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Review article

The significance of cadmium entering the human food chain via livestock ingestion from the agricultural use of biosolids, with special reference to the UK

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ABSTRACT

When biosolids are applied to agricultural soil, potentially toxic elements (PTEs) accumulate in the topsoil, although it takes many repeated applications to reach soil limit values.

Two programmes of UK government-funded research were commissioned in the 1990s to investigate the transfer of PTEs to the food chain via ingestion by sheep grazing biosolids-amended soil. Here, we critically re-examine this evidence in the light of other published work and current food quality standards. This was particularly motivated by the need to determine the safety of biosolids controls on PTEs in relation to revised and stricter European food quality controls for PTEs in foodstuffs.

The major pathway for transfer of PTEs to grazing livestock is via direct ingestion of biosolids or biosolids-amended soil from the soil surface. The main elements of concern for the human diet are cadmium (Cd) and lead (Pb), with Cd being the focus of the current paper. Animal ingestion of plant tissue is also a potential pathway for Cd exposure, which, unlike Pb, can transfer to crop tissues.

The concentrations of Cd in the muscle tissue of sheep grazing biosolids-amended soil were generally small and similar to control values. Cadmium concentrations in sheep offal were below the maximum permitted concentration for human consumption. This was despite ingestion of soils exceeding the maximum permissible concentration for Cd in soil (3 mg kg^{-1} dry soil) by up to three times, at an ingestion rate of 10% total dietary dry matter intake. Grazing trials under practical conditions on high Cd soils demonstrated that the Cd concentrations in sheep offal remained below the food limit value for this element in offal from the combined intakes from biosolids-amended soil and herbage. Futhermore, given the substantial fall in environmental emissions and concentrations in biosolids of this element and, consequently, it cannot accumulate in soil to the limit value, biosolids Cd does not represent an issue for the safety of animal meat products.

1. Introduction

1.1. Background

Application of sewage sludge biosolids to short- and long-term pasture has been an important route for beneficial recycling of biosolids in agriculture. It provides an outlet during periods when cultivated arable areas are not available due to the presence of established crops (Smith, 1996).

There are three possible routes for the transfer of potentially toxic elements (PTEs) to tissues of animals grazing biosolids amended pasture systems:

(i) ingestion of PTEs in biosolids adhered to herbage;
(ii) ingestion of biosolids/biosolids-amended soil from the soil surface; and
(iii) ingestion of herbage with elevated concentrations of PTEs as a consequence of plant uptake (Hill et al., 1998).

Direct ingestion of biosolids/biosolids-amended soil from the soil surface represents the most significant route for the transfer of PTEs to the tissues of animals grazing biosolids-amended soils (Carrington et al., 1989; Carrington et al., 1998a,b; Smith, 1996). Currently, given that the major use of biosolids on grassland is prior to establishing grass leys, ingestion of biosolids-amended soil is more likely than direct ingestion of biosolids from the soil surface.

The soil limits for agricultural recycling of biosolids (CEC, 1986; SI, 1989) vary according to the behaviour of the particular element and its

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critical environmental end-points (Carrington et al., 1998a) (Section S1, Supplementary Material), and it is Cd, Pb and mercury (Hg) that are controlled due to their potential risk to human health. Cadmium is readily absorbed by crops and can also accumulate in the offal of grazing livestock via ingestion of biosolids or biosolids-amended soil. Lead is not absorbed by plants, but can enter the human food chain via animal ingestion of biosolids or biosolids-amended soil and transfer to offal. Mercury is also a potential risk to the food chain, however, it is not taken up by crops and usually occurs in very small concentrations in biosolids. The soil limit for Cd is 3 mg kg^{-1} at all soil pH values ≥ 5.0 (Defra, 2018) (Table S1, Supplementary Material).

Cadmium absorption by livestock grazing biosolids-amended soil is complex and influenced by interactions between other soil, biosolids or dietary constituents (Stark et al., 1996). Soil characteristics influencing Cd absorption may include: soil electrical conductivity; clay, humus and sand contents; calcium (Ca), phosphorus (P) and sulphur (S) contents, and soil temperature (Sims and Kline, 1991; Phillips and Tudoreaunu, 2011). The presence of iron (Fe), zinc (Zn) or Ca in biosolids, soil or grass may also be antagonistic towards the absorption of biosolids-borne Cd. Indeed, the decline in Cd relative to Zn in biosolids has reduced the Cd to Zn ratio, significantly lowering the potential for Cd accumulation (Smith, 1996).

1.2 Research to investigate animal ingestion of PTEs by livestock grazing biosolids-amended pasture

In the 1980s, a review of the ingestion of biosolids-amended grassland soils by grazing ruminants (Stark, 1988), found the published literature concerning the accumulation of PTEs in animal tissues was limited. Hence, the UK Government funded two major programmes of research on animal ingestion of PTEs by sheep from biosolids-amended pasture. The then Ministry of Agriculture, Fisheries and Food (MAFF) and the Department of the Environment (DoE) separately commissioned two complementary series of indoor feeding trials, in 1990 and 1991, respectively. The aims of both programmes were:

(i) to provide data on the transfer of PTEs from ingested biosolids-amended soil to the liver, kidney and muscle tissues of sheep (Stark et al., 1996; Stark and Wilkinson, 1994); and
(ii) provide assurance that the limit values of PTEs in biosolids-amended agricultural soils (SI, 1989; DoE, 1989) avoided unacceptable transfers of PTEs to meat or other edible products, and to determine the margins of safety.

