliser resource within current guidelines and controls on agricultural utilisation is a safe and sustainable practice. The scientific evidence indicates that human health, the environment and soil quality and fertility are protected by the current regime of controls used to regulate agricultural recycling in Europe. Furthermore, the chemical quality of sludge is continually improving from what is an already high standard compared with 20 or 30 years ago and this further strengthens the case to support the use of sludge on agricultural land as the main outlet for the product from urban wastewater treatment. Sludge treatment and management practices also provide effective multi-barriers that prevent the spread of infectious disease to humans and farm animals when sludge is applied to the soil as a fertiliser. Inevitably, in an environmental situation as complex as this there are a number of issues that emerge that require consideration and attention to further strengthen the scientific basis underpinning the practice. This review identified a number of

key areas that require further work, and these include:

Pathogens

- Further research is recommended to distinguish the different sources of enteric microbes entering soil including sludge organisms, to quantify the decay of enteric viruses in sludge-treated agricultural soil, quantify the effects of seasonal conditions on the decay of enteric pathogens in soil and also the role and mechanisms of ecological suppression of enteric microorganisms applied to soil in sewage sludge.
- The mechanisms of pathogen inactivation during mesophilic anaerobic digestion of sludge, one of the principal methods of stabilizing sewage sludge for agricultural spreading adopted by the Water Industry, are poorly understood. Therefore, more work is recommended to determine the fundamental processes responsible for pathogen destruction during anaerobic treatment so that this important sludge stabilization process may be optimized for pathogen removal.
- Recent research has shown that centrifuge conditioning of sludge may significantly increase indicator numbers in digested and pasteurised sludge and time-temperature exposure during sludge pasteurisation may transfer *E. coli* to a viable, but non-culturable state, that is reactivated by centrifuge-dewatering. The effects of both these processes on recoveries of enteric bacteria in sludge have implications for compliance with microbiological reduction criteria for sludge and require further investigation.

Chemical contaminants

- There remains scope to further reduce the concentrations of problematic contaminants, and PTEs in particular, in sludge. This should continue to be a priority and pursued proactively by environmental regulators and the Water Industry as improving the chemical quality of sludge as far as practicable is central to ensuring the long-term sustainability of recycling sewage sludge in agriculture.
- A microbiological risk assessment (MRA) should be completed to confirm that antibiotic resistant microorganisms in sludge-treated soil represent a negligible risk to human health.
- In contrast to other persistent organic pollutants, chlorinated paraffins (PCAs) are in active production and occur in sludge in much larger concentrations than PCBs and PCNs, for example. Consequently, further work is recommended to assess the potential transfer to the foodchain and significance for human health of PCAs in sewage sludge-amended agricultural soil.
- A risk assessment of DEHP in sludge-treated agricultural soil is recommended, following the protocol developed by Schowanek *et al*. (2004), to clarify the potential impact of this plasticiser compound on human health, soil quality and the environment.
- A number of emerging compounds have been identified belonging to the group of chemicals described as body care products eg triclosan that may potentially impact soil microbes and further research is required to determine the significance of these chemicals for soil quality.
- Further research is recommended to elucidate the physico-chemical and biological mechanisms responsible for the ecotoxicological response to high-metal cake sludges observed in ongoing long-term field trials designed to determine the effects of sewage sludge applications to agricultural land on soil microbial activity and the implications for agricultural productivity and long-term soil fertility. However, at this stage the evidence from the long-term field experiments does not warrant any immediate change to the maximum permissible soil limit values for Zn, Cu or Cd.

Nutrients

- Phosphorus inputs to soil in sludge are increasing at agronomic rates of sludge application calculated on a N basis due to the expansion in P removal from wastewater. More information is therefore required on the long-term fate and release of P in sludge-treated agricultural soil to crop rotations to assess the agronomic benefit and efficiency of utilisation of P to ensure accumulation in soil and risk to the water environment are minimised.

Odour

- The treatment and land application of sewage sludge is managed to minimize odour nuisance. However, it may be a transient potential source and a potential cause of complaint and annoyance. Odour is one of the key factors raising public concerns and negative perceptions about the agricultural use of sludge. Odour emissions are generally considered primarily in terms of the nuisance caused to communities close to the source, but recently it has been reported that malodour can have direct and indirect impacts on human health (Schiffman and Williams, 2005). This has been documented for communities close to static odour emitters, such as intensive livestock production centres. Therefore, measures to prevent odour when sludge is used in agriculture should be reinforced and could become part of the controls on agricultural use, to ensure odour emissions from sludge are minimized as far as practicable.

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1. INTRODUCTION

1.1 Background

This report examines the potential implications for human health and the environment of recycling sewage sludge (biosolids) to agricultural land. In such an assessment, human health forms a primary consideration. However, soil fertility and other environmental endpoints are also important for the overall long-term sustainability of spreading sewage sludge on farmland. Encouraging the application of organic residuals on land to improve soil and recycle nutrients for crop production is recognised as a basic requirement of sustainable development (Bergström *et al*., 2005) and the agricultural use of sewage sludge has an important role to play in this. For example, the Green Alliance (2007) recently published a pamphlet to raise political awareness of the importance of closing nutrient cycles and the need for better management and increased recycling of nutrients through the foodchain.

Sewage sludge is an essential and inevitable residual by-product of urban wastewater treatment. Sludge is produced during wastewater treatment processes as a consequence of removing suspended, colloidal and dissolved solids from urban wastewater, discharged from homes, industry and collected as drainage from paved surfaces, so that the treated effluent can be safely returned to the water environment. Sewage sludge is further processed by a number of possible treatment methods before it can be spread on farmland to reduce or eliminate microbial pathogens of enteric origin that may be present in the sludge and to reduce odour nuisance and its attractiveness to insects and other vectors. Sewage sludge which is treated to a standard that is acceptable for use in agriculture is also referred to as 'biosolids', to distinguish this as a product from material that is unsuitable for recycling on land. In this report, all references to sewage sludge relate to material that is acceptable for agricultural application.

Agronomically, sewage sludge is an effective replacement for conventional inorganic nitrogen (N) and phosphate (P) fertilisers in crop production. The fertilizer value of sludge has been extensively researched (Coker, 1966; Coker *et al*., 1987) and guidance is available (eg WRc, 1986; MAFF, 2000) to maximise the utilisation of the nutrient resources contained in sludge and to minimise potential impacts and wasteful losses to the water environment (Shepherd, 1993; UKWIR, 1995a; Shepherd, 1996; Misselbrook *et al*., 1996; Smith *et al*., 1998a,b,c; UKWIR, 2000; Withers *et al*., 2001; UKWIR, 2005). Recent research has also been completed to provide information on the nutrient behaviour of the expanding range of different treated sludge types available for recycling on farmland produced by modern sludge treatment processes (eg Cogger *et al*., 1999; Smith and Durham, 2002; Smith *et al*., 2002a; Smith *et al*., 2002b; Morris *et al*., 2003; Smith *et al*., 2003, O'Connor *et al*., 2004). In addition to N and P, sludge provides a valuable agronomic source of sulphur (S) and magnesium (Mg) (UKWIR, 1997; Nicholson *et al*., 1997). The beneficial effects of sludge application on soil organic matter status, structural properties and soil moisture retention are also well documented (Hall and Coker, 1983; UKWIR, 1999; UKWIR, 2001; Gibbs *et al*., 2006a). In addition to the agronomic and soil quality benefits of supplying organic carbon (C) to soil in sludge, this practice has the advantage of increasing C sequestration and thus contributing to C mitigation measures to meet European climate change commitments under the Kyoto Protocol (Smith *et al*., 2000; Smith *et al*., 2001)

The importance of recycling P in the foodchain is arguably one of the most critical issues in favour of using sewage sludge in agricultural systems. Sludge provides a long-term maintenance dressing for this nutrient and can entirely replace P inputs from inorganic fertilisers in a crop rotation. World reserves of currently exploitable phosphate rock are estimated to be 40 billion t and, at the present rate of consumption (150 million t per year), this will be exhausted within 250 years (CEEP, 1997). The reserves of high quality P minerals, low in Cd, are also likely to be exhausted within several decades (Steen and Agro, 1998). Recycling P to soil in biosolids therefore has a global benefit by contributing to the conservation of geological mineral P reserves and also reducing external inputs of geogenic Cd (Cd) into the foodchain from phosphatic fertiliser sources (Loganathan *et al*., 1996). Therefore, recycling P in biosolids has to be considered as a key prerogative for long-term sustainability. Phosphorus recovery during wastewater treatment is highly efficient and sludge is an effective P fertiliser source that helps to close the nutrient loop through the food chain, provided it is carefully managed.

Whilst recognising its significant value as a resource, recycling sewage sludge to farmland requires careful and responsible management to avoid potential impacts on human health and the environment from infectious pathogenic microorganisms that may be present in sludge and from chemical contaminants, including potentially toxic elements (PTEs) and organic chemicals, that transfer to sludge as a normal consequence of the wastewater treatment process. Most European countries developed guidelines to ensure sewage sludge was used safely on farmland and the controls were regulated and unified at an early stage by European Directive 86/278/EEC (CEC, 1986). In particular, the Directive set maximum limit values for PTEs in sludge or sludge-treated soil and specified general land use, harvesting, and grazing restrictions, to provide protection against health risks from residual pathogens. The development of guidelines and statutory controls has been an ongoing process at a national level since the Directive was enforced and most countries have subsequently adopted more stringent limits and management practices than were originally specified by the Directive in 1986. For example, in the UK, a voluntary code has been agreed between the UK Water Industry and British Retail Consortium, known as the Safe Sludge Matrix (ADAS, 2001), that requires more rigorous control over sludge treatment, pathogen removal and use on land than was originally required by the guidelines in the *Code of Practice for Agricultural Use of Sewage Sludge* and the Statutory Instrument (DoE, 1989; UK SI, 1989) implementing the Directive. Importantly, the Matrix also introduced a two tier system of treatment for sludge with regard to the extent of pathogen removal, and the subsequent stringency of land use controls that was analogous to the US EPA's Class A and B pathogen reduction requirements in the *Part 503 Standards for the Use or Disposal of Sewage Sludge* for agricultural use of sludge (US EPA, 1993).

Agricultural use continues to be widely regarded as the best practicable environmental option (BPEO) for sludge management in most circumstances (Defra, 2007a). The European Commission considers that the use of sewage sludge on agricultural soils as a fertiliser is the best environmental option provided that it does not pose any threat to the environment as well as to animal and human health (EC, 2003a). In its most recent report to the European Parliament on the implementation of Community waste legislation (EC, 2006), the Commission confirmed that there are no reported cases of human, animal or crop contamination due to the use of sludge on agricultural soils following the provisions of Directive 86/278/EEC. Furthermore, it considers that the provisions of the Directive have effectively prevented the spreading of pollution from the use of sludge and that a monitored and well-regulated land spreading of sludge should be encouraged and sustained. The current Directive 86/278/EEC sets the minimum standard required for the use of sludge in agriculture and Member States have gone on to set additional controls as appropriate for local circumstances. The provisions of the Directive have generally clearly worked well achieving the aim of protecting the environment and human health whilst facilitating a practical basis to support the recycling of sludge (Blackmore *et al*., 2006). It is therefore in the interests of the regulator, Water Industry and society in general that any revision of the Directive should retain this effective, flexible and pragmatic approach. However, concerns about the potential consequences for human health and the environment of potentially toxic substances and harmful microorganisms in sludge have led to the banning of the use of sludge in agriculture in Switzerland, despite the recognition that there is no conclusive scientific evidence that the practice is harmful in anyway (FOEN, 2003). Given the evidence that will be reviewed in this report, this is seemingly an unnecessary and extreme action contravening European policy relating to waste management that aims to encourage the recovery of value from waste products.

With the growth in sludge production in Europe in response to the implementation of environmental legislation, and the Urban Wastewater Treatment Directive 91/271/EEC (CEC, 1991) in particular, and recognising the need to ensure stakeholder confidence in the reuse of sludge in agricultural soils, the Commission plans to undertake a comprehensive review of the provisions contained in the Directive. In particular, this is to ensure they take account of any long-term effects on soil quality.

The health, environmental and soil quality aspects of recycling sewage sludge to agricultural land have been extensively researched internationally. Different aspects of the subject have been reviewed many times and risk assessments have also been completed, most recently concerning the microbiological quality of sludge (Gale, 2003a,b; 2005). Collectively, all of this evidence can be used to formulate the basis of a consensus view on the measures required to manage sewage sludge on land in a safe and sustainable manner. The purpose of this report, therefore, is to give a holistic overview summarising the scientific understanding of the health and environmental effects associated with the practice to assist in the assessment of suitable, technically-based management practices and controls that ensure human health and the environment are protected when sewage sludge is recycled on farmland.

1.2 Aims and objectives

- To summarise the scientific evidence relating to the potential effects of recycling sewage sludge in agriculture on human health, soil quality and the environment.
- To review current sludge production within the EU and the extent of agricultural use in different countries and identify the main drivers influencing sludge management practices.
- To quantify the nature and extent of the contaminants present in contemporary sludges that may give rise to potential health and environmental concerns, and the trends in these concentrations with time.
- To objectively assess the protection and safety offered by the current regulatory framework for recycling sewage sludge in agriculture.
- To identify areas where human health and the environment may be potentially at risk when sewage sludge is recycled to farmland within the current regulatory controls and guidance.

2. SLUDGE PRODUCTION AND RECYCLING/DISPOSAL OUTLETS IN EUROPEAN COUNTRIES

Sludge production in individual EU Member States and the quantity recycled to agricultural land is presented in Table 1 for 1999/2000 (EC, 2003a). The total amount of sludge produced has increased in the past ten years in the EU, due to the introduction of new environmental legislation, and implementation of the Urban Wastewater Treatment Directive 91/271/EEC in particular (CEC, 1991). In England and Wales, for example, sludge production has increased by almost 40 % in the past 10 years owing to the expansion in wastewater treatment services to improve the quality of treated effluent discharges to the environment (Figure 1). In 1999/2000, approximately 7 million t DS were produced in the EU, of which approximately 40 %, almost 3 million t DS, was recycled in agriculture (EC, 2003a). However, the proportion recycled as a resource varies widely between different Member States (Table 2). Certain countries eg Belgium-Wallonia, Denmark, Spain, France, Ireland, the UK and Hungary, apply ≥50 % of the sludge generated on land (EC, 2006). In England and Wales, 73 % of sludge was used as an agricultural fertiliser in 2005 (Figure 1), almost doubling the amount recycled to land compared to 1996. The figure for the proportion of sludge recycled to land in the UK as a whole was equivalent to 64 % of the total production in 2004 (Table 2). In other Member States the recycling rate is much smaller and, in the period 2001-2003, Finland, Sweden and Slovenia applied <17 % of the sludge they generated on land. Greece, the Netherlands, Belgium (Flemish region), Slovakia and the Czech Republic spread very little, or no, sludge on agricultural land.

Agricultural application has been effectively prevented in some countries due to prohibitively stringent national limit values for heavy metals (eg the Netherlands, Belgium (Flemish region)). Alternatively, the land application of sludge may not be a developed practice, or there is no experience in agricultural utilisation, as is the case in Greece, for example. Incineration or landfilling are the main alternative methods to agricultural recycling for sludge management. Incineration may be the best practicable environmental option (BPEO) for sludge under certain circumstances, eg, when there is no land accessible for agricultural use, or the sludge is compromised for recycling perhaps due to large concentrations of contaminants. The objective of EU environmental policy is to reduce the disposal of biodegradable wastes in landfill and this is being implemented through the Landfill Directive 99/31/EC (CEU, 1999). Therefore, landfill is not considered a sustainable approach to sludge management in the long-term, and relatively little sludge is landfilled generally (Table 2), although some countries still depend heavily, or entirely on this outlet as a means of sludge disposal (eg Greece and Hungary – see Table 2). Consequently, most Member States dispose of a proportion of their sludge by incineration. In the UK, for example, 19 % of the total sludge production is disposed by incineration (and landfilling the residual ash), but only 1 % of sludge is currently disposed of in landfill (Figure 2). However, the amount of sludge that is incinerated significantly increases when recycling is constrained or prevented. In Belgium (Flanders), for instance, 76 % of sludge production was incinerated in 2005 (Table 2). In Austria, Denmark and Germany approximately 40 % of sludge is incinerated. The European Commission considers that the decline in sludge use in agriculture in favour of incineration is contrary to the waste hierarchy. Sludge incineration does not generate a surplus of energy as all the calorific value contained within the material is consumed in running the plant and due to the high energy inputs required to remove water and combust the material autothermically (Mininni *et al*., 1997; Thierbach and Hanssen, 2002). Therefore, sludge incineration should not be considered as a renewable energy source.

Table 1 **Sludge production and agricultural utilisation in EU Member States for 1999-2000 (EC, 2003a)**

Member State	Sludge production (t DS)	Sludge used in agriculture (t DS)	Sludge used in agriculture (%)	Year data recorded
Austria	401,867	40,455	10	2000
Belgium - Wallonia	18,228	10,733	59	2000
Belgium – Flemish[(1)]	80,708	0	0	2000
Germany	2,297,460	858,801	37	2000
Denmark	155,621	95,500	61	1999
Spain	853,482	454,251	53	2000
Greece	66,335	0	0	2000
France	855,000	552,000	65	1999
Finland	160,000	19,000	12	2000
Italy	779,220	217,805	28	2000
Ireland	35,039	14,109	40	2000
Luxembourg	7,000	5,600	80	1999
Netherlands[(2)]	242	36	15	1999
Portugal	238,680	37,176	16	2000
Sweden	220,000	35,000	16	2000
United Kingdom	1,066,176	584,233	55	2000
Total	7,235,058	2,924,699	40.4	

[(1)]No sludge from sewage treatment has been used in agriculture since December 1999 due to imposed national restrictions.
[(2)]No sludge from municipal treatment works has been used in agriculture since 1995 and the values relate only to sludge produced by private facilities.

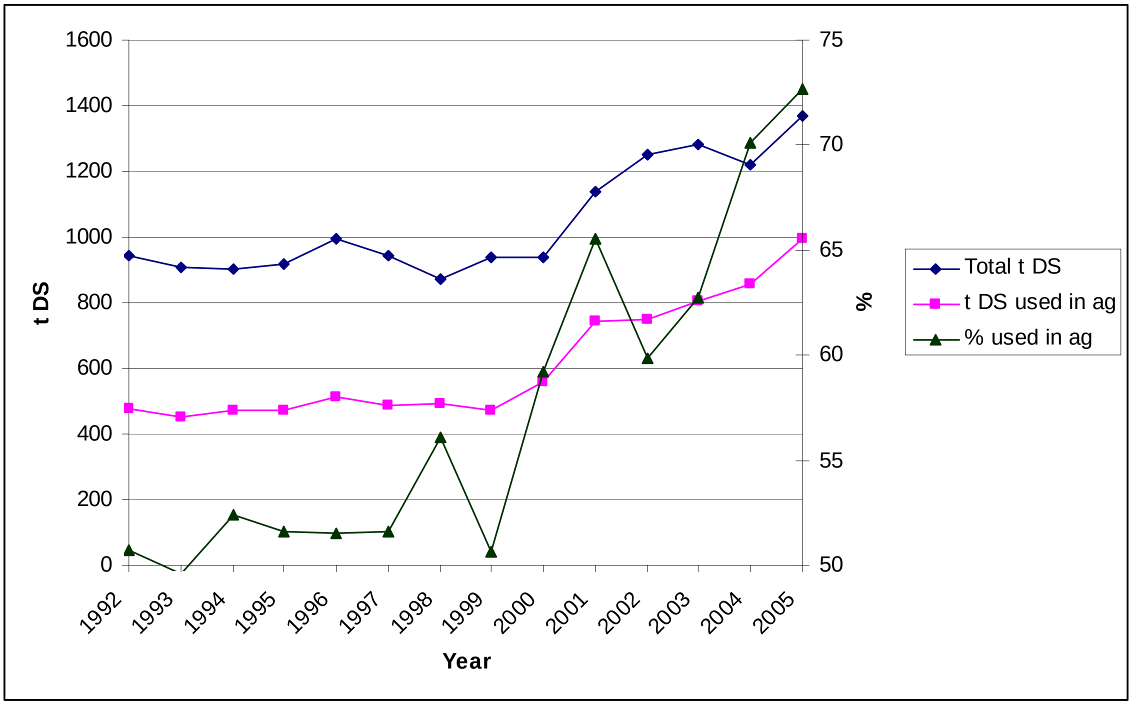

Figure 1 **Increase in sludge production in England and Wales and the amount used in agriculture (Defra, 2006)**

Table 2 Management routes for sewage sludge in EU Member States (Eureau, 2006)

Member State	Agriculture	Landfill	Incineration	Landfill cover	Land-scaping	Land reclamation	Other	Year of data
Austria	15	13	37				35	2005
Denmark [c]	55 [a]	2	43 [b]					2002
Finland	12	6		27	53			
Flanders	9		76	14				2005
Germany [d]	30	3	38		26		3	2003
Greece [e]		>90%						
Hungary	48	52 [f]						
Ireland	90						10	
Italy	32	37	8				22 [g]	
Luxembourg	47		20				33[h]	2004
Norway	62	2		7	12	14 [i]	3	2005
Poland [j]								
Sweden	10-15		2	10		60-65 [K]	10 [l]	
UK	64.1	1.1	19.4			11.0	4.4 [m]	2004

[a] *Agriculture* including sludge mineralisation plants, composting, long time-storage.
[b] *incineration*: including recovery, eg. cement or sand blasting agents (58% of incinerated sludge is recovered by alternative methods).
[c] There is a target for 2008 saying 50% recycling through agriculture, 45% incineration corresponding to 25% incineration with recycling of ashes in industrial processes and 20% "normal" incineration.
[d] Three of 16 federal states intend to stop agricultural sludge use.
[e] no specific figures supplied. Stated that most goes to landfill due to joint ownership of WWTP and landfills by municipalities but this now increasing problem.
[f] recent legislation regarding maximum water content of landfilled sludge could limit this outlet. Currently only about 3% of sludge meets the dry matter standard.
[g] including 19% which goes to composting, no final outlet given.
[h] 33% goes to composting, no final outlet given.
[i] listed in survey response as soil/aggregate production.
[j] no figures supplied, principle outlets listed as agriculture, composting, incineration, landfill.
[k] listed in survey response as construction soil.
[l] listed in survey response as vegetation material.
[m] including compost and industrial crops.

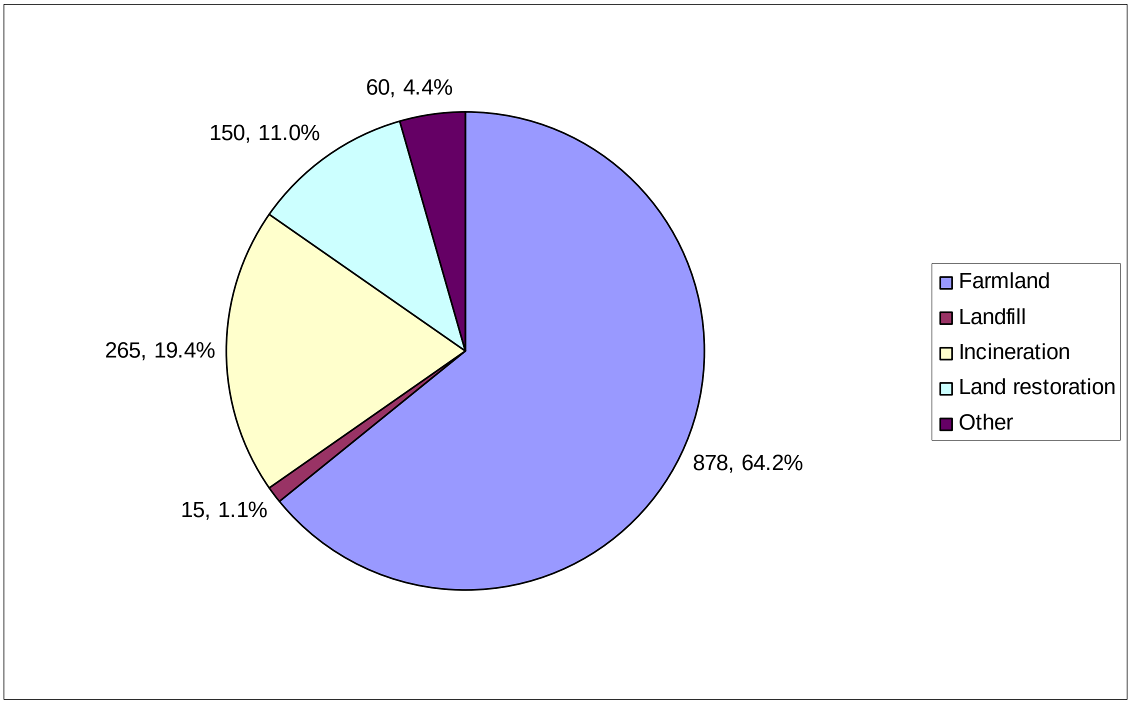

Figure 2 **The main outlets for sewage sludge management in the UK for 2004 (values denote t DS x 1000 and %) (Defra, 2006)**

3. SEWAGE SLUDGE TREATMENT

Sewage sludge is treated to enable it to be used in agriculture and this is done for three key reasons:

1. To stabilize the putrescible organic fraction to reduce nuisance and the potential offensiveness of sludge and particularly to reduce odour;
2. To significantly reduce or eliminate the potential content of infectious pathogenic organisms in sludge; in modern society with advanced treated water supply and foul water treatment the main organisms of concern are enteric bacteria and viruses and not helminths or protozoa;
3. To reduce vector attraction.

Article 7a of the Sludge Directive 86/278/EEC (CEC, 1986) states that sludge shall be treated before being used in agriculture. The use of untreated sludge may be authorized under specific conditions, although this practice has effectively ceased due to concerns about the potential risks to human health (eg RCEP, 1996). In the UK, land spreading of raw, untreated sludge to food crops was banned by the Safe Sludge Matrix from December 1999, and on land used to grow non-food crops from December 2005 (ADAS, 2001). Directive 86/278/EEC did not specify treatment processes for sludge other than a general definition that treated sludge should undergo biological, chemical or heat treatment, long-term storage or any other appropriate process so as to significantly reduce its fermentability and the health hazards associated with its use.

These overall requirements were interpreted and implemented within individual Member States according to local conditions and circumstances. For example, the UK developed a set of ‘effective sludge treatment processes’ that were published in a Code of Practice (DoE, 1989) at the same time as the Statutory Instrument (UK SI, 1989) implementing the Directive. In relation to point 2 above, the treatment processes were developed in particular to reduce the potential health hazards associated with salmonellae, the eggs of the human-beef tapeworm, *Taenia saginata*, potato cyst nematodes and a range of viruses that may be present in sludge (DoE, 1989, 1996). For a detailed review of the background to the research underpinning these recommendations see Carrington *et al*. (1998a). Recent detailed experimental investigations on the inactivation of pathogens and compliance of the UK Code of Practice (DoE, 1996) treatment conditions with numerical pathogen reduction requirements and limits in treated sludge sludge specified in the Safe Sludge Matrix (ADAS, 2001) are presented by Horan and Lowe (2002) and Horan *et al*. (2004). Further attention is given to this work in Sections 4 and 5 of the report.

There are a range of well established processes that are available for treating sludge and detailed reviews of the principles, engineering, economics and environmental aspects of these are provided by Frost *et al*. (1990) and Clark *et al*. (1999).

A brief description and assessment of some of the main treatment processes for sludge follows:

Mesophilic anaerobic digestion (MAD). The anaerobic digestion of sludge involves the microbiological degradation of the biodegradable organic matter contained in raw sludge in the absence of oxygen to produce a biogas comprising 50 % methane and carbon dioxide, that is valuable as a renewable source of energy. The stabilized sludge usually has no offensive odour. Digestion is usually performed in two stages to maximize pathogen removal. The first stage is in an enclosed vessel that is mixed and heated to 35 ^{O}C, with a retention time typically of 12-14 days. This is operated semi-continuously on a feed-draw basis. The secondary stage is either batch liquid storage typically for ≥14 days or storage of dewatered cake for several months is required to meet the reduction requirements for *Escherichia coli* stipulated in the Safe Sludge Matrix for agricultural use of conventionally treated sludge (2 $\log_{10}$ reduction and maximum number of 10^5 g^{-1} DS; ADAS, 2001; Sweet *et al*., 2001; Bellemaine and Bagnall, 2002; Horan *et al*., 2004).

Pasteurisation followed by MAD. This process effectively eliminates the pathogen content of sludge, by heating for a minimum period of 30 minutes at 70 ^oC or a minimum of 4 hours at 55 ^oC, and produces sludge that is compliant with the enhanced treatment status conditions stipulated in the Safe Sludge Matrix (6 log removal of *E. coli*, containing ≤10^3 *E. coli* g^{-1} DS and no *Salmonella* sp. in 2 g DS; ADAS, 2001; Sweet *et al*., 2001). Sludge is heated for the required time period and temperature in batches to ensure all the material is exposed to the necessary time-temperature conditions to eliminate pathogens. The biological phase by MAD is to ensure the organic matter content in the sludge is stabilized to prevent potential regrowth of pathogenic bacteria.

Thermophilic aerobic digestion (TAD). TAD produces an enhanced treated product for land application that is both pasteurized and stabilised. The minimum treatment requirements (DoE, 1996) are to maintain a minimum temperature of 55 ^oC for a period of at least 4 hours and have a total mean retention period of at least 7 days. The principle of TAD is that the sludge is aerated and aerobic microbes oxidize the organic matter and in so doing generate metabolic heat, which raises the temperature without an external heat source (as is required for MAD) typically to 60-70 ^oC, thus enabling pasteurization of the sludge.

Alkaline stabilization. The minimum recommended treatment requirements for alkaline stabilization are to raise the sludge pH to >pH 12 for a minimum period of 2 hours. This is generally done by blending quicklime to mechanically dewatered sludge (typically 25 % DS).

Composting. When dewatered sludge is blended with a suitable bulking agent (eg greenwaste, straw, woodchips), to reduce the moisture content to an optimum value of 60 % and increase the porosity, aerobic microbial activity occurs stabilizing the putrescible organic fraction and producing metabolically generated heat by the composting process. The temperature increases and is usually controlled at a optimal ceiling value of 55 ^oC. Composting may be undertaken in mechanically turned windrows usually in the open, or by using aerated static pile or invessel systems. Improved temperature control achieved by static piles or invessel techniques increases the effectiveness of pathogen inactivation compared to mechanical windrow turning, although all approaches can produce a sludge product compliant with stringent USEPA Class A (US EPA, 1993) and enhanced treated sludge (ADAS, 2001; Sweet *et al*., 2001) microbiological requirements (Pereira Neto *et al*., 1986).

Thermal drying. This process has become established as an enhanced treatment method for sludge. High temperature drying is performed using direct or indirect heating methods that raise the temperature of the sludge to 80 -140 ^oC, pasteurising the product and raising the DS content to 90 – 96 % (Donovan and Shea, 2004). The US EPA (1993) requirement to achieve Class A pathogen reduction status by drying is the sludge or exhaust gas should be heated to 80 ℃.

Thermal hydrolysis. In this process, sludge cake (12 % DS) is heated under pressure (7 – 10 bar) directly by steam injection to 130 - 180 ^oC (typically 165 ^oC) (Kepp *et al*., 1999a; Kepp *et al*., 1999b; Kopp and Ewert, 2006). The high temperature treatment destroys pathogens and breaksdown sludge organic matter increasing the digestibility and biogas production by MAD.

Treatment processes for sludge in the UK are managed according to the principles of HACCP (Hazard Analysis and Critical Control Point) (Water UK, 2004). HACCP applies risk management and control procedures to manage and reduce potential risks to human health and the environment. The approach has been adopted and applied to sludge treatment for agricultural application to provide reassurance that the microbiological requirements set out in the Safe Sludge Matrix are met by demonstrating that risk management and reduction combined with appropriate quality assurance procedures are in place and effective to prevent the use of sludge on farmland that does not comply with the microbiological standards.

4. CONTAMINANTS IN SEWAGE SLUDGE

4.1 Pathogens

4.1.1 Barriers to transmission and inactivation by sludge treatment processes

Sewage sludge is a product of urban wastewater treatment and can therefore potentially contain a large range of pathogenic enteric organisms discharged in the faeces of infected individuals. The principal pathogens of concern in sludge are listed in Table 3. The reality, however, is that the human population in a modern, industrialised society served by advanced water and wastewater services are usually healthy and are not normally hosts to pathogens and therefore carry very little enteric disease, and infections by parasitic protozoa and helminths are virtually absent. Thus, the numbers of potentially infective parasites in sewage are usually small (Bukhari *et al*., 1997; Robertson *et* al., 2000). Under these circumstances, the risk of infection from enteric bacteria and viruses are arguably the main issue concerning the agricultural use of sludge. Of the pathogenic bacteria of importance in sludge recycling, *Salmonella* spp. have historically been of the greatest concern in the UK, but only in respect of the risk to grazing animals (WHO, 1981). Surveys of *Salmonella* spp. in sludges in the UK showed that numbers were typically small or absent (Pike and Fernandes, 1981; Pike *et al.,* 1988). Whilst the recent work of Lang *et al*. (2002) and Lang *et al*. (2007) cannot be regarded as a survey of salmonellas in sludge, it does confirm that the organism is usually undetectable or only present in very small numbers in representative samples of mesophilic anaerobically digested (MAD) sludge containing typical numbers of the indicator bacteria *E. coli* that were compliant with the Safe Sludge Matrix. *Salmonella* spp. are ubiquitous in the environment, however, and proper control of human salmonellosis is most effectively achieved through food hygiene (Coker, 1983). Sewage sludge properly used in agriculture is not involved in the transmission of human salmonellosis (Coker, 1983) or transfers to grazing livestock (Jones, 1984; Evans, 1996). Indeed, the absence of pathogenic organisms from sludge under these circumstances poses a problem to researchers investigating the effectiveness of sludge treatment processes at pathogen destruction (Horan and Lowe, 2002), or there survival in the environment (Smith *et al*., 2006) and, consequently, target organisms under study are required to be artificially spiked into sludge. This is in contrast to farm livestock that are a significant reservoir of infectious enteric microorganisms, which are present in the manure of farm animals and therefore do represent a potential risk to human health (Nicholson *et al*., 2004; Hutchinson *et al*., 2005; Nicholson *et al*., 2005).

