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THE INFLUENCE OF SEWAGE SLUDGE APPLICATIONS TO AGRICULTURAL LAND ON HUMAN EXPOSURE TO POLYCHLORINATED DIBENZO-*P*-DIOXINS (PCDDs) AND -FURANS (PCDFs)

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Abstract

*Polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs) are organic chemicals of suspected extreme toxicity. PCDD/Fs are ubiquitous in the environment and relatively enriched in sewage sludges. The deliberate application of sewage sludge to agricultural land can therefore potentially increase exposure of the human population to these compounds via transfers through the foodchain. This paper presents a protocol for assessing the background human exposure to all seventeen 2,3,7,8-substituted PCDD/Fs for which toxic equivalent factors (TEFs) are available. Background daily human exposure in the UK to 2,3,7,8-T_4CDD and I-TEQ are calculated to be 0·0239 ng and 0·203 ng, respectively. The potential increases in crop plant and livestock tissue PCDD/F concentrations, and hence in human exposure, following applications of sewage sludge to agricultural land are quantified for different sludge application scenarios. The key role of PCDD/F transfer into livestock via ingestion of sludge adhered to vegetation in determining increases in human exposure is demonstrated.*

Keywords: sewage sludges, organic contaminants, PCDDs, PCDFs, agricultural soils.

INTRODUCTION

Due to European Community (EC) regulations, sea disposal of sewage sludge in the UK will cease in 1998. This, coupled with increased sewage sludge production caused by alterations in treatment of wastewater, will result in major shifts in sewage sludge disposal practices both nationally and internationally. Application to agricultural land currently accounts for about 43% of the 1·22 million tonnes (dry wt) of sludge produced annually in the UK (Powlesland & Frost, 1990). Recently, concerns have been expressed regarding the chemical content of sewage sludges. Concerns regarding the heavy metal content of sewage sludges relate to possible adverse effects on soil microbial processes, such as nitrogen fixation. Whilst organic chemicals in sludge are not perceived as affecting soil processes, concerns have been voiced over whether some toxic organics can transfer from the soil and into the human foodchain. The precise extent of the threat to human health posed by sludge borne organics disposed of to land remains unknown. The extreme toxicity of certain PCDD/F congeners, coupled with their propensity for bioaccumulation has attracted particular attention. To date, however, assessments of the risks posed by the organic contaminants in sewage sludges have not considered PCDD/Fs (Naylor & Loehr, 1982*a*,*b*; Connor 1984; Chaney *et al.*, 1991). Human exposure assessments which have regarded these compounds concentrate on 2,3,7,8-T_4CDD alone and have ignored sludge application as a potential source (Travis & Hattemer-Frey, 1987; Jones & Bennett, 1989). While assessing background human exposure to 2,3,7,8-T_4CDD has provided useful information, it fails to take account of the threat posed by the sixteen other 2,3,7,8-substituted PCDD/F congeners which, although less toxic than 2,3,7,8-T_4CDD, may be more toxicologically important owing to their greater environmental abundance. In addition, the environmental behaviour of PCDD/Fs is congener-specific and a risk assessment model centred on one congener cannot easily be extrapolated to others.

This paper initially assesses the background human exposure to 2,3,7,8-T_4CDD from diet, water and air intakes. Using the same protocol, background human exposure to all seventeen 2,3,7,8-substituted PCDD/Fs has then been estimated. Exposure is also given in terms of toxic equivalents (TEQs). Toxicity equivalency factors (TEFs) are weighting factors which express toxicity of individual PCDD/Fs in terms of the most toxic

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Table 1. Input data for the background exposure assessments. (See text for sample details)

Compound	Log K_{ow}[a]	Soil[d] (dry wt) (ng kg^{-1})	Herbage[d] (dry wt) (ng kg^{-1})	Air[f] (ng m^{-3})	Urban air[f] (ng m^{-3})	River water[h] (ng l^{-1})	Drinking water[h] (ng l^{-1})	Eggs[i] (wet wt) (ng kg^{-1})	Fish[j] (wet wt) (ng kg^{-1})	Chicken[i] (wet wt) (ng kg^{-1})
2,3,7,8-T_4CDD	6·80	0·50[a]	0·032	4·30E−07	4·30E−07	3·10E−06	5·00E−07	0·050	0·13	0·10
1,2,3,7,8-P_5CDD	7·40	0·50[e]	0·14	6·00E−06	6·00E−06	4·00E−07	5·00E−07	0·075	0·20	0·10
1,2,3,4,7,8-H_xCDD	7·80	0·37	0·14	4·00E−06	4·00E−06	6·00E−07	8·00E−07	0·123[l]	0·02[l]	0·16[l]
1,2,3,6,7,8-H_xCDD	7·80	0·62	3·00	7·60E−06	7·60E−06	4·60E−06	1·00E−06	0·132[l]	0·10[l]	0·48[l]
1,2,3,7,8,9-H_xCDD	7·80	0·48	1·40	nd[k]	nd	2·70E−06	9·00E−07	0·055	0·04	0·09
1,2,3,4,6,7,8-H_pCDD	8·00	27·0[e]	5·90	1·00E−04	6·56E−04[g]	3·60E−05	4·40E−06	0·845	0·32	2·50
OCDD	8·20	1·43[e]	24·00	2·30E−04	1·25E−03[g]	1·40E−04	1·70E−05	3·800	2·62	3·50
2,3,7,8-T_4CDF	6·53[c]	0·95	0·46	5·10E−05	5·10E−05	2·60E−05	9·60E−06	0·105	1·21	0·19
2,3,4,7,8-P_5CDF	6·92[c]	0·93	0·20	1·80E−05	3·60E−05[g]	8·50E−06	2·30E−06	0·075	0·66	0·07
1,2,3,7,8-P_5CDF	6·92[c]	1·06	0·18	1·90E−06	2·50E−05[g]	7·90E−06	3·00E−07	0·020	0·18	0·04
1,2,3,4,7,8-H_xCDF	8·53[b]	1·27	0·32	1·20E−06	7·50E−05[g]	9·50E−06	7·00E−07	0·065	0·08	0·07
1,2,3,7,8,9-H_xCDF	8·53[b]	0·01	0·023	7·90E−07	7·90E−07	6·00E−07	5·00E−07	0·000	0·04	nd
1,2,3,6,7,8-H_xCDF	8·53[b]	0·82	0·16	7·80E−06	6·70E−05[g]	3·10E−06	6·00E−07	0·035	0·07	0·04
2,3,4,6,7,8-H_xCDF	8·53[b]	0·73	0·15	5·40E−06	7·65E−05[g]	2·00E−06	4·00E−07	0·030	0·08	0·05
1,2,3,4,6,7,8-H_pCDF	7·92[c]	4·13	1·90	8·70E−05	3·43E−04[g]	9·90E−05	3·00E−07	0·110	0·09	1·11
1,2,3,4,7,8,9-H_pCDF	7·92[c]	0·38	0·14	1·00E−06	2·35E−05[g]	9·00E−07	1·00E−06	0·000	0·09	0·06
OCDF	7·97	15·0[a]	2·00	1·60E−05	1·60E−05	1·00E−04	6·00E−06	0·040	0·20	0·52
I-TEQ		2.27	0·88	2·24E−05	6·24E−05	1·45E−05	3·45E−06	0·194	0·74	3·32E−01

[a] Doucette & Andren (1987)
[b] Fiedler & Schramm (1990)
[c] Sijm *et al.* (1989)
[d] Kjeller *et al.* (1991)
[e] Creaser *et al.* (1989)
[f] Näf *et al.* (1990)
[g] Warren Spring data
[h] Rappe *et al.* (1990)
[i] MAFF (1992)
[j] Startin *et al.* (1990)
[k] nd = not detected
[l] See text for details of derivation

individual compound, 2,3,7,8-T_4CDD. Multiplication of the TEFs and the 2,3,7,8-substituted PCDD/F concentrations gives a TEQ. These can be summed to give a total TEQ value. The TEFs used here are those developed by the NATO Committee on Challenges to Modern Science and referred to as the International TEQ (I-TEQ) system. Using realistic PCDD/F concentrations in sewage sludge and a defined sludge application rate, the projected effects of sewage sludge application on estimates of the background human exposure are made. The effects of altering input parameters are also investigated.