The trials were conducted with sheep to represent a worst-case condition for ingestion of PTEs due to their greater soil ingestion rate compared to cattle (Thornton and Abrahams, 1983). A range of grazing situations for weaned lambs (MAFF and DoE) and breeding ewes (DoE) on pasture that had received applications of biosolids were simulated using indoor feeding trials. Biosolids-amended soils were pelletised with dried grass to provide a range of PTE intakes. The influence of the relative proportions of biosolids-amended soil in the diet, whether the soil was recently or historically amended with biosolids, and soil characteristics, were also investigated. Following the indoor feeding trials, MAFF commissioned a series of sheep grazing trials to assess the transfer of PTEs from biosolids-amended grassland under practical farming conditions. After completion of the research, Stark et al. (1998) reviewed the integrated findings of the MAFF/DoE research programmes. Further to this, the evidence underpinning the 1989 (revised in 1996) Code of Practice for Agricultural Use of Sewage Sludge (DoE, 1989, 1996), and the evidence since 1989 was reviewed by Carrington et al. (1998a,b). A main focus was the scientific basis to the PTE limits and potential for transfer to the food chain, including by animal ingestion. However, an integrated assessment of the findings of these major feeding and grazing experiments has not been published in the scientific literature. Therefore, it is important to re-visit this research when considering the potential risk to the food chain from PTEs in biosolids recycled to agricultural soil, in relation to current food safety standards.

Only a small number of directly relevant studies have been conducted since the MAFF/DoE funded programmes (Rhind et al., 2005). The overall aim here was to examine the evidence from the MAFF/DoE research programmes in the light of other published work on animal ingestion of Cd. This was to assess and update the conclusions on the significance of Cd in biosolids entering the human diet from animal food products in relation to current food quality standards for Cd.

The specific objectives of this critical review are to:

(i) give a general context to the presence of Cd in food and describe the legislation relevant to controlling Cd in the food chain in the UK and the EU;
(ii) examine the presence of Cd in biosolids and in soil;
(iii) discuss the uptake of Cd by pasture species;
(iv) examine the uptake of Cd to food products by sheep (in the MAFF/DoE trials) and other livestock grazing biosolids-amended soil, in the UK and internationally;
(v) consider the relative availabilities and bioaccumulation factors for dietary Cd from different sources, and
(vi) draw conclusions on the implications of research investigating the transfer of Cd from biosolids-amended soil to animal food products, in relation to food quality standards and Cd soil limits.

2. Cadmium in food and animal feed and legislation controlling cadmium in the food chain

2.1. EU and UK legislation controlling cadmium in food and animal feed

The underlying principles of EU legislation on contaminants in food are outlined in Regulation 315/93/EEC (CEC, 1993), which states that:

(i) food containing a contaminant at a toxicological level, or to an amount unacceptable to public health, is not to be placed on the market;
(ii) contaminant concentrations must be kept as low as can reasonably be achieved following the precautionary principle and recommended good working practices, and
(iii) maximum concentrations must be set for certain contaminants to protect public health. The maximum limits on contaminants in foodstuffs are controlled by Commission Regulation (EC) No. 1881/2006 (EC, 2006), and were implemented in the UK by The Contaminants in Food (England) Regulations 2013 (SI, 2013).

Commission Directive 2002/32/EC sets maximum limits for contaminants in animal feed. The maximum permitted Cd concentration in complete feeding-stuffs for cattle, sheep and goats is 1.0 mg kg^{-1} feed at 12% moisture (EC, 2002). The US National Research Council (NRC, 2005) reviewed the maximum tolerable concentrations of PTEs in the diets of domestic animals, and values for PTEs of concern in biosolids are summarised in Table S2 (Supplementary Material). For Cd, the maximum tolerable concentration in the diet is 10 mg kg^{-1} dry matter (DM) in cattle, sheep, swine and poultry feed.

2.2. Cadmium in food

Cadmium has accumulated in the environment mainly as a consequence of the agricultural use of Cd containing P fertilisers, and from historic mining activities (FSA, 2009). Foodstuffs are the main source of Cd exposure for the non-smoking general population (EFSA, 2012).

Whilst not a specific benchmark, UK total diet studies (TDSs) (FSA, 2015) provide a snapshot of the average concentrations of contaminants in composite samples of different food groups available from UK retail outlets. For example, in the 2014 TDS, Cd was present in small concentrations in 28 food groups, and was below the limit of detection (0.0012–0.006 mg kg^{-1}; note elemental concentrations in food are reported as fresh weight unless specifically stated) in carcass meat, poultry, oils and fats, eggs, fresh fruits, fruit products, beverages, milk and dairy products (FSA, 2015). Cadmium concentrations were largest in offal (0.067 mg kg^{-1}) and snacks (0.059 mg kg^{-1}). As expected, food consumed in the greatest quantities made the most significant contribution to the total dietary intake of Cd. For example, for the mean intake of Cd for adults aged 19 and over, potatoes (0.025 μg kg^{-1} body weight (bw) day^{-1}), cereals (0.026 μg kg^{-1} bw day^{-1}) and bread (0.024 μg kg^{-1} bw day^{-1}) were the most significant groups. The mean estimated Cd dietary intakes of different consumer groups calculated by the UK Food Standards Agency (FSA) were 0.12–0.54 μg kg^{-1} bw day^{-1}, and the maximum exposures of each group were in the range 0.21–0.85 μg kg^{-1} bw day^{-1} (FSA, 2015).

The Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a provisional tolerable weekly intake (PTWI) for Cd of 7 μg kg^{-1} bw in 1988 (JECFA, 2001). This was reviewed in 2010 and a provisional tolerable monthly intake (PTMI) of 25 μg kg^{-1} was established, corresponding to a lower weekly intake of 5.8 μg kg^{-1} (JECFA, 2011). However, the European Food Safety Authority (EFSA) panel on contaminants in the food chain (CONTAM) recommended that the PTWI should be reduced to a tolerable weekly intake (TWI) of 2.5 μg kg^{-1} bw, to ensure protection for exposed and vulnerable members of the population (EFSA, 2009).