The potential risk to human health from pathogens in sludge is managed by either:

1. Adopting multiple barriers to pathogen transmission, or
2. Providing a single barrier to prevent human exposure

The multi-barrier approach to protecting human and animal health from pathogens that may potentially be contained in sludge for agricultural use has a well established scientific basis (WHO, 1981; Pike and Davis, 1984; US EPA, 1992a, 2003). It relies upon reducing the density of pathogenic organisms by sludge treatment (sludge meeting these criteria are synomynously described as Class B by US EPA (1993) or conventionally treated by the UK Safe Sludge Matrix (ADAS, 2001)) and also adopting land use restrictions. The restrictions on end-use, for example, prevent application to soil that could be used to grow crops that are in direct contact with sludge and could be consumed raw, or ‘ready-to-eat’. Following this approach, it is not necessary for the sludge treatment process to eliminate pathogens, but numbers are significantly reduced and the restrictions on how the treated sludge may be used are designed to allow the natural attenuation of the residual pathogens to take place in the environment. Time periods are set before sensitive crop types may be grown, for example, the Safe Sludge Matrix requires a period of 12 months to elapse before vegetables may be grown on land amended with conventionally treated sludge and salad crops cannot be grown for a period of 30 months after soil application. The plant species within the different crop type categories (fruit, salad – includes ready to eat crops, vegetables, horticultural, combinable and animal feed crops, grassland and forage) are specified to avoid ambiguity (ADAS, 2001). The scientific basis underpinning the requirements specified in the Safe Sludge Matrix can be found in Carrington *et al*. (1998b). The absence of any evidence linking the controlled use of sewage sludge with the outbreak of disease in the human/animal population provides strong justification for the barrier approach (RCEP, 1996). The Advisory Committee on Microbial Safety of Food (ACMSF) in the UK, an independent technical group of experts that advise government on the microbiological aspects of food safety, considered that sewage sludge applied to agricultural land in accordance with the requirements of the Safe Sludge Matrix should not present any unacceptable risks to food safety.

Table 3 Principal pathogens of concern in sewage sludge (Pepper *et al*., 2006)

Bacteria	**Protozoa**
Salmonella spp.	*Cryptosporidium*
Shigella spp.	*Entamoeba histolytica*
Yersinia	*Giardia lamblia*
Vibrio cholerae	*Balantidium coli*
Campylobacter jejuni	*Toxoplasma gondii*
Escherichia coli (pathogenic strains eg O157)	
Listeria spp.	
Enteric viruses	**Helminth worms**
Hepatitis A virus	*Ascaris lumbricoides*
Adenovirus	*Ascaris suum*
Norovirus	*Trichuris trichirua*
Sapporovirus	*Toxocara canis*
Rotavirus	*Taenia saginata*
Enteroviruses	*Taenia solium*
Polio viruses	*Necator americanus*
Coxsackie viruses	*Hymenolepis nana*
Echoviruses	
Enteroviruses 68-91	
Reoviruses	
Astroviruses	
Hepatitis E virus	
Picobirnavirus	

The barrier approach protects human health when sludge is used in the context of mechanised agriculture. However, there are circumstances where the provision of a single barrier is justified or necessary by treating sludge to virtually eliminate its potential pathogen content to background environmental values. This level of treatment is generally much more demanding in terms of engineering and resource requirements compared to conventional treatment processes (Section 3). Sludge achieving this extent of pathogen removal is synonymously described as Class A by US EPA (1993) or enhanced treated by the UK Safe Sludge Matrix (ADAS, 2001). The types of circumstances where this level of treatment and additional cost are justified may include, for example:

- Use in intensive cropping systems, eg horticulture;
- Amenity uses (eg golf course management);
- Where agricultural practices rely on manual labour so that farmers and workers come in direct contact with the material, as is often the case in developing countries (see Hall and Smith, 1998);
- Where the end-use cannot be controlled or guaranteed;
- Use in domestic gardens.

Prior to the introduction of the European Sludge Directive 86/278/EEC (CEC, 1986), many European countries, notably the Scandinavian countries, Germany, Switzerland and Italy had introduced national legislation controlling the treatment and use of sludge that was mostly restrictive and requiring active disinfection. In particular, much work on disinfection of sludge was carried out in Switzerland and Germany to identify processes which would yield sludges free from detectable pathogens. The main principle behind these treatment processes is the time-temperature exposure to permit protein denaturation and microbial kill, as shown in Figure 3. The ‘Safety zone’ lies above and to the right of the highest curve demonstrating non-detection of the most resistant pathogen in experimental studies. The operating conditions in the ‘Safety zone’ are those which will produce a demonstrably pathogen-free sludge.

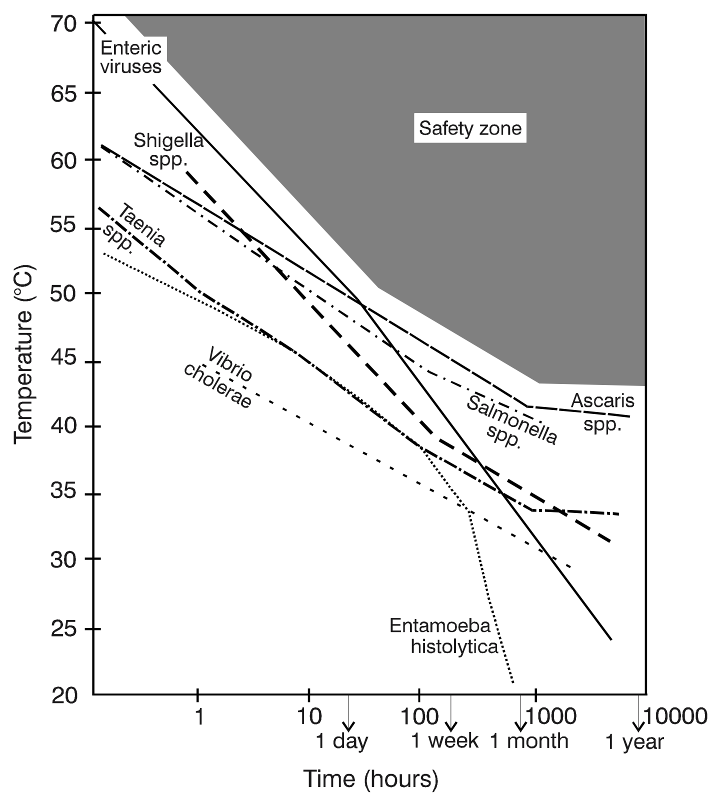

Figure 3 Time-temperature requirements to derive sludge which is virtually pathogen-free (Strauch, 1991)

The scientific principles behind the sludge treatment conditions specified in the UK (DoE, 1989, 1996) were based mainly on survival studies of *Salmonella* and the relatively resistant helminth organism, *Taenia saginata*, the human-beef tapeworm (Feachem *et al*., 1983;

Carrington *et al*., 1998a). The original work with *Taenia* reported by Pike (1986), in particular, was an elegant and comprehensive study involving the feeding of calves with cysts of *Taenia* variously exposed to sludge treatment processes and infectivity was determined directly by measuring the extent of cysticercosis infections in the animals. The relative effectiveness of the sludge treatments at *Taenia* removal was determined by calculating the ratio of cysts produced by treated and untreated eggs (Figure 4). The ranking of treatment processes emphasised the importance of two-stage anaerobic digestion to effectively reduce *Taenia* numbers in mesophilic anaerobically digested sludge (MAD). The relatively low removals during the first stage of MAD is explained due to the feed-draw nature of operation and potential for by-pass flow of untreated or only partially treated material. The second, unheated stage of the process is operated on a batch basis and this greatly improves inactivation rates for pathogens. Interestingly, the work also demonstrated the effectiveness of lagoon storage of liquid raw sludge at ambient temperatures at removing *Taenia*. Evidently, liquid sludge storage provides an aggressive environment towards pathogen survival and is effective at reducing pathogen viability. This can therefore provide the basis of an effective treatment strategy for sludge, coupled with air-drying for instance, in developing countries or where climate and space allow lagooning, storage and drying to be practiced (Hall and Smith, 1998).

Later research on the survival during sludge treatment of the emerging protozoan parasite, *Cryptosporidium parvum*, was performed in the 1990s (for example see, Whitmore and Robertson, 1995; Stadterman *et al*., 1995). Oocyst viability was shown to decrease by 81 - 88 % after 18 days exposure to mesophilic anaerobically digested sludge at 35 ^{o}C in the study by Whitmore and Robertson (1995), and complete inactivation occurred after 24 h by aerobic thermophilic digestion and pasterurisation at 55 ^{o}C. Stadterman *et al*. (1995) also reported that *C. parvum* oocysts exposed to anaerobically digested sludge at 37 ^{o}C disappeared at a fast rate and showed that 99.98 % of oocyts were inactivated after 24 h.

In practice, it is not feasible to monitor all the pathogens that could be potentially present in sludge, and because of the absence or only very small numbers actually present. Therefore, the effectiveness of a treatment process at reducing survival, should they be present, is determined by stipulating the appropriate operational parameters of temperature and process retention time (eg DoE 1996, or variant determined using HACCP (Water UK, 2004)), for instance, and also by measuring the reduction across the process and final total numbers in the treated product of suitable indicator species (Carrington, 2001). *Escherichia coli* is the organism most widely preferred and adopted for this purpose, due to its similarity in survival to vegetative bacteria, e.g. *Salmonella*, *Shigella*, *Listeria* and *Vibrio*, and also most viruses (Carrington, 2001; Lang and Smith, 2007a). Faecal streptococci (enterococci) are an alternative indicator organism, and are also marginally more resistant than *E. coli* and most pathogenic viruses and bacteria (Figure 5), although microbiological standards based on *E. coli* are regarded as precautionary and robust for protecting human health (US EPA, 2003). For instance, regular non-verotoxigenic *E. coli* has been shown to be an effective surrogate to monitor the survival of toxigenic *E. coli* O157 during sludge treatment and its decay in the environment (Horan *et al*., 2000; Smith *et al*., 2006).

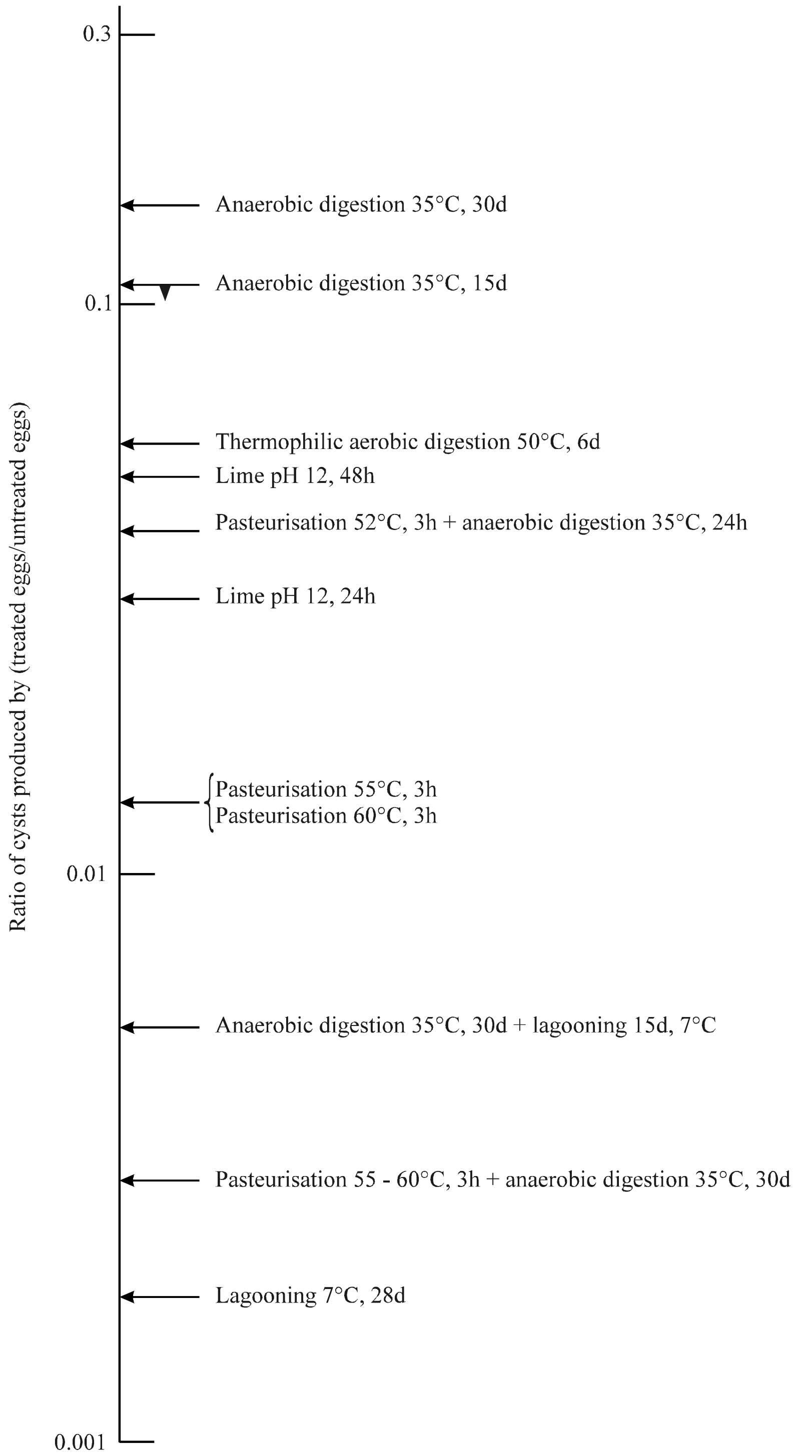

Figure 4 **Ranking of sludge treatment processes by their ability to reduce infectivity of *Taenia saginata* (Pike, 1986)**

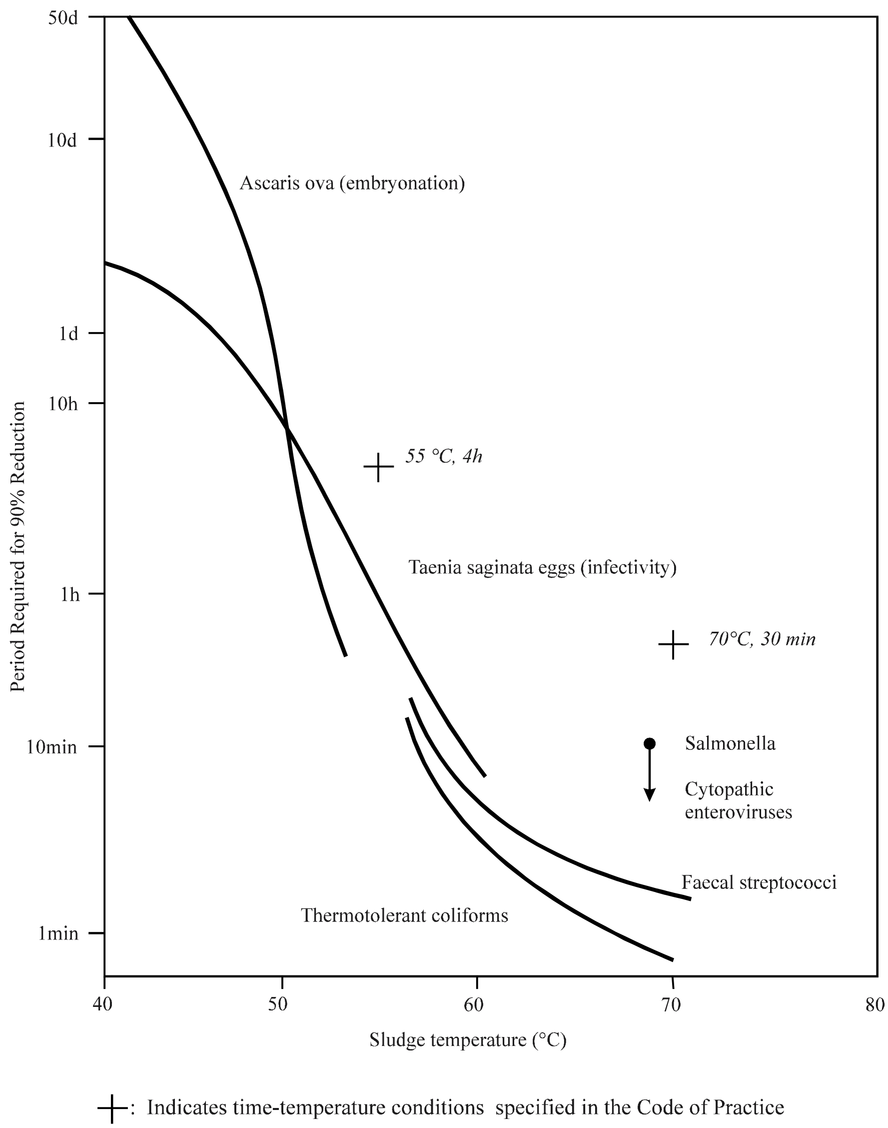

Figure 5 **Time-temperature conditions required to reduce the level of pathogens and Code of Practice conditions as specified in DoE (1996) (Pike and Carrington, 1986; Carrington *et al*., 1998a)**

Clostridium spp. (eg *C. perfringens*) are common in raw sludge and are more resistant to heat and other sludge treatment methods than vegetative bacteria. Langeland and Paulsrud (1985) indicated that numbers similar to those of *E. coli* were present in untreated sludge. Their destruction would confirm that spore bearing bacteria, eg *Bacillus* spp., and resistant ova, eg *Ascaris*, were destroyed during treatment. However, there are no reported studies on the effects of sludge treatment upon their survival. Carrington (2001) considered that appropriate indicators of pathogen removal by sludge treatment were, therefore, *E. coli*, and also *C. perfringens*, and that maximum numbers of *E. coli* of 1000 g^{-1} of sludge DS and a tentative maximum concentration of *C. perfringens* spores of 3000 g^{-1} of sludge DS, would indicate almost complete pathogen destruction. The basis of the microbiological limit value of 1000 *E. coli* g^{-1} DS can be traced to the work of Yanko (1987) and Farrell (1993), which show that this number of faecal coliforms correlates with a very low level of *Salmonella* spp.

detection in composted sludge (US EPA, 1992a). However, as intestinal parasite infections are rare in the human population where high standards of hygiene operate, and growth of infectious *Bacillus* spp., eg *B. botulinum*, is not favoured under nutrient elutriated conditions in urban wastewater treatment systems there would appear to be no justification to monitor *Clostridium* routinely as an indicator organism for sludge.

Research on the survival of enteric indicator and pathogenic bacteria, viruses and the protozoa, *Cryptosporidium* and *Giardia* during sludge treatment has been completed recently (Horan *et al*., 2000; Horan and Lowe, 2002; Horan *et al.*, 2004; also see Lang and Smith, 2007a), to further underpin the process conditions for sludge treatment specified in the current UK *Code of Practice for Agricultural Use of Sewage Sludge* (Figure 5; Pike and Carrington, 1986; DoE, 1996; Carrington *et al*., 1998a). Although these recommendations were in existence long before the Safe Sludge Matrix (ADAS, 2001), it was necessary to demonstrate that the treatment conditions given in the Code, and new processes introduced since the Code was published (eg thermal drying), were effective at reducing pathogens to the new numerical limits required by the Matrix. A summary of the results from this work is presented in Table 4 and show removals were in excess of the microbiological limits (Enhanced treated: 6 log removal of *E. coli* and $\leq 10^3$ *E. coli* g^{-1} DS, no *Salmonella* spp. in 2 g DS; Conventionally treated: 2 $\log_{10}$ reduction and $\leq 10^5$ g^{-1} DS of *E. coli*). Horan *et al*. (2004) concluded that two-stage mesophilic anaerobic digestion (MAD) is able to achieve >2 $\log_{10}$ removal of *E. coli*, *Listeria monocytogenes* and *Salmonella senftenberg*, but not *Campylobacter jejuni*. Nevertheless, for all bacterial pathogens evaluated, die-off by MAD was in excess of the numbers routinely encountered in sludge. However, the mechanisms of pathogen removal during MAD are poorly understood and it would benefit the Water Industry greatly in terms of optimising digester efficiency for pathogen destruction as well as sludge stabilisation, if a programme of fundamental research was performed in this area (Smith *et al*., 2005).

Recent research has demonstrated significant reductions in concentrations of *Cryptosporidium parvum* and *Cryptosporidium hominis* oocysts, *Giardia lamblia* cysts, and spores of human-virulent microsporidia (*Enterocytozoon bieneusi* and *Encephalitozoon intestinalis*) occur in dewatered, activated sewage sludge cake endproducts compared to those of the respective pathogens in corresponding samples collected during the activated sludge process (Graczyk *et al*., 2007). This demonstrates the aggressive nature of sludge from biological wastewater treatment processes towards reducing pathogen survival (eg Curds and Fey, 1969). However, it should be emphasized that sludge produced in this way would not be regarded as 'treated' sludge suitable for land application and the product would have to undergo further treatment to be compliant with the pathogen reduction and other requirements stipulated for agricultural use (US EPA, 1993; ADAS, 2001).

Thus, in conclusion, the effects of sludge treatment on pathogen removal have been well characterized and defined by laboratory and operational process monitoring investigations. These show that specified treatment process conditions are effective at reducing pathogen densities and thus prevent the transmission of infectious diseases within a risk management system based on multi-barriers. Alternatively, treatment options are also available that provide a complete barrier by essentially eliminating the pathogen content of sludge. From a public health control perspective, however, it is emphasized that there is no technical justification for adopting this high level of treatment where sewage sludge is applied for general agricultural use; under this type of end-use multi-barriers are effective at controlling the spread of infectious disease to protect human health. The research has also confirmed that *E. coli* remains a good choice of indicator for other enteric bacteria and viruses in sludge, which are the principal organisms of concern in context of Western European society.

Table 4 Summary of observed decimal (Log) inactivation in numbers of organisms (per g DS) for sewage sludge treatment processes (Horan and Lowe, 2002)

Organism	Process						
	MAD	Pasteurisation + MAD		Lime Stabilization	Composting**		Thermal drying (benchmarking only)
		70°C/30 min	55°C/240 min		55°C/4h	40°C/5 days	
Escherichia coli	3.38 3.36	8.33* 7.48*	8.33* 7.48*	4.76* 4.35*	6.18*	6.18*	7.26
Listeria monocytogenes	2.23† 2.23	9.00* 7.24*	9.00* 3.12*	6.75*	3.05 3.15	2.50 2.38	
Campylobacter jejuni	0.36 0.32	8.22* 5.31*	5.31* 3.82	7.49* 7.23*	5.72* 5.68*	5.72* 5.68*	
Salmonella senftenberg	4.33 4.03	7.97* 7.81*	7.97* 5.74	7.95* 6.11* 4.71	2.39	2.09	
Salmonella typhimurium	NP	9.33* 6.72* 6.12*	6.72* 6.12*	9.67* 8.75*	NP	NP	
Salmonella enteriditis	NP	8.24*	8.24*	NP	5.68*	5.68*	
Salmonella dublin	NP	NP	NP	7.58* 6.84*	5.58*	5.58*	
Poliovirus	4.49 4.43	8.43* 8.17*	8.43* 8.41*	6.82 6.50	7.85*	7.85*	
Cryptosporidium	3.20 2.15	Viability reduced by up to 97 % by pasteurisation and then to zero within 1 day by MAD		1.98 0.00	NP	NP	

*Complete removal
NP = Not performed

**laboratory-scale process under temperature controlled conditions
† Primary sludge digestion only

Nevertheless, continued vigilance and research is necessary to ensure that the specified process conditions and microbiological indicators are sufficiently robust to accommodate any new and emerging organisms that may be identified as a potential risk to human health in future. However, in overall assessment, the current technical understanding of pathogen behaviour and the engineered treatment processes available for sludge provide an effective and robust basis for managing the possible risks to health from enteric diseases when sludge is used in agriculture, should there be any pathogens present in the sludge. This level of control is also accompanied with appropriate monitoring and quality assurance systems to demonstrate that the barriers to pathogen transmission are in place and work effectively. Assurance that only microbiologically compliant sludge is used for agricultural purposes is provided, for example, by the introduction of HACCP management systems for sludge treatment processes (Water UK, 2004; Westrell *et al*., 2004).

4.1.2 Regrowth and effects of sludge conditioning on recovery of indicator bacteria

Two other issues relating to pathogens that require consideration are the potential regrowth of enteric bacteria in treated sludge (Zaleski *et al*., 2005) and also the apparent increase in numbers of the indicator bacteria, *E. coli*, after mechanically dewatering sludge by centrifugation (Qi *et al*., 2004; Higgins *et al*., 2007; WERF, 2007). One of the purposes of treating sludge is to stabilize the organic matter fraction so that it is less available for microbial growth to restrict the potential for regrowth and recolonisation by enteric bacteria (Pike and Davis, 1984). In some circumstances, regrowth of enterics, such as *Salmonella* has been observed in composted sludge, but this is unlikely when the treatment process is well managed, external sources of contamination, eg from faeces of wild animals and birds, are restricted, and the organic residue supports an active suppressive and predatory microbial population (Zaleski *et al*., 2005).

Significant increases in *E. coli* numbers, as much as 1 - 2 $\log_{10}$ g^{-1} DS, have been measured in digested sludge samples collected immediately after centrifugation (Cheung *et al*., 2003; Qi *et al*., 2004) and larger increases have been measured after centrifuge dewatering of prepasteurised digested sludge (for a review of recent research, see Panter (2007)). This has potentially important implications for compliance with the *E. coli* reduction requirements for conventional and enhanced treated sludge (EC, 2000; ADAS, 2001) and, by inference, it implies that numbers of human pathogens may also increase sludge. Storage of the dewatered cake for periods of up to 120 days is therefore usually employed to reduce *E. coli* numbers to the maximum limit value of 10^5 g^{-1} DS permitted for agricultural use of conventionally-treated sludge in the UK (Bellemaine and Bagnall, 2002). By contrast, Cooper *et al*. (2005) found that other passive mechanical dewatering methods eg belt press, did not increase *E. coli* numbers and there was only minimal effect of centrifuging liquid digested sludge after secondary digestion on bacterial counts. Reactivation of bacterial cells, ie changing from a viable but non-culturable (VBNC) to a culturable state, following exposure to extreme physical conditions in the dewatering centrifuge is suggested as a possible mechanism for the apparent increases in *E. coli* observed after centrifugation (Qi *et al.*, 2004; WERF, 2007). However, this does not seem to be an entirely plausible explanation as it implies that current culture based enumeration techniques may only measure about 1 % of the total population of enteric bacteria in environmental matrices and there is no evidence indicating that this is the case. An alternative explanation for the increased counts measured after centrifugation may be that cell colonies in primary digested sludge are disrupted by exposure to shear forces in the centrifuge (Cooper *et al*., 2005). To test this, Cheung *et al.* (2003) and Qi *et al*. (2004) examined the effects of increased shear and severity of sample disruption on enumeration of *E. coli* in digested sludge in the laboratory by a combination of blending, extended stomaching times, surfactant addition, shaking and sonification, but found that none of them had any effect on bacterial enumeration. However, these techniques do not simulate all the conditions existing in a dewatering centrifuge and further controlled experiments are required to examine the effects of shear forces on colony behaviour in digested sludge to provide a more robust explanation of the processes and mechanisms involved. Cooper *et al*. (2005) suggested that, after secondary batch storage of liquid digested sludge, which reduces the numbers of *E. coli*, it is possible that colony structure is also degraded so there is apparently a much smaller impact of exposure to shear in the centrifuge on bacterial counts for secondary digested sludge. The reactivation observed for prepasteurised sludge implies that the pasteurisation conditions usually adopted (70 ^{o}C for 30 minutes) may stress the indicator organism so it enters a VBNC state, but is not sufficient to eliminate them. Thus, the implications of the pasteurisation time-temperature exposure on the viability of *E. coli* should be further investigated. The mechanisms responsible for the observed increases in *E. coli* numbers after centrifuge dewatering are uncertain and further research in this area is required. However, this phenomena is unlikely to have significant microbiological implications for human health, but it is a critical operational issue to enable compliance with the microbiological standards for sludge. Sludge management and HACCP practices (Water UK, 2004) ensure that sludge microbiological quality meets the standard for agricultural application. If *E. coli* numbers increase as a result of centrifuge dewatering then additional measures (for instance sludge storage) are put in place to ensure sludge is compliant with the numerical limits on *E. coli* for use in agriculture (Bellemaine and Bagnall, 2002).

4.2 Potentially toxic elements

4.2.1 PTE content in sludge

Sewage sludges always contain potentially toxic elements (PTEs), including heavy metals, even if of entirely domestic origin. Non-point, diffuse sources, including those of domestic origin, and surface run-off contribute to the background concentrations of heavy metals in sludge and, in many cases, represent the main inputs of metals to sewer (Comber and Gunn, 1994; Bowen *et al*., 2003). Heavy metals are ubiquitous and occur naturally in the environment so it is inevitable and normal to find them in sludge. Heavy metals are a normal constituent of all types of food stuffs and are present in human faeces. For example, human faecal matter is reported to contain: 250 mg Zn kg^{-1}, 68 mg Cu kg^{-1}, 4.7 mg Ni kg^{-1}, 2 mg Cd kg^{-1} and 11 mg Pb kg^{-1} (IC Consultants, 2001). Important domestic inputs include leaching from plumbing systems (a significant source of Cu, Zn and Pb) and use in body care products (Zn is used in anti-dandruff shampoo and skin care products, for example). The other major source of heavy metals in sludge originates from direct discharges by industry connected to the sewer. However, these sources have declined drastically due to environmental legislation controlling the use and discharge to the environment of dangerous substances, such as Cd, within the EU (eg CEC, 1976; CEC, 1978), through active trade effluent control measures implemented by the Water Industry, and improved industrial practices leading to more efficient use of chemicals in industrial processes (Goodenough, 2006). Cadmium, Pb and Hg are designated Priority Hazardous substances under the Water Framework Directive (WFD) 2000/60/EC (EPCEU, 2000a), and so will be subject to further measures leading to the cessation or phasing out of discharges, emissions and losses of these PTEs to the environment as far as possible. This indirectly benefits sludge quality by reducing their concentrations in sludge.

The metals content of sludge is important in defining its quality and suitability for agricultural use and controls to restrict industrial and other discharges as far as practicable are a priority for the long-term sustainability and security of the outlet. Where trade effluent controls are rigorously enforced, metal concentrations in sludge have fallen markedly (Table 5, Figure 6 and 7). This has been so effective at improving sludge quality that, in the EU and UK, the mean Cd content in sludge applied to farmland reflects the background content discharged to sewer in faeces. As industrial discharges decline, diffuse inputs of PTEs from domestic sources, small-to-medium sized enterprises and institutions such as hospitals become more significant, but these are also more difficult to identify and control. Detailed reviews of the scientific literature and recent monitoring data of sources and inputs of metals to urban wastewater and sludge can be found in IC Consultants (2001), Bowen *et al*. (2003) and Rule *et al*. (2006).