Table 1 shows the input data used in this study. Congener specific data are limited, thus the input data are a composite from a variety of sources. Contemporary UK data have been used wherever possible. However, this paper aims to give an overview of the whole 'sludge-to-land' debate, rather than specifically of the UK situation. Thus overseas data have been included where UK data are unavailable. In the cited papers, the 1,2,3,4,7,8-H_xCDD, 1,2,3,6,7,8-H_xCDD data reported for eggs, fish and poultry were given as a combined concentration. Individual data have been derived by taking ratios between these compounds as indicated by other papers. For example, the fish data given by Startin *et al.* (1990) give a 1,2,3,4,7,8-H_xCDD and 1,2,3,6,7,8-H_xCDD combined concentration of 0·12 ng kg^{-1} (fresh wt). However, Fürst *et al.* (1990) gave individual concentrations of these congeners for two fish samples with a ratio of 0·138. This ratio has been used to estimate 1,2,3,4,7,8-H_xCDD and 1,2,3,6,7,8-H_xCDD concentrations from the data of Startin *et al.* (1990), giving 0·02 ng kg^{-1} and 0·10 ng kg^{-1} (FW), respectively. The same method has been used to estimate individual 1,2,3,4,7,8-H_xCDD and 1,2,3,6,7,8-H_xCDD concentrations for poultry and eggs, using ratios derived from Beck *et al.* (1989) and Fürst *et al.* (1990).

ASSESSING BACKGROUND HUMAN EXPOSURE TO PCDD/Fs

PCDD/Fs in soils

UK rural soils have been shown to contain PCDD/Fs. Creaser *et al.* (1989) reported concentrations of 2,3,7,8-T_4CDD in rural UK soils to lie between <0·5–2·1 ng kg^{-1}, with a median of <0·5 ng kg^{-1} dry wt, while OCDD concentrations can reach levels over 1300 ng kg^{-1}. Concentrations of PCDD/Fs are higher in urban soils, where levels of 2,3,7,8-T_4CDD can reach 4·2 ng kg^{-1} (Creaser *et al.*, 1990). Here it is assumed that agricultural soils contain 0·5 ng kg^{-1} dry wt. Most of the Creaser *et al.* (1989, 1990) data are not congener specific, thus soil concentrations (Table 1) for some of the PCDD/Fs considered here come from the study by Kjeller *et al.* (1991), which appears to be one of the only sources of congener specific data for the UK.

Transfers to plants

Introduction

Organic chemicals in soil may become incorporated into plant tissues. Generally, the uptake of chemicals by plants can be considered as two separate mechanisms; root uptake of compounds from the soil solution and foliar uptake of compounds volatilised from the soil surface. Bell *et al.* (1988) and Wild & Jones (1992*b*)

did not highlight 2,3,7,8-T_4CDD as being significantly susceptible to either process, although McCrady *et al.* (1990) found that the foliar route may be significant. There is very little information in the literature regarding transfer rates of PCDD/Fs from soil to plants. However, below we consider the movement of PCDD/Fs into above-ground and below-ground plant matter.

Above-ground plant matter

PCDD/Fs are lipophilic/hydrophobic and hence in soil they are strongly adsorbed to the soil organic matter. This severely restricts processes such as root uptake. Even if these compounds could be incorporated into plant roots they would not be efficiently translocated to above ground plant parts due to their inappropriate physico-chemical properties (Briggs *et al.*, 1982, McCrady *et al.*, 1990). However, there may be the potential for PCDD/Fs to become incorporated into plant leaves via volatilisation from the soil surface (McCrady *et al.*, 1990). Whilst experimentally obtained data would be preferable, concentrations of 2,3,7,8-T_4CDD in above-ground plant parts can be estimated from soil concentrations using the equation derived by Travis & Arms (1988) using log K_{ow};

$$\log B_v = 1{\cdot}588 - 0{\cdot}578 \log K_{ow}$$

where B_v = concentration of organic in above ground plant (dry wt)/concentration of organic in soil concentration (dry wt).

The congener 2,3,7,8-T_4CDD has a log K_{ow} value of 6·8 (Doucette & Andren, 1987). Using the regression equation above, a bioconcentration factor of 0·004546 is obtained. Thus a concentration of 0·00227 ng 2,3,7,8-T_4CDD kg^{-1} dry wt is calculated for plants grown on rural UK soils. Since plants are approximately 85% water this concentration reduces to 0·00034 ng kg^{-1} fresh wt.

The above plant 2,3,7,8-T_4CDD concentration estimate is extremely low compared to plant concentrations reported in the literature (Kjeller *et al.*, 1991; Startin *et al.*, 1989). Table 1 shows PCDD/F concentrations in UK plant samples (composite samples made from yearly grass cuttings between 1979 and 1988). Concentrations of 2,3,7,8-T_4CDD are in the range of 0·032 ng kg^{-1} dry wt, equivalent to 0·0048 ng kg^{-1} fresh wt assuming the vegetation is 15% dry matter (Kjeller *et al.*, 1991). The disparity between calculated and measured 2,3,7,8-T_4CDD levels is due to the overriding importance of atmospheric inputs of PCDD/Fs relative to soil inputs (Prinz *et al.*, 1990).

For the purposes of this paper we assume that rural vegetation grown on rural soil will contain the measured quantity as reported by Kjeller *et al.* (1991), i.e. 0·0048 ng kg^{-1} fresh wt 2,3,7,8-T_4CDD, and that this has been delivered exclusively by the atmosphere.

Below-ground plant matter

Some lipophilic organics such as PAHs and PCBs, which are very poorly translocated from plant roots to shoots, are known to associate with root tissues (Wild & Jones, 1992*a*). McCrady *et al.* (1990) found that about 70% of the 2,3,7,8-T_4CDD originally in solution adsorbed to root surfaces. The study indicated the potential importance of this process, but cannot be directly related to uptake from soils since it was conducted in a nutrient solution. However, movement of PCDD/Fs from soil into root crops has been highlighted as being a possible major source of human food-chain contamination (Prinz *et al.*, 1990). From studies reported in the literature it is considered that concentrations of PCDD/Fs in the tubers of root crops are unaffected by soil concentrations, but that some accumulation may occur in the peel (Hülster & Marschner, 1991). In peeled potatoes grown on uncontaminated soil, no PCDD/F concentrations were higher than 4 ng kg^{-1} dry wt. However, peel concentrations did appear to increase with increasing soil PCDD/F levels, although the relationship was not linear. Overall, it appears that transfer from soil to root crops is inefficient and that the transfer factor of 0·05 and probably less would appear to be realistic. This figure has been used here for all 2,3,7,8-substituted PCDD/Fs, although this is clearly an over-simplification. It is important to mention that concentrations in harvested materials from below ground, such as potatoes and carrots, are significantly lower than respective concentrations found in above ground plant parts, due to the lack of atmospheric inputs (Prinz *et al.*, 1990).

A worst case concentration of 0·025 ng 2,3,7,8-T_4CDD kg^{-1} dry wt is assumed here for unpeeled root crops grown on UK rural soil. Since root crops are approximately 10% dry solid, a wet weight concentration of 0·0025 ng 2,3,7,8-T_4CDD kg^{-1} is derived. In a later section, Table 3 gives the estimated plant root concentrations for all the 2,3,7,8-substituted PCDD/Fs.