Food limits for Cd provided by Commission Regulation (EC) No. 1881/2006 are close to the background values found in food and the limits for carcass meat, liver and kidney are shown in Table 1. In 2009, the CONTAM panel concluded that certain sub-groups of the population may be at risk of exceeding the PTWI for Cd, hence, in 2014 the European Commission (EC) adopted a Recommendation on the Reduction of the Presence of Cd in Foodstuffs (EC, 2014).

3. Cadmium contents in biosolids and soil

The median concentration of Cd in UK soils is 0.33 mg kg^{-1} dry soil (ds), with a range between 0.14 and 0.48 mg kg^{-1} ds (Rawlins et al., 2012). Geogenic Cd is potentially the major source of Cd in UK soils (Birke et al., 2017). However, in addition, the soil is a long-term sink for cationic PTEs because they are strongly adsorbed by the soil matrix (Violante et al., 2010), and leaching losses and plant uptake are usually relatively small compared to the total quantities entering the soil from different sources. In England and Wales, for example, inorganic fertilizers account for 39% of total Cd inputs, 34% were from atmospheric deposition, 10% from biosolids and 9% from livestock manures (ADAS, 2010). One of the major sources of Cd enrichment of agricultural soils internationally is inorganic P fertilisers (ADAS, 2010; Kelliher et al., 2017; Roberts and Longhurst, 2002; Stafford et al., 2016, 2018). Hence, PTEs may slowly accumulate in soil and, over the long-term, have implications for soil quality and function, and animal and human health. Consequently, reducing PTE inputs to soils is a key aim of UK and EU soil protection policies (EC, 2012; Defra, 2018; Defra, 2009).

Potentially toxic elements occur in biosolids as a consequence of inputs from domestic sources, road run-off and industrial inputs to sewers. They typically accumulate in biosolids in higher concentrations than they are present in soil (Carrington et al., 1989a,b). For instance, at the time the Code was drafted the median concentration of Cd in biosolids used in agriculture was 9 mg kg^{-1} dry solids (DS) (Carrington et al., 1989a), which was an order of magnitude greater than the median concentration of Cd in soils in England and Wales of 0.33 mg kg^{-1} ds (Rawlins et al., 2012).

Strict source controls on industrial contaminants have been implemented since the 1970s, coupled with a reduction in PTEs used in industry, and the decline in environmental emissions is reflected in the significant reductions in PTEs observed in biosolids (Smith et al., 2018) (Table S3, Supplementary Material). For example, Gendenbien *et al.* (2008) reported a median Cd concentration of 1.3 mg kg^{-1} DS in UK sludge and similar Cd concentrations of 1.2 and 2.4 mg kg^{-1} DS (UK; Rigby et al. 2015), 1.4 mg kg^{-1} DS (Brazil; Campos et al., 2019), and 3–3.4 mg kg^{-1} DS (Romania; Ilie et al., 2018), 3 mg kg^{-1} DS (median value; range 0.8–155 mg kg^{-1} DS) (Australia, Brazil, Canada, China, India, Iran, Poland, Romania, Spain, Sweden, UK, USA; Wang and Flury, 2019) have been reported internationally. The most recent data for the UK, for biosolids produced by three major UK water utilities (Smith et al., 2018), indicated the mean Cd content had decreased to 1.0 mg kg^{-1} DS.

Metal additions to soil associated with repeated annual applications of biosolids of 5.7 t DS ha^{-1} (compliant with nitrogen (N) application restrictions) are presented in Table S4 (Supplementary Material) and are all within permissible limits (Table S1). Table S4 shows that, given the relative concentrations of PTEs in biosolids (Table S3) and the current metal limits in soil, the PTEs that are the most limiting for the majority of biosolids are Zn and Cu. This trend has also been observed in other more recent studies (Rigby et al., 2015; Smith et al., 2018; Campos et al., 2019; Eid et al., 2019; Wang and Flury, 2019). The mass balance model shows that the soil metal limit values for Zn and Cu would be reached following 75 and 73 applications of biosolids, respectively (assuming median background concentrations of these elements in soil). The biosolids Cd concentration is significantly below the maximum limit in soil (3 mg kg^{-1} ds, Table S1, Supplementary Material). Consequently, the limit value for Cd, which would require 722 repeat annual applications is practically impossible to achieve (again, assuming a median background concentration suitable for agronomic applications of biosolids). However, this simple soil accumulation model is arguably precautionary because it assumes that metals are added as a mass and does not account for the biosolids concentration and blending effect in soil. Nevertheless, it shows that maximum Cd accumulations in agricultural soils receiving contemporary biosolids applications may only potentially increase to less than 10% of the maximum permissible limit for this element, due to the relatively larger Cu and Zn accumulation rates. Thus, the maximum permissible soil limit value for Cd can never be attained under practical biosolids application conditions, even if biosolids were supplied indefinitely to the same area of land.

4. Uptake of cadmium by herbage

In contrast to Pb, Cd is relatively mobile in soil and its absorption by crops is closely related to the Cd concentration in soil and the soil pH value (Arvas et al., 2013; Chaney et al., 1978; Dowdy et al., 1983; Reiser et al., 2014; Smith, 1994; Smith et al., 2009; Stark, 1988) (also see Table S5, Supplementary Material). A decrease in soil pH increases Cd solubility and, hence, bioavailability to crops (Smith, 1994; Carrington et al., 1989a; Reiser et al., 2014). Plant uptake is reduced when the soil contains elevated Cd concentrations to a depth of 7.5 cm, representative of surface applications of biosolids to pasture and no cultivation, compared to the same Cd concentration at a depth of 25 cm in cultivated land, because grass roots extend through the cultivation depth (Stark et al., 1998). Cadmium concentrations in ryegrass are typically approximately 10% of the concentration in soil; however,

Table 1
Limits for cadmium in animal products for human consumption at the time of the MAFF/DoE trials and currently (FW basis).