Table 5 **Decline in content of potentially toxic elements (PTE) (works size weighted mean, mg kg^{-1} ds) in sewage sludge recycled to agricultural land in the UK from published surveys ([1]Sleeman, 1984; [2]DoE, 1993; [3]EA, 1999)**

PTE	[1]1983	[2]1990	[3]1996	Reduction in 1996 relative to 1983 (%)
Zn	1319	922	792	40
Cu	703	574	568	19
Ni	107	65	57	47
Cd	14	5.0	3.3	76
Pb	462	201	221	52
Hg	5.0	3.5	2.4	52
Cr	312	208	157	50

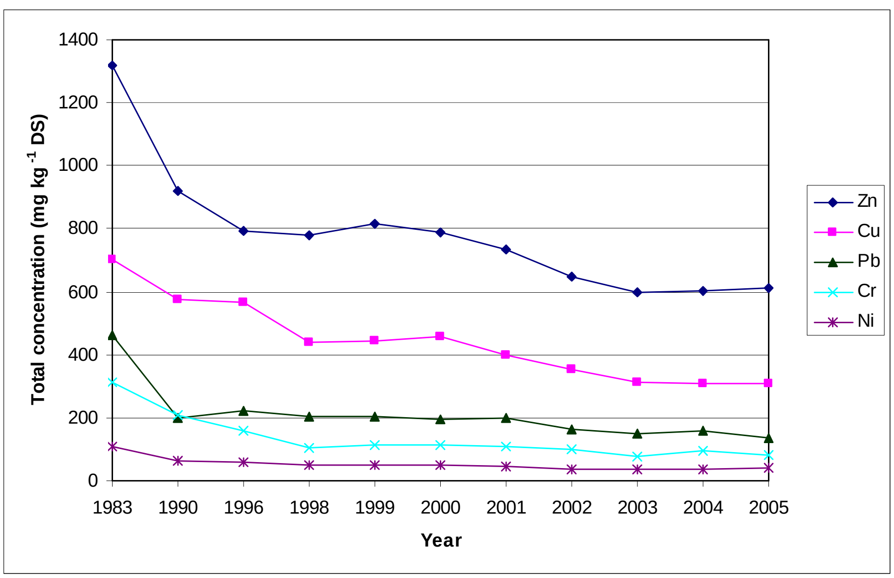

Figure 6 **Decline in mean total concentrations of Zn, Cu, Pb, Cr and Ni in sludge recycled to agricultural land in England and Wales (Note: 1983 - 2000 2000 are UK mean data) (pers. com. V. Sturt, Environment Agency)**

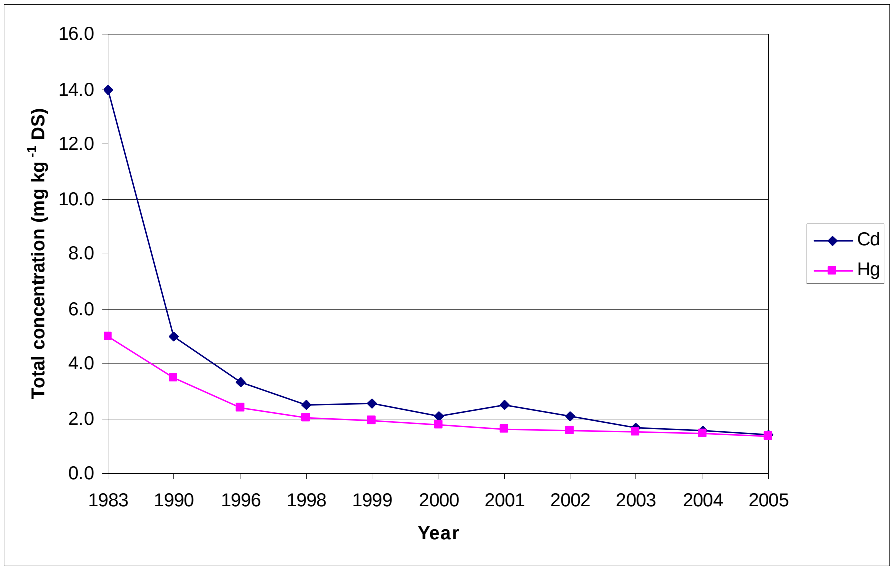

Figure 7 **Decline in mean total concentrations of Cd and Hg in sludge recycled to agricultural land in England and Wales (Note: 1983 - 2000 are UK mean data) (pers. com. V. Sturt, Environment Agency, UK)**

Nevertheless, whilst recognising the very significant progress that has been, the range of metal concentrations present in sludge (Table 6) suggest there remain sources of metal inputs that could be further reduced. Reporting to the European Commission on *Pollutants in Urban Wastewater and Sewage Sludge*, IC Consultants (2001) identified a number of opportunities to lower metal concentrations in sludge further, particularly from small to medium sized enterprises, as these inputs can be potentially significant (Smith and Koonlinthip, 2001). It must be stressed that continued vigilance in identifying and reducing these potential sources of metals as far as practicable to the wastewater collection system is vital for the long-term environmental, as well the operational, sustainability of recycling sewage sludge on agricultural soil.

Table 6 **Comparison of PTE concentrations in sludge from medium sized sewage treatment works in the UK (Smith and Koonlinthip, 2001)**

PTE	10 %ile (mg kg^{-1} ds)	50 %ile (mg kg^{-1} ds)	10 %ile, increase rel to 10 %ile (%)	90 %ile (mg kg^{-1} ds)	90 %ile, increase rel to 10 %ile
Zn	379	639	0.7	1156	2.0
Cu	203	405	1.0	810	3.0
Ni	14	27	0.9	93	5.6
Cd	0.7	2.0	1.9	3.8	4.4

4.2.2 Platinum group metals

Platinum and Pd are components of vehicle catalytic converters and emissions occur as the autocatalyst deteriorates. Catalytic converters are the main emission source of these metals in the environment and releases have increased with the expansion in use of autocatalysts. Platinum group metals potentially enter urban wastewater in run-off and transfer to sewage sludge in a similar way to other heavy metals. Thus, in contrast to the majority of other PTEs in sludge, whose concentrations are decreasing, those of the Pt group elements have shown an upward trend. The Pt content in sludge is typically in the range 0.1 – 0.3 mg kg^{-1} (DS) and the background value for soil is 1 $\mu g\ kg^{-1}$ (IC Consultants, 2001). From a recent review of literature, IC Consultants (2001) concluded that Pt group metals, emitted as autocatalyst particles, are probably inactive and immobile in soil and are unlikely to present a risk to human health or soil quality from the agricultural use of sludge.

4.2.3 Technical basis to regulating PTEs in sewage sludge-treated soil

A characteristic of heavy metals is that, once applied to the soil, they are effectively retained by the soil matrix indefinitely in the cultivated layer. Therefore, metals tend to accumulate when the concentration in a material that is applied to the soil is greater than the background amount present in the soil itself. This has implications for the long-term quality of sludge-treated soil for crop production, the foodchain and soil microbial activity. Therefore, controls are in place to ensure that PTE inputs are well below upper tolerable toxicological and ecotoxicological thresholds to protect sensitive environmental end-points and human health. The critical environmental end-points for setting limits for agricultural recycling sewage sludge vary according to the behaviour of particular elements and include (Smith, 1996):

- Phytotoxicity (Zn, Cu, Ni; Cr may also be listed here, but there is no evidence of crop damage due to Cr when sludge is used in agriculture);
- Human foodchain *via* crop uptake (Cd);
- Human foodchain *via* offal meat from animals ingesting sewage sludge or sludge-treated soil (Cd and Pb);
- Animal health (Cu, As, Se, Mo and F);
- Soil fertility (Zn).

Some PTEs are beneficial as trace elements such as Cu and Zn, whereas others have no known biological function. The environment can safely tolerate accumulations of PTEs up to certain limits before there is the potential to cause an adverse effect. The scientific approach to controlling PTEs in sludge-treated soil has been based on the quantitative toxicological/ecotoxicological dose-response relationship of the most sensitive environmental receptor to a particular element and establishes a limit value below the Lowest Observed Adverse Effect Concentration - LOAEC that allows for a further margin of safety (Figure 8).

The numerical standards for PTEs adopted in different countries vary considerably (Table 7) and reflect a political decision to adopt one of the following strategies, with increasingly limiting implications for land application of sludge:

- Risk/technical basis (US EPA, 1993; Chang *et al*., 2002);
- Technically based, but precautionary (UK SI, 1989; DoE, 1989; upper limits in Directive 86/278/EEC (CEC, 1986));
- No technical basis, highly precautionary.

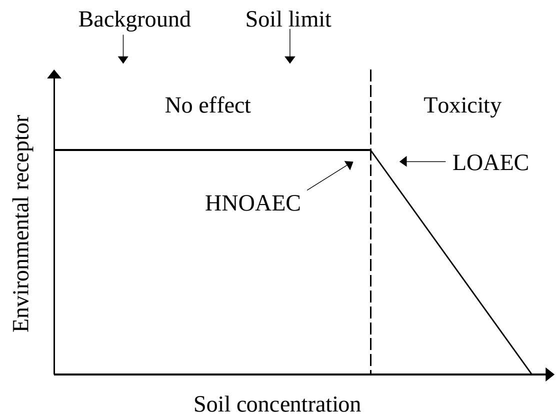

Figure 8 **Defining concentration limits for PTEs in soil (HNOAEC = Highest No Observed Adverse Effect Concentration; LOAEC = Lowest Observed Adverse Effect Concentration)**

Table 7 **Maximum permissible concentrations of potentially toxic elements in sludge-treated soils (mg kg^{-1} dry soil) in EC Member States and US**

PTE	EC	UK[(1)]	Germany	Denmark	US[(2)]
Zn	150-300	200	200	100	1500
Cu	50-140	135	60	40	775
Ni	30-75	75	50	15	230
Cd	1-3	3	1.5	0.5	20
Pb	50-300	300[(3)]	100	40	190
Hg	1-1.5	1	1	0.5	9
Cr	100-150[(4)]	400[(5)]	100	30	1540

[(1)] For soil of pH ≥5.0, except Cu and Ni are for pH range 6.0 – 7.0; above pH 7.0 Zn = 300 mg kg^{-1} ds (DoE, 1996);
[(2)] Approximate values calculated from the cumulative pollutant loading rates from Final Part 503 Rule (US, EPA 1993);
[(3)] Reduction to 200 mg kg^{-1} proposed as a precautionary measure, see Section 5.2.3;
[(4)] EC (1990) – proposed but not adopted;
[(5)] Provisional value (DoE ,1989);
For some elements smaller concentrations may be set a low soil pH value.

4.3 <u>Organic contaminants</u>

4.3.1 Occurrence in sewage sludge

The range of organic contaminants (OCs) known to exist in sludge is extensive and diverse and the major groups and their concentration ranges and properties are listed in Table 8 and a recent list of concentration data, compiled by Smith and Riddell-Black (2007) is presented in Table 9. For example, Drescher-Kaden *et al*. (1992) reported that 332 organic substances, with the potential to exert a health or environmental hazard, had been identified in German sludges and 42 of these were regularly detected in sludge. Compared with the 10 PTEs of concern in sludge, which are routinely monitored and controlled, regulating this diverse range of organic contaminants would appear to present a much more daunting prospect.

The compounds entering sludge may be generally categorised as follows (IC Consultants, 2001):

- Persistent compounds from incomplete combustion of fossil fuels that enter the urban wastewater (UWW) collection system through deposition onto paved surfaces via run-off (eg PAHs and PCDD/Fs);
- Persistent compounds that are associated impurities in wood preservatives such as creosote (PAHs) and pentachlorophenol (PCDD/Fs) and enter UWW in run-off;
- Controlled persistent compounds mobilised by volatisation from soil, deposition and transfer to UWW in run-off (eg PAHs, PCBs and PCDD/Fs);
- Persistent compounds generated by cooking food that are discharged from domestic sources (eg PAHs);
- Persistent compounds that are prohibited from use/manufacture, but domestic sources may exist and can transfer to UWW via run-off (eg chlorinated pesticides);
- Compounds discharged to sewer used directly in industrial processes or domestically including solvents, flame retardents or compounds that leach from plastics and surfaces during end-use and are carried in run-off (eg DEHP, PBDEs);
- Detergent residues (eg LAS, NPE);
- Pharmaceuticals, antibiotics, endogenous hormones and synthetic steroids;
- Compounds from the various above groups with endocrine disrupting potential.

A detailed overview of the significance of OCs in urban wastewater and sludge was undertaken by IC Consultants (2001), on behalf of DG Environment and a principal output from that work was an assessment of the significance of OCs and of potential opportunities to reduce contamination and this is shown in Table 10.

4.3.2 Approaches to regulating organic contaminants in sludge

In contrast to the general agreement about the specific PTEs of concern in sludge requiring controls and regulation, there is no consistent approach to the statutory measures on OCs in sludge between different countries (Table 11). This is mainly because the increasing level of investigation in recent years has not identified the ecotoxicological significance of organic contaminants in the soil-plant-water system and in the food chain (Sauerbeck and Leschber, 1992). European regulations on polyhalgenated compounds, for example, have therefore set highly precautionary values which have no toxicological basis (Sauerbeck and Leschber, 1992). Some countries, such as the UK, US and Canada, have therefore argued that there is no technical justification for setting limits on OCs in sludge, on the basis that research has shown that the concentrations present are not hazardous to soil quality, human health or the environment (US EPA, 1992b,c; WEAO, 2001; Blackmore *et al*., 2006). The evidence base on the impacts of OCs on human health and the environment are discussed further in the following sections. However, other countries have established limits for different groups of OCs. For example, in Germany, limits are set for the persistent compounds: PCBs and PCDD/Fs, but not PAHs, whereas France regulates PAHs and PCBs, but not PCDD/Fs. Denmark, on the other hand has established controls for the bulk volume chemicals including DEHP, LAS and NP/NPE. Even the European Commission has had different opinions about which compounds to regulate and limit values to adopt in the proposals to revise Directive

Table 8 Properties, occurrence, degradation and transfer of organic contaminant groups found in sewage sludge compared to data of biowaste and greenwaste compost (Smith, 1996, quoted from Arthur Andersen on behalf of DG Environment, 2001; cited by Amlinger *et al*., 2004)

Compound Group	Physico-chemical properties	Conc. in compost	Conc. in sludge	Degradation	Leaching Potent.	Plant uptake	Transfer to animals
Polynuclear aromatic hydrocarbons (PAHs)	Water soluble/volatile to lipophilic	BWC: 0.8-4.5 mg kg^{-1} GC: 1.7-3.8 mg kg^{-1}	1-10 mg kg^{-1}	Weeks → 16 years Strongly adsorbed by soil O.M.	None	Very poor	Possible but rapidly metabolised
Polychlorinated Biphenyls (PCBs)	Complex, > 200 congeners low water solubility, highly lipophilic and semi-volatile	BWC: 0.03-0.86 mg kg^{-1} GC: 0.015-0.48 mg kg^{-1} mixed BWC/MC: 0.02-1.68 mg kg^{-1}	1-20 mg kg^{-1}	Very persistent Half-life ca. 12 years Strongly adsorbed by soil O.M.	None	Root retention Foliar absorption Minimal root uptake and translocation	Possible into milk/tissues via soil ingestion Long half-life
Polychlorinated dibenzo-P-dioxins and Furans (PCDD/Fs)	Complex, 75 PCDD congeners, 135 PCDF congeners, Low water solubility, highly lipophilic and semi-volatile	BWC: 4.3-17.5 ng I-Teq kg^{-1} GC: 3.6-13 ng I-Teq kg^{-1}	Very low <few µg kg^{-1}	Very persistent Half life several years Strongly adsorbed by soil O.M.	None	Root retention Foliar absorption Minimal root uptake and translocation	Possible into milk/tissues Via soil ingestion Long half-life
Phtalate acid esters	Generally lipophilic, hydrophobic and non-volatile	DEHP: 0–69.6 mg kg^{-1} Means: BWC: 1.4–9.6 mg kg^{-1} GC: 0.4–2.9 mg kg^{-1}	High 1-100 mg kg^{-1}	Rapid Half-life <50 days	None	Root retention Not translocated	Very limited
Linear alkybenzene-solphonates (LAS)	Lipophilic	Only one investigation > 0: 9–396 mg kg^{-1} Mean: 41 mg kg^{-1}	Very high 50-15000 mg kg^{-1}	Very rapid in aerobic environment	None	None	None
Alkylphenols	Lipophilic	~ 200 µg kg^{-1}	100-3,000 mg kg^{-1}	Rapid < 10 days	None	Minimal	Minimal
Organoclorine pesticides	Varied, lipophilic to hydrophilic, some volatile	0.6 – 100 µg kg^{-1}	<Few mg kg^{-1}	Slow> 1 year Loss by volatilisation	None	Root retention Translocation not important Foliar absorption	Via soil ingestion persistent in tissues
Monocyclic aromatics	Water soluble and volatile	n.i.	<1-10 mg kg^{-1}	Rapid	Moderate	Limited due to low persistence Rapidly metabolised	Rapidly metabolised
Chlorobenzenes	Water soluble/volatile to lipophilic	HCB 0-156 µg kg^{-1}	<0.1-50 mg kg^{-1}	Lower mol wt types lost by volatilisation Higher mol wt types persistent	High to low	Possible via roots and foliage Maybe metabolised	Important for persistent compounds
Short-chained halogenated aliphatics	Water soluble and volatile	n.i.	0-5 mg kg^{-1}	Lower mol wt types lost by volatilisation Higher mol wt types persistent	Moderate	Foliar absorption Possible translocation	Low
Aromatic and alkyl amines	Water soluble and low volatility	n.i.	0-1 mg kg^{-1}	Slow	High	Possible	Low
Phenol	Varied, lipophilic high water solubility and volatile	n.i.	0-5 mg kg^{-1}	Rapid	Moderate to low	Possible via roots and foliage	None

BWC…Biowaste Compost (including source separated organic household/kitchen waste); GC…Green Compost (from garden and park waste only); n.i. … not identified in literature

Table 9 **Summary of organic compounds found in sewage sludge in the UK (mg kg^{-1} DS except where indicated) (see Smith and Riddell-Black (2007) for full inventory of data)**

Substance	Minimum	Maximum	Mean
Non-halogenated monocyclic aromatics	0.025	22.1	2.29
Monocyclic aromatic (chloro- and nitro-anilines)	0.0001	192000	125.83
Monocyclic aromatics	0.004	2.6	0.1427
Alkyl and aromatic amines/imines	0.01	3.8	1.34
Organotin	0.01	1.3	0.36
Halogenated aliphatics	0.00002	16.6	1.0178
Carbonyl	1.4	23	10.1
∑Polycyclic aromatic hydrocarbons	6.4	72	130
∑Polychlorinated biphenyls	0.054	0.93	0.22
PCDD/Fs	2.4 pg kg^{-1}	80000 ng kg^{-1}	2178 ng kg^{-1}
TEQ contribution attributable to the PCDD/Fs & PCBs	0.7	680	36.5
Aroclor mixtures	0.00002	23.1	0.023
Organochlorine pesticide	0.00001	2.2	0.035
Linear alkybenzene sulphonate [(1)]	2100	10500	5560
Di(2-ethylhexyl)phthalate	0.3	1020	110 (median)
Nonylphenols and nonylphenol ethoxylates	256	824	351
Chlorinated paraffins	0.9	6000	171
Polycyclic musks (ΣHHCB, AHTN)	2.0	97	32
Chlorinated phenols	0.09	52.6	No value
Polychlorinated naphthalenes	0.05	0.19	0.083
Polychlorinated *n*-alkanes C10-13	6.9	200	42
Polychlorinated *n*-alkanes C14-17	30	9700	1800
∑Polybrominated diphenyl ethers (6 congeners)[(2)]	12.5 μg kg^{-1}	288 μg kg^{-1}	108 μg kg^{-1} (median)
Triclosan[(3)]	0.09	16.8	2.3 (median)
Triclocarban[(4)]	36	66	51

Notes: [(1)] Schowanek *et al*. (2007) for anaerobically digested sludge
[(2)] Knoth *et al*. (2007)
[(3)] Ying and Kookana (2006)
[(4)] Heidler *et al*. (2006)

Table 10 Assessment of the significance of organic contaminants entering urban wastewater and sewage sludge (IC Consultants, 2001)

Contaminant	[1]Content in WW/sludge	[2]Priority hazardous substance	[3]Destruction in treatment		Accumulation (Yes, Y; No, N)		Background inputs	[4]Overall significance	
			WW	Sludge	Biological	Soil	(Yes, Y; No, N)	WW	Sludge
LAS	H	N	H	L Anaerobic H Aerobic	N	N	N	H	L
NPE	M - H	Y	M	L Anaerobic H Aerobic	N	N	N	H	L
DEHP	M	(Y)	M	L Anaerobic M Aerobic	N	N	N	H	L
PAHs	L - M	Y	L	L	Y	Y	Y	L	L
PCBs	L	N	L	L	Y	Y	Y	L	L
PCDD/Fs	L	N	L	L	Y	Y	Y	L	L
Pharmaceuticals	L	N	M	M	N	N	N	M	L
Oestrogenic:									
Endogenous	L	N	M - H	M - H	Y	N	Y	H	L
Synthetic	L	N	L - M	L - M	Y	N	N	H	L

Notes: [1]Concentration ranges for sludge: L<1 mg kg^{-1} DS; M<100 mg kg^{-1} DS; H>100 mg kg^{-1} DS. Concentrations in wastewater are small (mg l^{-1}) and highly variable, but will follow a similar general pattern to sewage sludge;
[2]EPCEU (2001): N = No; Y = Yes; (Y); Substance under review
[3]Approximate indicative ranges: L < 20 %; M = 20 – 60 %; H > 60 %
[4]Significance rating: Low, L; Moderate, M; High, H

LAS	Linear alkylbenzene sulphonates
NPE	Nonylphenolethoxylates
DEHP	Di-(2-ethylhexyl)phthalate
PAHs	Polycyclic aromatic hydrocarbons
PCBs	Polychlorinated biphenyls
PCDDs	Polychlorinated dibenzo-p-dioxins
PCDFs	Polychlorinated dibenzo-p-furans

86/278/EEC (EC, 2000; 2003b) – Table 11. This incoherency has occurred despite the generally consistent chemical composition of sewage sludge produced in different countries (Table 8). Consequently, agreement on regulating OCs in sludge is likely to emerge as one of the most controversial aspects of the consultation on the revised Sludge Directive (86/278/EEC).

Table 11 Standards for maximum concentrations of organic contaminants in sewage sludge (mg kg^{-1} DS except PCDD/F: ng TEQ kg^{-1} DS) (EC, 2000, 2003b; Smith, 2000a; Düring and Gath 2002; US EPA 2003b)

	AOX	DEHP	LAS	NP/NPE	PAH	PCB	PCDD/F
EC (2000)[a]	500	100	2600	50	6[b]	0.8[c]	100
EC (2003b)[a]			5000	450	6[b]	0.8[c]	100
Denmark		50	1300	10	3[b]		
Sweden				50	3[d]	0.4[c]	
Lower Austria	500					0.2[e]	100
Germany	500					0.2[e]	100
France					9.5[f]	0.8[cg]	
USA							300[h]

Notes:
a proposed but withdrawn and basis subject to review
b sum of 9 congeners: acenapthene, fluorene, phenanthrene, fluoranthene, pyrene, benzo(b+j+k)fluoranthene, benzo(a)pyrene, benzo(ghi)perylene, indeno(1,2,3-c,d)pyrene.
c sum of 7 congeners: PCB 28, 52, 101, 118, 138, 153, 180
d sum of 6 congeners
e each of the 6 congeners: PCB 28, 52, 101, 138, 153, 180
f sum of 3 congeners: fluoranthene, benzofluoranthen(b), benzo(a)pyrene
g for pasture the limit is 0.5 mg kg^{-1} DS
h following detailed risk assessment US EPA final decision was not to regulate PCDD/Fs (US EPA, 2003b)

4.3.3 Bulk volume and industrial compounds

Organic contaminants tend to accumulate in sludge due to their lipophilic and hydrophobic properties and consequently they sorb onto the sludge organic matter. Readily volatile OCs, eg solvent compounds (benzene, tetrachloroethane, tetrachloroethylene, toluene, and trichloroethane) are lost by volatilisation during wastewater and sludge treatment and land spreading and do not present a hazard to the soil or human health (Webber and Goodin, 1992). For example, Wilson *et al.* (1994) measured the concentrations of 15 important volatile organic compounds (VOCs) (e.g. chloroform, benzene, toluene) in samples of 12 liquid digested sewage sludges obtained from rural, urban and industrial treatment works in north-west England. No apparent relationship was found between sludge VOC concentrations and the volume of industrial input to the sewage treatment works, influent treatment, population served and sludge dry solids content. Wilson *et al.* (1994) concluded, therefore, that normal rates of sludge application to agricultural land were unlikely to increase the VOC content of the soil to levels which may cause concern for human health and the environment.

The principal route of loss of OCs during wastewater and sludge treatment, however, is through microbially mediated biodegradation processes. Destruction during anaerobic or aerobic digestion and composting of sludge significantly reduces the concentrations of many types of OCs in sludge. It is difficult to generalise but destruction rates by anaerobic digestion may be typically in the range 15 – 35 %. Aerobic treatment processes, including composting, may lead to greater destruction rates compared to anaerobic digestion. This is due to the increased rates of microbial activity at the normally higher operating temperatures of aerobic compared to anaerobic treatment processes, and also because aerobic conditions provide metabolic pathways of destruction for a greater range of organic substrates compared to anaerobic respiration. For example, LAS, an important high volume usage surfactant compound, is not degraded to any significant extent in anaerobic sludge digesters (AISE-CESIO, 1999). LAS concentrations are considerably higher in anaerobically digested sludges (1000–30,000 mg kg^{-1} dry solids (DS)), compared to fresh aerobic sludges (typically < 1000 mg kg^{-1} DM), or in aerobically stabilised sludges (100–500 mg kg^{-1} DS) (De Wolf and Feijtel, 1998; Ducray and Huyard, 2001; HERA, 2004; Jensen and Jepsen, 2005). There are several reasons for the elevated concentrations of LAS in anaerobically digested sludges: high usage volumes, sorption to primary sludge, precipitation as insoluble Mg/Ca salts in the primary settler, minimal degradation under anaerobic condidtions, and the solids mass loss effect caused by the digestion process.

Other compounds may be formed during anaerobic digestion of sludge. For example, nonylphenol (NP) and nonyl-phenol ethoxylates (NPEOs) comprise a group of surfactants that were used in large amounts in industry and agriculture. During anaerobic digestion of sewage sludge, mono- and di-ethoxylates are degraded to 4-nonylphenol and this treatment method increases the concentration of 4-NP in sludge. Digested sewage sludge samples collected at 2 UK WWTPs contained 250-820 mg kg^{-1} DS of NP (Jones and Northcott, 2000).

However, both LAS and NP degrade rapidly in soil. Under normal conditions LAS disappears rapidly from the soil as a result of biodegradation, with half-lives of approximately 1-3 weeks, and typically <10 days, depending on microbial adaptation, bioavailability and soil characteristics (Jones and Northcott, 2000). The half-life of NP in soil is typically 20 days. Thus, neither compound transfers through the foodchain or poses a risk to terrestrial organisms at the loadings applied to soils (De Wolfe and Fiejtel, 1997; Sjöström *et al*., 2004). The ecotoxicological risk assessment of LAS is described in more detail in Section 6.2.2.

In contrast to the low toxicity of LAS, however, NP is an identified endocrine disrupting compound (EDC) (IC Consultants, 2001). Within the EU, the use of NP and NPEOs has been restricted by Directive 2003/52/EC (EPCEU, 2003a), which stipulates that these compounds may not be placed on the market or used as a substance or constituent of preparations in concentrations equal or higher than 0.1 % by mass for a specified range of purposes. Nonyl phenols are also classed as a Priority Hazardous Substance under the Water Framework Directive (EPCEU, 2001) and will therefore be subject to further source controls to ultimately phase out emissions to the environment. In the UK, the Chemical Stakeholders Forum has reached a voluntary agreement within the chemical industry to phase out the use of nonyl phenols (DEFRA, 2004). Consequently these measures will ultimately lead to the further reduction of NP in sewage sludge to very low levels in future.

The other major bulk chemical found in sludge is di(2-ethylhexyl)phthalate (DEHP), which is also unchanged by anaerobic sludge digestion (IC Consultants, 2001). Phthalate esters are, however, rapidly destroyed under aerobic conditions and biological wastewater treatment (eg activated sludge process) can usually achieve >90% removal in 24 h (IC Consultants, 2001). In soil, the reported half-life of DEHP is <50 d. DEHP is an important chemical constituent of plastics, and can form upto 50 % of plastic products and PVC. It confers the unique properties and flexibility of plastics and is therefore an essential constituent of plastic products. It is not chemically bound to the polymer and so slowly leaches from the plastic, and hence can be transferred to wastewater. The household contribution of phthalates is reported to be as high as 70% of the total load, emphasising the ubiquity of DEHP and related compounds in the built environment and the difficulty that would be presented if controls to reduce emissions were required (IC Consultants, 2001). Two major sources of domestic releases to wastewater are floor and wall coverings and textiles with PVC prints (IC Consultants, 2001). In contrast to LAS and NP, DEHP has received much less attention in terms of the toxicological and ecotoxicological implications of recycling sewage sludge containing DEHP to agricultural land. In many respects, if controls were imposed on this compound in sludge or to reduce its emission to the environment (for example, DEHP is subject to a review for identification as a possible priority hazardous substance under review within the WFD), it would be much more difficult to manage because of its ubiquitous and widespread use in so many aspects of the built environment.

Polychlorinated *n*-alkanes (PCAs) refered to as chlorinated paraffins are in active production and widespread use eg as extreme pressure lubricant additives, plasticisers, flame retardants and paint additives, and therefore occur in sludge in much larger concentrations than other related families of persistent organic pollutants (POPs), such as PCBs and PCNs, whose use has been extensively phased out. PCAs are also persistent and bioaccumulatory. Stevens *et al*. (2003) measured total concentrations of the short-chained and medium-chained PCAs in the range 7-200 mg kg^{-1} DS and 37-9700 mg kg^{-1} DS, respectively. PCAs are widely used industrial chemicals and the high concentrations found in sludge were indicative of chemicals with numerous and ongoing diffuse sources.

Modern society is dependent on chemicals including plastics and their constituents and detergents, as well as many other types of organic compounds, which may therefore be present in sludge due to the extent of use and the amounts discharged in wastewater. It is prudent to ensure the compounds are not persistent or pose any toxicological or ecotoxicological risk and further improvements in sludge chemical quality should be expected from implementation of the EU REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals) Regulation 1907/2006 (EPCEU, 2006a, EC, 2007). Neverheless, the presence of apparently large concentrations of these chemicals in sewage sludge does not itself constitute a hazard to health or the environment, as this may be perfectly acceptable and safe. The impacts of alternative compounds, introduced as replacements for existing chemicals, should be carefully assessed as the net advantage of the alternatives may be uncertain.

4.3.4 Persistent organic pollutants

Concerns about the widespread presence of persistent, bioaccumulative, carcinogenic and mutagenic compounds in the environment resulted in the introduction of mitigation measures for PAHs, PCBs and PCDD/Fs in 1980-90s and these have significantly reduced the primary sources of persistent organic pollutants (POPs) in the environment (IC Consultants, 2001; Smith and Riddell-Black, 2007), as illustrated in Figure 9 and Figure 10. The main combustion emission sources of PAH and PCDD/F emissions, such as waste incineration (Figure 11) for example, are subject to stringent air quality emission standards (EPCEU, 2000b). Consequently, there has been a very significant concomitant decrease in POP concentrations found in sewage sludge, which reflect the background levels of these compounds in the environment in many cases (Table 12, 13 and 14, Figure 12). PAHs are also WFD Priority Hazardous Substances and will therefore be subject to further measures that will aim to cease emissions and losses of these compounds by 2020 (EPCEU, 2001). Given the substantial progress that has been made in controlling emissions of POPs in the environment, there is probably little scope for further reductions in PAH, PCB and PCDD/F concentrations in sewage sludge. Soil is the main repository of POPs present within the environment and recycling by volatilisation from the soil and deposition onto paved surfaces and runoff is a major mechanism of entry into the wastewater collection system and hence, into sludge (eg see Harrad *et al*., 1994; Wild and Jones, 1995).

Inputs of POPs to sewage sludge now principally reflect (Smith and Riddell-Black, 2007):

- Background inputs to the sewer from normal dietary sources;

- Background inputs by atmospheric deposition due to remobilisation/volatilisation from soil and cycling in the environment (e.g. PCBs, PCDD/Fs and PAHs);
- Atmospheric deposition from waste incineration (eg PCDD/Fs);
- Atmospheric deposition from domestic combustion of coal;
- Biodegradation during sludge treatment;
- Volatile solids destruction during sludge treatment.

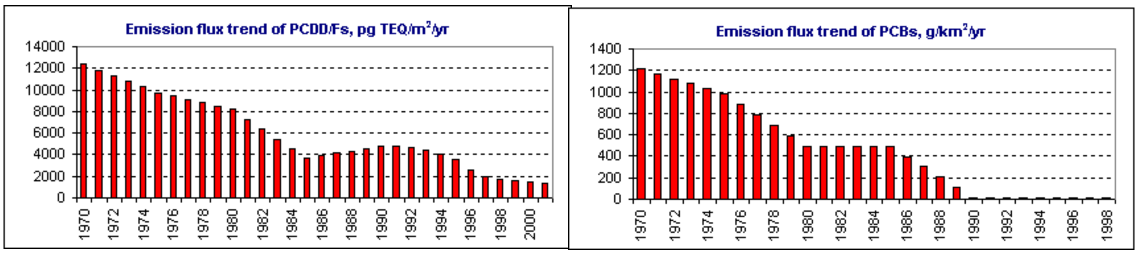

Figure 9 Long-term trends in the emissions of POPs to the UK environment (MSC-E, 2004)

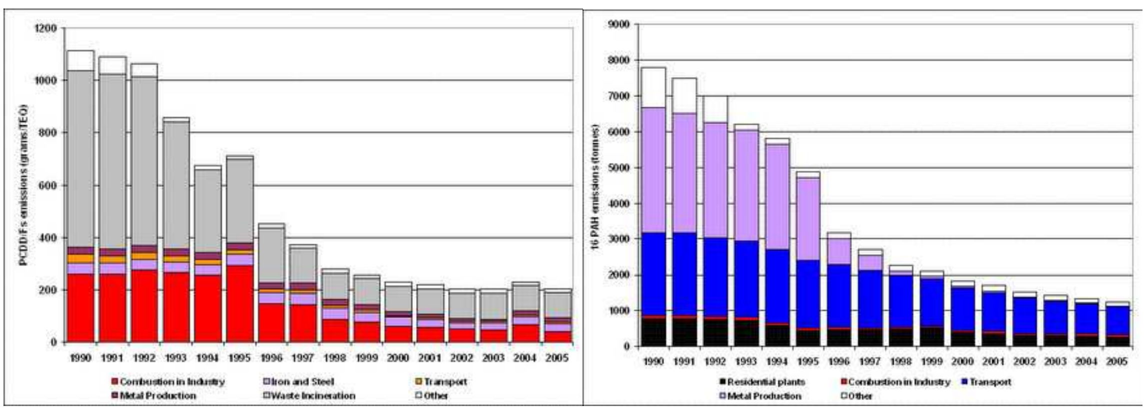

Figure 10 Long-term trends and principal sources of PCDD/F and PAH USEPA 16) emissions in the UK (NIAE, 2007)

(a) Austria (b) United Kingdom

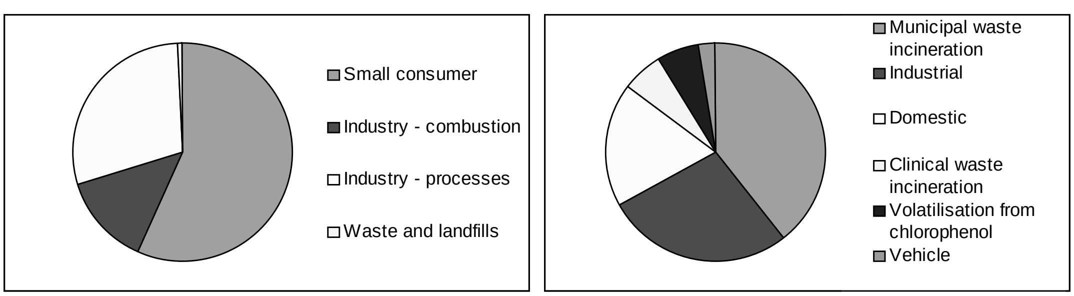

Figure 11 Sources of dioxin emissions (% of total emitted) in (a) Austria in 1998 and (b) UK in 1991 (IC Consultants, 2001)

Table 12 **Trends in concentrations of organic contaminants in European sewage sludge (Amlinger *et al.*, 2004)**

	AOX	PCB	PAH	DEHP	NPE	PCDD/F
	mg kg^{-1} DS					**ng TEQ kg^{-1} DS**
1988/89 range of means	250 – 350	<0.1	0.25 – 0.75	50 – 130	60 – 120	<50
1991/96 range of means	140 – 280	0.01 – 0.04	0.1 – 0.6	20 – 60	–	15 – 45
2002 range	60 – 400	0.001 – 0.232	0.01 – 246	0.2 – 170	0.02 – 824	0.2 – 128
2002 range of means	150 – 350	0.04 – 0.113	0.57 – 34	3.16 – 67.3	0.02 – 25.8	4.2 – 20.5
2004 range	13 – 510	0.013 – 0.379	0.77 – 8.97	<0.095 – 47	0.46 – 65	2.87 – 24.7
2004 range of means/medians	157	0.054	3.64	7.2	25	5.5

Table 13 Decline in PCDD/F and PCB concentrations in Spanish sewage sludge (Eljarrat *et al*., 2003)

WWTP	1980s	1990s	2000
pg TEQ PCDD/F g^{-1} *DS*			
Figueres	277	23	21
Olot	1028	7.8	5.4
Roses	78	13	7.2
Tossa	99	158	4.9
Vilafrance	56	108	7.2
ng PCB g^{-1} *DS*(1)			
Figueres	62		23
Reus		653	40
Roses	80		50
Tossa	121		37
Vilafranca		69	73

(1) ΣPCB 28, 52, 101, 118, 153, 138 and 180.