Transfers to livestock

Introduction

PCDD/Fs can be transferred from the terrestrial environment into livestock by a variety of pathways. These include soil ingestion, plant ingestion and intakes associated with water and air, each of which are dealt with below.

Soil ingestion

Soil is consumed by grazing livestock as part of their diet. Consumption of soil results in direct ingestion of organic contaminants. The amount of soil ingested varies between different species and climatic conditions, but is thought to constitute on average about 6% of the diet by weight (Fries, 1991; Stark & Hall, 1992). However, under certain circumstances (i.e. drought, over-grazing, etc.) soil ingestion rates can be considerably higher. For the purposes of this paper, transfers of PCDD/Fs from soil to cattle are estimated and it is assumed that other livestock accumulate these compounds to the same extent. If a lactating cow grazing a rural pasture consumes 15 kg dry wt food per day, a 6% soil ingestion rate results in the cow ingesting 900 g

of soil, equivalent to 0·45 ng of associated 2,3,7,8-T_4CDD. A non-lactating cow consumes approximately 8 kg dry wt food per day, thereby consuming 0·24 ng of 2,3,7,8-T_4CDD associated with 480 g of soil.

Vegetation ingestion

Using an average above ground vegetation 2,3,7,8-T_4CDD concentration of 0·032 ng kg^{-1} dry wt (Table 1), then 0·4512 ng kg^{-1} are consumed associated with the 14·1 kg of vegetation consumed per day by a lactating cow. A non-lactating cow consumes 0·24064 ng 2,3,7,8-T_4CDD day^{-1} in the 7·52 kg of vegetation consumed.

Clearly these calculations assume that the cow only eats vegetation from the field in question, and that PCDD/F inputs from other forms of feed are negligible.

Other sources

A cow ingests PCDD/Fs via inhalation and water. Assuming an inhalation rate of 150 m^3 per day (McLachlan *et al.*, 1990), and a 2,3,7,8-T_4CDD air concentration of $4{\cdot}3 \times 10^{-7}$ ng m^3 (Table 1) gives a daily 2,3,7,8-T_4CDD intake of $6{\cdot}45 \times 10^{-5}$ ng. A cow drinks about 30 litres of water per day (McLachlan *et al.*, 1990). Assuming a water 2,3,7,8-T_4CDD concentration of $3{\cdot}1 \times 10^{-6}$ ng kg^{-1} (Table 1) gives a daily intake of $9{\cdot}3 \times 10^{-5}$ ng.

Summary

From the above it is estimated that a lactating cow grazing a rural pasture takes in 0·45, 0·4512, $6{\cdot}45 \times 10^{-5}$ and $9{\cdot}3 \times 10^{-5}$ ng 2,3,7,8-T_4CDD per day associated with soil, vegetation, air and water intake respectively, giving a total of 0·9014 ng day^{-1}. This figure reduces to 0·4808 ng 2,3,7,8-T_4CDD day^{-1} if the cow is not lactating. It is illustrated that diet and soil ingestion both account for approximately 50% of 2,3,7,8-T_4CDD exposure experienced by cattle. This agrees well with the estimates of McLachlan *et al.* (1990) who reported daily 2,3,7,8-T_4CDD intakes by lactating cattle of 1·32, 0·01 and 4×10^{-4} ng day^{-1} from feed, water and air respectively.

Effects on livestock tissue concentrations

In mammals, ingested organics may be excreted, metabolised, stored or secreted in milk or transferred into offspring. McLachlan *et al.* (1990) found that about 75% of ingested 2,3,7,8-T_4CDD was excreted in the faeces with little or none in the urine. Some workers have estimated the effect of PCDD/F intake on animal tissues using equations such as those developed by Travis & Arms (1988), which use biotransfer rates (BT) to relate beef/milk concentrations to physico-chemical properties. Such equations assume a positive linear relationship between log K_{ow} and transfer coefficients into beef and milk. However, Fries (1991) and McLachlan *et al.* (1990) both show that bioconcentration (fat/diet) generally decreases with the degree of PCDD/F chlorination. Therefore, transfer coefficients decrease with log K_{ow}. The reason for this probably concerns the lower bioavailability of the higher

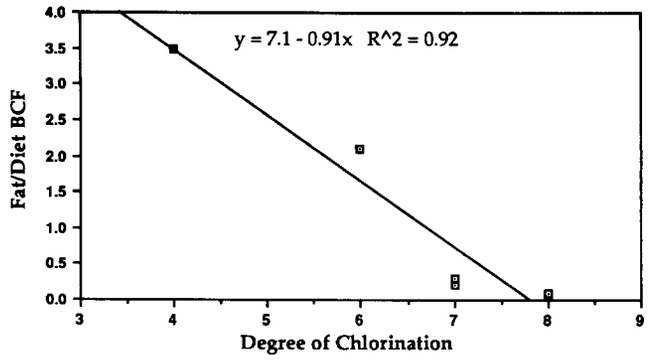

Fig. 1. Relationship between degree of PCDD/F chlorination and animal fat/diet bioconcentration factors (data from Fries, 1991).

chlorinated PCDD/Fs across the gastro-intestinal tract (Fries, 1991). Therefore, the Travis & Arms (1988) equations are deemed to be inappropriate for these compounds. Reported transfer coefficients summarised by Fries (1991) have been used here to predict beef concentrations from dietary intake estimates. These have been extrapolated to predict coefficients for compounds where no literature estimates have yet been made. The approach adopted has been to use the degree of PCDD/F chlorination, since McLachlan *et al.* (1990) found a good relationship between this parameter and transfers of PCDD/Fs to milk from diet. Figure 1 shows the relationship between degree of PCDD/F chlorination and animal fat (ng kg^{-1})/diet (ng kg^{-1}) bioconcentration factors (data from Fries, 1991).

Using this relationship, the fat/diet BCFs for compounds where no estimates are available can be predicted. The relationship shown assumes identical BCFs for all PCDD/Fs of a given degree of chlorination, i.e. that 2,3,7,8-T_4CDD behaves the same as 2,3,7,8-T_4CDF. However, this is probably not the case. Table 2 shows the meat fat/diet BCFs derived by this method and those measured milk fat/diet BCFs. Table 2 illustrates that the estimated meat fat/diet BCFs are generally similar to measured milk fat/diet BCFs, implying that the equation above is reliable. However, there are two exceptions, 2,3,7,8-T_4CDF and 1,2,3,7,8-P_5CDF, for which the meat fat/diet BCFs are considerably higher than milk fat/diet BCFs. This may be an artefact of the estimation method. Therefore, the milk fat/diet BCFs for these two compounds have been assumed to apply to the meat fat/diet BCFs.

For 2,3,7,8-T_4CDD the measured fat/diet BCF of 3·5 has been used. This provides estimates of beef fat concentrations of 0·2103 ng kg^{-1} and 0·2104 ng kg^{-1} if lactating and non-lactating cattle are grazing rural pasture. These are both equivalent to fresh meat values of 0·0526 ng kg^{-1}, on the assumption that animal meat is 25% fat.