	Limits during the MAFF/DoE trials (1990–91)	Current limits
	(mg kg^{-1})	
Source	Draft EC Regulations	CEC (2006)
Muscle	0.05–0.1	0.05
Liver	nls	0.5
Kidney	1	1

nls; no limit specified.

livestock usually ingest far greater quantities of plant DM compared to soil so, under certain circumstances, Cd intakes in herbage may represent a significant source of Cd to livestock (Stark et al., 1998).

Grass swards generally contain a number of herbage species other than ryegrass (other grass species, clover and other dicotyledonous plants) and the relative bioavailabilities and retention of organic and inorganic forms of Cd by animals ingesting soil, biosolids and plant material have been investigated (Prankel et al., 2004; Phillips et al., 2005) (see Section 7). In other parts of the world, forage species other than traditional pasture grasses may also be integrated into grazing systems (Brekken and Steinnes, 2004; Madejón et al., 2012; Stafford et al., 2016). In New Zealand, for example, this may include summer and winter grazed forage brassicas or chicory and plantain, which are beneficial due to their growth and resistance during hot, dry summers (Stafford et al., 2016). Stafford et al. (2016) investigated the uptake of Cd by these forage species in a glasshouse trial grown in soil with a Cd concentration of 0.43 mg kg^{-1}ds, and found that chicory and plaintain had a significantly greater Cd concentrations (1.64 and 0.73 mg kg^{-1} DM respectively) than other species (0.04–0.53 mg kg^{-1} DM) including turnip, kale and perennial ryegrass amongst others.

In another glasshouse trial, Gray and McLaren (2005) found up to a 2–fold range in Cd plant tissue concentrations between different ryegrass varieties, with short rotation/annual varieties accumulating significantly smaller amounts of Cd (and other PTEs) than the other varieties investigated. However, Loganathan et al. (1997) found less Cd uptake by clover compared to pasture grass species and, as the yield was also lower, its contribution to pasture Cd uptake was relatively small (< 7%). The apparent lack of data on the variation in Cd uptake between different pasture species was recently highlighted by Reiser et al. (2014). Indeed, in contrast to the relatively comprehensive animal feeding investigations conducted on biosolids, there remains uncertainty about the potential significance of Cd intakes from different dietary sources by animals grazing fertilised pastures in general and particularly from long-term inputs of Cd in mineral phosphate fertilizers (Nicholson et al., 2003).

5. Cadmium uptake into the tissues of sheep and other livestock grazing biosolids-amended pasture

5.1. Cadmium uptake into muscle tissue in UK direct feeding studies

The experimental details of the MAFF/DoE feeding trials are described fully in Section S5.1 of the Supplementary Material. Lambs and ewes were fed diets containing between 0.04 and 1.04 mg Cd kg^{-1} (dry basis) (Table 2). In MAFF Experiment 1, three different soil types (loamy sand – Soil S, sandy clay loam – Soil L and a clay loam – Soil C) were blended with four rates of highly contaminated biosolids and added to the basal diet at a constant rate of 10% of total DM intake. MAFF Experiment 2 used soils from historical biosolids experimental sites (Cassington and Royston) each with three different Cd concentrations, plus an unamended control soil, that were added to the basal diet at a rate of 10% DM intake. The basal diet was dried grass with background concentrations of Cd (and other PTEs). In the DoE trials, lambs or ewes were fed dried grass blended with one of three biosolids-amended soils (two clay loam soils and one clay soil), containing a range of Cd concentrations (Table 2), from long-term biosolids-treated sites at rates of 2.5, 5.0 or 10% of total dietary DM.

Cadmium did not accumulate in muscle tissue of lambs and ewes in any of the MAFF/DoE indoor controlled feeding trials, and concentrations of this element remained below the detection limit of 0.04 mg Cd kg^{-1} fresh weight (FW) in muscle tissue (Table 2). Hence, concentrations were below the current maximum permissible concentration for carcass meat of 0.05 mg Cd kg^{-1} FW (CEC, 2006).

Concentrations of Cd in carcass meat in the 2014 UK TDS were below the detection limit, which was considerably smaller than in the earlier MAFF/DoE studies, and equivalent to < 0.006 mg kg^{-1} (FSA, 2015). Therefore, it is not possible to determine whether the Cd concentrations in the muscle tissue were in a similar range to the values currently found in carcass meat in the UK.

5.2. Cadmium uptake into liver and kidney in UK direct feeding studies

In general, liver Cd concentrations in the MAFF/DoE trials reflected the Cd concentrations in the diet (Table 2) (Stark et al., 1998). For lambs fed control diets containing no soil or unamended soil, concentrations were low, and similar to or less than the detection limit.

The largest liver Cd concentrations (0.15–0.20 mg Cd kg^{-1} FW) were observed for groups of lambs ingesting the highest rates of recently or historically biosolids-amended soils at two to three times the statutory soil limit concentration for Cd (diets S3, C3 and Cassington soil) (Table 2). However, in all cases, concentrations of Cd in the liver of lambs remained below the current maximum food quality standard for Cd in liver of 0.5 mg Cd kg^{-1} FW (CEC, 2006).

The ingestion of biosolids-amended soil also increased liver Cd concentrations for groups of ewes (Table 2) (Stark et al., 1998). The raised concentration of Cd in the liver of aged control ewes in DoE Study 2 (0.056 mg kg^{-1} FW) compared to younger animals was not unexpected. This suggested that Cd accumulation in offal tissue had occurred over the lifetime of the animals, prior to the commencement of the DoE feeding trial, from background dietary sources (Stark et al., 1998).