Table 14 Total PAH concentrations in sewage sludge surveyed in the UK in 2000/01 and 2002/03

Source	Approx. year	n	Min		Max		mean	
			EU	Total	EU	Total	EU	Total
Stevens *et al*. (2003)	2000/01	14 STWs	18	67	50	370	36	130
pers com S. Comber (Atkins Ltd, UK)	2002/03	7 from 3 STWs	0.09	0.34	29.4	41.3	4.8	7.0

Note: EU is the sum of 9 congeners: acenapthene, fluorene, phenanthrene, fluoranthene, pyrene, benzo(b+j+k)fluoranthene, benzo(a)pyrene, benzo(ghi)perylene, indeno(1,2,3-c,d)pyrene proposed by EC (2000, 2003b)

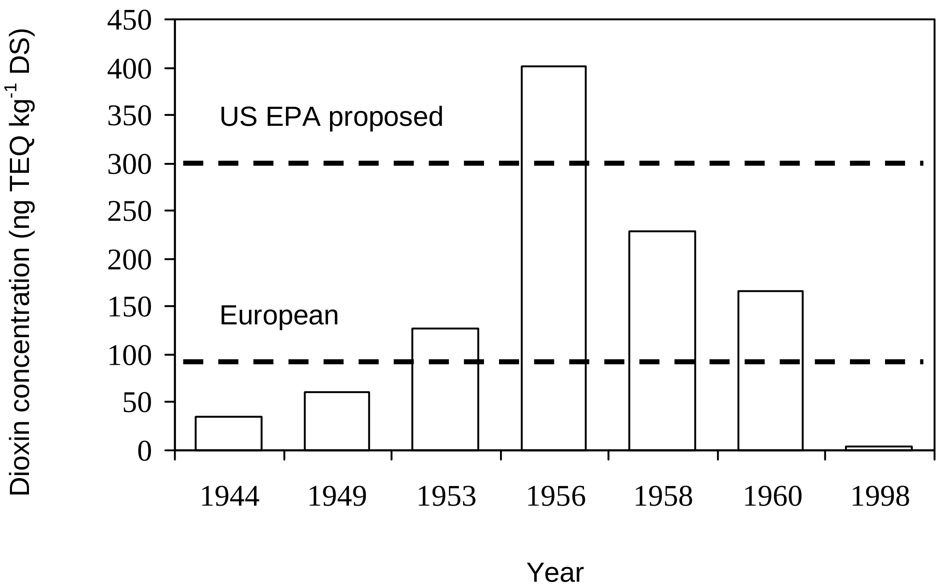

Figure 12 Dioxin content of archived samples of sewage sludge from a major wastewater treatment works in West London, UK (Note: following detailed risk assessment US EPA final decision was not to regulate dioxins in sludge – see USEPA (2003b)) (Smith, 2000a)

The polybrominated diphenyl ethers (PBDEs) are a group of compounds used for flame retardation in furnishings, textiles and electrical insulation and their use has expanded due to fire regulation requirements and the increased use of plastic material and synthetic fibres. These compounds have similar properties to dioxin, they are persistent and have the potential to bioaccumulate. The consequences of brominated flame retardants for health and the environment has received increasing attention in the scientific literature (de Wit, 2002; Letcher, 2003). A recent survey of PBDE concentrations in German sewage sludge (Knoth *et al*., 2007) indicated the total content of the six significant BDE congeners (28, 47, 99, 153, 154 and 183) was in the range 12.5 and 288 $\mu g\ kg^{-1}$ DS, with a median value of 108 $\mu g\ kg^{-1}$ DS. PentaBDE is on the WFD list of proposed priority hazardous substances, the Chemical Stakeholder Forum list of chemicals of concern (Defra, 2005) and the EU Pollutant Release and Transfer Registers (PRTR) for water (EPCEU, 2006). Action was taken in Europe to significantly restrict the use of pentaBDE and octaBDE and the placing on the market of articles containing one or both of these substances taking effect from 15 August 2004 (EPCEU, 2003b). Therefore, emissions to wastewater and presence in sludge are expected to decrease.

Data on pharmaceutical and antibiotic concentrations in sludge applied to land are very limited, but the available evidence indicates that the pharmaceuticals present vary greatly in biodegradability. There is a wide range of removal efficiencies during wastewater treatment and, typically, 60 - 90 % of the compounds in the influent may be removed (Ternes, 1998). Many common compounds such as the analgesics, ibuprofen and naxoproxen, are readily biodegradable with removal efficiencies between 22-90 % and 15-78 %, respectively (IC Consultants, 2001). For example, Hilton *et al.* (2003) showed that paracetamol is readily degradable and hence rarely found in sewage effluents.

4.3.5 Endocrine disruptors, pharmaceuticals, antibiotics and personal care products

Endocrine disrupting chemicals (EDCs) are synthetic or naturally occurring compounds that affect the balance of normal oestrogen hormone functions in animals, potentially resulting in reproductive impairment or disorders. There are about 100 OCs that are released to the environment that are suspected EDCs (Keith, 1998) and most, if not all, are likely to be found in sewage sludge. The categories of substances with endocrine-disrupting activities are listed in Table 15.

As discussed above, polychlorinated compounds are controlled at source, as are organochlorine pesticides, which are barely detectable in sewage effluents or sludge (Bowen *et al*., 2003), and are therefore no longer a concern for agricultural recycling of sludge. Alkylphenols and ethoxylates are currrently being phased out under the WFD as Priority Hazardous Substances and through restrictions imposed on their marketing and use in the EU by other dangerous substances legislation and national measures (EPCEU, 2003b; Defra, 2004) and therefore will no longer be significant. Biphenols have very short half-lives in soil of a few days and therefore cannot transfer to the human foodchain. Consequently It is the natural, endogenous hormones, oestrone and 17β-oestradiol, and to a much lesser extent the synthetic hormone ethinyloestradiol, that are considered primarily responsible for oestrogenic activity observed within treated sewage effluents. Approximately, 90 % of potential oestrogenic activity (based on 17β-oestradiol equivalency) in urban wastewater is reduced by wastewater treatment and <3 % may be transferred to the sewage sludge (Young *et al*., 1997). Oestrogenic compounds partition onto particulates and may be associated with sewage sludge, but there is no information on the amounts of endogenous oestrogens or ethinyloestradiol in sludge and only limited information on their biodegradation (Young *et al*., 1997). However, this is unlikely to have significant environmental implications for use of sludge in agriculture (IC Consultants, 2001). Oestrogenic substances excreted in the wastes of farm livestock are likely to represent a much greater loading onto soil compared to recycling sewage sludge (Boxall *et al*., 2004) and they are potentially more of a concern for the aquatic environment than for soil.

Table 15 Categories of substances with endocrine-disrupting activities (EA, 1998 cited IC Consultants, 2001)

Category	Examples	Uses	Modes of action
Natural			
phytoestrogens	Isoflavones, lignans	Present in plants	Oestrogenic and anti-oestrogenic
Female sex hormones	17β-oestradiol, oestrone	Produced in animals	Oestrogenic
Man-made			
Polychlorinated organic compounds	PCBs, dioxins	By-products from incineration and chemical processes	Anti-oestrogenic
Organochlorine pesticides	DDT, dieldrin, lindane	Insecticides	Oestrogenic and anti-oestrogenic
Alkylphenols	Nonylphenol	Production of NPE and polymers	Oestrogenic
Alklphenol ethoxylates	NonylPhenol Ethoxylate (NPE)	Surfactant	Oestrogenic
Phthalates	Dibutyl phthalate	Plasticiser	Oestrogenic
Bi-phenolic compounds	Bisphenol A	In polycarbonate plastics and epoxy resins	Oestrogenic
Synthetic steroids	Ethinyl oestradiol	Contraceptives	Oestogenic

Triclosan is a chlorinated phenoxyphenol and is used as an antimicrobial agent in a wide range of medicinal and consumer care products. European consumption of triclosan is about 350 t per annum (Singer *et al.*, 2002). The widespread use of triclosan means that it is inevitably discharged to wastewater treatment works. Singer *et al.* (2002) examined the transformation of triclosan through wastewater treatment processes and found 79% was biologically degraded, 15% associated with the sludge and 6% was released in final effluent. Degradation in soil is also rapid with reported half-lives in the range 17.4 – 35.2 days (Samsøe-Petersen, 2003). Concentrations in activated sludge range from 0.028 to 4.2 mg kg^{-1} DS and values up to 15.6 mg kg^{-1} DS are reported (Singer *et al*., 2002). Concentrations in the range 0.09 – 16.8 mg kg^{-1} DS with a median value of 2.32 mg kg^{-1} DS were reported in Australian sludge by Ying and Kookana (2006). The range reported in US sludge is 0.53 – 15.6 mg kg^{-1} DS with a mean of 6.97 mg kg^{-1}DS (McAvoy *et al*., 2002) and in Germany sludge contains 0.4 – 12 mg kg^{-1} of triclosan (Bester, 2003).
Triclocarban (TCC) is also a widely used topical antiseptic agent that is a common additive in many antimicrobial household consumables, including soaps and other personal care products. From a mass balance of the compound behaviour through a typical activated sludge wastewater treatment plant in the US, Heidler *et al*. (2006) concluded that approximately 21 % of the mass of TCC entering the plant was degraded, 3 % was discharged in the effluent and 76 % was transferred to the sewage sludge. Anaerobic digestion for 19 days did not promote TCC transformation, resulting in an accumulation of the antiseptic compound in dewatered, digested sludge to 51 +/- 15 mg kg^{-1} DS.

4.3.6 Polyacrylamide

The mechanical dewatering of sludge is becoming increasingly widely practiced, which, amongst other advantages, reduces the volume and therefore the transportation of sludge, it increases operational flexibility and aids storage until agricultural land is accessible for application. The sludge solids are consolidated to improve the efficiency of mechanical dewatering by the addition of cationic polyacrylamide (PAM) polymer, which is a chemical conditioning agent (Dentel, 2001). Polymer is added typically at a rate of up to 10 kg t^{-1} DS and, therefore, the concentration in sludge may be relatively high and equivalent to up to 1 % on a DS basis. PAMs are a class of compounds formed by the polymerizarion of acrylamide (AMD) and related monomers. The polymer therefore contains small traces of unpolymerized AMD. However, the concentrations of AMD in PAM are small and it is rapidly degraded in soil so, despite early concerns about the potential toxicity of AMD applied to soil in PAM, these are unfounded (Sojka *et al*., 2007). Anionic PAM is used extensively for chemical soil conditioning for water infiltration management and runoff control, and has very low environmental toxicity (Sojka *et al*., 2007). Cationic PAM has shown negative effects on aquatic organisms in pure water, and microorganisms and plants in *in vitro* and hydroponic tests, but its potential toxicity is greatly reduced in the presence of organic matter and clay minerals and the relevance of standard laboratory protocols for assessing the toxicity of cationic PAM has been questioned (Dentel, 2001). Biodegradation in the environment of the cationic monomer units in cationic PAM causes a transient release of trimethylamine which decomposes to dimethylamine that may form N-nitrosodimethylamine under certain conditions. However, though of potential concern to human health in principal, the rapid biodegradation of these intermediate products in soil and absence of plant uptake, or rapid metabolism and absence of accumulation in plant tissues eliminates possible transmission to the human foodchain (Smith, 2007a). The environmentally benign backbone backbone of the polymer molecule is not degraded by microbial attack, but is broken down slowly by mechanical disnitegration and becomes part of the residual organic matter content in soil (Sojka *et al*., 2007). Therefore, although PAM is present in large concentrations in mechanically dewatered sewage sldge, it does not pose a risk to human health, soil quality or the environment when sludge cake is recycled to farmland.

4.4 Radionuclides

Background radioisotope levels in sewage sludge were reported by Miller *et al*. (1996) for 25 municipal wastewater treatment plant in the US to determine the amount of man-made, gamma-ray emitting radioisotopes present in the environment. Dried sludge was found to have 0.0016 +/-0.0022 Bq g^{-1} of ^{137}Cs and 0.001 +/- 0.003 Bq g^{-1} of ^{60}Co. These data were intended to provide reference values to determine whether sewage effluents from nuclear facilities have levels of radioactivity above the background values expected from the environment.

Radionuclide levels in sludge from wastewater treatment plants near to nuclear power plants (NPP) were measured by Puhakainen *et al*. (2000). The main contributor was the discharge of radionuclides from NPPs into the atmosphere, although workers may transmit small amounts through their clothes or skin, or from internal contamination. The most significant activities due to discharges into the air were from ^{131}I and ^{110}Ag. The other nuclides probably originating from the NPP were ^{51}Cr, ^{54}Mn, ^{58}Co, ^{59}Fe, ^{60}Co, ^{124}Sb.

Martin and Fenner (1997) measured radioactivity levels in sludge from the Ann Arbor, Michigan, waste water treatment plant following radioiodine treatments of two patients at the University of Michigan hospital. Approximately 1.1% of the radioactive ^{131}I administered therapeutically to patients was measured in the primary sludge. The residence time of radioiodine in the sewer was longer than expected possibly due to absorption of iodine by organic material in the sewer, which contributed to its natural decay. Martin and Fenner (1997) considered there was no radiological health concerns from agricultural use of the sludge due to the short half-life of ^{131}I. Prichard *et al*. (1981) reached a similar conclusion from an investigation of ^{131}I levels in dried sludge used as a soil amendment, from a sewage treatment works serving a catchment containing the Texas Medical Centre, Houston, since the treatment time and period before harvesting crops grown on sludge-amended soil greatly exceeded the 8-day half-life of ^{131}I. However, incineration of sludge, which was performed in the winter months at Ann Arbor, directly released ^{131}I from sewage sludge to the atmosphere, and even though exposures to both workers and the public were found to be considerably lower than 1% of the natural background level, incineration of sludge is a pathway for public exposure. Martin and Fenner (1997) considered that the behaviour of radioiodine and other radioactive materials released into municipal sewage systems, such as those from large medical facilities, was not well understood.

5. IMPLICATIONS FOR HUMAN HEALTH

5.1 Pathogens

5.1.1 Principal exposure routes

The main potential pathway of exposure for the general population to infectious microorganisms that may be present in sludge is when it is used as a fertiliser on food crops that may be consumed raw (Gerba and Smith, 2005). This is managed and controlled by introducing multiple barriers (through sludge treatment processes to reduce pathogen numbers and restricting the uses of sludge on land combined with harvesting restrictions) or single barriers that rely on the treatment of sludge to effectively eliminate the pathogenic content. Another potential route of exposure, which has been recently examined, is from bioaerosol emissions from sludge handling and spreading activities on fields adjacent to residential areas (Brooks *et al*., 2005; Tanner *et al*., 2005). Although permitted under Directive 86/278/EEC (CEC, 1986), the practice of surface applying conventionally-treated sludge to grazed pasture has stopped at different times in individual European Member States, including the UK, for pasture hygiene considerations. Therefore, this no longer represents a potential route of infectious disease exposure and transmission to grazing livestock and the human foodchain. Nevertheless, there is no evidence that sludge was responsible for causing outbreaks of disease in farm animals when the controls for surface spreading to grazed pasture were followed (Jones, 1984; Evans, 1996).

5.1.2 Exposure through food

The potential risks of exposure to pathogenic organisms from land spreading of sludge have been recently quantified by the application of microbiological risk assessment (MRA) techniques (Gale 2003a,b; Gale, 2005). The risk assessment approach was developed to predict the additional risks to consumers from consumption of crops and cereal-based foodstuffs grown on agricultural land to which sewage sludge has been applied according to the Safe Sludge Matrix. Under the Matrix the use of untreated (raw) sludge is prohibited. The risk assessment was based on a highly conservative approach embodying large margins of safety; for example, the consumption of root crops grown on agricultural land receiving treated sewage sludge. Root crops are considered to have the greatest soil loading at point of harvest. Furthermore, it was assumed that 50% of vegetable crops are not only eaten raw (and unpeeled), but also only have a 12 month harvest interval (as opposed to the 30 months specified by the Matrix for ready-to-eat crops) after application of conventionally treated sludge. Uncertainty was dealt with in general, by using worst-case assumptions. Event trees formed the basis of the risk assessment calculation and an example of an event tree estimating the number of salmonellas in a harvested tonne of a root crop grown on sludge-amended soil amended with enhanced treated sludge is shown in Figure 13.

For conventionally-treated sludges applied to land with a 12 month harvest period, the predicted annual numbers of human infections from salmonellas, *Listeria monocytogenes*, *E. coil* 0157, Campylobacters, *Giardia* and enteroviruses, approach zero in the UK (Gale, 2003a). The highest pathogen levels were predicted for *L. monocytogenes* and *Giardia*, while the lowest were predicted for *E. coli* O157 and enteroviruses (Gale, 2005). These were calculated from laboratory-derived removal ratios for mesophilic anaerobically digestion, and from the point of view of decay in the soil, are worst case because the decay times are not extrapolated to the full 12 or 30-month harvest intervals specified by the Safe Sludge Matrix. Nevertheless, the predicted risks of infection to individuals are remote (Table 16) for salmonellas, *L. monocytogenes* and enteroviruses. Thus, the predicted risk of infection from salmonellas is 7.9×10^{-9} per person per year, giving one infection in the UK population every 111 years, or 0.009 $year^{-1}$. The highest risk predicted is that for *Giardia* at 4.3×10^{-5} per person per year, which translates into 50 infections per year in the UK. The risk of infection from *Cryptosporidium* is 4.2 x 10^{-7} per person per year (Table 16) and would give one infection in the UK population every 2 years (0.48 $year^{-1}$). The predicted risk of illness from *E. coli* O157 is remote and using conservative decay data one illness may occur in the UK every 4 years, i.e. 0.24 $year^{-1}$. It is emphasised, however, that considering the 12 month waiting period as stipulated in the Safe Sludge Matrix, significantly reduces the microbiological risk and predicted numbers of human infections from salmonellas, *L. monocytogenes*, *E. coli* O157, *Campylobacters*, *Giardia* and enteroviruses approach zero, with an estimated infection occurring once every ten million years in the UK based on linear extrapolation of pathogen decay data in soil (Gale, 2003a). The highest risk, in this case, was for *Cryptosporidium*, and the model estimated one infection every 45 years on average in the UK population as a whole.

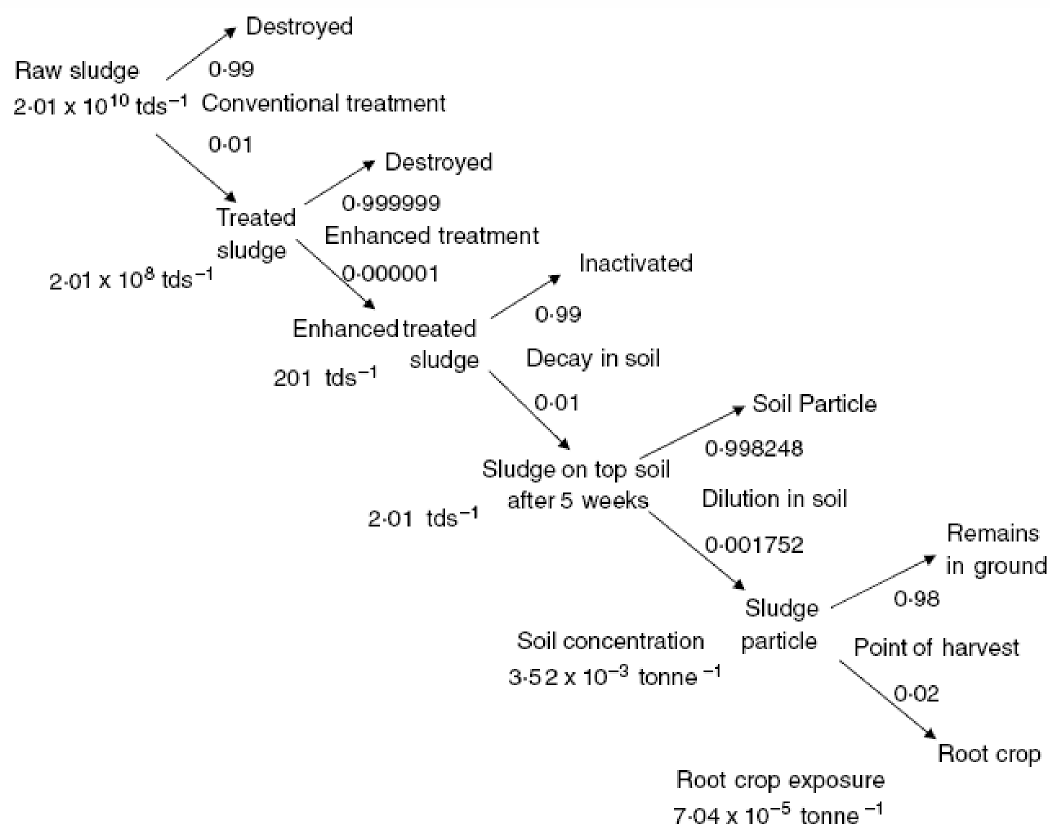

Figure 13 Event tree for transmission of salmonellas from sewage sludge to root crops (Gale, 2005)

For enhanced-treated sludges (e.g. pasteurisation) applied to land with a 10 month harvest period, the predicted numbers of infections annually from salmonellas, *Listeria monocytogenes*, *E. coli* 0157, Campylobacters, *Giardia* and enteroviruses also approach zero (Figure 13). In the case of *Cryptosporidium*, the model predicted a single infection per year for pasteurised sludge. The 12-month harvest interval, (30 months for salad crops) as specified by the Matrix was important for *Giardia*, particularly in the event of sludge treatment operating at less than 100% efficiency. Consequently, the operational efficiency of sludge treatment processes (and enhanced treatment processes in particular) is critical in determining the net level of pathogen destruction achieved. The risk assessment therefore supported the HACCP approach (Water UK, 2004) to ensure operational efficiency is maintained. Nevertheless, the risk assessment also demonstrated that the minimum 10-

month harvest interval required by Directive 86/278/EEC for all uses of sludge on land for food crop production compensates for the possibility of enhanced treatment processes not working to full efficiency at all times.

The risk assessment also identified significant data gaps in certain areas. These included data on levels of *E. coli* 0157 in sewage and sewage sludge, operational efficiency of sludge treatment, and the nature of the decay characteristics of pathogens on soil overtime periods of 10—30 months. These have been addressed to a significant extent in the work of Horan and Lowe (2002) and Horan *et al*. (2004) on pathogen removal during sludge treatment processes and by, for example, Lang *et al*. (2007) and Cass *et al*. (2007) on bacterial indicator and pathogen survival in field soil.

Table 16 Predicted risks of infection and pathogen levels in MAD-treated sludge, soil and on root crops (Gale, 2005)

Pathogen	Pathogen levels (arithmetic mean, kg^{-1})			Exposure†	
	MAD-treated sludge	Soil (decay time)	Root crops at point of harvest	(pathogen particles per person per day)	Risk of infection (per person per year)
Salmonellas	$5{\cdot}8 \times 10^{2}$	$3{\cdot}0 \times 10^{-3}$ (42 days)	$6{\cdot}0 \times 10^{-5}$	$2{\cdot}1 \times 10^{-7}$	$7{\cdot}9 \times 10^{-9}$
Listeria monocytogenes	$8{\cdot}7 \times 10^{5}$	$9{\cdot}6 \times 10^{-2}$ (42 days)	$1{\cdot}9 \times 10^{-3}$	$6{\cdot}6 \times 10^{-6}$	$1{\cdot}15 \times 10^{-8}$
Cryptosporidium	$2{\cdot}2 \times 10^{3}$	$4{\cdot}0 \times 10^{-3}$ (63 days)	$7{\cdot}8 \times 10^{-5}$	$2{\cdot}7 \times 10^{-7}$	$4{\cdot}2 \times 10^{-7}$
Giardia	$2{\cdot}9 \times 10^{4}$	$8{\cdot}5 \times 10^{-2}$ (84 days)	$1{\cdot}7 \times 10^{-3}$	$6{\cdot}0 \times 10^{-6}$	$4{\cdot}3 \times 10^{-5}$
Escherichia coli O157	$5{\cdot}0$	$2{\cdot}8 \times 10^{-7}$ (50 days)*	$5{\cdot}5 \times 10^{-9}$	$1{\cdot}9 \times 10^{-11}$	$7{\cdot}5 \times 10^{-11}$‡
Campylobacters	$1{\cdot}3 \times 10^{6}$	$1{\cdot}1 \times 10^{-3}$ (42 days)	$2{\cdot}2 \times 10^{-5}$	$7{\cdot}9 \times 10^{-8}$	$5{\cdot}5 \times 10^{-7}$
Enteroviruses	$0{\cdot}7$	$1{\cdot}1 \times 10^{-7}$ (90 days)	$2{\cdot}4 \times 10^{-9}$	$8{\cdot}3 \times 10^{-12}$	$1{\cdot}8 \times 10^{-9}$

†Assumes washing removes 90% of soil from root crops.
‡Illness.

Critically, the MRA for sludge received extensive peer review. In particular, it was the view of the Advisory Committee on the Microbial Safety of Food (ACMSF), that advises the UK Food Standards Agency on any matters relating to the microbiological safety of food, that the properly treated and applied sewage sludge represents a minimal food safety risk.

Gale and Stanfield (2001) have also applied MRA for bovine spongiform encephalopathy (BSE) in sludge and estimated that the risks to humans, based on a worst-case exposure scenario through consumption of vegetable crops, are acceptably low. Based on worst-case assumptions, the model also suggested that the application of sewage sludge to agricultural land used for grazing cattle would not sustain endemic levels of BSE in the UK cattle herd. The model emphasised the importance of containment of brain and spinal cord within the abattoir as the most effective mechanism of minimising the risk from BSE to cattle and humans. Thus, there may be circumstances where treatment and land use restrictions, whilst providing barriers to disease transmission, should also be complemented by measures to prevent infectious material from entering the wastewater collection system, in a similar manner to the source control of undesirable chemical compounds.

The multiple barrier approach to preventing pathogenic infections depends on the natural attenuation and decay of enteric organisms in the soil environment. Cropping and harvesting restrictions are based on a knowledge of the survival times of enteric organisms in soil and on plants (Table 17). The general effects of soil, environmental and management factors on the survival of enteric microorganisms in soil are reasonably well understood (Table 18). However, information on the specific, quantitiative relationships between these factors and decay of enteric species in soil has been lacking (Carrington, 2001). Therefore, recent research has also been undertaken to further understanding of the behaviour of enteric pathogens in the soil environment (Lang *et al*., 2007; Lang and Smith, 2007b; Rogers *et al*., 2006, 2007; Rogers and Smith, 2007). Pathogen and indicator decay was measured under

Table 17 Survival times of pathogens in soil and on plants (Gerba and Smith, 2005)

Pathogen	Soil		Plants	
	Absolute maximum	Common maximum	Absolute maximum	Common maximum
Bacteria	1 year	2 months	6 months	1 month
Viruses	6 months	3 months	2 months	1 month
Protozoa	10 days	2 days	5 days	2 days
Helminths	7 years	2 years	5 months	1 month

Table 18 Factors influencing pathogen decay in sludge amended soil

Factor	Comments	Source
Temperature	• 10 ^{o}C rise increased die-off twofold • Longer survival at lower temperatures and in winter than summer	Tierney *et al*. (1977) Bell & Bole (1978) Reddy *et al*. (1981) Andrews *et al*. (1983)
Moisture content	• Decreasing moisture increased die-off	Boyd *et al*. (1969)
Water holding capacity (WHC)	• Poor WHC decreased survival	Mallman & Litsky (1951)
Soil-water suction	• Maximum survival in saturated soil	Kibbey *et al*. (1978)
Application method	• Conflicting reports for die-off rates for sludge applied to the soil surface and incorporated sludge • Injected sludge may protect microorganisms • Discing may re-contaminate land	Giddens *et al*. (1973) Andrews *et al.* (1983) Wallis *et al*. (1985)
pH	• Shorter survival times in acid soils (pH 3-5)	Beard (1940) McFeters & Stuart (1972) Lambert (1974) Ellis & McCalla (1976)
Low nutrient availability and competing flora	• Soil is a hostile environment for pathogen survival	Baubinas & Vlodavets (1974) Gudding & Krogstad (1975)
Sunlight	• Shorter survival times at soil surface	Van Donsel *et al*. (1967) Tannock & Smith (1971) Gerba *et al*. (1975) Ellis & McCalla (1976)
Toxic substances	• Antibiotics produced by indigenous soil flora may inhibit growth of other microorganisms	Grossard (1952) Pinck *et al*. (1961) Soulides *et al*.(1961)

field and laboratory conditions. *E. coli* decayed and was indistinguishable from the background numbers measured in an agricultural sandy loam soil within 3 months following the application of conventional, dewatered digested sludge (Figure 14). Interestingly, the timing of sludge incorporation (spring or autumn) or initial numbers of *E. coli* measured in the sludge were not important factors governing the rate of decay of the organism. This confirmed the assumption of Gale (2003) that the 10 month waiting period required before harvesting food crops grown on sludge amended soil (CEC, 1986) provides a significant margin of safety and barrier to the transmission of pathogenic organisms to the human foodchain. Under laboratory conditions Lang and Smith (2007b) observed that *E. coli* decayed at a similar rate in two contrasting soil types maintained in a moist condition and the T_{90} value was 20 days. However, air-drying the soil before sludge amendment significantly reduced the rate of decay of *E. coli* and, in this case, the T_{90} was 100 – 200 days. This information, and other evidence reviewed by Rogers and Smith (2007) strongly point to the role of soil ecological processes in the active removal of enteric bacteria from soil. Ongoing research (Smith *et al*., 2006; Perez-Viana *et al*., 2007) is examining the effects of bacteriophagous protozoa on elimination of pathogenic bacteria from soil amended with conventionally treated sludge. It would appear that sludge application provides a self-limiting mechanism that actively destroys pathogenic bacteria in soil by stimulating the growth and activity of protozoa. These mechanisms, and coupled with the fact that enteric bacteria applied to the soil in sludge are poorly adapted to survival in this environment, restrict the potential for regrowth of enteric bacterial pathogens in sludge-amended agricultural soil (Zaleski *et al*., 2005). Similar mechanisms would also appear to be important in modulating the survival of parasites, such as *Cryptosporidium*, as oocysts of this organism degraded more rapidly in soil containing a native, background microbial population compared to sterile soil (Olson *et al*., 1999; Guan and Holley, 2003). The interaction between enteric parasites, such as *Cryptosporidium*, and soil ecological processes, that influence their survival in the environment, is poorly understood and is an area requiring further research.

Survival times for *Cryptosporidium* are longer than for other enteric protozoa eg *Entamoeba* and *Giardia* (see Feachem *et al*., 1983; Olson *et al*., 1999; Guan and Holley, 2003) and are temperature dependent. In cold (4 – 6 ^{o}C) and warm (20 – 30 ^{o}C) soils inactivation occurred in approximately 60 and 30 days, respectively (Olson *et al*., 1999). However, much longer survival times in soil have been reported in the literature for this parasite. Jenkins *et al*. (2002) predicted the number of days to reach 99.9 % inactivation in soil was in the range 2300-4000 days at 4 ^{o}C and 600 – 2300 days at 20 ^{o}C and considered this to represent the extreme limits of oocyst survival in soil.