PCDD/Fs after consumption may be transferred into milk. As with animal fat, the higher chlorinated PCDD/Fs are less bioavailable and therefore less likely to be transferred to milk (McLachlan *et al.*, 1990). McLachlan *et al.* (1990) found that 35% of ingested 2,3,7,8-T_4CDD is transferred into milk, equivalent to a

Table 2. Measured and predicted bioconcentration factors (BCFs) for animal meat and milk fat from diet (kg DW) for PCDD/Fs

Compound	Measured and predicted meat fat/dw diet	Measured milk fat/dw diet
2,3,7,8-T_4CDD	3·50[a]	3·75[c]
1,2,3,7,8-P_5CDD	2·57[b]	3·54[c]
1,2,3,4,7,8-H_xCDD	1·65[b]	1·82[c]
1,2,3,6,7,8-H_xCDD	1·65[b]	1·50[c]
1,2,3,7,8,9-H_xCDD	1·65[b]	1·93[c]
1,2,3,4,6,7,8-H_pCDD	0·20[a]	0·32[c]
OCDD	0·05[a]	0·43[c]
2,3,7,8-T_4CDF	3·50[b]*	0·75[c]
2,3,4,7,8-P_5CDF	2·57[b]	2·68[c]
1,2,3,7,8-P_5CDF	2·57[b]*	0·64[c]
1,2,3,4,7,8-H_xCDF	1·65[b]	2·04[c]
1,2,3,7,8,9-H_xCDF	1·65[b]	2·04[c]
1,2,3,6,7,8-H_xCDF	1·65[b]	1·71[c]
2,3,4,6,7,8-H_xCDF	1·65[b]	1·50[c]
1,2,3,4,6,7,8-H_pCDF	0·30[a]	0·32[c]
1,2,3,4,7,8,9-H_pCDF	0·74[b]	0·86[c]
OCDF	0·10[a]	0·11[c]

[a] Fries (1991).
[b] Calculated (see Fig. 1).
[c] McLachlan *et al.* (1990).
* These estimates have not been used, see text for explanation.

Table 3. Calculated fresh weight concentrations of PCDD/Fs in root crops, animal meat and milk

Compound	Root crops (ng kg^{-1})	Milk (ng l^{-1})	Meat (ng kg^{-1})
2,3,7,8-T_4CDD	0·002 5	0·011 3	0·052 6
1,2,3,7,8-P_5CDD	0·002 5	0·028 6	0·104 0
1,2,3,4,7,8-H_xCDD	0·001 9	0·014 0	0·063 5
1,2,3,6,7,8-H_xCDD	0·003 1	0·214 0	1·180 0
1,2,3,7,8,9-H_xCDD	0·002 4	0·130 0	0·555 0
1,2,3,4,6,7,8-H_pCDD	0·135 0	0·115 0	0·358 0
OCDD	0·715 0	0·670 0	0·389 0
2,3,7,8-T_4CDF	0·004 8	0·018 4	0·091 9
2,3,4,7,8-P_5CDF	0·004 7	0·032 7	0·157 0
1,2,3,7,8-P_5CDF	0·005 3	0·007 5	0·037 3
1,2,3,4,7,8-H_xCDF	0·006 4	0·0385	0·156 0
1,2,3,7,8,9-H_xCDF	0·000 1	0·002 3	0·009 2
1,2,3,6,7,8-H_xCDF	0·004 1	0·017 1	0·082 4
2,3,4,6,7,8-H_xCDF	0·003 7	0·013 9	0·076 3
1,2,3,4,6,7,8-H_pCDF	0·020 7	0·032 6	0·153 0
1,2,3,4,7,8,9-H_pCDF	0·001 9	0·006 6	0·028 6
OCDF	0·075 0	0·015 3	0·069 5
I-TEQ	0·011 4	0·089 4	0·412 0

milk fat/diet BCF of 3·75. From the diet data derived here 0·3155 ng 2,3,7,8-T_4CDD is estimated to enter milk in the daily production of 28 litres, if the lactating cow is grazing rural pasture. A fresh weight milk 2,3,7,8-T_4CDD concentration of 0·0113 ng $litre^{-1}$ is derived, equivalent to 0·225 ng kg^{-1} fat, given that milk is 5% fat. This milk concentration is similar to values reported by Rappe *et al.* (1987) (~0·02 ng 2,3,7,8-T_4CDD kg^{-1} fresh wt and by Startin *et al.* (1990) (0·009 ng 2,3,7,8-T_4CDD kg^{-1} fresh wt). The other transfer rates calculated from the figures reported by McLachlan *et al.*, (1990) shown in Table 2 have been used to estimate the milk concentrations for the other PCDD/Fs. Most of these estimates are within the range given by Rappe *et al.* (1987) and those given by MAFF (1992). The I-TEQ of 0·0894 ng kg^{-1} is well below the maximum tolerable concentration (MTC) of 0·7 ng I-TEQ kg^{-1} whole product given by MAFF (1992).

Summary of predicted values

Table 3 shows a summary of the plant root, animal meat and milk concentrations for the 2,3,7,8-substituted PCDD/Fs predicted by the methods described above.

Transfers to humans

This section uses the PCDD/F concentration estimates given above to calculate background 2,3,7,8-T_4CDD human intake values.

Some previous exposure assessments have considered human ingestion of soil as an important exposure pathway (Naylor & Loehr, 1982*a*,*b*; Connor, 1984). However, recent studies suggest that soil ingestion by normal adults is minimal and is not important with respect to 2,3,7,8-T_4CDD exposure (Paustenbach & Murray, 1986). Human exposure to PCDD/Fs here is considered to result from eating conventional foodstuffs and intakes associated with water and air, and excludes soil ingestion. Additional intakes via skin absorption and consumer products are also considered as negligible.

Intakes can be calculated by relating the food PCDD/F concentrations to the corresponding consumption rate. Yearly fresh weight consumption rates in the UK have been measured by MAFF (1988) and are shown in Table 4 which summarises the estimated background human 2,3,7,8-T_4CDD exposure.

The estimate uses the fresh weight meat concentrations as calculated here and assumes that pork and lamb meat contains the same PCDD/F load as beef. The milk and vegetation 2,3,7,8-T_4CDD concentrations are those calculated here. However, the poultry, fish and egg data are taken from Table 1. Concentrations of 2,3,7,8-T_4CDD in dairy products are derived using milk fat PCDD/F concentrations as Fürst *et al.* (1990) found similar PCDD/F concentrations in cow's milk, cheese and butter on a fat content basis. The milk fat PCDD/F concentrations derived from the McLachlan *et al.* (1990) data assumed that milk was 5% fat, whereas milk consumed in the UK is generally only about 3·5% fat. Therefore, it is assumed that the PCDD/Fs within milk are associated with 3·5% fat. These values have then been used to estimate PCDD/F concentrations in other dairy products. We have assumed that cheese, butter, cream, ice cream and yoghurt are approximately 35%, 80%, 25%, 3·5% and 3·0% fat, respectively. The resultant PCDD/F concentrations are similar to those reported by MAFF (1992). It is assumed here that vegetable oil contains the same concentrations as those found in dry weight vegetation (0·032 ng 2,3,7,8-T_4CDD kg^{-1}) and that animal oil con-

Table 4. Calculation of background human exposure to 2,3,7,8-T_4CDD

Media	Amount consumed (kg yr^{-1})	Concentration (ng kg^{-1} fresh wt)	Daily intake (ng day^{-1})	% Contribution to total
Water	730	5·000E−07	1·00E−06	0·004
Air	7300	4·300E−07	8·60E−06	0·04
Meat				
Beef/pork/lamb	29·93	5·260E−02	4·31E−03	18·0
Poultry	6·21	1·000E−01	1·70E−03	7·1
Milk	109·14	1·130E−02	3·37E−03	14·1
Dairy products				
Cheese	4·51	1·010E−01	1·25E−03	5·2
Butter	3·43	2·310E−01	2·17E−03	9·1
Milk products	2·05	1·130E−02	6·34E−05	0·3
Ice cream	1·96	1·010E−02	5·44E−05	0·2
Yoghurt	2·76	8·670E−03	6·55E−05	0·3
Cream	0·61	7·220E−02	1·21E−04	0·5
Eggs	9·49	5·000E−02	1·30E−03	5·4
Oil				
Animal	9·22	2·100E−01	5·31E−03	22·2
Vegetable	1·37	3·200E−02	1·20E−04	0·5
Vegetation	115·39	4·800E−03	1·52E−03	6·3
Root crops	65·65	2·500E−03	4·50E−04	1·9
Fish	5·84	1·300E−01	2·08E−03	8·7
Total			0·0239	

centrations are the same as those in animal meat fat (i.e. 0·2104 ng kg^{-1} 2,3,7,8-T_4CDD).