Ewes in the DoE Study 2, fed a diet containing soil at 1–1.5 times the statutory Cd limit (10% total DM intake) (N100), had similar median liver concentrations (0.15 mg kg^{-1} FW) to lambs fed the same diet or a diet containing soil with twice the statutory Cd limit (10% total DM intake) (W100) (Table 2). These liver Cd concentrations were greater than for the control ewes or for any of the ewes in DoE Study 1 (0.036–0.067 mg kg^{-1} FW). Interestingly, Cd concentrations in livers of the Study 1 ewes, which had been fed biosolids-amended soils for 3 years since weaning, appeared to reach a plateau at values similar to those in the lambs (Stark et al., 1998). These animals were continuously fed a diet containing 5% of their DM intake, and the soil contained up to approximately three times the maximum permissible limit value for Cd, representing an extreme worse-case. Therefore, the results suggested that, in the long-term, dietary factors, animal husbandry and Cd transport from the liver may restrict the overall upper accumulation of Cd in liver tissue of sheep (Table 2), below values that would be considered a concern to the human diet (Table 1).

The kidney is the critical organ for long-term Cd exposure in both animals and humans, where it irreversibly impairs re-absorption of proteins, sugars and amino acids (de Vries and McLaughlin, 2013). The kidney Cd concentrations for finished lambs observed in the MAFF/DoE trials were generally in the same range to those for liver and followed a similar pattern (Stark et al., 1998). However, for the ewes in the DoE study, Cd concentrations were between 2 and 6 times greater in the kidney than in the liver (Stark et al., 1998) (Table 2). Nevertheless, a maximum concentration of 0.39 mg kg^{-1} FW was observed in kidney tissue of sheep ingesting biosolids-amended soil W (twice the statutory soil Cd concentration) at 10% of their total dietary DM intake (Table 2), which is well below the current maximum concentration for Cd in kidney of 1.0 mg kg^{-1} (CEC, 2006).

In general, liver Cd concentrations were higher at the final slaughter point compared to the intermediate slaughter stage in MAFF Experiment 1, although the rate of increase diminished with the period of dietary exposure. This behaviour was also confirmed by liver biopsies, which were taken at approximately two-monthly intervals during the DoE trials and showed reductions in the rates of Cd accumulation as the trials progressed.

For kidney, a significant increase in Cd concentration occurred between the two slaughter ages, but, unlike liver, no plateau was observed in the rate of accumulation of Cd in the kidney (Stark et al., 1998). This reflected the normal accumulation of larger concentrations of PTEs in

Table 2
Cadmium (Cd) concentrations in control and biosolids-amended soils and diets fed to lambs and ewes, and the resulting concentrations in muscle, liver and kidney tissue during DoE (Stark et al., 1996) and MAFF (Stark and Wilkinson, 1994) indoor feeding trials (tissue concentrations are for lambs unless indicated otherwise).

Treatment	Rate of soil addition to diet (% DM)	Cd concentration (mg kg^{-1}) (dry basis)		Cd concentration in animal tissue (mg kg^{-1} FW)		
		Soil	Diet	Muscle	Liver	Kidney
MAFF Experiment 1						
Soil S	10	0.25	0.08	< 0.04	< 0.018	0.033
Soil L	10	0.45	0.18	< 0.04	< 0.017	0.033
Soil C	10	0.23	0.14	< 0.04	< 0.021	0.038
Biosolids-amended S1[a]	10	1.62	0.23	< 0.04	0.023	0.052
Biosolids-amended S2[b]	10	3.01	0.36	< 0.04	0.085	0.11
Biosolids-amended S3[c]	10	6.06	0.64	< 0.04	0.20	0.17
Biosolids-amended soil L1[a]	10	1.70	0.23	< 0.04	< 0.022	0.058
Biosolids-amended soil L2[b]	10	3.47	0.36	< 0.04	0.11	0.088
Biosolids-amended soil L3[c]	10	6.10	0.63	< 0.04	0.20	0.18
Biosolids-amended soil C1[a]	10	1.81	0.22	< 0.04	0.027	0.071
Biosolids-amended soil C2[b]	10	3.66	0.35	< 0.04	0.14	0.12
Biosolids-amended soil C3[c]	10	6.01	0.63	< 0.04	0.15	0.12
MAFF Experiment 2						
Royston control soil	10	0.75	0.19	0.04	0.018	0.020
Cassington control soil	10	0.62	0.13	< 0.032	0.021	0.023
Royston Guide	10	4.16	0.49	< 0.40	0.057	0.061
Royston Medium	10	7.03	0.77	< 0.037	0.061	0.071
Cassington Guide	10	2.93	0.38	< 0.037	0.074	0.048
Cassington Medium	10	6.23	0.70	< 0.033	0.12	0.087
Cassington High	10	8.82	1.04	< 0.038	0.15	0.14
Wye control (grass only)	N/A	N/A	0.11	< 0.035	0.023	0.029
Treatment	**Rate of soil addition to diet (% DM)**	**Cd concentration (mg kg^{-1}) (dry basis)**		**Cd concentration in animal tissue (mg kg^{-1} FW)**		
		Soil	**Diet**	**Muscle**	**Liver**	**Kidney**
DoE, Study 1 (ewes)						
Control (no soil)			0.057/0.09/0.13	≤0.04	0.042	0.173
Biosolids-amended soil H	5	1.65/1.66	0.12/0.16/0.18	≤0.04	0.036	0.163
Biosolids-amended soil N	5	3.30/4.46	0.19/0.25/0.32	≤0.04	0.047	0.238
Biosolids-amended soil W	5	6.02/8.55	0.31/0.40/0.39	≤0.04	0.067	0.389
DoE, Study 2						
Control (no soil)			0.044		≤0.02	0.03
Biosolids amended soil N	10	3.30/4.46	0.35		0.07	0.09
DoE, Study 2 (ewes)						
Control			0.044	≤0.04	0.056	0.19
Biosolids-amended soil N	10	3.30/4.46	0.35	≤0.04	0.152	0.29

[a] Biosolids added to soil at a rate of 7.5% dry weight.
[b] Biosolids added to soil at rate of 15% dry weight.
[c] Biosolids added to soil at a rate of 30% dry weight.
[d]Dietary PTE concentrations differed slightly during the course of DoE Study 1 due to small differences in in the PTE content of dried grass and/or batches of soils used.

the body tissues of older animals compared to younger livestock with the lifetime intake of PTEs (Baxter et al., 1982; Stark, 1988; Lee et al. 1996).