The long-term decay of a range of pathogens including *Salmonella*, *Campylobacter*, *Listeria*, *E. coli* 0157, as well as *Clostridium* has been recently measured (Cass *et al*., 2007; Rogers *et al*., 2007) in soil amended with digested and untreated sludge types following inoculation with a large, artificial spike of these organisms (Figure 15). Overall survival times varied between the organisms, mainly due to differences in the background numbers that were detected by the selective enumeration techniques in the unamended soil. The maximum survival time recorded was approximately 160 days until the organisms either declined below the limit of microbial detection or approached the background numbers measured in soil. This is well with the waiting periods stipulated for the agricultural use of sludge (CEC, 1986; ADAS, 2001). However, it should be stressed that this repesents an extreme and highly conservative estimation of the time required for enteric bacteria to loose viability. This is explained for two reasons. Firstly, it is normally difficult to detect any pathogenic bacteria in sludge-amended soil in practice because the numbers in treated sludge are small or not detectable (Lang *et al*., 2007). Secondly, if small numbers were present in soil these would decay to background or undetectable levels in a much shorter time period, compared to when large numbers of a target organism are introduced artificially into soil for experimental purposes to quantify the decay kinetics. For example, Figure 15 shows a 2 $\log_{10}$ decline in *Salmonella* occurs in approximately 20 days from sludge incorporation. This equates to an initial population of about 4 $\log_{10}$ g^{-1} DS of *Salmonella* initially in sludge (conservatively assuming 10 t DS ha^{-1} are applied and diluted in soil by incorporation to a depth of 10 cm), which represents an extreme number of this or of the other major bacterial pathogens potentially found in sludge in practice. Comparison of the data in Figures 14 and 15 also emphasise the suitability of *E. coli* as an indicator of decay of pathogenic bacteria in sludge-amended soil systems.

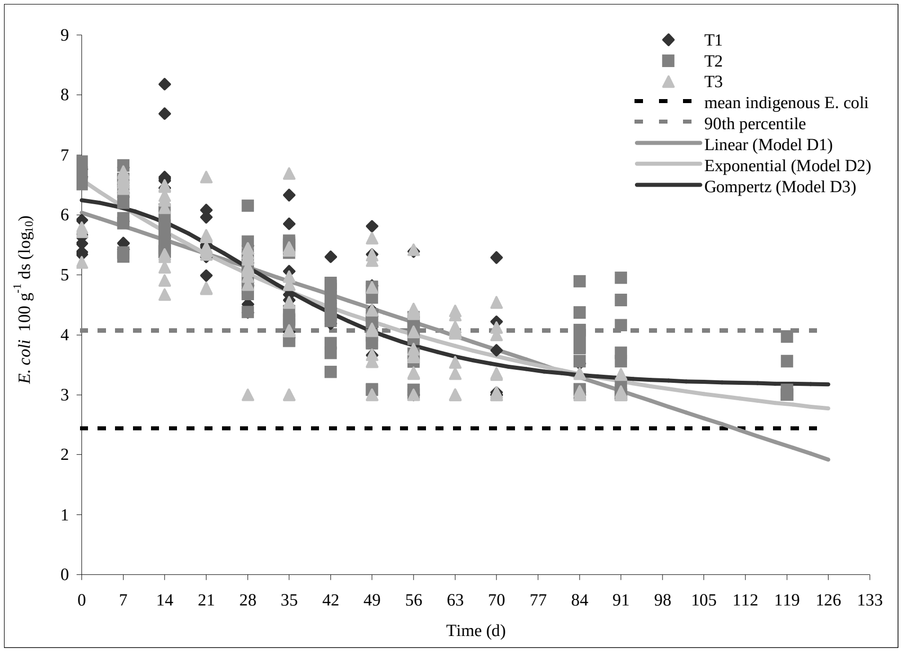

Figure 14 ***E. coli* decay in sandy loam field soil amended with digested sludge cake at a rate of 10 t DS ha^{-1} and incorporated to a depth of 10 cm; mean ( ) and 90th percentile ( ) background numbers of *e. coli* in the soil are also shown (Lang *et al*., 2007)**

5.1.3 Exposure from bioaerosols when sludge is applied to land

The possible health impacts arising from potential dispersal of bioaerosols from land application of sludge was recently reported, based on a major national site network investigation in the US (Brooks *et al*., 2005; Tanner *et al*., 2005). This work was undertaken following the recommendation of NRC (2002) based on anecdotal evidence in the US suggesting that illness within local communities adjacent to sludge amended fields was linked to emissions from sludge application activities (Lewis and Gattie, 2002; Gattie and Lewis, 2004). However, whilst this had a major negative influence on public perception in the US, from an entirely scientific perspective the reports were fundamentally flawed because they did not include an equivalent control group living near to farmland that was managed in a similar way, but where sludge had not been applied. It was further suggested that infections by the opportunistic pathogen, *Staphylococcus aureus*, were linked to the application of sludge nearby to residential housing (Lewis *et al.,* 2002). However, *S. aureus* has been subsequently shown to be absent from sewage sludge (Rusin *et al*., 2003). No coliphages or *E. coli* were detected in air in close proximity (2 m) downwind of the spray application of liquid mesophilic anaerobic digested sludge (Tanner *et al*., 2005). The community risk of infection from bioaerosols to residents living near sludge application sites was determined by Brooks *et al*. (2005) from a national monitoring network of bioaerosol emissions at 10 sites located in the

USA. A suite of indicator and pathogenic viruses and bacteria were monitored downwind of sludge application and handling activities. Total coliforms, *E. coli*, *C. perfringens* and coliphage were rarely detected at any sites. These results confirmed earlier field investigations (Pillai *et al*., 1996) that land application of sludge poses little risk of airborne transmission of bacterial pathogens. Sludge cake loading operations into spreading equipment resulted in the largest concentrations of the aerosolized microbial indicators, but they were only detected at distances within 15 m (Tanner *et al*., 2005). The greatest overall risk of infection occurred during loading operations from potential inhalation of coxsackievirus A21, estimated on a boundary distance of 30.5 m. Pillai *et al*. (1996) also found that physical agitation during handling of dewatered digested sludge stockpiles in the field caused very localised elevations in airborne bacteria within the immediate vicinity of activities such as spreader loading operations. Other research on the significance of bioaerosol emissions, for example, from waste composting operations, indicates that bioaerosols are reduced to background values within 250 m from the source (Swan *et al*., 2003; TCA, 2004). In practice, most potential residential exposures from sludge application sites are located at greater distances and particularly from handling operations, which usually take place well inside field boundaries. In overall conclusion, therefore, the community risk for bioaerosol infections is negligible (Pepper *et al*., 2006).

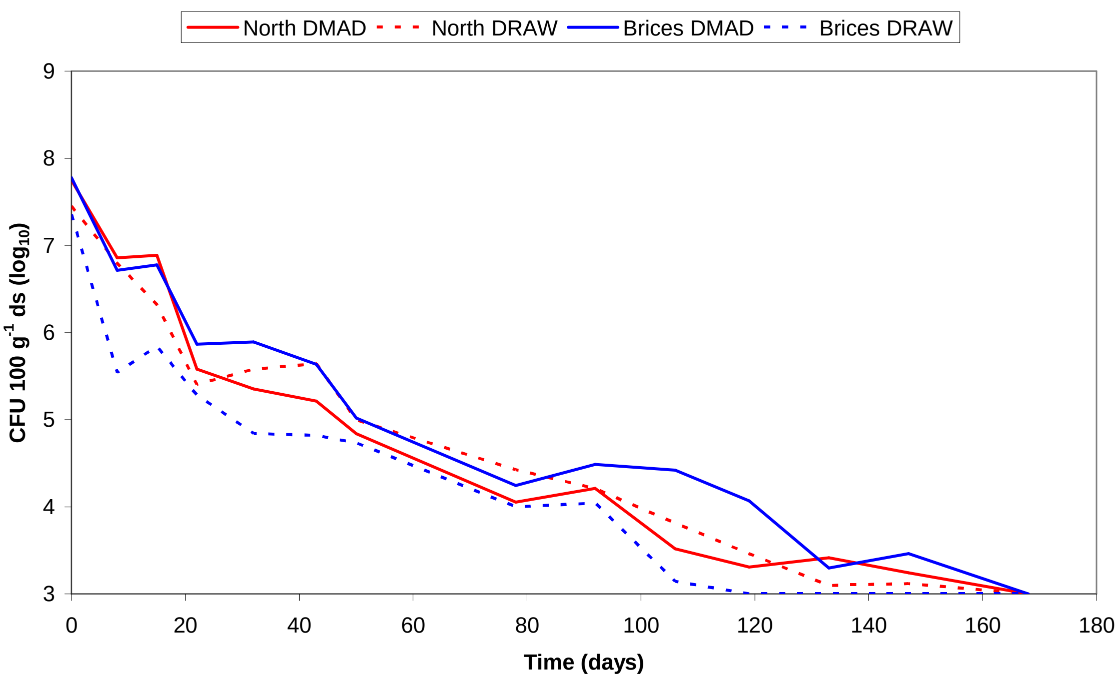

Figure 15 **Decay of *Salmonella enterica* in two adjacent agricultural fields with contrasting soil types (North: clay loam; Brices: clay) amended with inoculated dewatered digested (DMAD) and untreated (DRAW) sludge applied in October at a rate of 10 t DS ha^{-1} and incorporated to a depth of 10 cm (Cass *et al*., 2007)**

5.1.4 Summary remarks

There is no evidence linking the application of sewage sludge to farmland with disease in the human population when biosolids are treated and applied according to the regulatory controls and guidance (RCEP, 1996; Carrington *et al*., 1998b; Pepper *et al*., 2006). Following an independent evaluation of US EPA's Part 503 rule governing land application of sewage sludge, the US National Research Council (NRC, 2002) came to the overarching conclusion that there was no documented scientific evidence to

indicate that the controls had failed to protect public health, although there were areas where a further commitment to research was considered necessary, especially to complete a microbiological risk assessment (MRA). The US EPA is currently undertaking a MRA for pathogens in sludge (pers. com. J. Smith, US EPA), which will help to refine that completed in the UK by Gale (2003a,b, 2005). Nevertheless, the UK ACMSF considered that the risks to food safety were minimal following the critical peer review of the MRA of sludge management practices performed by Gale (2003a). The absence of disease in humans or agricultural livestock (eg Jones, 1984; Evans, 1996; Carrington *et al*., 1998b) arising from the agricultural utilisation of sewage sludge may therefore be regarded as strong evidence demonstrating the effectiveness of multi-barriers at preventing disease transmission when sludge is applied to farmland as a fertilizer and soil conditioner. The community risks to health from bioaerosols emitted during sludge handling and application operations are also negligible (Pepper *et al.*, 2006).

5.2 Potentially toxic elements

5.2.1 General aspects of foodchain exposure

Detrimental effects on human health due to the application of heavy metals to soil and subsequent transfer to the foodchain from the agricultural use of sludge are extremely unlikely (MAFF/DoE, 1993a; Chaney, 1994; Smith, 1996). This is because foodchain transfer mechanisms are well understood and appropriate controls are in place restricting the accumulation of zootoxic elements in soil amended with sludge. Furthermore, the concentrations of the principal zootoxic metals of concern in sludge: Cd, Pb and Hg have declined significantly over the past 30 years owing to effective trade effluent and emission control measures (Section 4.2). Indeed, the total concentrations of these elements is sludge have declined to such an extent that the average amounts of Cd and Pb measured in contemporary sewage sludges (Figures 6 and 7) applied to agricultural land are smaller than the maximum permissible soil limits (Table 7) themselves. Under these circumstances, the application of high quality modern sewage sludge products to soil cannot result in the accumulation of these elements above the safety limits. This is further emphasized because these elements are not the most restrictive elements present in sludge as the total soil limits for Cu or Zn, which are the most abundant metals present in sludge (Figure 6), are usually reached first, so that accumulations of Cd, Pb and Hg will always remain below there respective maximum permissible values in soil in the practical field situation. Pb and Hg are not absorbed by crops from sludge-amended soil and consequently do not pose a risk to the diet from crop uptake (Carlton-Smith, 1987; Smith *et al*., 1993). Cadmium, on the other hand, can accumulate in the edible parts of crops, but dietary intake models show that the human food chain is protected by the EU maximum soil limit for Cd in sludge-treated agricultural land, equivalent to 3 mg Cd kg^{-1} ds (CEC, 1986; Carlton-Smith, 1987; MAFF/DoE, 1993a). Indeed, individuals consuming a well balanced diet, including plant foods that may be grown on sludge-treated soil, actually represent a low risk group of exposure because improved mineral intake in vegetables reduces Cd adsorption by the gut (Logan and Chaney, 1983).

5.2.2 Cd transfers to the diet through crop uptake

Cd is the most labile and transferable of the potentially zootoxic elements present in sludge to crop tissues. In contrast to Cd, Pb and Hg, the other heavy metals of concern to human health, are so strongly adsorbed in sludge-treated soil they show little or no transfer to crop plants (Logan and Chaney, 1983; Carlton-Smith, 1987; Aldridge and Alloway, 1993). By contrast, Cd readily traverses the soil-plant barrier and can potentially accumulate in staple food crops and transfer to the human foodchain. Cadmium is not an acute toxin, but represents a potential chronic risk to health from life-time exposure. Therefore, the Food and Agriculture Organization and World Health Organization (1978) have set a provisional limit for the daily intake of Cd by humans of 70 μg per day for a 70 kg person (equivalent to 1 μg kg^{-1} body weight per day) and this is used as the basis for assessing the _toxicological risk_ of potential exposures to Cd from the agricultural use of sludge. The amount of Cd ingested in plant food depends on a number of factors including:

- Soil Cd content;
- Soil properties, particularly pH value;
- Crop species sensitivity to Cd accumulation;
- Edible crop component eg grain, root, tuber or leaf;
- The amounts of various crop types consumed in the diet;
- The proportion of food in the diet that is grown on sludge-treated soil.

Crops types vary in sensitivity to Cd accumulation (Figure 16). However, Cd uptake and concentrations in the staple food crops including, for example, potato tubers and cereal grain are much less than for certain leafy crop types, which usually represent a relatively small fraction of the total food intake.

Estimated intakes of Cd for typical UK diets are well within the FAO/WHO (1978) tolerable limit at the EU maximum permissible soil concentration for Cd of 3 mg kg^{-1} (Figure 17). These calculations conservatively assume that the vegetable portion of the diet is obtained entirely from sludge-treated soil. The proportion of cereal foods in the diet grown on soil receiving sludge is assumed to be 2 %, to reflect the relative area of sludge-treated arable land in the UK and the marketing and distribution of cereal crops. The Steering Group on Chemical Aspects of Food Surveillance (MAFF/DoE, 1993a) in the UK considered that the marginal impact of the agricultural use of sludge on the Cd intake by the general populaton, compared with the background intake from the foodchain in the UK of 18.8 μg Cd d^{-1}, was inconsequential to human health.

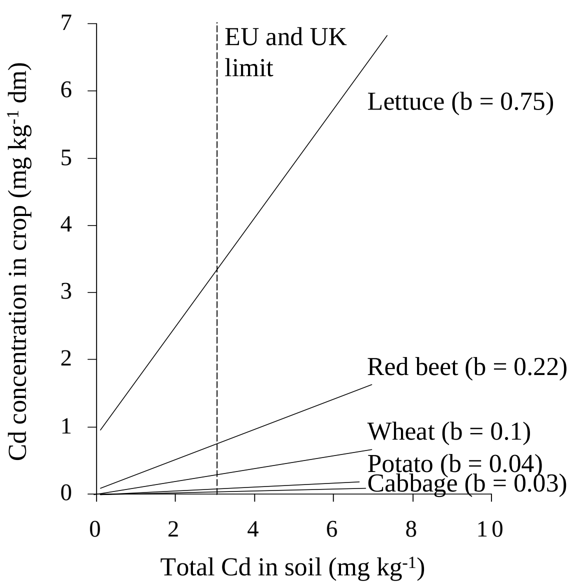

Figure 16 Cd uptake by different crops in relation to content in loamy sand soil, b represents the slope of the linear regression relation (Carlton-Smith, 1987)

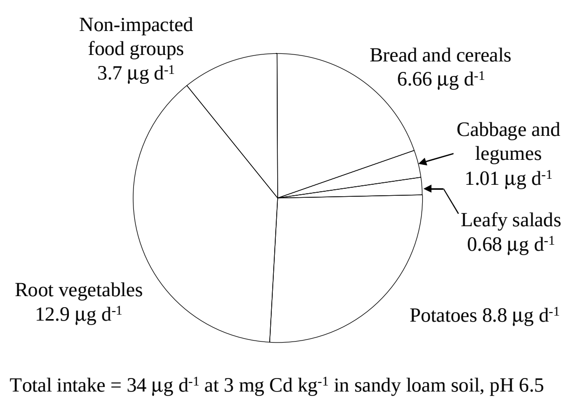

Figure 17 Dietary intake of Cd from sludge-treated soil (Carlton-Smith, 1987)

Assumptions relating to the proportion of food in the diet that is obtained from sludge-treated soil are a major factor influencing the absolute values of risk-derived limits for Cd applied to farmland in sludge. Individuals consuming a large share of their total plant food intake from land receiving sewage sludge are potentially the most exposed group to Cd uptake by crops and this provides a conservative bias in estimating limit values that protect the human diet. For example, in the US EPA critical environmental pathway analysis for Cd in sewage sludge-treated soil (US EPA, 1992b), linear soil-crop transfer factors for the major plant food groups were used to estimate the allowable intake based on the difference between the maximum average intake recommended by WHO of 70 μg Cd day^{-1} for a 70 kg person and the background ingestion level in the US of 16.14 μg Cd day^{-1}. This analysis showed that 120 kg Cd ha^{-1} could be safely applied to soil in sewage sludge without adverse effects. Ryan and Chaney (1994) argue that, from a technical perspective, no individual would be harmed even if sludge-amended soil reached a soil concentration of 60 mg Cd kg^{-1}.

Stern (1993) examined the intake of Cd by highly exposed individuals who consume a large proportion of their own food (59 % of their fruit and vegetable intake and 37 % of their potato intake) grown on soil affected by sludge-Cd. Stochastic techniques were applied using Monte Carlo probabilistic analysis of the exposure variables, to take account of the uncertainties and variability within the modelling process. This provided upper confidence limits to indicate the probability of exceeding the acceptable daily dietary exposure to Cd with increasing Cd cumulative application rate. The probabilistic analysis of the exposure variables was also restricted to Cd uptake slopes for soils with pH $\leq$6.5. The pH restriction had a moderate influence on the cumulative Cd loading compared to no pH restriction. Thus, without pH restriction, the 90th and 95th upper confidence limits for the acceptable cumulative loading rate for Cd were 30 and 17 kg ha^{-1}, respectively, and with pH restriction to $\leq$6.5, the 90th and 95th upper confidence limits were 23 and 13 kg ha^{-1}, respectively. In acid soils, the Cd loading rate at the 95th upper confidence limit therefore equates to a total soil concentration of 4-6.5 mg Cd kg^{-1} (assuming conversion factors of 3.25 or 2, respectively; MAFF/DoE, 1993b). Consequently, this provides further independent evidence indicating that the European maximum permissible limit for Cd in sludge-amended agricultural soil is inherently protective of the human foodchain (CEC, 1986).

In agricultural situations, the amount of food consumed from sludge-treated soil is significantly smaller than by individuals who grow most of their own plant produce. Indeed, the EPA (1992b) estimated that 2.5 % of the diet in the general population may be affected by the agricultural use of sludge in the US. This further emphasises the extent of protection against potentially harmful accumulations of Cd in the human diet from the agricultural use of sludge.

The adoption recently in the EU (EC, 2001) of mandatory limits on the maximum concentrations of particular metals permitted in certain foodstuffs, such as for Cd in cereal grains (eg 0.1 mg kg^{-1} wet weight in barley and 0.2 mg Cd kg^{-1} wet weight in wheat; equivalent to 0.235 mg kg^{-1} dry weight in wheat grain – Chaudri *et al*., 2007) will require soil concentrations to be assessed based on their influence on the Cd content in crop parts rather than on a toxicological risk basis as described above. Using data on soil crop transfers (eg Figure 15) it is a straightforward task to estimate the appropriate soil Cd limit concentration to avoid exceeding the grain limits for Cd. Thus, the relationship for wheat determined by Carlton-Smith (1987), presented in Figure 15, indicated that a reduction in the current maximum soil limit to 2 mg kg^{-1} would be necessary to comply with the grain quality standards for wheat crops grown on potentially high risk soils (eg course-textured and low pH). Smith (1994a) recommended reducing the maximum permissible soil limit for Cd from 3.0 mg kg^{-1} ds to 2.0 and 2.5 mg Cd kg^{-1} ds for soils with pH values in the ranges 5.0 - ≤5.5 and 5.5 - ≤ 6.0, respectively, to account for the increased crop uptake and potential transfer of the element to the human diet under low soil pH conditions. However, this proposal was not accepted by the UK Steering Group on Chemical Aspects of Food Surveillance (MAFF/DoE, 1993a) due to the negligible overall impact of sludge-borne Cd on total human dietary intake. More recently, based on soil and crop analyses from an extensive programme of sludge-amended field experiments in the British Isles, Chaudri *et al.* (2007) reported that the current total Cd limit of 3 mg kg^{-1} in sludge-treated soil would not ensure Cd contents in wheat grain would be below the food quality standard unless the soil pH was maintained above 6.8. In addition to the effects of soil properties on Cd availability, there are also strong differences in the sensitivity to soil Cd between plant species (Davis and Carlton-Smith, 1980) and also between wheat cultivars, which accumulate different amounts of Cd at equivalent soil Cd contents (Chaudri *et al*., 2001).

Adams *et al*. (2004) developed a simple multiple regression model to predict grain Cd concentrations, which had the form: log(grain Cd) = $a + b$ log(soil Cd) – c(soil pH). The model was used to estimate the total Cd content in soil that would be acceptable to comply with the limit in wheat grain, with 95 % confidence. Thus, predicted soil concentrations at soil pH values of 5, 6 and 7 were estimated as <1.0, 2.4 and 5.8 mg Cd kg^{-1} ds. The model predicted that the soil pH would require maintenance at ≥pH 6.3 to ensure compliance with limit for Cd in wheat grain at the current maximum permissible limit value for this element in soil (3 mg Cd kg^{-1} ds), with 95 % confidence. It should be noted, however, that Adams *et al*. (2004) and Chaudri *et al*. (2007) used high Cd sludges to define the relationship between soil and grain Cd contents (for example, in the case of Chaudri *et al*. (2007) they applied sludge containing 44 mg Cd kg^{-1} DS – see Gibbs *et al*. (2006c)) and this may increase Cd accumulation in plant tissues compared to equivalent soil concentrations obtained with low Cd sludge (De Brouwere and Smolders, 2006). The mean Cd concentration in sludge applied to agricultural soil in the UK is currently only 1.5 mg kg^{-1} DS (Figure 7). Consequently, these estimates of the upper soil Cd concentration necessary to comply with the grain Cd standard may be overly conservative. Barley accumulates much lower concentrations of Cd in the grain than wheat under comparable soil conditions. Adams *et al*. (2004) concluded that barley grown in Britain under typical field conditions and management regimes is unlikely to exceed the current EU maximum limit for Cd in barley grain.

Chang *et al*. (2002) have developed human-health related guidelines for the World Health Organization, Geneva for the application of sewage sludge in agriculture that may be generally applicable to different regions. Plant uptake was selected as the principal exposure route of concern and a 'global' representative diet was developed to provide the criteria for calculating technically-based standards. The model also conservatively assumed that 100 % of all plant foods in the diet were obtained from sludge-amended soil and that acceptable exposure to a pollutant was limited to 50 % of the WHO maximum tolerable intake for an adult with a body weight of 60 kg. This analysis indicated that, as a general guide, a conservative soil limit value of 4 mg kg^{-1} was appropriate to protect the foodchain from Cd applied to soil in sewage sludge. Although this value is consistent with the EU soil limit for this element (Table 7), the precautionary calculations by Chang *et al*. (2002) further emphasize the minimal risk to the human foodchain due to Cd applied in sludge to agricultural soil. The soil limit concentrations derived for the other potentially zootoxic elements were: 84 mg Pb kg^{-1}, 7 mg Hg kg^{-1}, 8 mg As kg^{-1}, 6 mg Se $kg^{-1}$and 635 mg F kg^{-1}. These values are influenced by the highly conservative dietary exposure scenario assumed by Chang *et al*. (2002). In the case of Pb, for example, the consensus is that this element is relatively unavailable for uptake by food crops from sludge-amended soil and, therefore, does not pose a potential toxicological hazard through plant uptake (Aldridge and Alloway, 1993). Indeed, Pb availability is usually reduced after sludge application compared to most non-sludged mineral soils due to the increased binding of Pb by the sludge matrix.

5.2.3 Livestock ingestion route

The direct ingestion by grazing livestock of soil and surface applied sludge is another potential route of transmission of metals from sludge-amended soil to the human foodchain. Extensive experimental investigations were conducted in the UK for the Ministry of Agriculture, Fisheries and Food (MAFF) and Department of the Environment (DoE) (Stark and Wilkinson, 1994; Stark *et al*., 1995) on the effects and implications for the human foodchain of heavy metal ingestion by sheep grazing sludge-amended pasture. For an integrated summary of this work and other international literature relating to animal ingestion see Smith (1996); a detailed review of the MAFF and DoE studies is also provided by Stark *et al*. (1998). Sheep were selected for study as they represent a potential worse-case of soil ingestion compared to other gazing animals (Stark, 1989). The main risks to the human diet were identified as the accumulation of Cd, and particularly Pb, in offal meat. Importantly, however, heavy metals do not accumulate in muscle tissue. A potential risk of exceeding the food limit for Pb in offal meat[1] was identified (Stark *et al*., 1998). Consequently, Carrington *et al*. (1998b) recommended reducing the maximum permissible soil limit for Pb from 300 mg Pb kg^{-1} ds to 200 mg Pb kg^{-1} ds and this was incorporated into proposals to amend the statutory controls for the agricultural use of sludge in the UK (DEFRA/WA, 2002), although this has not yet been enacted. The concentrations of Cd in offal meats are unlikely to exceed the food standards[2] for this element at the mandatory soil limit. Offal represents only a very small component of the diet (0.5% fresh weight) compared with staple plant foods such as potatoes or cereal crops, and is therefore unlikely to have much impact on total dietary intake of these elements overall. Market dilution factors further minimize the dietary intake of Cd and Pb in offal and also of Cd in plant produce.

5.2.4 Summary remarks

The potential transfer pathways of entry of zootoxic elements to the human foodchain have been extensively researched and quantified, and conservative, maximum permissible soil limits for Cd and Pb have been set and adjusted accordingly to ensure human health is protected. There is no evidence for the transfer of Hg, the other zootoxic element of concern, to the human diet and a precautionary soil limit is set for this element (Carrington *et al*., 1998a). Following an extensive review of scientific evidence, the Committee on the Use of Treated Municipal Wastewater Effluents and Sludge in the Production of Crops for Human Consumption (NRC, 1996) in the US concluded that no adverse human acute or chronic toxicity effects have been reported from ingestion of food plants grown in sludge-amended soils.

Whilst understanding the extent of human exposure to potentially toxic metals is critical, this could be regarded increasingly as an academic exercise under contemporary circumstances due to the major reductions in the concentrations of these elements in sludge that have occurred in the past 30 years. Indeed, trade effluent controls have been so effective that, for Cd and Pb, the average concentrations in sludge are smaller than the total limits in soil. Furthermore, it is impossible for the concentrations of these elements to accumulate up to their respective limit values in soil receiving modern high quality sludge as the limits for the most abundant elements in sludge (Cu or Zn) are always reached first. Historically, Cd has been the main zootoxic element of concern due to its potential uptake and accumulation by crop plants. However, whilst the potential risk to human health from Cd is a recognised concern associated with the land application of sewage sludge, it has become a theoretical risk due to these reasons and, in the practical field situation, the

[1] The EU food limit for Pb in edible offal of cattle, sheep, pig and poultry is 0.5 mg kg^{-1} wet weight (EC, 2001)
[2] The EU food limit for Cd in liver and kidney of cattle, sheep, pig and poultry is 0.5 and 1.0 mg kg^{-1} wet weight, respectively (EC, 2001)

agricultural use of sludge has minimal impact on Cd in the foodchain overall. Nevertheless, some adjustment to the current limit value for Cd in sludge-amended agricultural soil may be required, perhaps according to different pH bandings (eg 5.0-5.5, 5.5-6.0 and 6.0-7.0), to ensure that the total soil limit is consistent with complying with the maximum concentration permitted in wheat grain in the EU.

5.3 Organic contaminants

5.3.1 Evidence base and risk assessment

The toxicological and environmental effects of organic contaminants (OCs) in sludge and sludge-treated soil have received an increasing level of research attention in the past 20 years and many of the earlier uncertainties and concerns have largely been resolved. Details can be found in a number of recent critical reviews which provide an in depth analysis and assessment of the literature. For example, IC Consultants (2001) examined the fate and significance of pollutants, including organic compounds, in wastewater and sludge for the European Commission DG Environment. That report provided an exhaustive assessment of concentration data in sludge and fate during sludge treatment processes and detailed case studies on surfactants, polyacrylamide (used extensively as a polyelectrolyte to aid mechanical dewatering sludge) and potentially important emerging groups of compounds such as pharmaceutical and bodycare products, are presented in the Appendix. In a related report for DG Environment, Erhardt and Prüeβ (2001) examined the occurrence of organic compounds in sludge, related toxicological data, their persistence in soil and risk assessments for various exposure pathways. In North America, a detailed review for the Water Environment Association of Ontaria (2001) examined the fate of selected contaminants in sewage biosolids applied to agricultural land based on scientific literature and consultation with stakeholder groups. The compounds considered included:

- Volatile organic contaminants (VOCs), polychlorinated biphenyls (PCBs), polynuclear aromatic hydrocarbons (PAHs) and pesticides;
- Linear alkylbenzene sulphonate (LAS) surfactants;
- Endocrine disrupter compounds (EDCs) including alkylphenol surfactants (APs) and oestrogenic hormones;
- Dioxins and furans (PCDD/Fs)

Jones and Northcott (2001) completed a survey of organic contaminants in UK sewage sludge samples and considered their potential significance in a report for the UK Gorvenment (also see Stevens *et al*., 2003). More recently, a major study for Defra by Smith and Riddel-Black (2007) reviewed the evidence relating to OC inputs to soil from all sources, including sludge, and their potential environmental consequences. The behaviour and environmental effects of organic contaminants in sludge-amended soil were examined in detail in a series of papers by international authors published in a special edition of The Science of the Total Environment (Jones, 1996). In Europe, OCs were also considered by Working Party 2 – Chemical Contamination of Sludges and Soils of the COST 681 programme on Treatment and Use of Sewage Sludge and Liquid Agricultural Wastes (Davis *et al*., 1983; Hall *et al*., 1992a; Hall *et al*., 1992b). These selected examples demonstrate that, whilst it is always possible to argue that the knowledge base on the diverse range of compounds potentially present in sludge is incomplete (Chaney *et al*., 1996; Harrison *et al*., 2006), a considerable amount is known about the fate and behaviour of the main and most significant compounds, or groups of compounds identified in sludge to enable their potential significance for human health and the environment to be quantified.

The available research and monitoring of the fate and potential effects of OCs in sewage sludge used on agricultural soil has identified specific pathways of possible transfer of OCs to humans, livestock, plants and other potentially sensitive environmental endpoints (Chaney *et al*., 1996). Transfer coefficients from sludge and sludge-amended soil to plants, animals and humans have been derived for many organic substances and these formed the basis of the first quantitative pathway analysis and risk assessment of OCs in sewage sludge amended agricultural soil completed by US EPA (1992b,c). Fourteen environmental pathways were identified, and the most important for human health were:

Pathway 1: Sludge-Soil-Plant-Human (from agricultural application)
(Pathway 2 was also the Sludge-Soil-Plant-Human route, but represented the domestic use of sludge by home gardeners)
Pathway 3: Sludge-Soil-Human
Pathway 4: Sludge-Soil-Plant-Animal-Human
Pathway 5: Sludge-Soil-Animal-Human

The US EPA screened 200 pollutants and selected 18 OCs of principal concern for further evaluation by the pathway risk analysis of environmental exposure. The health impact was assessed in relation to the cancer causing potency of the chemicals in humans from continual exposure of an individual to a specified concentration over a life time period of 70 years. The compounds that were evaluated included:

- Aldrin/dieldrin
- Benzo(a)anthracene
- Benzo(a)pyrene
- Bis(2-ethylhexyl)phthalate
- Chlordane
- DDD/DDE/DDT
- Heptachlor
- Hexachlorobenzene
- Hexachlorobutadiene
- Lindane
- Methyl bis(2-chloro-aniline)
- Methylene chloride
- n-Nitrosodimethylamine
- Pentachlorophenol
- Polychlorinated biphenyls
- Toxaphene
- Trichloroethylene
- Tricresyl phosphate

The US EPA evaluated these contaminants by the pathway risk analysis and decided that their regulation was unnecessary to protect human health and the environment when sewage sludge was used as a soil amendment (US EPA, 1992b,c). Therefore, organics were deleted from the Final Part 503 Standards for the Use or Disposal of Sewage Sludge (US EPA, 1993). For an organic pollutant to be deleted from the regulation, one of the following criteria had to be satisfied (Ryan, 1993):

1. The pollutant was banned from use in the US; has restricted use or is not manufactured for use in the US.

2. Based on the results of the National Sewage Sludge Survey (NSSS), the pollutant has a low level of detection in sewage sludge.

3. Based on data from the NSSS, the limit for an organic pollutant in the Part 503 exposure assessment (US EPA, 1992b) was not expected to exceed the concentration in sewage sludge.