Table 4 illustrates that the estimated 2,3,7,8-T_4CDD dietary intake for a human consuming produce from rural areas is 8·712 ng $year^{-1}$. Humans are also exposed to PCDD/Fs in water and air. Assuming the water and air consumption rates shown in Table 4 and the 2,3,7,8-T_4CDD concentrations given in Table 1, it is estimated that an overall yearly human background 2,3,7,8-T_4CDD intake is 8·723 ng, equivalent to 0·0239 ng per day.

Overall, 99·96% of total 2,3,7,8-T_4CDD exposure comes from diet, with air inhalation and water consumption accounting for 0·04% and 0·004%, respectively. Consumption of meat and animal fat accounts for about 47% of the 2,3,7,8-T_4CDD exposure, with most of the remainder being due to dairy products and fish. Consumption of fish accounts for 8·7% of the 2,3,7,8-T_4CDD ingested. The importance of fish in the diet has been reported by other workers (Svensson *et al.* 1991). It is important to note that the exposure assessment uses food PCDD/F concentrations in uncooked produce. The effect of cooking and cooking method on PCDD/F levels is unquantified at present. Charcoal grilling may, for example increase meat PCDD/F levels, whereas loss of meat fat during cooking may decrease levels.

The intake estimate derived here can be compared to other reported exposure assessments. Exposure to 2,3,7,8-T_4CDD has been estimated to be 0·05 ng day^{-1} to an average US person by Travis & Hattemer-Frey (1987); Jones & Bennett (1989) reported an exposure estimate of 0·1 ng day^{-1}, while Graham *et al.* (1985) estimated the daily 2,3,7,8-T_4CDD dose to be in the region of 0·1–0·2 ng day^{-1}. The lower value calculated here is probably the result of using background PCDD/F concentrations for the exposure assessment. For example, using the 2,3,7,8-T_4CDD soil concentration used by Travis & Hattemer-Frey (1987) of 2·2 ng kg^{-1} elevates the exposure assessment here to 0·054 ng 2,3,7,8-T_4CDD day^{-1}. The assessment by Beck *et al.* (1989), regarded as one of the most accurate assessments to date, found that daily 2,3,7,8-T_4CDD exposure to West Germans was 0·0246 ng, thus being very close to the estimate made here.

It is considered that the exposure assessment protocol adopted here provides realistic estimates for 2,3,7,8-T_4CDD in rural areas and will provide reliable assessments of human exposure to all 2,3,7,8-substituted PCDD/Fs. Furthermore the protocol can readily be adapted to consider various sludge application scenarios.

Human exposure to 2,3,7,8-substituted PCDD/Fs

Table 5 shows the estimated background daily human exposure to all 2,3,7,8-substituted PCDD/Fs calculated using the above protocol. The highest exposure estimate for a single congener is 2·075 ng day^{-1} calculated for OCDD. However, the compound which makes the greatest contribution to the daily I-TEQ of 0·2027 ng (202·7 pg) is 1,2,3,6,7,8-H_xCDD which contributes around 26% of the total. Beck *et al.* (1989) estimated a TEQ daily exposure of 93·5 pg (West German TEQ); the German 1984 TEQ system generally gives lower figures than the I-TEQ system. MAFF (1992) estimated UK I-TEQ human exposure to be 125 pg day^{-1}, relatively close to the estimate derived here. The WHO/EURO Expert group TDI guideline value of 10 pg I-TEQ kg^{-1} (body weight) per day is equivalent to 600 pg I-TEQ day^{-1} for a 60 kg person (MAFF, 1992). The

Table 5. Calculated daily human exposure (ng day^{-1}) to 2,3,7,8-substituted PCDD/Fs

Compound	Daily exposure (ng day^{-1})	I-TEQ (ng day^{-1})	% Contribution to total I-TEQ
2,3,7,8-T_4CDD	0·023 9	0·023 9	11·8
1,2,3,7,8-P_5CDD	0·051 6	0·025 8	12·7
1,2,3,4,7,8-H_xCDD	0·034 3	0·003 4	1·7
1,2,3,6,7,8-H_xCDD	0·518 0	0·051 8	25·6
1,2,3,7,8,9-H_xCDD	0·259 0	0·025 9	12·8
1,2,3,4,6,7,8-H_pCDD	0·547 0	0·005 5	2·7
OCDD	2·075 0	0·002 1	1·0
2,3,7,8-T_4CDF	0·079 2	0·007 9	3·9
2,3,4,7,8-P_5CDF	0·074 8	0·037 4	18·4
1,2,3,7,8-P_5CDF	0·026 3	0·001 3	0·6
1,2,3,4,7,8-H_xCDF	0·075 9	0·007 6	3·7
1,2,3,7,8,9-H_xCDF	0·004 9	0·000 5	0·2
1,2,3,6,7,8-H_xCDF	0·038 8	0·003 9	1·9
2,3,4,6,7,8-H_xCDF	0·035 5	0·003 5	1·7
1,2,3,4,6,7,8-H_pCDF	0·179 0	0·001 8	0·9
1,2,3,4,7,8,9-H_pCDF	0·029 0	0·000 3	0·1
OCDF	0·152 0	0·000 2	0·1
I-TEQ	0·202 7		

background human exposure to all seventeen 2,3,7,8-substituted PCDD/Fs is therefore about 34% of the guideline values.

ASSESSING HUMAN EXPOSURE TO PCDD/Fs FOLLOWING SEWAGE SLUDGE APPLICATION TO AGRICULTURAL LAND

Sewage sludge PCDD/F content and effect on soil concentrations

PCDD/Fs are ubiquitous in sewage sludges, at μg kg^{-1}–ng kg^{-1} concentrations. Table 6 presents some of the PCDD/F sewage sludge concentrations reported in the literature.

Concentrations of 2,3,7,8-T_4CDD generally fall in the range 0–1·0 ng kg^{-1}, while levels of OCDD usually exceed 1000 ng kg^{-1}. PCDD/F concentrations selected for use in the following sludge application scenarios are highlighted in Table 6, those being the median of values reported by Näf *et al.* (1990), Rappe *et al.* (1989), and Broman *et al.* (1990). These concentrations are considered to be representative of PCDD/F concentrations in sewage sludge routinely applied to agricultural land.

At present, 43% of the 1·22 million t dry wt of sewage sludge annually generated in the UK is applied to about 1·3% of available agricultural land. Application rates vary considerably, however, for the purposes of this scenario we use a dry weight sewage sludge application rate of 10 t ha^{-1} which is representative of current UK application rates.

The following sections assume that above and below ground vegetation products harvested for human consumption are grown in soil that has been amended with sewage sludge at a rate of 10 t ha^{-1} dry wt containing PCDD/F concentrations as indicated above, and that the sludge is ploughed into a depth of 15 cm. It is assumed that meat, milk and dairy products are produced in areas where sludge is applied at the same rate, but that it is surface applied and not ploughed in, thus only being incorporated into the soil surface. It is also assumed that there is no PCDD/F degradation in the soil.