No evidence of placental transfer of Cd was observed in the DoE trials, and liver and kidney Cd concentrations were below detection limits (equivalent to 0.017 mg kg^{-1} FW for liver and 0.023 mg kg^{-1} FW for kidney) in all newborn lambs (Stark et al., 1998). There was also little evidence of Cd transfer during suckling as concentrations of Cd in milk and in the liver and kidneys of lambs slaughtered at weaning were also below detection limits (equivalent to 0.019 mg kg^{-1} FW for liver and 0.023 mg kg^{-1} FW for kidney) (Stark et al., 1998). Similar findings have been observed for different animal species ingesting high Cd diets. For example, Telford et al. (1984) fed grass-legume silage grown on biosolids-amended soil containing as much as 135.5 mg Cd kg^{-1} ds to pregnant goats. The silage grass produced on Cd contaminated biosolids-amended soil had a concentration of 3.81 mg Cd kg^{-1} DM compared to 0.14 mg Cd kg^{-1} DM in the control. However, there was no evidence of placental or mammary transfer of Cd to the kids. Furthermore, no consistent differences in the Cd content in milk or in the body tissue of the kids were found compared to the control groups.

Direct feeding studies also emphasise the importance of soil type as a major factor modulating the extent of Cd transfer to livestock and higher Cd concentrations in the tissues of livestock grazing sandy soils are likely, compared to clay soils (Morcombe et al., 1994). Greater PTE uptakes were also observed for sandy soil in the MAFF experiments (Stark and Wilkinson, 1994). For example, in MAFF Experiment 1, kidney Cd concentrations were 40% larger for the same biosolids added to a sandy soil (Soil S) compared to the other two soil types examined with greater clay contents (Table 2).

5.3. *Cadmium retention by livestock in UK outdoor grazing studies*

An outdoor grazing study commissioned by the UK MAFF investigated the accumulation of PTEs in edible tissues of lambs reared on permanent pastures that had received repeated applications of biosolids between 1981 and 1994 (Wilkinson et al., 2001). The accumulation of PTEs in tissues of blocks of lambs allocated to biosolids-treated pasture and untreated control pasture was examined by monthly serial slaughter in two experiments over two consecutive grazing seasons in 1994 and 1995, with the main focus of the research being the accumulation of Cd by sheep. The concentration of Cd in soil samples taken to 25 cm depth from the treated sites was close to the UK statutory limit (3 mg Cd kg^{-1} ds), and exceeded the limit by 1.67 times at the soil surface; this was compared to concentrations of 0.38–2.53 mg Cd kg^{-1} ds on the rest of the farm. The concentrations of other PTEs in soil were below statutory limits for the 15 or 25 cm sampling depth (Table S1) and advisory limits for grassland at the 7.5 cm sampling depth (Table S1). Mean concentrations of Cd, Pb, Cu and Zn in herbage were higher at the treated site compared to the untreated control. In the second experiment, biosolids were applied to the treated site between the grazing seasons, five months before grazing commenced.

During the first grazing period, the Cd concentration in plant tissue from the biosolids-amended blocks was 0.09–0.46 mg kg^{-1} DM compared to < 0.04–0.26 mg kg^{-1} DM in the control. In the second experiment it was higher at 0.25–1.63 mg kg^{-1} DM in the biosolids treatment compared to 0.13–0.19 mg kg^{-1} DM in the control (Wilkinson et al., 1997). Therefore, during the first grazing period, in some cases the grass Cd concentration approached the NRC limit at the time of 0.5 mg Cd kg^{-1} DM in feeding stuffs (NRC, 1980), and the limit was exceeded in the second experiment by over 3 times in some cases. The EU Directive on undesirable substances in animal feed (EC, 2002) set a 1 mg kg^{-1} upper limit for Cd in complete feed, which was also exceeded in some cases in the second experiment. The study was conducted on swards with relatively low herbage mass, to encourage high rates of soil intake. Thus, the mean proportion of total DM ingested by the lambs as soil was 10% in the first grazing period and 5% in the second.

No significant differences in the PTE concentrations in muscle tissue were observed between lambs on the biosolids-treated and untreated site. However, Cd in kidneys of lambs grazing treated pasture increased as the growing season progressed from 0.028 to 0.066 mg kg^{-1} FW in the first grazing period and from 0.0041 mg kg^{-1} FW initially to 0.53 mg kg^{-1} FW in the second. Therefore, Cd remained below the maximum limit permitted in kidney (1 mg kg^{-1} FW) (CEC, 2006), under these representative worst-case conditions.

Similar trends to those for kidney Cd were also observed for liver Cd and in the second grazing period, Cd concentrations in liver increased to 0.305 mg kg^{-1} FW compared to a maximum permitted concentration of 0.5 mg Cd kg^{-1} FW. The differences in the accumulation of Cd observed between the two grazing periods were probably as a consequence of the application of biosolids to the site between the two trials. It may also have been due to an alteration of Cd bioavailability already present in the soil, although the mechanism was not fully understood. The amount of Cd ingested in the diet originating from different sources, including historically biosolids-amended soil, recently applied biosolids and grass was not known, and Stark et al. (1998) suggested that the results emphasise the need to quantify these sources and the relative availabilities of Cd to grazing animals from each source. Nevertheless, the results indicated overall that, under worst-case conditions and a surface soil Cd content almost 1.7 times the maximum permitted value, liver and kidney Cd were below the food limit for this element.