In December 1999, the US EPA proposed a limit for PCDD/Fs and certain dioxin-like PCBs of 300 ng kg^{-1} TEQ DS in sludge for agricultural use in the Round Two regulation 40 CFR Part 503 Standards for the Use or Disposal of Sewage Sludge (US EPA, 1999). However, following further evaluation of the pathways of greatest exposure from consumption of crops and meat products by farmers and their families, who were assumed to produce a significant proportion of their own food on sludge-treated land for an entire lifetime, US EPA determined that dioxins from this source do not pose a significant risk to human health or the environment. US EPA also noted the continuing decline of dioxin concentrations in sludge, particularly with more rigorous control on combustion practices. Therefore, US EPA decided that no numerical limits or specific management practices were required for dioxins in land-applied sewage sludge (US EPA, 2003b).

Risk assessment procedures for chemicals in sewage sludge will continue to undergo refinement. For example, Schowanek *et al*. (2004) recently presented a conceptual framework for risk assessment of OCs in sludge, shown in Figure 18, for all potential exposure pathways and environmental end-points. However, as further data are collected, and concentrations of the most toxic and persistent compounds continue to decline in the environment and in sludge, due to the source control measures operating in Europe (Section 4.3.4), the general trend is for the significance for human health of OCs in sewage sludge to continue to diminish in importance. Furthermore, the exposures to the major groups of high volume, bulk chemicals found in sludge (eg surfactants and plasticisers) from the agricultural use of sludge appear to be no greater than the normal background level. For example, Vikelsøe *et al*. (2002) showed that only small concentrations of a range of phthalate and NPs were detected in soil receiving typical doses of sludge at agronomic rates of application (4.3 t DS ha^{-1} y^{-1}) for at least three consecutive growing seasons and the values were within the normal background ranges measured for other cultivated and uncultivated soils receiving different fertilizer and manure regimes, but without sludge input. Consequently, in the practical field situation, using sewage sludge as an agricultural fertiliser does not increase the accumulation of these compounds in soil or their transfer to the human foodchain.

5.3.2 Human exposure from uptake into crop plants

One of the primary concerns associated with the presence of OCs in sludge is the potential for entry into the human foodchain from uptake into the edible parts of crop plants. However, despite the increasing amount of scientific investigation into the potential environmental consequences of OCs applied to farmland in sewage sludge, there is no evidence for soil-crop transfer and that the risk to human health from consuming food crops grown on soils amended with sludge is negligible (Jacobs *et al*., 1987; Schmitzer *et al*., 1988; Aranda *et al.*, 1989; Kampe and Leschber, 1989; O'Connor *et al*., 1991; Hembrock-Heger, 1992; Offenbacher, 1992; Sauerbeck and Leschber, 1992; Webber *et al*., 1994; NRC, 1996; Duarte-Davidson and Jones, 1996; O'Connor, 1996; McGrath, 2000). This is explained due to the behaviour of organic contaminants in soil and the presence of only very small concentrations of some of the more potentially toxic compounds in sludge, eg the dioxin, 2,3,7,8-TCDD (Sauerbeck and Leschber, 1992). Thus, organic compounds can be grouped into three broad categories according to their behaviour (Smith, 2000a):

1. Volatile compounds which are quickly lost to the atmosphere from sludge and sludge-treated soil;
2. Compounds which are rapidly mineralised by microorganisms and have little or no persistence;

3. Persistent compounds which are strongly adsorbed onto the sludge and soil organic matrix.

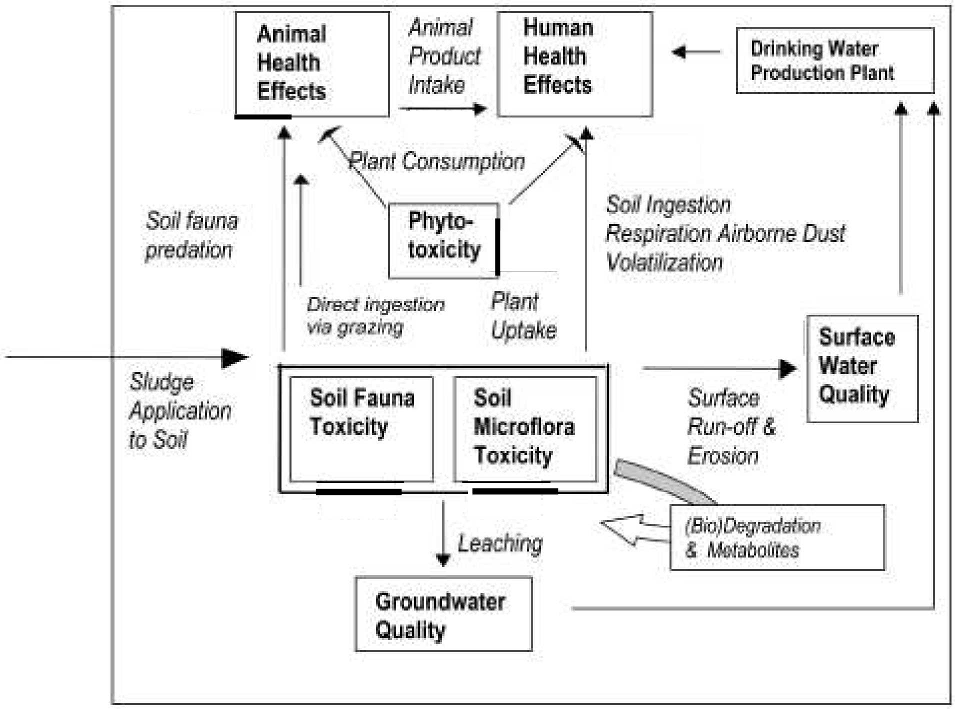

Figure 18 Conceptual framework for the risk assessment of organic chemicals in sewage sludge applied to agricultural land (Schowanek *et al*., 2004)

Compounds exhibiting some solubility and potential for plant uptake are also susceptible to rapid degradation processes in soil, or are lost through volatilisation or leaching processes (Webber and Goodin, 1992; Wilson *et al*., 1994; Duarte-Davidson and Jones, 1996; Wilson *et al*., 1996; Wilson and Jones, 1999). For example, nonylphenol (NP) and its precursors, the nonylphenolpolyethoxylates (NPEOs), have caused concern due to their ability to interfere with human and animal endocrine systems and have the potential to transfer to agricultural crops (Sjöström *et al*., 2004). However, their degradation in soil is rapid (half-life 20 – 60 days for NP for example – see Table 8) and enhanced by the addition of organic substrates such as biosolids and uptake by plants is very low (Kirchmann and Tengsved, 1991; Roberts *et al*., 2006) so the possibility of any transfer or risk to the human food chain in practice is very limited. Furthermore, NP is identified as a priority hazardous substance under the Water Framework Directive and is therefore subject to source control measures to phase out discharges, emissions and losses to the water environment (EPCEU, 2001). More persistent compounds, on the other hand, usually have very low solubilities, are present in very small concentrations, and are strongly adsorbed by the soil matrix in non-bioavailable forms and this prevents them transferring to plant tissues (O'Connor, 1996). The evidence suggests that the groups of compounds of emerging interest (Section 4.3.5) also follow the same behaviour and are likely to exhibit negligible transfer to crops for the above reasons. For example, Thiele-Bruhn (2003) concluded from a recent review that uptake into plants even of mobile antibiotic compounds was generally small, although effects on plant growth were apparent for some species and antibiotics. Endocrine disrupting chemicals are another emerging concern for human health. However, whilst uncertainties may remain regarding their potential significance for agricultural use of sludge, no link has been identified between environmental sources of endocrine disrupting chemicals (EDCs) and health effects in humans (EA, 1998). On balance it seems probable that they will have negligible relevance to human health (Section 4.3.5) since the most potent forms (eg endogenous oestrogens) are present in very small amounts in sludge and/or are likely to degrade very rapidly in soil. Furthermore, humans receive much greater exposures to EDCs through other pathways than are potentially possible from sludge. For example, one of the main identified exposure routes for humans to EDCs is through dietary intake of phytooestrogens (Phillips and Harrison, 1999). These naturally occurring chemicals are present in many common and staple foodstuffs and have structural similarities to 17β-oestradiol and are more potent oestrogens than many synthetic chemicals. Materials used for food packaging may also contain EDCs, which can migrate into the food in small amounts, for example, bisphenol-A, which is used in the resin linings of food cans.

5.3.3 Human exposure from ingestion by grazing livestock

The principal concern and theoretical mechanism of entry of OCs to the human foodchain from agricultural utilisation is by surface spreading liquid sludge to grassland and intake by livestock grazing treated pasture resulting in the potential accumulation of lipophilic compounds in meat fat and milk (Wild *et al*., 1994; Fries, 1996; Duarte-Davidson and Jones, 1996). Only lipophilic halogenated hydrocarbons accumulate in animal tissues and products (Fries, 1996). Compounds such as phthalate esters, PAHs, acid phenolics, nitrosamines, volatile aromatics and aromatic surfactants are metabolized and do not accumulate (Fries, 1996). In practice, there has been no demonstrable link between sludge application and transfer of organics to the human foodchain via this pathway (NRC, 1996). This theoretical exposure pathway does not operate when surface spreading to pasture is not permitted or when liquid sludge is injected into the soil (Stark and Hall, 1992; Wild *et al*., 1994).

Recent studies in the UK (Erhard and Rhind, 2004; Rhind, 2004; Paul *et al.*, 2005) with sheep grazing sludge-amended pasture soils have suggested a relationship between the apparent exposure of the animals to environmental contaminants in sludge and their behavioural and reproductive development and that this could indicate potential implications for human health from exposures to these chemicals in sludge. Careful scrutiny of the basic design of the grazing trials, however, shows that they were not sufficiently critically controlled to be able to draw such a profound conclusion. For example, there were major differences in the amounts and characteristics of the diets fed to the animals due to variations in fertiliser nitrogen supply and this could be a major factor influencing animal performance and development. In the published reports (Erhard and Rhind, 2004; Paul *et al.*, 2005), no supporting chemical analysis of the applied sludge, soil or herbage, or measurement of direct ingestion of soil or sludge by the experimental animals is presented making conclusive interpretation impossible. In the unpublished report (Rhind, 2004), DEHP was indicated as an important EDC, however, this is not supported by *in vitro* and *in vivo* tests completed by the US FDA, which demonstrated that DEHP and other phthalates were not oestrogenic (US FDA, 2001). DEHP concentrations in soil were affected by seasonal factors, but not by sludge application and, furthermore, sludge application did not increase the body burden of DEHP above the control animals. The fact that EDC concentrations (DEHP and alkyl phenols) did not accumulate in soil following repeated applications of sludge supports the view that they are rapidly degraded in the soil by microbial activity and that the soil environment is very efficient at destroying these compounds through natural attenuation processes.

Attempting to understand and quantify the impacts of subtle physiological responses to trace amounts of organic substances in the diet of grazing animals in a complex field grazing environment is very difficult in practice. The only practicable and quantitatively robust way to do this would be through controlled feeding experiments where animals are managed under identical conditions and receive exactly the same diet with the only variable being the addition or absence of the target compound.

5.3.4 Particular compounds of interest

5.3.4.1 Phthalates

Plasticizers have been identified in this review as deserving special mention because there is apparent misunderstanding regarding their potential impacts on human health, and consequently, also in relation to their significance in sewage sludge-amended agricultural soil.

Plasticisers are added to plastic polymers to give plastics useful properties such as colour, resistance to fire, strength and flexibility. Over 90% of plasticizers used are phthalate based compounds and in Western Europe about one million t of phthalates are produced each year, of which approximately 90% are used to plasticize PVC (polyvinyl chloride) (CSTEE, 1999). The annual global production of DEHP has been estimated to be approximately 2 million t (Koch *et al.*, 2003a,b). Concentrations of phthalates in PVC range from 15-50%. There are five phthalate plasticizers: di(2-ethylhexyl) phthalate (DEHP, sometimes also referred to as DOP), diisodecyl phthalate (DIDP) and diisononyl phthalate (DINP), butyl benzyl phthalate (BBP) and di-n-butyl phthalate (DBP). DEHP is one of the most commonly used plasticisers and accounts for approximately 20% of all plasticiser usage in Western Europe (ECPI, 2007). Phthalates are not chemically combined with PVC and are slowly released to the environment by leaching, for example, and consequently transfer to soil by the water and sludge application route. DEHP is a priority substance under a review within the WFD (EPCEU, 2001) and a EU draft limit was proposed for sludge (EC, 2000), although DEHP was not included in the Draft Directive for agricultural use of sludge proposed by the European Commission (EC, 2003b). DEHP is also regulated under Danish legislation for agricultural use of sludge (Table 11). DEHP is listed as a pollutant with a maximum threshold value for release to land under the new European Regulation on Pollutant Release and Transfer Registers (E-PRTR Regulation) (EPCEU, 2006). A further reason for giving DEHP special consideration is that it is one of the most frequently detected, high concentration compounds found in municipal sludges. Mattson *et al.* (1991) (cited in Paxéus, 1996) estimated the household contribution of phthalates to municipal wastewater was 70% of the total load emphasising the ubiquity of the compounds and difficulty of control. Concentrations in US sludge are reported to range from 0.3 to 1020 mg kg^{-1} DS with a weighted average value of 110 mg kg^{-1} DS (US EPA, 1992d). Given the ubiquitous use of phthalates and their potential to transfer to the environment, and the fact that they have been regulated in sludge and are under review with regard to the WFD, it is surprising that their significance in the environment has not been more fully considered, compared to surfactant compounds for example.

Initial concerns that DEHP may be potentially carcinogenic to humans were based on animal testing with rodents which developed liver tumours when exposed to the compound. However, the mechanisms by which DEHP increases the incidence of hepatocellular tumours in rats and mice is not relevant to humans. Apparently, primate species do not absorb DEHP as efficiently as rodents, nor do they convert it to mono (ethylhexyl) phthalate (MEHP) as readily as rats or mice (Huber *et al*., 1996), which is the chemical thought to initiate the process leading to liver cancer in rodents. Taking due regard of this scientific evidence, the International Agency for Research on Cancer (IARC) downgraded the carcinogenicity classification of DEHP from Group 2B, *as possibly carcinogenic to humans* to Group 3, *not classifiable as to carcinogenicity in humans* (IARC, 2000).

There is a wealth of information in animals administered DEHP for periods ranging from a few days to lifetime studies that show DEHP is a developmental and reproductive toxicant by mechanisms not yet completely understood (ATSDR, 2002). However, the reproductive effects observed in some rodents exposed to DEHP during fetal development and as adults are not thought to be caused by DEHP docking to an oestrogen receptor. Rather, as with carcinogenicity, these effects are apparently related to unique mechanisms and sensitivity in susceptible rodents (Davis *et al*., 1994). Indeed, results from *in vivo* and *in vitro* studies indicate that DEHP has negligible estrogenic potency relative to the endogenous hormone, 17β-estradiol. From a comprehensive toxicological profile of the compound, the US Agency for Toxic Substances and Disease Registry (2002) concluded that there is no evidence that DEHP is an endocrine disruptor in humans at the levels found in the environment.

Humans are exposed to phthalates in numerous ways, such as migration of phthalates into foodstuff from packaging materials, by dermal contact of cosmetics and inhalation, and young children can also be exposed through mouthing of soft PVC toys, for example (Koch *et al*., 2003ab). Nevertheless, exposure of the general population to DEHP is considered to be significantly below (at least 280 times) levels that may place human health at risk (ECPI, 2007). Furthermore, large exposures to DEHP occur through medical procedures due to leaching of DEHP out of plastic medical devices into solutions that come in contact with the plastic and this can result in an aggregate exposure when multiple devices are used (US FDA, 2001). However, there is no evidence of health abnormalities in these highly exposed and sensitive groups (US FDA, 2001) despite the widespread use of PVC plasticized with DEHP in medical situations. In contrast to these possible pathways of exposure to DEHP, the risk to human health arising from diffuse environmental sources, such as from DEHP inputs to soil from recycling sewage sludge, would appear to be very minor indeed. This is reinforced by the degradability and biotransformation of DEHP in aerobic soil (Figure 19) and also the absence or minimal transfer of DEHP to crops and the foodchain (Staples *et al*., 1997; Schmitzer *et al*., 1988; Aranda *et al.*, 1989; Madsen *et al*., 1999; van Wezel *et al*., 2000; Yin *et al*., 2003). Thus, phthalates have no toxicological significance in sewage sludge-treated agricultural soil and sludge is unlikely to contribute to the accumulative human exposure from multiple sources of DEHP.

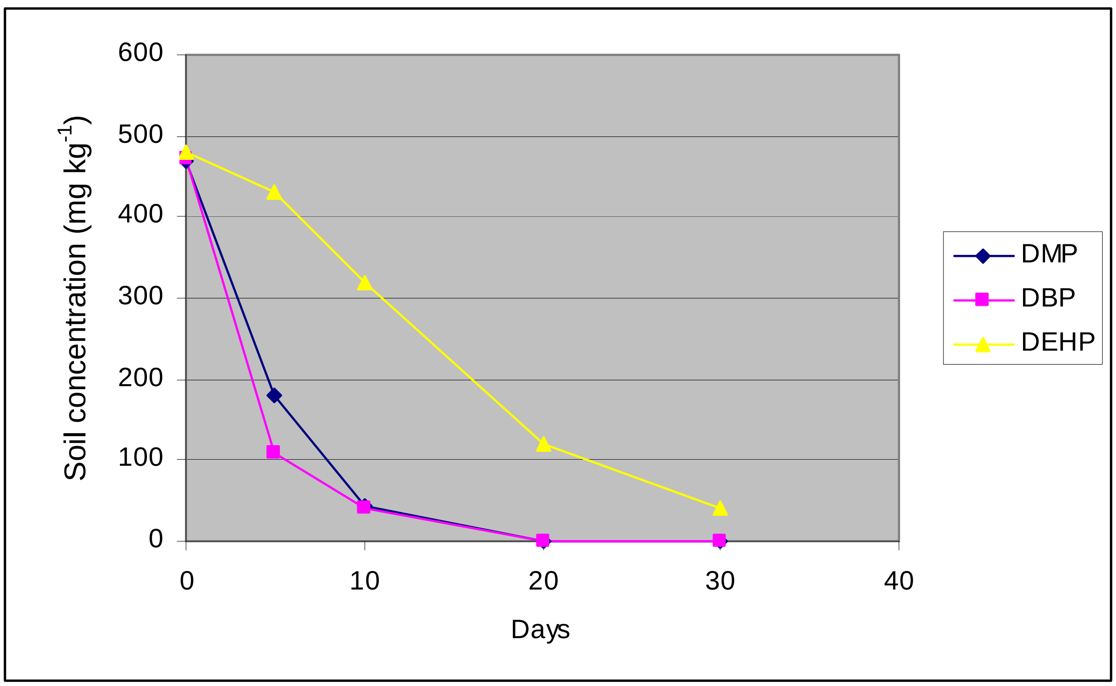

Figure 19 Decay of plasticizers in soil (Shanker *et al*., 1985)

5.3.4.2 Antibiotics

Many pharmaceutical compounds have similar physico-chemical characteristics as other organic compounds, such as persistence and lipophilicity with the potential to bioaccumulate, but much less is known about their entry into the environment and subsequent fate (Alcock *et al*., 1999; Smith and Riddell-Black, 2007). However, based on the general models of behaviour identified for OCs in Section 5.3.2, pharmaceuticals are also likely to follow the same general fate and behaviour as the other organic compounds with these properties that have already been reasonably well characterised in sludge-treated soils. Thus, it is unlikely for antibiotics (and indeed, other pharmaceutical compounds) to have a direct toxicological impact on humans from agricultural use of sludge due to the very small concentrations present in sludge, but more importantly because environmental transfer pathways to humans do not operate (eg crop plant uptake).

Of considerably greater potential importance than direct toxicological impacts of environmental sources of antibiotics on human health is the potential spread of antibiotic resistant human pathogens from emissions of these compounds to the environment (Jørgensen and Halling-Sørensen, 2000). The development of antibiotic resistant populations of bacteria in soil, in particular, has been directly linked to the use of antibiotics in animal husbandry and from spreading livestock manures on land (Jørgensen and Halling-Sørensen, 2000; Nwosu, 2001; Haller *et al.*, 2002; Onan and LaPara, 2003; Thiel-Bruhn, 2003) and is also potentially a concern for the agricultural use of sewage sludge since residues of these compounds may also be exoected to occur in sludge (Kinney *et al*., 2006).

Only trace concentrations of active antibiotic substances in soil and other environmental media are required to provoke resistance development (Halling-Sørensen *et al*., 1998). Because human and animal microbial ecosystems are closely related, microbial antibiotic resistance readily crosses species boundaries (Witte, 1998). In this way, multiple-resistant strains of microorganisms have found their way into the food chain (Berger *et al*., 1996 cited in Halling-Sørensen *et al*., 1998). The ability of antibiotics to exert these effects at very low concentrations appears to be a unique characteristic of antibiotic compounds as this type of behaviour is not apparent for any other chemical (Jørgensen and Halling-Sørensen, 2000).

However, the development of resistance characteristics in soil bacterial populations appears to be only a short-lived, transient response to the presence of antibiotics in soil. Recent studies show that, once the selection pressure from inputs of antibiotic compounds in livestock manures is removed, the resistance profiles of the soil community return to pre-treatment values, or are equivalent to untreated control soil (Sengeløv *et al*., 2003; Rysz and Alvarez, 2004). Thus, resistance characteristics are rapidly lost because, once the selective advantage of maintaining antibiotic resistant genes is removed the acquired resistance is depleted from the population since the maintenance requirements set the resistant organisms at a disadvantage to the general population. Consequently, natural attenuation mechanisms operate in the soil microbial community that mitigate the spread of resistance vectors.

5.3.5 Summary Remarks

The WHO Working Group on the Risk to Health of Chemicals in Sewage Sludge Applied to Land (Dean and Suess, 1985) considered that organic chemicals may be ingested with soil by grazing animals, but concluded that: "the total human intake of identified organic pollutants from sludge application to land is minor and is unlikely to cause adverse health effects". Sauerbeck and Leschber (1992) also commented that despite an increasing level of investigation, the ecotoxicological significance of OCs in the soil-plant-water system and in the food chain had not been identified. The consensus view that emerges from the increasing body of scientific evidence concerning organic chemicals in sewage sludge-treated soil is that the risk they pose to human health and the environment is effectively negligible (Lester, 1983; Overcash, 1983; Davis *et al*., 1984; Dean and Suess, 1985; Jacobs *et al*., 1987; Rogers, 1987; O'Connor *et al*., 1991; Sweetman, 1991; Wild and Jones, 1991; USEPA, 1992b; UKWIR, 1995b; NRC, 1996; O'Connor 1996; Smith, 1996; Carrington *et al*., 1998b; Jones and Northcott, 2000; Smith, 2000a; Erhardt and Prüeβ, 2001; WEAO, 2001; Blackmore *et al*., 2006; Smith and Riddell-Black, 2007).

With improving methods of analysis and detection of organic chemicals it is possible that other OCs could emerge as a potential concern in the future (Muir and Howard, 2006). Consequently, it is important to adopt a vigilant, proactive and responsible approach to research in this area so that the significance of new developments and the potential risk to human health and the environment can be rationally assessed.

5.4 Radionuclides

Recycling sludge to land provides a potential pathway for radionuclides to enter soil and transfer to the human foodchain. Investigations of the potential impacts have been undertaken by Ham *et al*. (2003), Thorne and Stansby (2002) and Thorne (2003). In their study of the potential for sewage sludge to affect radionuclide concentrations in soil, Ham *et al*. (2003) introduced a screening approach based on radioisotopes expected to be discharged in waste water and their associated half lives. Drawing on data compiled by Titley *et al*. (2000) an estimate was made of the potential impact of radionuclides of sewage sludge from the Beckton sewage treatment works in the UK, if it were to be applied to land (rather than incinerated as is currently the case). These results are shown in Table 19 and indicate that the effect, in general, is likely to be undetectable or a few times above the detection limit (Butler, 2007).

Table 19 Potential increases in radioactivity arising from the disposal of Becton sewage sludge to soil (Ham *et al*., 2003)

Isotope	Conc. (sludge) (Bq kg^{-1})	Bq m^{-2} applied $^{(1)}$	Equiv. Soil conc. (Bq kg^{-1})	$^{(2)}$Estimated detection limit (Bq kg^{-1})
^{14}C	100	150	3.1	5
^{3}H	140	210	4.4	5
^{35}S	50	75	1.6	2
^{32}P	155	232	4.8	1
^{51}Cr	10	15	0.3	2
^{125}I	0.33	0.5	0.01	0.05
^{131}I	420	630	13.1	2

Notes: $^{(1)}$Based on application rate of 15 t ha^{-1}
$^{(2)}$Calculated at the 95% certainty level

5.5 Odour

Unpleasant odours can be a significant nuisance and sludge, which, given the nature of the material is potentially a source of malodour, is managed to mitigate this problem by stabilisation treatment to reduce its fermentability and by rapid incorporation into the soil (CIWEM, 1995). Malodour emissions often create negative reaction by the general public – one reason for this is that foul odour is associated with warning signs or indicators of potential risks to human health (Schiffman and Williams, 2005), but they are not necessarily considered to be direct triggers of health effects. However, odour sensations themselves have been recently linked to health symptoms. From a review of literature, Schiffman and Williams (2005) concluded that malodors emitted from large animal production facilities and wastewater treatment plants, for example, elicit complaints of eye, nose, and throat irritation, headache, nausea, diarrhea, hoarseness, sore throat, cough, chest tightness, nasal congestion, palpitations, shortness of breath, stress, drowsiness, and alterations in mood. At least three mechanisms by which ambient odours may produce health symptoms were identified. Firstly, symptoms can be induced by exposure to odourants (compounds with odour properties) at levels that also cause irritation or other toxicological effects. That is, irritation - rather than the odour itself - is the cause of the health symptoms, and odour (the sensation) simply serves as an exposure marker. Secondly, health symptoms from odourants at nonirritant concentrations can be due to innate (genetically coded) or learned aversions. Thirdly, symptoms may be due to a copollutant (such as endotoxin) that is part of an odourant mixture. For example, coomon sites of irritation and injury from odourants are the respiratory organs and the nose and increased frequencies of such respiratory and stress-related symptoms are found in the vicinity of animal production facilites (Nimmermark, 2004). Hydrogen sulphide and a number of other gases released from animal production units with hazardous properties are considered as possible contributory factors to the impact of odour on health, despite their low concentrations. The Water Industry recognises the importance of controlling odour emissions from sewage sludge treatment and agricultural application operations. In the first instance these largely dictate public acceptance. However, odourous emissions potentially released when sludge is applied to farmland are transient, and whilst these should be minimised as far as possible, the potential exposure and impact on health would appear to be minor compared to sustained exposures to individuals living in close proximity to centres of livestock production, for example.

6. ECOTOXICOLOGICAL AND ENVIRONMENTAL EFFECTS

6.1 Potentially toxic elements

6.1.1 Phytotoxicity

The potential effects of heavy metals on crop yields are an important concern when sewage sludge is recycled on agricultural land. Zinc, Cu and Ni are the principal phytotoxic elements applied to soil in sludge. There is an extensive literature defining critical plant tissue and soil limit concentrations for the phytotoxic elements (Kabata-Pendias and Pendias, 1992). For example, the risk assessment models for Zn, Cu and Ni, that formed the basis of the standards for these elements in the US EPA Part 503 Rule (US EPA, 1993) included an assessment of 271 articles on crop phytotoxicity (Chang *et al*., 1992; US EPA, 1992b,c). In Europe, soil limit values for these elements (CEC, 1986) were developed to ensure uptake into crops remains below critical toxic concentration thresholds in plant tissues (for comprehensive reviews, see Smith, 1996 and Carrington *et al*., 1998a,b). The permissible soil concentrations were established using sensitive crop species and coarse textured soils in field trials and plant pot experiments and therefore protect all crops grown on a range of soil types from phytotoxicity (eg Figure 20). Zinc and Ni uptake is pH sensitive, and increases with declining soil pH value, whereas Cu bioavailability is modulated to a much lesser degree by soil pH over the range of pH conditions acceptable for sludge application (≥5.0). Soil limits may also therefore be adjusted according to soil pH value to further minimise the risk of phytotoxicity under low pH conditions (Smith, 1994b).

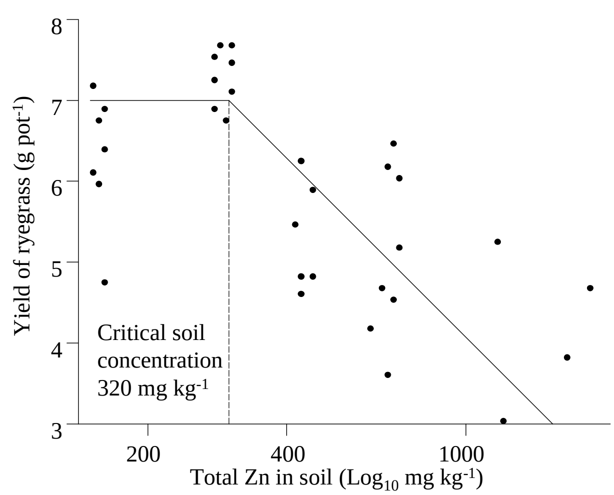

Figure 20 Yield of ryegrass in relation to soil Zn in a pot experiment (Davis and Carlton-Smith, 1984)

In the limited number of cases where reported, phytotoxicity in sludge-treated soils is almost exclusively associated with elevated Zn concentrations, albeit above the European maximum permissible limit (Carrington *et al*., 1998b). Only in exceptional circumstances is Ni phytotoxic and Cu toxicity in sludge-amended soil has been rarely reported (Chang *et al.*, 1987; Carrington *et al*., 1998b). Indeed, the risk assessment by US EPA (1992b) could identify no field evidence of phytotoxicity due to Cu or Ni in sludge-treated soil at any of the loading rates of Cu and Ni or soil conditions tested in different field experiments.

Evidence from long-term field investigations (Carlton-Smith and Stark, 1987) demonstrate that the current and revised maximum permissible soil limits for Zn are inherently protective of crop yields. The strength of the available technical information on phytotoxicity is further demonstrated by the consistency in upper critical phytotoxic soil limits obtained by independent field studies (Carlton-Smith and Stark, 1987; Carrington, 1998b) and the risk assessment of Zn, Cu and Ni (US EPA 1992b; US EPA, 1993) (Table 20). On the basis of this analysis, sludge could be applied indefinitely to an area of land without exceeding the upper critical phytotoxic concentrations of Zn, Cu and Ni in soil at the metal contents present in operationally produced sludges (Figure 6). From a technical stand-point, the EU soil limits for Zn, Cu and Ni (Table 7) are evidently highly precautionary and would protect crop yields even if soil conditions were to change once sludge applications had ceased, for instance, a significant decrease in soil pH value.

The current Directive, 86/278/EEC, regulating the agricultural use of sludge does not set an upper maximum limit for Cr in sludge-amended soil (CEC, 1986). Cr is identified as being potentially phytotoxic and Williams (1988) suggested that root crops may be potentially sensitive to Cr toxicity. However, the Cr(III) form that predominates in sludge-treated soil is virtually insoluble and is not taken up by plants (Cary *et al*., 1977; McGrath and Cegarra, 1992). Thus, studies of forage and root crops grown in soils of high total Cr content, up to 7000-15,300 mg Cr kg^{-1} (Carlton-Smith and Davis, 1983; Carlton-Smith and Stark, 1987; Smith *et al*., 1993), showed no effects on yield performance or uptake of Cr into crop tissues. A provisional soil limit for Cr is currently set in the UK of 400 mg Cr kg^{-1} ds (DoE, 1996) and the available scientific evidence on the effects of this element in sludge-treated agricultural soil on crops indicates that this a highly precautionary value.

6.1.2 Long-term impacts on soil fertility

Early investigations of the possible effects of PTEs on C and N mineralisation and nitrification processes found no detrimental impacts of heavy metals on microbial activity in sludge-treated soil (Premi and Cornfield, 1969; Premi and Cornfield 1971; Cornfield *et al*., 1976). Therefore, the argument that the current rules controlling agricultural utilization of sludge do not take account of soil microbiology (McGrath *et al*., 1994) is not strictly correct. Later research on sludge-amended soil from the Woburn Market Garden Experiment in the UK confirmed that C mineralization was not influenced by soil metal contamination (Brookes and McGrath, 1984). However, impacts were observed on the total soil microbial biomass and symbiotic N_2-fixation, at metal concentrations close to the EU soil limits (Brookes and McGrath, 1984; McGrath *et al*., 1988). Because there were a range of metals in the sludge-treated plots at Woburn, it was not possible to determine which metals were toxic to rhizobia (Chaudri *et al*., 1993). However, the high Cd content of the sludge-amended soil at Woburn could be a possible contributory factor to the apparent negative impacts on soil microorganisms observed at this site compared to soils supplied with contemporary, clean sewage sludges with low metals and Cd contents (McGrath, 1984; MAFF/DoE, 1993b; EA, 1999; also see: Chaudri *et al*., 1992; Giller *et al*., 1993; Chander *et al*., 1995). For example, McGrath *et al.* (1988) reported a Cd concentration in the soil receiving sewage sludge of 15.4 mg kg^{-1}, more than 5 times the EU legal limit in sludge-treated agricultural soil (Table 7; CEC, 1986). In a later review, McGrath *et al*. (1995) concluded that the upper range of lowest observed adverse effect concentrations (LOAEC) for Cd on soil microorganisms found in soils of six long-term field experiments was 6 mg Cd kg^{-1} ds, which further enforces the view that Cd may have contributed to the negative effects of heavy metals on soil microorganisms observed at this field site (Smith, 2000b). To put this into context, contemporary sewage sludges applied to farmland on average contain approximately 1.5 mg Cd kg^{-1}ds (Figure 7). By contrast, the maximum mean Cd concentration in the sludge applied to the Woburn field experiment recorded for the period 1957-61 and was 137 mg kg^{-1} DS (McGrath, 1984).