Assuming a 2,3,7,8-T_4CDD sludge concentration of 0·72 ng kg^{-1} dry wt and a sludge application rate of 10 t ha^{-1} dry wt, then 7200 ng 2,3,7,8-T_4CDD are applied per hectare, equivalent to 0·72 ng m^{-2}. For the crop production scenario we assume that the soil is ploughed to a depth of 15 cm which weighs an estimated 150 kg m^{-2}. Sludge application would thus increase soil 2,3,7,8-T_4CDD concentrations by 0·0048 ng kg^{-1} (0·96%) to 0·5048 ng kg^{-1}, if agricultural soil is assumed to contain 0·5 ng kg^{-1} of 2,3,7,8-T_4CDD dry wt. The soil I-TEQ is raised by 9·6% under this scenario. Sludge applied to the surface of a pasture can become incorporated into the soil surface by infiltration with mixing caused by rain and the action of livestock hooves. These processes are assumed to result in applied sludge being evenly distributed within the surface 0–5 cm soil layer. This scenario elevates soil 2,3,7,8-T_4CDD concentrations by 0·0144 ng kg^{-1} (2·9%) to 0·5144 ng kg^{-1}. Surface sludge application increases the soil I-TEQ by 144%. Whichever application method is adopted the soil 1,2,3,7,8,9-H_xCDF and OCDD concentrations show the greatest elevations due to their relative enrichment in sludge compared to soil.

Transfers to plants

Above-ground matter

As in an earlier section the 2,3,7,8-T_4CDD concentration in above-ground herbage can be estimated from the equation given by Travis & Arms (1988). This equation gives a dry plant 2,3,7,8-T_4CDD concentration of 0·00229 ng kg^{-1} dry wt (0·000344 ng kg^{-1} fresh wt) for plants grown on sludge amended soil, assuming the soil 2,3,7,8-T_4CDD concentration is 0·5048 ng kg^{-1}. Thus it is predicted that sludge application and incorporation into soil (0–15 cm) would increase plant tissue concentrations by $3{\cdot}3 \times 10^{-6}$ ng kg^{-1} (i.e. 0·96%). Since background plant concentrations arising from atmospheric deposition are 0·0048 ng kg^{-1} fresh wt, it is assumed here that vegetation on sludge treated soil contains 0·004803 ng 2,3,7,8-T_4CDD kg^{-1} fresh wt under this scenario. Estimating the other 2,3,7,8-substituted PCDD/F plant concentrations by this method is difficult owing to the inconsistency of literature reported log K_{ow} values, especially the PCDFs. Table 1 shows the log K_{ow} values used here and includes measured values as well as QSAR estimates. However, given that the transfer of PCDD/Fs from soil to above ground plants is inefficient and that atmospheric inputs dominate, the inconsistency of these log K_{ow} values is not critical to the overall exposure assessment.

Below-ground matter

The soil to root crop transfer coefficient detailed above can be used to predict that 2,3,7,8-T_4CDD concentrations in crop roots grown on sludge-amended soil

Table 6. PCDD/F concentrations reported in sewage sludges (ng kg^{-1} dry wt). Data are congener specific except that from Hagenmaier *et al.* (1985) where figures are homologue specific. Values selected for inclusion in sludge scenario are indicated

Compound	Näf *et al.* (1990) (mean of 3 samples)	Rappe *et al.* (1989) (mean of 2 samples)	Broman *et al.* (1990) (mean of 4 samples)	Hengstmann *et al.* (1990) (median of 3 samples)	Hagenmaier *et al.* (1986) (median of 15 samples)	Lamparski *et al.* (1984) (mean of 2 samples)
2,3,7,8-T_4CDD	0·64	0·72[a]	0·79	12	<10	13·5
1,2,3,7,8-P_5CDD	2·93[a]	2·53	3·43	17	170	—
1,2,3,4,7,8-H_xCDD	2·77[a]	1·35	3·55	219	—	—
1,2,3,6,7,8-H_xCDD	26·0[a]	10·62	39·59	—	940	233
1,2,3,7,8,9-H_xCDD	9·60[a]	3·70	9·96	—	—	—
1,2,3,4,6,7,8-H_pCDD	1 087[a]	720	1 303	4 184	2 300	4370
OCDD	9 833[a]	8 250	10 906	15 567	7 700	55 000
2,3,7,8-T_4CDF	4·73	4·75[a]	5·28	119	160	—
2,3,4,7,8-P_5CDF	4·77	3·55	4·59[a]	64	250	—
1,2,3,7,8-P_5CDF	3·47	2·75	3·10[a]	—	—	—
1,2,3,4,7,8-H_xCDF	6·37[a]	6·65	6·06	121	—	—
1,2,3,7,8,9-H_xCDF	0·93[a]	3·60	0·66	—	—	—
1,2,3,6,7,8-H_xCDF	11·2	2·80	6·70[a]	—	300	—
2,3,4,6,7,8-H_xCDF	6·12[a]	3·60	7·92	—	—	—
1,2,3,4,6,7,8-H_pCDF	128·7	88·0[a]	35·19	161	420	—
1,2,3,4,7,8,9-H_pCDF	2·43[a]	6·10	0·72	—	—	—
OCDF	65·7[a]	83·5	2·54	433	400	—
I-TEQ	33·52	24·08	37·22	—	—	—

[a] Value selected for inclusion in sludge scenario.

(ploughed to 15 cm) will be 0·0252 ng kg^{-1} dry wt or 0·00252 ng kg^{-1} fresh wt, thus being approximately $2{\cdot}1 \times 10^{-5}$ (0·96%) ng kg^{-1} higher than root crops grown on unsludged rural soil.

Transfers to livestock

Soil ingestion

Soil ingestion at 6% of the diet of a lactating cow grazing pasture surface amended with sewage sludge (incorporated into the surface 5 cm) results in ingestion of 0·463 ng 2,3,7,8-T_4CDD per day. A non-lactating cow would consume 0·247 ng of 2,3,7,8-T_4CDD per day. These values are both approximately 2·9% higher than ingestion rates on non-sludge amended soil.

Vegetation ingestion

Ingestion of 2,3,7,8-T_4CDD associated with consumption of sludge grown vegetation can be derived as 0·452 ng kg^{-1} per day by a lactating cow and 0·241 ng kg^{-1} per day for a non-lactating cow. This figure is obtained by estimating above ground herbage concentrations as in the section above. Concentrations of 2,3,7,8- T_4CDD in vegetation ingested by livestock are estimated to be 0·00481 ng kg^{-1} fresh wt (0·0321 ng kg^{-1} dry wt).

Following sludge application, plant surfaces can be contaminated by sludge. Sludge adherence to crops after sludge spray application has been reported by Chaney & Lloyd (1979) and Buttigieg *et al.* (1990). Chaney & Lloyd (1979) reported that, immediately following spray application of liquid sewage sludge, plant material could be over 20% sludge by weight, reducing to about 5% 30 days after application. Buttigieg *et al.* (1990) found that application of liquid sewage sludge at a rate of 67 m^3 ha^{-1} resulted in 35% of the harvested crop being sludge. If sludge containing 12% dry solids was applied at the same rate then the harvested crop immediately after application was 54% sludge by weight. From the time of application, sludge contamination fell due to a combination of vegetation growth and wash-off by rainfall. Decontamination was found to be dependent on rainfall frequency, whether the grass was cut and whether the sludge had time to dry onto the grass. In the study by Buttigieg *et al.* (1990), herbage contamination by sludge was generally reduced to below about 10% after 21 days. Since the EC mandatory no-grazing interval is three weeks (DoE, 1989*b*), it seems inevitable that livestock allowed to graze sludge amended pastures will consume sludge associated with vegetation. The rate of sludge adherence is clearly dependent on many environmental as well as operational variables. However, for this scenario a sludge contamination of 5% (by weight) is taken. It is assumed that the grazing livestock have been consuming sludge at this rate until both milk and body fat are in steady-state with dietary intake. This could take up to 60 days for milk and even longer for body fat (Fries, 1991).