Rhind et al. (2005) also reported a sheep grazing trial on biosolids-amended pasture, which was conducted over a 4–5 year period. The Cd concentration in the biosolids was 0.85 mg kg^{-1} DS, and was therefore more representative of contemporary concentrations in biosolids (Table S3). Cadmium concentrations in the biosolids-amended soil were correspondingly small and equivalent to 0.35 mg kg^{-1} ds, which was relatively similar to the control soil (0.32 mg kg^{-1} ds). Hence, Cd concentrations in liver of ewes grazed on the biosolids-amended pasture were small, at 0.041 mg kg^{-1} FW, and were similar to, albeit less than, the control group, which had a mean liver Cd concentration of 0.073 mg kg^{-1} FW.

5.4. Other international studies on Cd transfers to grazing livestock from biosolids

Reported international studies investigating the influence of ingesting biosolids, biosolids-amended soil and crops grown on biosolids-amended soil by livestock consistently show the absence of significant Cd accumulation in muscle tissue (Table S6, Supplementary Material). Johnson et al. (1981) fed six Hereford steers a diet containing 11.5% DM of a very high Cd biosolids (98 mg Cd kg^{-1} DS in biosolids, and 13.1 mg Cd kg^{-1} DM in the pelletised feed containing biosolids) for 106 days (Table S6, Supplementary Material). Nevertheless, there was no significant difference between the Cd concentration in the muscle tissue of treated steers and control steers, despite the very high Cd exposure in the diet, which was 13 times larger than that fed in the MAFF/DoE trials (1.04 mg Cd kg^{-1} DM, Cassington High treatment in MAFF Experiment 2, Table 2). Other investigations of Cd accumulation in muscle tissue of livestock fed crops grown on biosolids-amended soil (Telford et al., 1984; Bray et al., 1985) or diets containing high Cd biosolids (Sanson et al., 1984) similarly found: (1) only low Cd concentrations in muscle tissue (0.03- < 0.12 mg kg^{-1} DM) and (2) no statistically significant differences between animals fed high Cd diets from biosolids-amended soil compared to control diets (Table S6, Supplementary Material).

They also report elevated Cd in the kidney and liver tissues of livestock, but PTE concentrations were far greater than those present in contemporary biosolids (Table S3) (e.g. Bertrand et al., 1980; Baxter et al., 1982; Johnson et al., 1981) or soil concentrations were many times larger than the current maximum permissible limits in the UK and Europe and, under these extreme conditions, food limits for Cd in offal meat may be exceeded.

Several extreme cases of livestock ingestion exposure to Cd from biosolids are reported in the literature. For example, Fitzgerald (1980) grazed cows on pasture treated with liquid anaerobically digested biosolids containing 13.3–14.5 mg Cd l^{-1} (239–272 mg Cd kg^{-1} DS) over 4 years, which was exceptionally contaminated compared to the UK median concentration of 1.3 mg kg^{-1} DS (Table S3). The amended soil contained 22–36 mg Cd kg^{-1} ds, exceeding the maximum permissible soil limit value (3 mg Cd kg^{-1} ds) by 7–10 times, compared to 2–3 mg kg^{-1} ds in the control. Indeed, the Cd concentration measured in the control soil was also significantly elevated compared to typical background values (Table S3) and approached the maximum permissible concentration for Cd in biosolids-amended soil in the UK. This suggested a high background content (for example, the median Cd concentration in topsoil in England and Wales is 0.33 mg kg^{-1} ds (Rawlins et al., 2012)) from other contamination inputs or geogenic Cd. As may be expected under such extreme conditions, Cd concentrations were significantly increased in the liver of the cows from 1.8 to 7.1 mg kg^{-1} DM (0.5 to 2.0 mg kg^{-1} FW) and 14.4 to 40.7 mg kg^{-1} DM (3.2 to11.8 mg kg^{-1} FW) in kidney tissue, and were also increased in the liver and kidneys of calves, although overall they had lower concentrations of Cd compared to older animals (Table S6, Supplementary Material).

In another interesting example, Telford et al. (1982) found the kidneys of sheep fed silage corn with a high Cd concentration (3.88 mg kg^{-1} DM), grown on biosolids-amended soil, exceeded the current food limit for Cd by nearly four times at 3.74 mg kg^{-1} FW (17.8 mg kg^{-1} DM). However, the tissue Cd concentration of sheep fed control silage was also relatively high at 2.91 mg kg^{-1} DS (0.61 mg kg^{-1} FW), suggesting a high background Cd intake. The Cd concentration in the silage diet fertilised with biosolids was nearly 4 times larger than the maximum dietary Cd supplied in the MAFF/DoE trials (Cassington High, 1.04 mg Cd kg^{-1} DM, Table 2), and almost seven times greater than the concentration of Cd expected in ryegrass grown on soil with a pH value of 5.0 at the statutory Cd limit (0.44 mg kg^{-1} DM, Table S5, Supplementary Material), emphasising the extreme exposure to dietary Cd.

Later, Telford et al. (1984) also fed goats grass-legume silage grown on biosolids-amended soil. The biosolids were highly contaminated, containing 135.5 mg Cd kg^{-1} DS (more than 100 times the current median value for Cd in biosolids, Table S3), resulting in silage grass with a concentration of 3.81 mg Cd kg^{-1} DM compared to 0.14 mg Cd kg^{-1} DM in the control (Table S6, Supplementary Material). In this case, the biosolids fertilised silage contained approximately 9 times more Cd than may be expected in ryegrass grown on soil with a pH value of 5.0 at the maximum permissible limit for Cd (0.44 mg kg^{-1} DM, Table S5, Supplementary Material). The livers of adult goats fed high Cd silage accumulated 0.39 mg kg^{-1} DM of Cd (0.11 mg kg^{-1} FW) compared to 0.1 mg kg^{-1} DM (0.029 mg kg^{-1} FW) in liver tissues of control animals (Table S6, Supplementary Material). Cadmium in kidney was increased to 1.65 mg kg^{-1} DM (0.35 mg kg^{-1} FW) compared to 1.06 mg kg^{-1} DM (0.22 mg kg^{-1} FW) in the control. These diets are arguably unrealistic of contemporary biosolids quality and, whilst the Cd concentrations in liver and kidney tissue were below the limit for human consumption of 0.5 and 1 mg kg^{-1} FW, respectively (Table 1), the exposure conditions to Cd in biosolids represented a potentially high risk of Cd accumulation in kidney and liver tissues.