Table 20 **Soil and sludge PTE concentrations which protect against phytotoxicity relative to concentrations in sludge and soil limit values (mg kg^{-1} ds) (Carrington *et al*., 1998b)**

PTE	Predicted[(1)] or max[(2)] soil concentrations at UK historic sites	EU maximum soil limits (pH 6-7)	Soil content based on 503 cumulative application	503 Pollutant concentration	Mean UK sludge content (2005 data)
Zn	1290[(1)]	300	1500	2800	612
Cu	850[(2)]	140	775	1500	309
Ni	515[(2)]	75	230	420	39

[(1)]Based on a crop (ryegrass) uptake model derived from field data, an upper critical tissue concentration of 200 mg kg^{-1} dm and soil pH value of 6.0 where Plant Zn_{log} = 1.5SoilZn_{log} + 0.21pH – 0.17SoilZn_{log}.pH – 0.53 (r^2 = 0.68, n = 156) (see Smith, 1997a)

[(2)]Ryegrass contained less than the upper critical tissue concentrations of Cu and Ni (40 and 90 mg kg^{-1} dm) in all cases and soil concentration values are those associated with the highest recorded plant tissue contents.

From a recent review of literature, Vig *et al*. (2003) concluded that there were conflicting results regarding the apparent toxicity of Cd to soil microorganisms. Evidently there are potentially negative impacts of Cd on soil microbial parameters, but these only become apparent at soil Cd concentrations significantly above the current EU limit of 3 mg Cd kg^{-1} (Vig *et al*., 2003).

An Independent Scientific Committee, convened by the UK Government to review the evidence on soil fertility aspects of PTEs in sludge-treated soil (MAFF/DoE, 1993b), concluded that Zn was the element potentially responsible for the observed effects of sludge on soil microbial processes. A decision to reduce the UK soil limit from 300 mg kg^{-1} (pH 6-7) to 200 mg kg^{-1} (pH 5-7) was taken on precautionary grounds largely based on results from a sludge-treated field trial at Braunschweig, Germany, which showed declining numbers of free-living *Rhizobium leguminosarum* biovar *trifolii* above a soil concentration of 200 mg Zn kg^{-1} (Chaudri *et al*., 1993). However, the potential significance and impacts of PTEs on the fertility of sludge-treated agricultural soil has been a controversial subject (McGrath and Chaudri, 1999; Smith, 2000b). This is because research on a wide range of other sludge-treated soils has not supported the link between metal contamination of soil and toxicity to the microbial biomass or *Rhizobium*. For example, no adverse effects of Zn on the microbial biomass content of soil were detected in potentially vulnerable, course-textured soils containing up to 450-600 mg Zn kg^{-1} across the pH range 5.4-7.0 (Figure 21; Smith, 1995; Smith *et al*., 1999), which is up to 3 times the EU maximum permissible limit for this element (Table 7; CEC, 1986). Furthermore, no consistent effects of soil PTEs on the absence of *Rhizobium* were apparent in long-term, sludge-amended field soils and free-living bacteria were routinely isolated from sites with significantly elevated PTE concentrations above the soil limits (Smith, 1997b). Rhizobia are adapted to live in the free state and can survive for a long periods in soil (McGrath *et al*., 1995). However, many factors influence the presence/absence of *Rhizobium* sp in soil in the practical field situation and the initial assumption that clover rhizobia are ubiquitous in UK soils has been shown to be incorrect (Giller *et al*., 1998). One of the most important factors governing the presence of the free-living bacteria in soil is the period of time that has lapsed since the host crop was grown at a particular site (Nutman, 1975). However, rhizobia are always present in soil and fix N effectively when the host plant is present, irrespective of the soil metal content (Smith and Giller, 1992; Obbard and Jones, 1993), and they may become absent simply because the host plant has not been established at the site for a long period. Other species of symbiotic N fixing bacteria appear to have much lower sensitivities to metals compared with the white-clover bacteria and the extent to which indigenous strains are tolerant to metals may be highly site specific (El Aziz *et al*., 1991; Angle *et al*., 1993; Smith and Giller, 1992). Some of the key issues to emerge from the research on the effects of metals on microorganisms in sludge-treated soil are that:

- Critical nutrient mineralisation and nitrification processes are not affected in soils with elevated PTE concentrations in excess of EU soil limits;
- Zn has emerged as the only element of potential concern with regard to long-term effects on soil fertility in the context of European legislation;
- The microbial biomass content of soil is not impacted by Zn above the EU maximum limit value;
- The variation and unique characteristics of particular species, groups or types of microorganism at specific sites mean that results from one field site or experiment may not be applicable for general extrapolation to other soils;
- There is no definitive assessment of the agronomic or ecological significance of microbial-PTE relations in sludge-treated soil.

More recently, Speir *et al*. (2004) examined the effects of PTEs in sewage sludge treated soil on a broad range of soil ecological indicators in a 4 year field investigation. Composted sewage sludge was supplied up to a maximum rate of 200 t ha^{-1} y^{-1} DS to a sandy loam soil at two pH ranges (5.0-5.5 and 6.0-6.5). The maximum total metal concentrations in the soil were: 184 mg Zn kg^{-1}, 101 mg Cu kg^{-1}; 19 mg Ni kg^{-1} and 0.63 mg Cd kg^{-1}. The mean background concentrations in the unamended control soil were: 45 mg Zn kg^{-1}, 14.5 mg Cu kg^{-1}, 12 mg Ni kg^{-1}, 0.18 mg Cd kg^{-1}. Soil basal respiration, microbial biomass C and anaerobically mineralisable N were significantly increased in the compost-amended plots relative to the control. Sulphatase and phosphatase activities also increased, although not significantly. There were no effects of sludge addition on the numbers of *Rhizobium*, free-living in the soil, or on sensitive microbial biosensors (Rhizotox C and *lux*-marked *Escherichia coli*). The application of sewage sludge was shown to enhance soil fertility, productivity and microbial biomass and activity, with no apparent adverse effects attributable to heavy metals. The importance of this work is particularly emphasised because the soil Zn concentration was within the range of the UK soil limit values (200 mg Zn kg^{-1} at pH 5.0 – 7.0; DoE, 1996).

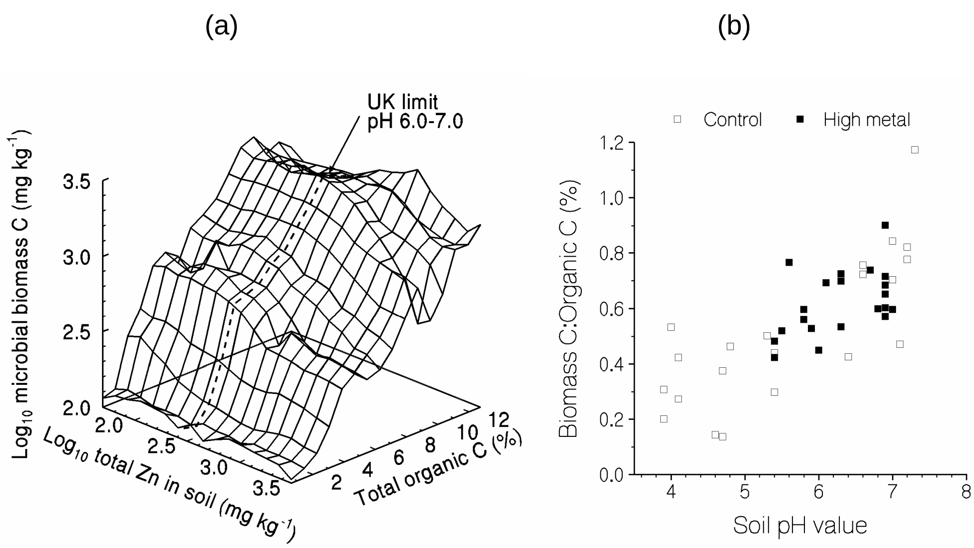

Figure 21 **Microbial biomass content of long-term sludge amended soils in relation to total Zn concentration and (a) organic C content in 13 operationally treated historic sites, n=211, (Smith, 1995), (b) pH value in a field experiment with low and high Zn soils (450 mg Zn kg^{-1} ds) of equivalent loamy sand textural class (Smith *et al*., 1999)**

Broos *et al*. (2005a) examined the survival of rhizobia in soils with elevated Zn and Cd concentrations from equilibrated soils that had received applications of sewage sludge or metal salts at least 10 years previously. Cd had no effect on numbers of rhizobia in soil at the maximum concentration of this element studied of 118 mg Cd kg^{-1} in a neutral pH soil containing about 34 % clay. Increasing Zn, on the other hand, significantly reduced survival, but this occurred at total concentrations above, or well above current EU and UK soil limits. Thus, rhizobia numbers declined significantly at 233 mg Zn kg^{-1} at pH 5.6 and 876 mg Zn kg^{-1} at pH 6.3. Therefore, the results appear to support the current soil limits for sewage sludge amended agricultural soil and indicate the limit values for Cd and Zn protect sensitive rhizobia bacteria from metal toxicity. Furthermore, these thresholds were lower than corresponding values for toxicity to symbiotic N_2-fixation by white clover (Broos *et al*., 2004).

In Australia, a major national programme of field investigations aims to derive metal limit standards for sludge-treated agricultural soil appropriate to Australian conditions (the National Biosolids Research Program), and this has also included measurements of soil microbial parameters (Broos *et al*., 2006; Broos *et al*., 2007). Data are reported for two microbial indices, including substrate induced nitrification (SIN) and substrate induced respiration (SIR) from twelve field experiments on representative soil types from around Australia amended directly with $ZnSO_4$ and $CuSO_4$ salts. The microbial techniques were selected as they have been considered to combine the advantages of relative sensitivity and robustness (Broos *et al*., 2005b) and also have been applied in recent European projects on the microbial ecotoxicity of heavy metals in soil (Smolders *et al*., 2004; Oorts *et al*., 2006). Broos *et al*. (2007) calculated median EC_{50} values for sites where a significant toxic effect was detected. The median EC_{50} values for SIN and SIR for Zn were approximately 560 and 2000 mg Zn kg^{-1} ds, respectively. The median EC_{50} for SIN and SIR for Cu was 560 and 900 mg Cu kg^{-1}, respectively. Using the EC_{50} value as the end-point for deriving soil limits could be criticised, however, as not being sufficiently protective of the soil microbial community since it represents a 50 % loss in performance, which is a apparently a large reduction in activity. However, this is necessary to determine the toxicity-threshold soil concentration for an element because the intrinsic variability apparent in measuring soil microbial parameters requires relatively large changes in the microbial dose-response to obtain a statistically significant reduction in activity compared to the untreated control. Indeed, Broos *et al*. (2006) earlier commented that only in one case was there a statistically significant decrease in the EC_{10} value for SIR and SIN reported directly after application compared to the control, despite the extreme increases in soil Zn and Cu concentrations from salt addition up to maximum values of 7490 mg Zn kg^{-1} ds and 5899 mg Cu kg^{-1}. The lowest observed EC_{50} values recorded were 107 mg Zn kg^{-1} ds and 108 mg Cu kg^{-1} ds. However, these are probably extreme and unrepresentative values compared to what may be expected under European conditions. This is because the dose-response was determined using metal salts, which have potentially greater bioavailability and ecotoxicity compared to sludge-bound metals, and many of the soil types in Australia are considerably more vulnerable to metal toxicity having low or extremely low organic matter contents and acidic pH. Interestingly, the toxicity thresholds were also found to increase over time presumably due to the reaction and transformation of the metals to less bioavailable forms with time (Broos *et al*., 2006). On balance, therefore, the median EC_{50} values obtained from this work would seem to provide inherently conservative estimates of the toxicity-thresholds of Zn and Cu, and a tentative comparison with European soil limits suggests that these microbial endpoints will be protected at the current maximum permissible concentrations for these elements in sludge-treated agricultural soil.

A series of long-term experiments were established in 1994 at nine sites throughout Britain, representing variations in climate and soil conditions to quantify the effects of heavy metals on the long-term fertility of sewage sludge-amended agricultural soil (Chambers, 2006; Gibbs *et al*., 2007; UKWIR, 2007). The programme was undertaken in response to one of the key recommendations from the *Review of the Rules for Sewage Sludge Application to Agricultural Land. Soil Fertility Aspects of Potentially Toxic Elements* by the Independent Scientific Committee in 1993 (MAFF/DoE, 1993b), that further research was needed to examine the effects of heavy metals from sewage sludge on soil microorganisms to provide a robust and informed basis to setting soil metal limits. Metal-rich sludge cakes, metal salt-amended liquid sludges and inorganic metal salts were applied directly to the soils to provide a range of soil concentrations that encompassed the current EU/UK maximum soil metal limits for Zn, Cu and Cd. An important feature of the experimental design was that the organic matter inputs to the cake sludge plots were balanced across all the treatment levels including the control, using a contemporary low metal sludge. The effects on soil microbial biomass, respiration rate and numbers of rhizobia, free-living in the soil, were measured. Metal salt additions to soil, whether directly applied or added to the liquid sludge were expected to provide a worse-case, and arguably unrepresentative, increase in bioavailability and potential toxicity compared with sludge application to soil in operational practice (Logan and Chaney, 1983; Giller *et al*., 1998; also see Smolders *et al*., 2004). There are often pronounced differences in the toxicity response of microorganisms to metals in long-term equilibrated soils compared with artificially 'spiked' soils, which are considerably more sensitive to the toxic effects of metals (Olson and Thornton, 1982; Doelman, 1986; Smolders *et al*., 2004; Laguerre *et al*., 2006). Nevertheless, no consistent effects of the Zn metal salt additions, either directly or applied with liquid sludge (Gibbs *et al*., 2006b), have been found on any of the soil microbial parameters measured, despite increasing the ammonium nitrate extractable metal fraction in the liquid sludge plots above that of the cake sludge-amended plots in the early years following application. By contrast, decreases in numbers of rhizobia and microbial biomass have occurred at a number of the sites receiving the high Zn sludge cake. The reason why the Zn cake sludge is apparently more toxic to soil microbes, but not the high bioavailability metal salt treatments, may be explained by the large metal content of the Zn cake sludge. The Zn sludge cake contained approximately 6000 mg Zn kg^{-1} DS, equivalent to almost 10 times the average concentration found in contemporary sewage sludges applied to farmland (Figure 6). The apparent total Zn concentration measured in soil amended with sludge cake is a function of the averaging effect of the high metal sludge particles mixing through the soil profile and are not a true reflection of the actual microbial exposure to Zn at the microscopic scale. The average Zn concentration measured in the bulk soil is not what the organisms actually 'see' - they are exposed to the high metal concentration particles of sludge containing 6000 mg kg^{-1} of Zn or at the 'micro' scale. In other words, the sludge itself may be toxic to soil microorganisms due to the excessive metal concentrations.

Copper also caused negative impacts on rhizobia numbers and microbial biomass in the British long-term field experiments, but in this case both metal salts and high Cu sludge cake were toxic. The Cu in the sludge cake applied to the trials was approximately 5050 mg Cu kg^{-1} DS, which is 17 times the average concentration found in sludge currently applied to farmland in the UK (Figure 6). Copper is biocidal to microorganisms, hence its use eg as a fungicidal agent (Bunemann *et al*., 2006). However, Cu readily complexes with the mineral and organic matter phases in soil (Chaudri *et al.*, 1992) and, provided that the soil and sludge binding capacity is not exceeded, its bioavailability is usually relatively low in sludge-treated soil (Smith, 1996). Therefore, it seems plausible that the toxic response to Cu applied in sludge cake was probably explained by the same mechanisms as for Zn (for example, see Landner *et al*., 2000) and the effects of inorganic salts were due to the toxicity of soluble inorganic forms of Cu to microorganisms (Bunemann *et al*., 2006). However, as may be expected from earlier field and laboratory investigations, no effects of Cd on the soil microbial biomass, respiration rate or rhizobial numbers have developed in the long-term field experiments at the maximum concentration of this element in soil of 4 mg Cd kg^{-1} ds.

The 'critical' concentration of Cu for agricultural soil of 60 mg kg^{-1} ds proposed by Witter (1992, also see the review by Landner *et al*., 2000) to protect soil microorganisms has not been considered generally applicable to sludge-amended agricultural soil. This is because Cu has not been identified as a major concern at the current maximum permissible EU limit value for this element in long-term field experiments with sensitive soil types and in laboratory studies under worse-conditions in soil spiked with metal salts or metal-enriched sewage sludge (MAFF/DoE, 1993b; Chaudri *et al.*, 1992; Chander *et al*., 1995). No significant effects on the soil microbial biomass content or metabolic quotient have been detected of increasing Cu concentrations up to 460 mg Cu kg^{-1} ds in sludge-treated soil or from long-term application of distillery waste, which, in contrast to sludge, only enriches soil with Cu (Smith, 1994c). From the review recently completed by Landner *et al*. (2000), it is apparent that the maximum loading rate for Cu stipulated in the EU Directive controlling the agricultural use of sludge of 12 kg Cu ha^{-1} (CEC, 1986) does not constitute any major risk for adverse effects on crop yields, soil invertebrate fauna, health of humans and domestic animals, nor on the fertility and microbial activity of soil. Consequently, the weight of scientific evidence indicates that Cu is unlikely to present a significant risk to soil microbial processes at the current EU limit value for this element.

Evidence from other field experiments show that sludges containing excessive concentrations of Zn are phytotoxic to crop plants (Marks *et al*., 1980; Bhogal *et al*., 2003; De Brouwere and Smolders, 2006). Corey *et al*. (1987) proposed that sludge properties, and the metal concentration in particular, strongly influenced metal release and bioavailability in soil and this is confirmed by recent literature (Merrington *et al*., 2003). De Brouwere and Smolders (2006) measured a significant yield reduction in wheat grain yield compared with the control after applying lime-amended sludge containing 6000 mg Zn kg^{-1} DS to the plots in the previous year and 1450 mg Zn kg^{-1} prior to drilling the crop. Interestingly, the sludge applied by Brouwere and Smolders (2006) had the same Zn content as that used in the long-term sludge experiments in Britain. However, there were no indications of phytotoxicity in a barley crop grown in the following year treated with sludge containing much smaller amounts of Zn (480 mg Zn kg^{-1} DS). The bulk soil total Zn concentration (130 mg Zn kg^{-1} ds) was too small to explain the yield reductions. Therefore, De Brouwere and Smolders (2006) concluded that, in the initial years, sludge and soil were not homogeneously mixed and plant roots could be exposed to locally elevated Zn sludge concentrations that were in the toxic range. A similar explanation could account for the observed toxicity to soil microbes of the high Zn sludge cake in the long-term field experiments in Britain. In contrast to the effects on plant growth, which occurred soon after sludge application, toxicity to microorganisms in the long-term experiments only manifest itself <u>after</u> the final Zn sludge cake application (Gibbs *et al*., 2006c; Chambers, 2006; Gibbs *et al*., 2007; UKWIR, 2007). Plant roots are likely to be affected by metals in large sludge particles, and the effects may diminish as the particles break down with soil cultivation and become increasingly diluted throughout the soil profile, thus reducing the exposure to the high metal concentrations. At the microscopic level, however, this process could have exactly the opposite effect. Relatively few microorganisms would be initially exposed to high metal concentrations when the sludge is present in large particles soon after it is applied. However, cultivation and the activity of soil invertebrates will cause breakdown into smaller particle sizes and gradually more of the microbial community will become exposed as the high-metal particles of sludge disperse and mix into the soil. It is therefore possible that a point would be reached where the level of dispersion of sludge particles and exposure to the localised high concentrations of metals will be sufficient to impact on the soil microbial community.

The lack of a toxic Zn response on the metal salt plots, where metals are uniformly distributed within the soil profile and not present in high concentration particles, represents the true 'average' effect of the soil metal concentration on soil microorganisms. Representative operationally produced sludges, with typical background concentrations of Zn and Cu (Figure 6), were also applied, however, there is no evidence of toxicity in these soil treatments, although total metal levels where not elevated to anywhere near the maximum soil Zn (or Cu) limit values. Therefore, since the metal salt treatments, and the application of operational sludge to soil, had no effects on the soil microbial community, it is possible to conclude from these long-term experiments that:

1. Current EU maximum permissible the soil limits are protective of soil microbial processes and long-term soil fertility;
2. Sludges produced and land applied under current operational practice are safe and non-hazardous to the soil microbial community.
3. The apparent toxicity of the cake sludges to soil microorganisms may be explained by localised exposure to high metal concentrations at the microscopic scale.

A document setting the results from the UK long-term field experiments into a policy context (Defra, 2007b) concluded reasonably that, overall, immediate regulatory changes as a result of the experimental results were unwarranted. As may be expected, the results would be considered when taking into account current policies relating to the sustainable application of sludge to land.

6.2 Organic contaminants

6.2.1 General Impacts on soil ecology

Toxic effects of OCs to plants can be induced under artificial laboratory culture, using *in vitro* and hydroponic test conditions (eg Harms and Kottutz, 1992; Sweetman *et al*., 1992; Harms, 1996; Bradel *et al*., 2000 cited in Jjemba, 2002). However, this does not reflect the actual behaviour and interactions that occur in sludge-amended soil systems, which significantly reduce bioavailability of OCs to soil organisms (Section 6.3.2). Consequently, there is no evidence that OCs are phytotoxic at the concentrations that are likely to occur in operationally sludge-treated agricultural soil (eg Shea *et al*., 1982; Overcash, 1983; de Wolf and Feijtel, 1998; Jensen, 1999; Roberts *et al*., 2006; Smith and Riddell-Black, 2007). For example, a recent three year programme of field and glasshouse investigations in Denmark (Petersen *et al.*, 2003) examined the effects of sludge and other organic residuals on soil properties and soil biota, the fate of selected OC (which included four principal groups or types of OC present in sludge: PAHs, DEHP, NP + NPE and LAS, and their potential for plant uptake. No accumulation of the OCs in soil or plant uptake were detected and no adverse effects of waste application on soil biota or crop growth were found (Petersen *et al.*, 2003).

There are very few reports of toxicity of OCs to soil microorganisms at representative environmental concentrations. For example, Chaudri *et al*. (1996) selected 10 organic compounds (LAS, NP, toluene, Aroclor1016, 2,4-dichlorophenol, ethylbenzene, phenol, anthracene, trichloroethylene and pentachlorophenol (PCP)) representing a wide range of chemical classes, including those that potentially present the highest risk to soil microorganisms, and added them directly to soil without sludge. Numbers of the N_2-fixing bacteria, *Rhizobium leguminosarum* bv. *trifolii*, free-living in the soil were measured after an incubation period and, of the different compounds tested, only PCP was toxic and significantly reduced the indigenous soil population. The conditions of the test were designed to represent a worse-case and, for PCP for example, the amount of compound added to the potentially vulnerable sandy loam soil was 200 mg kg^{-1}, which is the maximum concentration of PCP found in sewage sludge. However, PCP was not detected in any influent samples in a screening study of UK sewage treatment works with a detection limit of 2 $\mu g\ l^{-1}$ (Bowen *et al*., 2003), therefore its presence in sludge in significant or potentially toxic concentrations is unlikely. PCP has been principally used for timber treatment and as a textile preservative, however its production in the EC was banned in 1992 and its use as a chemical intermediate in the chemical industry was banned in 2000 (Bowen *et al*., 2003). PCP is also a WFD priority substance under review (EPCEU, 2001).

There is no evidence of a direct impact of persistent organic pollutants (POPs) on soil ecological systems (Smith and Riddell-Black, 2007). Roberts *et al*. (2006) reported no effects of NP on soil respiration except at exceptionally high soil concentrations (>10000 mg kg^{-1}). After reviewing the available scientific literature, De Wolf and Feijtel (1998) and Jensen (1999) concluded that LAS was not toxic to terrestrial biota. No ecotoxicological impacts of antibiotics or pharmaceutical compounds are expected at the small concentrations likely to occur in sewage sludge-amended agricultural (Smith and Riddell-Black, 2007). Therefore, overall the available evidence indicates that OCs do not present a ecotoxicological risk to soil microorganisms or fauna from agricultural recycling of sewage sludge (McGrath, 2000; Smith and Riddell-Black, 2007). This contrasts markedly with the biocidal effects on soil microorganisms reported for a range of pesticide compounds licensed for use in agriculture (Smith and Riddell-Black, 2007).

6.2.2 Linear alkylbenzenesulphonate (LAS)

The total consumption of surfactant chemicals for industrial and domestic purposes was 1.7 M t in 2000, 85 % of which were used in domestic products. The chemical LAS is a low to moderately toxic, non-bioaccumulative, high production volume anionic surfactant, employed in detergents and cleaning products. It was introduced in 1964 as the readily biodegradable replacement for highly branched alkylbenzene sulfonates (ABS) (www.lasinfo.org). It is presently used at rates of 350,000 - 400,000 t y^{-1} in Western Europe. Potentially large concentrations of LAS can occur in sludge due to: high usage per capita, sorption and precipitation of the compound onto sludge solids and low degradability during anaerobic digestion. However, LAS biodegrades rapidly in aerobic soil (Table 8) with primary and ultimate half-lives of up to 7 and 30 days, respectively (Holt *et al*., 1989; Figge and Schöberl, 1989; Jensen, 1999; Prats *et al*., 1999; HERA, 2004; Prats *et al*., 2006). LAS is not generally regarded as toxic; it is not on the 'list of priority substances in the field of water policy' of the Water Framework Directive (WFD) (EPCEU, 2000a), or the European Pollutant Release and Transfer Register (EPCEU, 2006) or recognised as a chemical of concern by the UK Chemical Stakeholder Forum (Defra, 2005).

Early uncertainties over the potential environmental impacts of LAS in sludge-treated soil prompted the Danish EPA to introduce a quality standard for agricultural application (2600 mg LAS kg^{-1} dry solids (DS) from 1997, which was reduced to 1300 mg LAS kg^{-1} DS in 2000) based on a risk assessment using the ecotoxicological data available at the time and a $PNEC_{Soil}$ (Predicted No Effect Concentration is the chronic HC_5 hazardous concentration protecting 95% of soil species) of 5.2 mg LAS kg^{-1} dry soil (ds) (SPT, 1999, also see Jensen *et al*., 2001a). This was a controversial decision in the light of an apparently substantial amount of evidence demonstrating that there were no tangible ecotoxicological effects of LAS (eg de Wolf and Feijtel, 1998). Another reason for the apparently conservative limit value estimated for LAS was that the rapid biodegradation of the compound was not considered in the initial risk assessment (Erhardt and Prüeβ, 2001). Nevertheless, the European Commission proposed to include the upper limit for LAS adopted in Danish regulations in a revision of Directive 86/278/EEC for agricultural use of sludge (CEC, 1986; EC, 2000) – see Table 11. However, a revised risk assessment on the compound has been recently published by the Expert Group on Risk Assessment of Sewage Sludge within the Environment and Health Task Force of the International Life Sciences Institute (ILSI) – Europe (Schowanek *et al*., 2007), based on the conceptual framework for risk assessment for organic chemicals in sewage sludge for agricultural use proposed by Schowanek *et al*. (2004). Fundamental to this is that the species-sensitivity distribution for LAS has been more comprehensively profiled, which this has lead to a new $PNEC_{Soil}$ value of 35 mg LAS kg^{-1} ds. Risk analysis procedures in Europe have also been refined and standardised (EC, 2003c) and these recommendations, including, for example, taking account of the biodegradability of a compound in soil, have been incorporated into the revised risk assessment for LAS (Table 21). A Monte Carlo procedure was used to deal with multiple sources of variability and the sludge limit value derived from these calculations was approximately 50,000 mg LAS kg^{-1} DS. This value is significantly higher than the European average concentration for LAS in anaerobically digested sludges of 5564 mg kg^{-1} DS (Table 9) and the maximum value reported recently for LAS in UK sludge of 10,500 mg kg^{-1} DM (Bowen *et al*., 2003). Therefore, this revised assessment emphasises that there is no risk identified for LAS in sludge. Indeed, Jensen *et al*. (2007) concluded that LAS does not represent an ecological risk in Western Europe when applied to soil in sewage sludge under normal practice.

Other comprehensive risk assessments on LAS have also been completed or updated recently (HERA, 2004; OECD, 2005). These independent studies also concluded that the ecotoxicological parameters of LAS have been adequately and sufficiently characterised and that the ecological risk of LAS is low.

HERA (2004) has completed a programme of risk assessments for compounds in household cleaning products, concluding they do not generally represent a risk to human health or the environment. However, policies or controls that would ultimately limit the use of LAS would inevitably lead to its substitution with alternative surfactant compounds that are potentially less well characterised or understood in terms of their possible impacts on soil.

Table 21 Summary of key risk assessment parameter values in the EU TGD (EC, 2003c), the initial agricultural soil risk assessment of LAS by Danish EPA and the recently revised calculation by ILSI[1] (the potential variability across the EU is also shown) (Jensen *et al*., 2007; Schowanek *et al*., 2007)

Parameter	TGD Parameters	Danish EPA	ILSI[1]: Distribution of parameters for sensitivity analysis
$PNEC_{Soil}$ LAS	Value determined by experimental investigation and soil testing	5.2 mg kg^{-1}	35 mg kg^{-1}
Sludge application rate	5 t DS ha^{-1} y^{-1}	6 t DS ha^{-1} y^{-1}	Triangular distribution (trigen)[2] with 4000 as mode, 2500 as 5th percentile and 10,000 kg/ha as 95th percentile (truncated at 1500 and 13,000 kg/ha)
Soil bulk density	1500 kg m^{-3}		Lognormal distribution with mean = 1350 kg m^{-3}, sd 150 (truncated at 1000 and 1700 kg m^{-3}
Soil depth	0.2 m (0.1 for grassland)(assumes homogeneous mixing)	0.1 m	0.15–0.25 m: trigen distribution with mean = 0.2 m (truncated at 0.15 and 0.25m)
Sludge/soil dilution factor	600	250	Calculated as dependent variable
Averaging time to calculate initial exposure	30 days	Not considered	30 days
Biodegradation half-life in soil	Primary biodegradation: $T_{0.5}$ = 10 d (k = 0.069 day^{-1})[2] Mineralisation: T0.5 = 30 d (k = 0.023 day^{-1})	Not considered	Primary biodegradation: lognormal distribution of half-life with mean = 10 days and sd = 5 days Mineralisation: lognormal distribution with mean = 30 days, sd= 15 days (for information, data not presently used in the simulations)
Sludge quality standard		1,300 mg kg^{-1} DS	50,000 mg kg^{-1} DS

(1) Interational Life Sciences Institute Europe (see Schowanek *et al*., 2007)
(2) Trigen is a type of triangular distribution in the @RISK software
(3) The TGD (EC, 2003c) uses a half-life of 30 days as a default value, which was adjusted to 10 days here based on field monitoring data

6.2.3 Phthalates

Phthalates enter sludge by leaching from plastic materials in the built environment and are present in relatively high concentrations (Tables 8 and 9). In the environment, phthalates are degraded by a wide range of bacteria and actinomycetes under both aerobic and anaerobic conditions (Staples *et al*., 1997). Kirchmann *et al*. (1991) reported there were no effects of DEHP in soil on important soil microbial parameters including respiration, N mineralization and nitrification. Cartwright *et al*. (2000) showed that, even at concentrations as high as 100 g kg^{-1} in soil, DEHP had no effect on the structural diversity (bacterial number and fatty acid methyl ester analysis) and functional diversity (BIOLOG) of the soil microbial community, or on cell membrane integrity, and concluded therefore that the DEHP would have no impact on soil microorganisms. Diethyl phthalate (DEP) had no impact on soil microbes at a soil concentration of 0.1 g kg^{-1}, but did cause perturbation at concentrations >1 g kg^{-1} soil. However, DEP was biodegraded very rapidly in soil with a half-life of 0.75 days at 20 ^{o}C and, in sludge-treated soil, phthalate concentrations would not be increased to this extent in practice (Vikelsøe *et al*., 2002). Accumulation of DBP and DEHP has been reported in some mesofauna suggesting a potential risk to these organisms (e.g. Hu *et al*., 2005) and the potential for bioaccumulation through natural foodchains, although the compounds were artificially introduced into soil by direct spiking without sludge, which may exaggerate their apparent bioavailability. However, no significant adverse effects of DEHP or DBP were observed by Jensen *et al*. (2001b) on soil collembolan.

Phthalates are suspected EDCs and whilst emissions of chemicals exhibiting this behaviour on surface water habitats have potentially important consequences for aquatic life (EA, 1988), the significance of endocrine disruption for soil ecology has not been identified, and is likely to be much less important. Van Wezel *et al*. (2000) considered how effects on the endocrine/reproductive system should be incorporated into Environmental Risk Limits (ERL) for phthalates and performed *in vitro* and *in vivo* tests with these endpoints for a range of phthalate compounds (ERLs are concentrations of a substance in water, air, sediment and soil that are expected to be protective of the environment). However, for those compounds which demonstrated endocrine disruptive effects (DBP, DEP, DHP, BBP, DEHP, DIBP, MEHP), the relative potency compared to the potency of the natural estrogen, 17β-estradiol, was very small low (10^{-4} – 10^{-8}). Van Wezel *et al*. (2000) concluded, therefore, that dose response relationships obtained from conventional ecotoxicity tests give sufficient protection against endocrine disruptive effects. The ERLs obtained for DBP and DEHP in soil/sediment were 0.7 and 1.0 mg kg^{-1} soil (fresh weight). The ERL for DEHP, for example, is more than 25 times greater, than the concentration measured by Vikelsøe *et al*. (2002) in soil regularly receiving applications of sludge at typical agronomic rates.

In conclusion, phthalates are one of the main groups of OC present in sewage sludge. However, research into their potential impacts on soil microorganisms and macrofauna has shown that, as with other groups of organic compounds present in sludge, they are not ecotoxic when applied to the soil at the concentrations found in contemporary, operationally produced sewage sludges.