Of the 14·1 kg of vegetation consumed per day by a lactating cow, some 705 g could be dry sludge. This would result in 0·5076 ng and 0·4295 ng 2,3,7,8-T_4CDD being ingested daily in association with sludge and vegetation, respectively, yielding a total 2,3,7,8-T_4CDD vegetation intake of 0·937 ng day^{-1}. A non-lactating cow may also consume 0·27 ng day^{-1} of 2,3,7,8-T_4CDD from sludge adhered to vegetation and 0·229 ng day^{-1} from vegetation, giving a total daily intake of 0·499 ng. This scenario results in higher cattle intake estimates than under background conditions.

Summary

Although it is possible that air concentrations are slightly higher due to volatilisation from the soil, and that some PCDD/Fs may be transferred into the water supply, the intakes of 2,3,7,8-T_4CDD associated with water and air are assumed to be identical whether the land has been sludge amended or not. Thus, a lactating cow which grazes a pasture that has been amended with a single application of sewage sludge at a rate of 10 t ha^{-1}, ingests 0·463, 0·937, $9{\cdot}3 \times 10^{-5}$ and $6{\cdot}45 \times 10^{-5}$ ng 2,3,7,8-T_4CDD per day associated with soil, vegetation, water and air intake, respectively. The total of 1·4 ng day^{-1} constitutes a 55% increase in exposure over background. Total daily exposure of a non-lactating cow is calculated to be 0·747 ng 2,3,7,8-T_4CDD day^{-1}.

Effects on livestock tissue concentrations

Using the fat/diet BCFs detailed in the initial exposure assessment it is possible to estimate that cattle meat fat and milk fat would contain 2,3,7,8-T_4CDD concentrations of 0·327 and 0·35 ng kg^{-1} respectively, equivalent to fresh weight beef and milk concentrations of 0·0817 ng kg^{-1} and 0·0175 ng $litre^{-1}$.

Summary of predicted values

Table 7 presents the fresh weight concentration estimates for the 2,3,7,8-substituted PCDD/Fs in plant herbage, root crops, cow's milk and meat. Milk and meat concentrations are still within the range of measured concentrations reported by MAFF (1992), except for the OCDD concentration in milk.

Transfers of 2,3,7,8-T_4CDD to humans

As in the previous exposure assessment it is possible to use the 2,3,7,8-T_4CDD concentrations predicted to estimate total human exposure, assuming all the produce consumed is derived from an agricultural land to which sewage sludge is applied at a rate of 10 t ha^{-1}. Using identical dietary intake values and the same method to estimate the 2,3,7,8-T_4CDD concentrations in dairy products, a human consuming exclusively produce derived from sludge amended land ingests 12·103 ng of 2,3,7,8-T_4CDD per year or 0·0332 ng per day. This is equivalent to 0·553 pg kg^{-1} BW day^{-1}, assuming an individual body weight of 60 kg. Therefore, this scenario increases human exposure to 2,3,7,8-T_4CDD by 39% over background conditions considered earlier. It should be stressed that the consumption of produce exclusively from sludge application sites is unlikely to occur. This scenario assumes that poultry, eggs and fish PCDD/F concentrations are unaffected by sludge application.

Table 7. Estimates of PCDD/F fresh weight concentrations in vegetation tops and root crops from a site receiving 10 t ha^{-1} of sewage sludge incorporated into the top 15 cm of soil. Meat and milk concentrations come from a site receiving surface sludge applications at 10 t ha^{-1} which becomes incorporated into the surface 5 cm

Compound	Plant herbage (ng kg^{-1})	Plant root (ng kg^{-1})	Milk (ng l^{-1})	Meat (ng kg^{-1})
2,3,7,8-T_4CDD	0·004 8	0·002 5	0·017 5	0·081 7
1,2,3,7,8-P_5CDD	0·021 0	0·002 6	0·052 5	0·191 0
1,2,3,4,7,8-H_xCDD	0·021 0	0·001 9	0·025 6	0·116 0
1,2,3,6,7,8-H_xCDD	0·450 0	0·004 0	0·298 0	1·640 0
1,2,3,7,8,9-H_xCDD	0·210 0	0·002 7	0·168 0	0·719 0
1,2,3,4,6,7,8-H_pCDD	0·886 0	0·171 0	0·949 0	2·970 0
OCDD	3·610 0	1·043 0	10·84 0	6·300 0
2,3,7,8-T_4CDF	0·069 0	0·004 9	0·026 2	0·131 0
2,3,4,7,8-P_5CDF	0·030 0	0·004 8	0·061 1	0·293 0
1,2,3,7,8-P_5CDF	0·027 0	0·005 4	0·012 0	0·059 8
1,2,3,4,7,8-H_xCDF	0·048 0	0·006 6	0·068 2	0·276 0
1,2,3,7,8,9-H_xCDF	0·003 5	0·000 1	0·006 7	0·027 2
1,2,3,6,7,8-H_xCDF	0·024 0	0·004 3	0·044 0	0·213 0
2,3,4,6,7,8-H_xCDF	0·022 5	0·003 9	0·035 5	0·195 0
1,2,3,4,6,7,8-H_pCDF	0·285 0	0·023 6	0·099 0	0·464 0
1,2,3,4,7,8,9-H_pCDF	0·021 0	0·002 0	0·011 4	0·049 0
OCDF	0·300 0	0·077 2	0·032 2	0·146 0
I-TEQ	0·132 0	0·012 4	0·164 0	0·699 0

Human exposure to other 2,3,7,8-substituted PCDD/Fs following sludge application

Table 8 shows the human exposure to all 2,3,7,8-substituted PCDD/Fs, assuming that the individual consumes vegetable products that are exclusively from fields amended with 10 t ha^{-1} dry wt of sewage sludge which has been ploughed to a depth of 15 cm. Meat, milk and dairy products are assumed to be exclusively from cattle grazing pasture which has received 10 t ha^{-1} dry wt surface applications of sludge incorporated into the top 5 cm of soil.

Table 8 shows that, under this sludge application scenario, human exposure to individual PCDD/F congeners is elevated by between 15–400% above background exposure. I-TEQ exposure is raised to 0·3020 ng day^{-1}, which is an increase of 49%. 1,2,3,6,7,8-H_xCDD and 2,3,4,7,8-P_5CDF make the highest contributions to the I-TEQ. The I-TEQ of 5·03 pg kg^{-1} BW day^{-1} (assuming a 60 kg human) derived is approximately 50% of the level of the 10 pg I-TEQ kg^{-1} BW day^{-1} TDI limit prescribed by the WHO/EURO Expert Committee.

Table 8. Human exposure to PCDD/Fs under the sludge application scenario. Percentage elevation over background exposure shown

Compound	Daily exposure (ng day^{-1})	I-TEQ (ng day^{-1})	Elevation (%)
2,3,7,8-T_4CDD	0·033 2	0·033 2	39
1,2,3,7,8-P_5CDD	0·082 5	0·041 3	60
1,2,3,4,7,8-H_xCDD	0·051 1	0·005 1	49
1,2,3,6,7,8-H_xCDD	0·655 0	0·065 5	26
1,2,3,7,8,9-H_xCDD	0·313 0	0·031 3	21
1,2,3,4,6,7,8-H_pCDD	1·560 0	0·015 6	185
OCDD	9·620 0	0·009 6	364
2,3,7,8-T_4CDF	0·091 2	0·009 1	15
2,3,4,7,8-P_5CDF	0·118 0	0·058 8	57
1,2,3,7,8-P_5CDF	0·033 3	0·001 7	27
1,2,3,4,7,8-H_xCDF	0·117 0	0·011 7	54
1,2,3,7,8,9-H_xCDF	0·011 1	0·001 1	124
1,2,3,6,7,8-H_xCDF	0·079 7	0·008	105
2,3,4,6,7,8-H_xCDF	0·070 8	0·007 1	100
1,2,3,4,6,7,8-H_pCDF	0·279 0	0·002 8	55
1,2,3,4,7,8,9-H_pCDF	0·035 8	0·000 4	23
OCDF	0·177 0	0·000 2	17
I-TEQ		0·302	49

General comments

It is important to emphasise that the above exposure estimates assume consumption of produce grown or reared exclusively on sludge-amended soil. Clearly, in reality people consume only a small proportion of sludge influenced produce. It is difficult to generalise on this issue, since intake of such produce will be highly variable. However, to illustrate, if 1·3% (i.e. the percentage of UK agricultural land currently amended with sludge) of the produce consumed is derived from sludge amended land the increase in background exposure to 2,3,7,8-T_4CDD and I-TEQ drop from 39% to 0·51% and from 49% to 0·64%.