6.2.4 Triclosan and other personal care products

Triclosan is a chlorophenol compound and is used for its antibacterial properties, as an ingredient in many detergents, dish-washing liquids, soaps, deodorants, cosmetics, lotions, anti-microbial creams, toothpastes, and an additive in plastics and textiles. Consequently it can be found in low to moderate concentrations in sewage sludge. For example, Ying and Kookana (2007) reported the maximum concentration of triclosan in Australian sewage sludges was 17 mg kg DS^{-1} with a median content of 2.3 mg kg^{-1} DS. The safety of triclosan has been questioned in regard to environmental and human health and a terrestrial risk assessment of tricolsan recently completed by the Danish EPA (Samsøe-Petersen *et al*., 2003) indicated that the concentrations measured in sewage sludge (Section 4.3.5) could have negative effects on soil organisms immediately after application. Other antimicrobial agents are also extensively used in personal care products and may also have potential implications for soil health when applied in sewage sludge. Consequently, these would seem to be an environmentally undesirable group of substances and there appear to be strong grounds to question the purpose and rationale for there use in domestic and other products and the possible need for source control measures to be implemented. Nevertheless, a recent laboratory investigation (Pipe, 2007) showed no significant effects of tricolsan and 2 other important biocidal personal care compounds (butylparaben and methylchloroisothiazolinone) on the microbial biomass content of a sensitive sandy loam soil after the first week of application at concentrations 100 and 1000 times greater than the highest recorded amounts of triclosan measured in sludge, and assuming an application rate of 10 t DS ha^{-1} and incorporation depth of 10 cm (Figure 22). No significant effects of the personal care chemicals on soil microbial biomass were detected in a sandy silt loam soil. A possible explanation for the apparent low toxicity of triclosan and the other personal care products in soil may be related to their strong adsorption to the soil organic and mineral phases and the rapid biodegradation of the compounds in the soil environment. Considering the 'averaging' times (30 days) accepted for environmental risk assessment of chemicals (EC, 2003c) and the high concentrations used in this study, there would appear to be little cause for concern from personal care products applied to soil in sewage sludge under normal operational conditions. However, further work is necessary to examine the impacts of antimicrobial personal care products on soil health, such as the common antiseptic agent triclocarban. Heidler *et al*. (2006) found the average concentration of triclocarban in sludge from a large representative sewage treatment works in the US was 51 +/- 15 mg kg^{-1}; approximately 75 % of the mass of triclocarban disposed of by consumers in the catchment of the plant was ultimately is released into the environment by application of sewage sludge on land including for agricultural use.

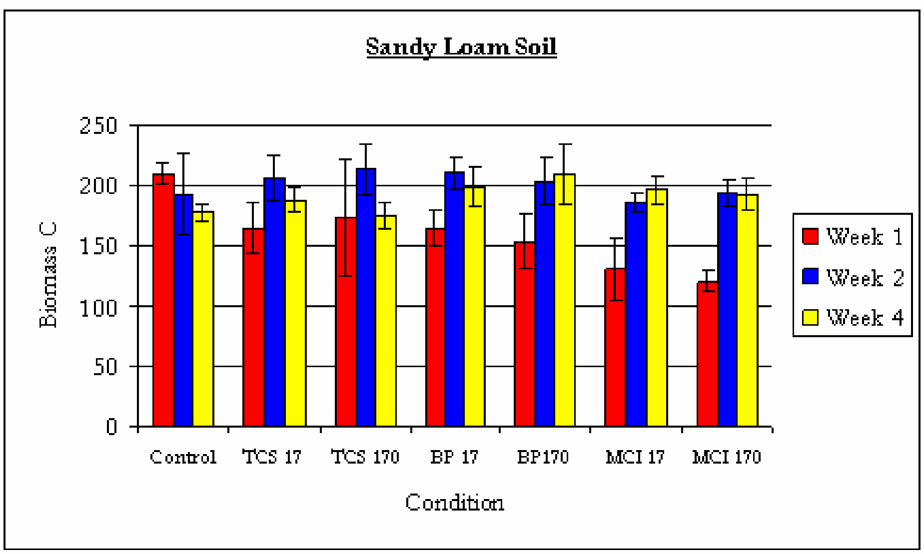

Figure 22 Microbial biomass C concentration in sandy loam soil (mg C kg^{-1} ds) after incubation for 1, 2 and 4 weeks with 3 antimicrobial personal care products (TCS: triclosan, BP: butylparaben, MCI: methylchloroisothiazolinone) applied at rates of 17 and 170 mg kg^{-1} ds (Pipe, 2007)

Nitro musks (chloronitrobenzenes), a group of synthetic dinitro- or trinitro-substituted benzene derivatives, and polycyclic musks, are used as synthetic fragrance ingredients in perfume and personal care products, washing agents etc. and therefore have obvious routes into the wastewater stream. Tas *et al*. (1997) completed an environmental risk assessment of the nitromusks: musk ketone and musk xylene, following EU-Technical Guidance Document guidelines. For soil organisms, the PEC/PNEC ratio (ie the Risk Quotient) was 0.5 for musk ketone, indicating no adverse effect of the compound on soil biota. However, the RQ value for musk xylene was >1, and equivalent to 1.3, indicating that this compound may have adverse effects on soil organisms. However, nitro musks have largely been replaced in Europe because of concerns about safety and bioaccumulation (Gatermann *et al*., 1999) and consequently this is reflected in the concentrations of these compounds measured in contemporary sewage sludges. Thus, Jones and Northcott (2000) and Stevens *et al.* (2003) did not detect any of the major nitro musk compounds (musk xylene, musk ketone, musk ambrette, musk moskene and musk tibetene) in UK sewage sludge samples. The two main compounds currently used as fragrance ingredients are the polycyclic musks: 7-acetyl-1,1,3,4,4,6-hexamethyl-1,2,3,4-tetrahydronaphthalene (AHTN) and 1,3,4,6,7,8-hexhydro-4,6,6,7,8,8-hexamethylcyclopenta-γ-2-benzopyran (HHCB). Consequently, Stevens *et al*. (2003) found that HHCB and AHTN were the most abundant synthetic musks in sludge with concentrations in the range 1.9 – 81 mg kg^{-1} DS and 0.12 – 16 mg kg^{-1} DS, respectively, and mean values of 27 and 4.7 mg kg^{-1} DS, respectively. This was consistent with use patterns, as together these compounds share >95 % of the market for polycyclic musks (Rimkus, 1999). Relatively little biodegradation and destruction of HHCB or AHTN occurs during wastewater treatment or anaerobic digestion of sewage sludge (Kupper *et al*., 2004; Yang and Metcalfe, 2006). However, Yang and Metcalfe (2006) reported the initial amounts of HHCB and AHTN in field soil immediately after sludge application were 1.0 and 1.3 µg kg^{-1}, respectively, but found that concentrations declined relatively rapidly over the next six weeks and AHTN was not detectable in the sludge-amended field soil after 6 months. Polycyclic musks are hydrophobic and sorb strongly to organic matter and sludge solids, therefore it is unlikely that they were lost from the soil by leaching or some other transport mechanism and a possible explanantion is that they were biodegraded by soil microbial activity. In the field study of Yang and Metcalfe (2006), sludge was applied just before the onset of winter, and it is plausible, therefore, that the rate of removal would be increased when warmer soil conditions support faster rates of microbial activity. The potential risks of human toxicity to synthetic musks from the use of sewage sludge on farmland is obviously a very minor consideration compared to the general exposure received from these compounds in body care and washing products. In relation to their potential ecotoxicological significance in soil, Balk and Ford (1999) estimated an adjusted PNEC value for AHTN and HHCB in soil of 0.32 mg kg^{-1} ds using a risk assessment factor of 50. However, the above observations, coupled with the soil ecotoxicological data and No Observed Effect Concentrations (NOECs) for earthworms and springtails of ≥45 mg kg^{-1} for both HHCB and AHTN (Balk and Ford, 1999) suggests there is likely to be minimal impact of polycyclic musks in practice under normal operational field conditions when sewage sludge is used as an agricultural fertiliser.

6.3 Plant Pathogens

The significance of plant pathogens for recycling sludge in agriculture is the possibility that infective agents, derived from household and industrial washing of vegetables and ingress of soil into the sewers during rainfall, could be transferred to the sludge during sewage treatment. The plant pathogens of greatest concern with regard to agricultural utilisation are: (1) the potato cyst nematodes (PCN), *Globodera rostochiensis* and *G. pallida*, (2) rhizomania disease of beet, caused by beet necrotic yellow vein virus that is transmitted by the agency of the primitive soil fungus Polymyxa betae, and (3) brown rot of potatotes caused by the bacterium, *Ralstonia* (=*Pseudomonas*) *solanacearum* (Carrington, *et al*., 1998a,b). The treatment processes specified in the UK DoE *Code of Practice for Agricultural Use of Sewage Sludge* (DoE 1989, revised 1996) were designed to be effective against PCN (Section 3). Spaull *et al*. (1988) showed that anaerobic digestion and thermal treatment of sludge reduced viability of PCN cysts almost completely, however, the effect of sludge treatment on the viability of *R. solanacearum* is not known. Codes of practice on the agricultural use of sludge should also prohibit application to land where introduction of the

PCN would have serious consequences (eg DoE, 1989 revised 1996). In the UK, the use of sludge on land used for growing seed potatoes and nursery stock (including bulbs) for export, including growing them in a crop rotation, is not permitted, for instance, to comply with European regulations to limit the introduction of harmful plant pathogens into other states. Measures to mitigate the spread of rhizomania and brown rot of potatoes include restrictions on the movement of infected crops and contaminated soil off the land and controlling the reservoirs of infection (potato ‘ground keepers’ or weeds) on infected land (Elphinstone, 1996). These measures aim to contain infection and permit decay of the pathogen on infected land. No additional provisions, above the normal requirements specified on the use of sludge in agriculture to prevent the dissemination of infectious disease (eg DoE, 1996), are considered necessary for sludge (Carrington *et al*., 1998b).

There is no evidence linking the spread of plant infections to the agricultural use of sewage sludge (Carrington *et al*., 1998b). However, there are reports (Stone and Powers, 1989; Castagnone-Sereno and Kermarrec, 1991; Lewis *et al*., 1992) suggesting sludge application to soil may beneficially stimulate the natural suppression of soil-borne plant pathogens, and reduce plant infections, which may be linked to increased microbial activity and antagonism in response to inputs to soil of organic matter and nutrient resources contained in sludge.

6.4 Phosphorus

The principal major plant nutrients contained in biosolids are N and P. The total concentrations of these nutrients in dewatered digested biosolids are typically in the range 4 – 5 % and 2 – 3 % dry solids, respectively (EA, 1999). However, P removal during wastewater treatment can more than double the concentration of this nutrient in sludge. The contribution of biosolids N to agricultural systems is equivalent to approximately 3 % of the total N applied to all crops in Great Britain in mineral fertilisers (Smith, 2007b). Phosphorus supplied in sludge, on the other hand, supplies a much larger contribution, equivalent to approximately 15 % of the total phosphorus in mineral fertilisers – a significant quantity relative to artificial fertiliser (Smith, 2007b).

In terms of the sustainability of the practice of using sludge in agriculture, recycling P in the foodchain is arguably more important than recycling N. Sludge provides a long-term maintenance dressing for this nutrient and can replace P inputs from inorganic fertilisers in a crop rotation. Recycling P to soil in biosolids has a much wider global benefit than simply reducing farmers’ costs, however, by contributing to the conservation of geological mineral P reserves and reducing external inputs of geogenic Cd into the foodchain. World reserves of currently exploitable phosphate rock are estimated to be 40 billion t and, at the present rate of consumption (150 million t per year), this will be exhausted within 250 years (CEEP, 1997). More seriously, however, current estimates suggest that low-Cd phosphate minerals from high-grade volcanic deposits may be exhausted within 25 years (Steen and Agro, 1998). Cadmium inputs to soil from rock phosphate fertiliser production is a concern with long-term implications for soil fertility and human health. Therefore, recycling P in biosolids is a key prerogative for long-term sustainability. Phosphorus recovery during wastewater treatment is highly efficient and the sludge is an effective P fertiliser source that closes the nutrient loop through the food chain, provided it is carefully managed.

Much research has been completed to determine the fertiliser replacement value of N, and to a lesser extent of P, in sludge (eg Coker and Carlton-Smith, 1986; Smith *et al*., 2002a; O’Connor *et al*., 2004). For example, the P availability in sludge is typically given as 50 % relative to mineral fertiliser. This information is readily available to operators and farmers in the form of advisory fertiliser recommendations (eg MAFF, 2000). Phosphorus inputs in sludge exceed crop demand and recommendations are given on the basis that a single dressing of dewatered biosolids will supply adequate phosphate for most 3 – 4 year crop rotations. This is justified on the basis that P is effectively retained in sludge-amended soil, but is available for crop uptake. Recent research shows there is lower risk of P transfer in land runoff following application of sludge compared with other agricultural P amendments at similar P rates (Withers *et al*., 2001). Based on an incubation study Withers and Flynn (2007) concluded that even when the equivalent of 20 years of typical biosolid P supply is incorporated into the soil, there may be no increase in runoff P, depending on the degree of soil P saturation, soil type and biosolid type. Iron-dosed and lime-treated biosolids in particular had a very low risk of dissolved P transfer to run-off. Leaching of P from agronomic rates of sewage sludge application to high risk sandy soils is not a major P loss pathway (Shepherd and Withers, 2001). Furthermore, no significant increase in P losses have been recorded in tile drain flow from annual applications of sludge to clay soil (Hodgkinson *et al*., 2002). However, improved understanding of the release of residual P applied to soil in biosolids in the growing seasons after application is an area requiring further attention to more precisely define the long-term P balance in sludge-amended soil. Some European countries have adopted P-based limits on the agronomic rates of application of biosolids to agricultural land (eg Ireland) and a similar approach is being considered for biosolids in the US (Shober and Sims, 2003). In the UK, however, nutrient recommendations for biosolids are N-based, which enables agronomic advantage to be taken of both of the major plant nutrients present in sludge. The available research indicates biosolids-P does not place the water environment at risk following a N-based nutrient regime when P supplied in sludge is managed on a crop rotation basis. However, as the UK moves towards implementing the EU Water Framework Directive (EPCEU, 2000a), which requires improvements in the ecological status of all inland and coastal waters, action is likely to be taken to reduce diffuse pollution from agricultural sources, focusing particularly on N and P. It is unclear at this stage what form these measures are likely to take and what their potential impact on recycling biosolids is likely to be. Recognising that land varies in terms of its potential risk of contributing nutrients, depending on the connectivity, to receiving waters may provide a pragmatic approach to managing nutrient inputs to soil in biosolids and avoiding unacceptable losses to the water environment (Withers and Flynn, 2007).

Ensuring that full account is taken of the nutrient value provided by biosolids, as well as by other organic manures, by adjusting supplementary mineral fertiliser applications to amended soil is critical to reduce wastage, impacts on crop quality and losses of nutrients to the environment. Further progress could be made in this area and would have direct benefits in terms of improved nutrient utilisation, reduced pollution as well as providing economic savings in fertiliser costs.

6.5 Summary remarks

In general, the ecotoxicological impacts of recycling sewage sludge on farmland are very limited. Current EU soil limits protect against phytotoxicity and the weight of scientific evidence indicates there is no serious negative influence on long-term soil fertility or microbial processes, such as the ability to perform symbiotic N fixation. Organic contaminants also have no effect on plant growth or on soil microorganisms. The only case that can be identified that warrants further investigation is the potential impact of certain bodycare products eg triclosan on soil ecological systems. Further research is also necessary to improve understanding of the chemical nature and long-term solubility and fate of P applied to soil in sludge and particularly in terms of its release during the crop rotation in the years following application.

7. CONCLUSIONS

7.1 Environmental assessment

An overall assessment of the potential environmental impacts caused by recycling sewage sludge to agricultural land is presented in Table 22 by ascribing a qualitative description of the apparent risk, based on the scientific information discussed in the report, for each of the principal sludge components, or groups of components, against potentially impacted environmental parameters. It has been assumed in this analysis that sludge is applied according to the regulations (CEC, 1986; UK SI, 1989) and codes of practice and other guidance (eg DoE, 1996; ADAS, 2001).

Table 22 Assessment of risks to health and the environment from recycling sewage sludge to agricultural land ([1]L = low risk, [2]P = possible risk)

Environmental parameter	PTEs	Organic contaminants	Pathogens	Nutrients	Odour
Human health	L	P[c]	L	L	P[b,d]
Crop yields	L	L	L	L	N/A
Animal health	L	L	L	L	N/A
Groundwater quality	L	L	L	P[c]	N/A
Surface water quality	L	L	L	P[c]	N/A
Air quality	L	L	L	L	P[b,d]
Soil fertility	P[a]	P[b,c]	L	L	N/A

[1] Risk is designated as 'low" (L) where environmental effects are minimized by current operational practice
[2] Risk is designated as 'possible' (P) where there is some reported evidence that current operational practice may result in a potential impact on the environment on the basis that one or more of the following conditions apply:
(a) published evidence of effects is contradictory
(b) effects may occur under certain extreme 'worst-case' conditions, given the current regulations and codes of practice
(c) there is uncertainty about the environmental implications of particular sludge components
(d) nuisance – see Section 5.5 and 7.3.5
N/A not applicable

7.2 Trends in contaminant concentrations in sludge

Heavy metal and POP concentrations in sludge have generally continued to decline in response to source controls and improved industrial practices and do not represent a threat to human health or the environment. The chemical quality of sludge is currently at the highest level it has ever been and this further ensures the sustainability of the agricultural recycling of sewage sludge. Nevertheless, there is still scope to further minimise the concentrations of problematic contaminants, and PTEs in particular, to underpin the long-term sustainability of the agricultural use of sewage sludge and this should continue to be a priority of environmental regulators and the Water Industry. The pathogen load to urban wastewater is very low due to the high general health status of the human population, therefore actual pathogen numbers in sludge are low and are further reduced or eliminated by treatment. This contrasts to the significant reservoirs of zoonotic pathogens in livestock and their wastes, which are not regulated or treated to manage the risks to human health associated with application to land. Recent research has shown that centrifuge conditioning of sludge may significantly increase indicator numbers in digested and pasteurised sludge and time-temperature exposure during sludge pasteurisation may transfer *E. coli* to a viable, but non-culturable state, that is reactivated by centrifuge-dewatering. The effects of both these processes on recoveries of enteric bacteria in sludge have implications for compliance with microbiological reduction criteria for sludge and require further investigation.

7.3 Human Health

7.3.1 Pathogens

The absence of disease in the human population relating to the agricultural utilization of sewage sludge, managed according to the provisions of Directive 86/278/EEC and national statutory controls and codes of practice, demonstrates the effectiveness of the barriers (sludge treatment and land use) in place to prevent disease transmission. In the UK, these measures have been further strengthened recently by the introduction of the Safe Sludge Matrix, which sets additional microbiological reduction criteria, and further clarifies land use controls for the use of sludge in agriculture. Quantitative MRA and recent experimental investigations of pathogen decay mechanisms in sewage sludge amended agricultural soil confirm the negligible risk from infectious pathogens to the human population when sewage sludge is applied to agricultural land according to recommended and statutory controls. Indeed application of organic matter and nutrient resources to soil in sludge stimulate soil ecological processes including predation by native soil protozoa providing an active mechanism for the destruction of enteric bacteria potentially applied to the soil in sludge. The results emphasise that the multi-barriers in place when sludge is used in agriculture enable the natural attenuation of residual numbers of enteric organisms to background values well within the cropping and harvesting restrictions stipulated for agricultural use and that the waiting periods specified in the controls are likely to provide a significant margin of safety. Further research is recommended, however, to provide more information on the different sources of enteric microbes entering soil to distinguish sludge organisms from other imputs, the decay of enteric viruses in sludge-treated agricultural soil, the quantitative effects of seasonal conditions on the decay of enteric pathogens and also the role and mechanisms of ecological suppression of enteric microorganisms applied to soil in sewage sludge. The mechanisms of pathogen inactivation during mesophilic anaerobic digestion of sludge, one of the principal methods of stabilizing sewage sludge for agricultural spreading adopted by the Water Industry, are poorly understood. Therefore, more work is recommended to determine the fundamental processes responsible for pathogen destruction during anaerobic treatment so that this important sludge stabilization process may be optimized for pathogen removal.

7.3.2 Potentially toxic elements

Detrimental effects on human health arising from the agricultural use of sludge are extremely unlikely. The principal PTEs of concern for human health are Cd, Pb and Hg. Pb and Hg are not absorbed to any extent by crops and consequently do not pose a risk through the dietary intake of plant foods grown in sludge-amended soil, which is the main pathway of exposure to dietary contaminants. Cadmium, on the other hand, is not subject to the ‘soil-plant barrier' and can accumulate in crops to concentrations which may be potentially hazardous to animals consuming them without appropriate control. However, dietary models of Cd intake from crops grown in sludge-treated soils indicate human health is protected by the controls on the potential accumulation of Cd in sludge-amended agricultural soil. Given the high margin of safety against potential dietary intake of Cd, which is intrinsic in the maximum permissible concentration of Cd in sludge-treated soil of 3 mg kg^{-1}, the detrimental accumulation of ingested Cd cannot occur. Indeed, individuals consuming a well balanced diet, including fresh vegetable produce that may be grown on sludged soil, actually represent a low risk group because improved mineral intake reduces Cd absorption. Dietary analyses show that the risk to human health from Cd in sludge-amended agricultural land is likely to remain at a very low level in practice even if a downward trend in soil pH status, which increases Cd bioavailability, became apparent in the future.

It is theoretically possible that the new EU grain limit for Cd could be exceeded for wheat grown on sludge-treated soil if the soil Cd limit were reached. In practice this is very unlikely however as Cd concentrations have consistently fallen over the past several decades and on average are now significantly below the soil limit value for this element.

Livestock ingestion of PTEs from sludge surface-applied to pasture soils is another possible pathway of food chain exposure. However, of crucial importance to the human diet is that Cd and Pb do not accumulate in muscle tissue (carcass meat) entering the food chain even under 'worst-case' conditions of PTE intake by grazing livestock from sludge-treated grassland. By contrast, Cd and Pb may potentially accumulate in the kidney and liver of livestock principally through the direct ingestion of soil and surface-applied sludge. However, feeding trials have shown that the concentrations of Cd and Pb in offal do not place the human diet at risk at the mandatory soil limit for Cd. A reduction in the advisory soil limit for Pb in the UK from 300 mg kg^{-1} to 200 mg kg^{-1} is proposed (DEFRA/WA, 2002) on the recommendation of Carrington *et al*. (1998b) as a precautionary measure to protect the quality of offal meat inrelation to the food standard for Pb.

7.3.3 Organic contaminants

Organic contaminants (OCs) present minimal risk to the human foodchain from land application of sewage sludge because the potentially most toxic compounds (eg 2,3,7,8-TCDD) cannot be be detected in sludge, and they are also influenced by a variety of physico-chemical and biological attenuation mechanisms that prevent the transfer to crop tissues, including:

- Rapid volatilisation and loss to atmosphere
- Rapid biodegradation and with minimal or no persistence, or
- Strong adsorption of persistent compounds

Therefore, risk to human health *via* dietary intake of OCs from crops grown on sludge-treated soils is minimal due to absence of crop uptake.

The potential impacts on the foodchain of persistent OCs in sludge, including: PAHs, PCDD/Fs or PCBs, has been a key concern for agricultural utlilisation. However, international emission controls on the main point sources of these priority persistent compounds have significantly reduced their entry into the environment and consequently also into the urban wastewater collection system. Thus, atmospheric deposition and environmental cycling are the main source of PCBs in sludge, and, consequently, the concentrations of this historically used chemical in sludge generally represent background environmental levels. The potential transfer to the human food chain of certain groups of OCs contained in sludge has been predicted based on known physico-chemical properties of the compounds. However, ongoing research has not identified any toxicological or ecotoxicological link with these compounds in sludge. Therefore, on balance, the importance of these contaminants has significantly diminished and there is no quantitative, scientific evidence to support the need for limits or controls on PAHs, PCBs or PCDD/Fs in sewage sludge. The EC Joint Research Centre concluded that (Erhardt and Prüeβ, 2001): 'organic contaminants in sludge are not expected to pose major health problems to the human population when sludge is re-used for agricultural purposes'. They also have stated that 'it does not make much sense to include PCDD/F, PCBs and PAHs in routine monitoring programmes'.

Brominated flame retardents are also an important group of persistent, lipophilic and bioaccumulative compounds potentially found in sludge. However, the use of the main types (PentaBDE and OctaBDE) has been restricted in the EU, therefore they also are expected to diminish in importance as contaminants in sewage sludge.

In contrast to other POPs, polychlorinated *n*-alkanes (PCAs), also known as chlorinated paraffins, are in active production and occur in sludge in much larger concentrations than PCBs and PCNs, for example. Consequently, further work is recommended to assess the potential transfer to the foodchain and significance for human health of PCAs in sewage sludge-amended agricultural soil. Therefore, because of this uncertainty, the risk to human health from OCs has been designated as ‘possible’ in Table 22.

Because they are lipophilic POPs characteristically show a high potential propensity to transfer and accumulate in animal fat tissue and milk fat from livestock ingesting surface-applied sludge adhering to herbage, which is theoretically the principal route of human exposure to potentially toxic OCs in sewage sludge. In practice, however, there is no evidence that organic pollutants transfer to the foodchain by this route, due to the low concentrations of POPs present and management factors (eg injecting sludge) avoiding surface application to grazed pastures for pathogen control.

There is some confusion about the apparent toxicity and significance of the phthalate plasticiser compound, DEHP, for human heath. However, DEHP has been recently down graded as carcinogen by IARC, and the US Agency for Toxic Substances and Disease Registry concluded that there is no evidence that DEHP is an endocrine disruptor in humans at the levels found in the environment. Exposures received through medical procedures from use of DEHP in PVC are significantly larger than they are likely to be through environmental routes. In the absence of any actual evidence of carcinogenic or reproductive abnormalities in highly exposed, sensitive medical situations, and also the direct ingestion of DEHP, for instance, by the general populaton transferred to food from plastic packaging materials, it would appear that the risk to human health from diffuse environmental sources, such as from DEHP inputs to soil from recycling sewage sludge, is very minor. This is reinforced by the rapid degradability of DEHP in soil and also the absence of significant transfers of DEHP to food crops. A risk assessment of DEHP in sludge-treated agricultural soil is recommended, following the protocol recommended by Schowanek *et al*. (2004), to clarify the minimal impact of this plasticiser compound on human health, soil quality and the environment.

7.3.4 Antibiotic resistance

The possibility that sludge may contain populations of antibiotic resistant bacteria and also trace concentrations of antibiotic compounds causing antibiotic resistance to develop in soil bacteria has raised concerns that this could have important consequences for human health. However, antibiotic resistance is a transient characteristic and resistance levels relatively quickly return to normal background values. This is because high maintenance requirements place the resistant organisms at a disadvantage therefore natural attenuation occurs when the selection pressure is removed. Application of sludge at normal agronomic rates, the relatively infrequent application of sludge to land and extended return periods are likely to permit natural attenuation of antibiotic resistant bacteria populations in soil. However, an MRA should be completed to confirm that antibiotic resistant microorganisms in sludge-treated soil represent a negligible risk to human health. This is also the basis for designating the risk to human health from OCs as ‘possible’ in Table 22.

7.3.5 Odour

Odour emissions are generally considered primarily in terms of the nuisance caused to communities close to the source, but recently it has been reported that malodour can have direct and indirect impacts on human health (Schiffman and Williams, 2005). This has been documented for communities close to static odour emitters, such as intensive livestock production centres. The treatment and land application of sewage sludge is managed to minimize odour nuisance, and is only a transient potential source, but this remains a potential cause of complaint and annoyance, and is one of the key factors raising public concerns and negative perceptions about the agricultural use of sludge. Therefore, measures to prevent odour when sludge is used in agriculture should be reinforced and could become part of the controls on agricultural use, to ensure odour emissions from sludge are minimized as far as practicable.

7.4 Crop yields

Zinc, Cu and Ni are the principal phytotoxic elements applied to soil in sludge. Current soil limit values in Directive 86/278/EEC for these elements ensure uptake into crops remains below the critical toxic concentration thresholds in plant tissues. These elements do not pose a dietary risk because they are subject to the `soil-plant barrier' since toxic concentrations in plant tissue are lower than the amounts which are potentially injurious to animals and man.

Organic contaminants have no phytotoxic activity at the concentrations found in sludge-treated soils. Plant pathogens are effectively destroyed by sludge treatment processes so effects on crop yield due to plant disease infection appear unlikely. There is no evidence which suggests that plant diseases are transmitted in sewage sludge.

7.5 Soil fertility

There is concern that the long-term sustainability of agricultural production may be impaired by the application of heavy metals to soil in sewage sludge due to effects on soil fertility. The principal decomposition and nutrient cycling processes are unaffected by concentrations of heavy metals exceeding those currently permitted in sludge-treated soils. Only Zn has been identified as potentially placing at risk certain sensitive groups of soil micro-organisms, and *Rhizobium leguminosarum* bv. *trifolii* in particular, at the statutory maximum soil limit values.

Recent results from a major programme of long-term field trials in the UK show no effects of elevated bioavailabilities of Zn from spiking soil with soluble salts on rhizobia or soil microbial biomass. Negative impacts were observed, however, due to application of exceptionally Zn-contaminated dewatered sludge. Copper salts and high-Cu cake sludge also caused toxic effects on these soil microbial indices. Copper is potentially biocidal when applied in bioavailable soluble form (hence its use as an agricultural fungicide, for example). However, it is proposed that the toxic effect of the high metal sludges is explained by the gradual disintegration of sludge particles from soil movement and cultivation causing increased exposure of the soil microbial population with time to the high metal toxic sludges at the micro-scale in soil. Other long-term field experiments have not identified significant effects of metals on important microbial indicators of soil fertility. Therefore the weight of evidence suggests that the UK advisory limit value for Zn in soil (200 mg Zn kg^{-1}) protects the soil microbial population. There was no effect of Cd on soil microorganisms observed in the UK long-term field experiments at the maximum soil concentration used of 4 mg kg^{-1} ds, but Cd may become toxic to soil microbes at higher concentrations than this. Further research is recommended to elucidate the physico-chemical and biological mechanisms responsible for the ecotoxicological response to the high-metal sludge observed in the long-term field trials. However, at this stage the evidence from the long-term field experiments does not warrant any immediate change to the maximum permissible soil limit values for Zn, Cu or Cd (Defra 2007b). As published evidence of effects of PTEs on soil fertility is contradictory however, the risk category has been designated as ‘possible’ in Table 22.

There is no evidence that the vast majority of sludge-borne OCs have a detrimental impact on soil microbial processes. Earlier concerns about the potential impact of linear alkylbenzene sulphonate (LAS), a detergent surfactant present in large concentrations in sludge, on soil ecological processes have been further elucidated and shown to be unfounded. Whilst the presence of large concentrations of certain high-volume bulk chemicals, like LAS, warrants careful investigation and assessment of the risks to the environment when sludge is used as an agricultural soil amendment, this does not necessarily represent a hazard to the soil ecological environment. However, if compounds like LAS were to be banned or their use restricted in future this would lead to product substitution, but there is uncertainty about the potential environmental consequences of the alternatives. Phthalates have not been found to cause any significant adverse effects on soil microbial processes or on soil fertility. In general, high volume useage compounds have very low toxicity and degrade rapidly in soil. A number of emerging compounds have been identified in the review as having a potential impact on soil microbes and these belong to the group of chemicals described as body care products eg triclosan and the significance of these warrants further investigation. Consequently the risk to soil fertility of OCs in sewage sludge spread on farmland has been designated as 'possible' in Table 22.

7.6 Phosphorus

Phosphorus concentrations in sludge are increasing with the expansion of P removal during wastewater treatment and require careful management of the nutrient inputs to soil in sludge is necessary to avoid over application. Phosphorus inputs to soil in sludge are therefore increasing at agronomic rates of sludge application calculated on a N basis. More information is required on the long-term fate and release of P in sludge-treated agricultural soil to crop rotations to assess the agronomic benefit and efficiency of utilisation of P to ensure accumulation in soil and risk to the water environment are minimised. Therefore, the environmental risk from nutrients applied to the soil in sludge has been designated as 'possible' in Table 22.

7.7 Regulating organic contaminants in sludge

Despite the extensive range of organic chemicals that can be present in sewage sludge, the expanding experimental evidence base indicates that these are not a significant limitation to the agricultural use of sewage sludge. This view is based on a technical evaluation of the situation, which acknowledges that the presence of effective source control measures and small concentrations of persistent contaminants in sludge, biodegradation and behaviour in soil, absence of crop uptake, and sludge application practices, minimise potential impacts of organics in sludge on soil quality, human health and the environment. The consensus view therefore is that there appears to be no scientific rationale for including numerical limits on OCs in quality assurance systems for the agricultural use of sewage sludge. Nevertheless, new issues and compounds of concern may emerge in the future, such as endocrine disruption, requiring an ongoing and proactive approach to research and vigilant monitoring and rational assessment of the significance of new developments as they arise.

There has been much debate about the relevance of quality standards for OCs in sludge. The general consensus that has emerged is that they are not a priority for regulation and proposals (EC, 2000) to revise the current Directive on agricultural use of sludge (86/278/EEC; CEC, 1986), which, amongst other issues included standards for organic chemicals (Table 11), have not been taken further, but could be revisited in a future revision of this legislation. Furthermore, the EC JRC recommended that routine monitoring of dioxins (PCDD/F), PCBs and PAHs in sludge used for agricultural purposes was unnecessary. However, the JRC did raise questions about the safety of detergent residues applied to soil in sludge, due to concerns about their solubility and potential to impact aquatic systems (Erhardt and Prüeβ, 2001). However, the consensus of scientific opinion is that detergent residues in sludge do not pose a significant environmental or health risk. The limit values for organic contaminants in sludge proposed by Leschber (2004) are not supported by this review of scientific evidence and do not appear to be justified on any technical grounds.

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