From the data obtained it is possible to assess which transfer processes exert greatest influence on human PCDD/F exposure. If sludge is applied only to arable land and not to pasture grassland, background human exposure to 2,3,7,8-T_4CDD and the I-TEQ exposure are increased by only 0·023% and 0·104%, respectively. Even under an extreme worst case situation, such as a sludge application rate of 100 t ha^{-1} dry wt, a plough depth of 5 cm and a soil to root crop transfer coefficient of 0·1, the human 2,3,7,8-T_4CDD and I-TEQ exposure are still increased by only 1·2% and 5·9%, respectively, over background. Again this assumes that the individual consumes products exclusively from sludge amended land. Thus, it appears that application of sludge to crop land will not significantly influence

human exposure to PCDD/Fs, due to the inefficiency of transfers from soil to plant tissues. In contrast, transfers of PCDD/Fs from sludge amended pasture into livestock via ingestion of soil and sludge adhered to vegetation are critical with respect to human exposure. These transfer processes are the principal pathways influencing the human diet, because they offer direct and efficient transfer from the soil/sludge to the grazing livestock, thus elevating meat and milk concentrations, which in turn increase human exposure. In the light of this, and given the lack of information regarding such transfer routes, especially sludge adherence to vegetation, it would seem prudent to investigate these processes further.

Human PCDD/F exposure under different sludge application scenarios

Factors such as sludge PCDD/F concentrations, sludge mixing depth, sludge adherence factors and soil ingestion rate affect human PCDD/F exposure. A few of these factors have been selected to briefly illustrate the possible role they play in PCDD/F transfer from sludge into the human foodchain. Alterations to input parameters on arable land have been shown to have an insignificant affect on human PCDD/F exposure and will, therefore, not be discussed further.

Sludge PCDD/F concentration

Concentrations of PCDD/Fs in sewage sludge vary considerably. If the lowest congener concentrations reported in Table 6 are used under the sludge-to-land scenario detailed above it is possible to calculate that human background 2,3,7,8-T_4CDD and I-TEQ human exposure are increased by 34% and 33%, respectively. Using the highest concentrations causes 2,3,7,8-T_4CDD background human exposure to increase by 760% to 0·205 ng day^{-1} (3·42 pg kg^{-1} BW day^{-1}). The I-TEQ is increased to 0·673 ng day^{-1} (11·2 pg I-TEQ kg^{-1} BW day^{-1}), an increase of about 230% over background exposure, thereby exceeding the I-TEQ WHO/EURO TDI guideline value by 12%. Thus, if sludge contains such high PCDD/F concentrations there is the potential that current guidelines may be exceeded. Again it must be stressed that this assumes that the individual consumes all his produce from agricultural land amended with sludge. If only 1·3% of produce comes from sludge amended land the I-TEQ reduces to below the WHO/EURO TDI guideline value.

Livestock soil ingestion

The amount of soil ingested by livestock has been reported to vary from 0·5% to 50% of the total daily intake (Stark & Hall, 1992). If soil ingestion on the sludge amended pasture is zero, human I-TEQ exposure is 37% higher than background. Thus exposure is reduced by a lack of soil in livestock diet. If 10% of the diet consumed by livestock is soil then I-TEQ is 46% higher than background.

Livestock ingestion of sludge adhered to vegetation

If no sludge associated with vegetation is consumed by livestock, human exposure to 2,3,7,8-T_4CDD and I-TEQ are both only about 1% higher than the background values. If a sludge adherence factor of 1% is used, human 2,3,7,8-T_4CDD and I-TEQ exposures are 9% and 11% higher than background. However, if 10% of the vegetation consumed by the livestock is sludge, values for human exposure to 2,3,7,8-T_4CDD and I-TEQ are 67% and 97% higher than background values. Clearly in the light of these findings this factor has an overriding influence on human exposure to sludge applied PCDD/Fs.

The effect of multiple sewage sludge applications

The above scenarios all deal with single applications of sewage sludge to land, whereas in reality sludges may be applied to soils for several years in succession. Half-lives for H_xCDD, H_pCDD/Fs and OCDD/Fs applied to one field site receiving sewage sludges were reported to be about 2 years (Harrad *et al.*, unpublished data). Here it is assumed that the half-lives of 2 years result in the soil containing 25% of the original sludge PCDD/F input after 4 years, with only 0·4% remaining 20 years after application. If the soil receives twenty 10 t ha^{-1} annual applications of sewage sludge the soil 2,3,7,8-T_4CDD and I-TEQ concentrations would be 0·517 ng kg^{-1} and 3·027 ng kg^{-1}, while the pasture receiving surface applications of sludge would have a 2,3,7,8-T_4CDD and I-TEQ concentrations of 0·55 ng kg^{-1} and 4·459 ng kg^{-1}, respectively.

After 20 successive years of sludge application at 10 t ha^{-1}, human exposure to PCDD/Fs is higher than following a single sludge application. Background daily 2,3,7,8-T_4CDD and I-TEQ exposure are increased by 42% and 53%. The increase is caused almost solely by the increase in the amount of PCDD/Fs ingested by livestock due to higher soil concentrations. For example, grazing livestock consume about 7% more 2,3,7,8-T_4CDD associated with soil. However, it is apparent that most exposure still comes from the ingestion of sludge adhered to vegetation which is a function of the last sludge application. Again it is important to emphasise that the above calculations assume that the individual human consumes produce exclusively from the sludge application site. If only 1·3% of consumed products are from the sludge site the I-TEQ exposure under the last scenario is only 0·7% higher than the background value.

CONCLUSIONS

Background human exposure to 2,3,7,8-substituted PCDD/Fs has been estimated as 0·2027 ng I-TEQ day^{-1} using a detailed assessment protocol. This protocol includes transfers of PCDD/Fs from soil into above-ground and below-ground vegetation, transfers into animal meat/milk and ingestion into the human system. Dietary intake of food is revealed to be the major

exposure route (>99%), whilst intakes from water and air are negligible.

Human PCDD/F exposure is elevated if sewage sludge containing typical PCDD/F concentrations is applied to soil. Application of sludge to arable land appears to have only a very minor effect on human exposure to PCDD/Fs. The most important transfer mechanism from sludge amended soil to the human foodchain is illustrated to be livestock ingestion of sludge adhered to vegetation. It is apparent that the current WHO/EURO TDI guideline value of 10 pg I-TEQ kg^{-1} BW day^{-1} is exceeded only when a sludge containing very high PCDD/F concentrations is considered and the individual concerned consumes products exclusively reared from agricultural areas receiving sludge. If only 1·3% of consumed produce came from such an area, then human exposure would remain below guideline levels. If sludge ingestion by livestock could be eliminated as a source of PCDD/Fs, I-TEQ exposure would be below 1% higher than the background level. In the light of these findings it would seem appropriate to gather more information on actual PCDD/F levels in sludge currently applied to land and to gain further insights into the amounts of sludge consumed by grazing livestock following sludge spraying to pasture. Sub-surface injection of sludge, rather than surface spraying would clearly reduce the potential of PCDD/Fs to enter the human foodchain.

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