Bray et al. (1985) also investigated the effects of feeding biosolids fertilised silage to goats for three years and observed kidney Cd concentrations of 2.3 mg kg^{-1} FW (10.9 mg kg^{-1} DM), exceeding the 1 mg kg^{-1} FW limit in food, compared to 0.65 mg kg^{-1} FW (3.09 mg kg^{-1} DM) in animals receiving a control diet. Again, the silage treated with biosolids had a high Cd content of 0.71–1.84 mg Cd kg^{-1} DM, 2–4 times larger than expected on high Cd availability soil with pH 5.0 at the statutory limit for Cd (3 mg kg^{-1} ds) (Table S5, Supplementary Material).

These findings indicate the potential for significant herbage contamination with Cd, and accumulation in kidney and liver of livestock, when exceptionally contaminated biosolids are applied to grassland. The Cd concentrations in silage were signficantly greater than predicted for grass grown under worst case conditions at the soil limit value for Cd (3 mg kg^{-1} ds) at pH 5.0. In addition to the excessive Cd contents in the diets, it is speculated that bioavailability and transfer to animal tissues could be exacerbated for this dietary source by the acidic pH of ensiled forage, which may increase Cd solubility and absorption. Nevertheless, the results emphasise the importance of controlling Cd accumulation in soil where there are significant Cd concentrations in biosolids, to prevent human dietary exposure to Cd from the livestock ingestion route and consumption of liver and kidney. However, the dietary exposure to Cd in these reports was significantly more than what is theoretically possible at the UK and European maximum permissible limit for Cd (3 mg kg^{-1} ds) in biosolids-amended soil (Table S1). Indeed, other studies investigating comparable Cd concentrations in livestock diets to those observed in the MAFF/DoE studies, which were intended to reflect the UK and EU Cd soil limit and worst-case grazing conditions, resulted in similar tissue Cd concentrations (Kienholz et al., 1979; Sanson et al. 1984), and these were consistently below the maximum values permitted in liver and kidney used for human consumption (CEC, 2006). For example, Sanson et al. (1984) found that the Cd concentrations in the liver tissue of ewes fed a diet containing 7% biosolids (with a dietary Cd concentration of 0.5 mg kg^{-1} DM) (Table S6, Supplementary Material) was 0.23 mg kg^{-1} FW, equivalent to 50% below the food limit for Cd in liver of 0.5 mg kg^{-1} FW (CEC, 2006).

6. Relative availabilities and bioaccumulation factors for cadmium in the diet of grazing livestock from different sources

The apparent availability of dietary PTEs to livestock can be calculated using Eq. (1). Phillips et al. (2005) investigated the effects of Cd source on its absorption by sheep, by comparing the apparent availability of Cd in contaminated soil, high Cd grass and inorganic Cd additions to the diet. Greater absorption of Cd occurred when it was incorporated into plant material than when it was added to the diet as Cd $(NO_3)_2$, indicating that Cd may be more bioaccessible (i.e. available for absorption in the gastrointestinal tract) when combined in organic compared to simple inorganic mineral forms. However, Hill et al. (1998) calculated the apparent availabilities of elements in biosolids-treated soil to sheep for MAFF Experiment 1 (indoor feeding trial) and found that Cd had a negative value (i.e. output of Cd in faecal DM was greater than the intake), and that there was no clear pattern in Cd accumulation with the rate of biosolids inclusion in the diet. Hence, this method of assessing Cd accumulation may not be reliable, particularly from short-term digestion trials. The accurate assessment of PTE accumulation rates in animal tissues was only possible based on PTE retention results from slaughter samples (Hill et al., 1998).

Apparent availability of PTE in DM = (intake of PTE in diet DM − output of PTE in faecal DM)/(intake of PTE in diet DM) (1)

The relative availability of PTEs from different dietary sources can be compared using bioaccumulation factors, calculated according to Eq. (2) (Rodrigues et al., 2012).

BAF (bioaccumulation factor)$_{\text{feed-animal}}$ = concentration of PTE in an animal organ (mg kg^{-1} in animal organ FW)/concentration of PTE in feed (mg kg^{-1} in feed DM) (2)

de Vries et al. (2007) reported the bioaccumulation factors for Cd from green fodder to kidney, liver and muscle tissue of sheep were: 2.99, 0.554 and 0.0033 mg kg^{-1} FW/mg kg^{-1} DM, respectively, indicating greater absorption by the kidney compared to the liver, and minimal absorption by the muscle tissue, consistent with the observations of the MAFF/DoE trials (Section 5).

Prankel et al. (2004) conducted a *meta*-analysis of feeding trials investigating Cd accumulation in the liver and kidney of sheep, integrating the results of 21 controlled randomised experiments in which sheep were fed diets with elevated Cd, and the concentration of Cd was measured in liver and kidney tissues after slaughter. The different diets represented inorganic and organic sources of Cd, including biosolids and fodder grown on biosolids-amended soil. The *meta*-analysis showed that the Cd concentration in the feed and the duration of exposure were significant predictors of Cd accumulation in liver and kidney. Prankel et al. (2004) therefore suggested that, to prevent Cd from animal products adversely affecting human health, livers and kidneys of older animals should be excluded from the human food chain. Furthermore, consistent with the later work of Phillips et al. (2005), organic forms of Cd in the diet increased accumulation in the animal tissue compared to inorganic sources. However, in this case, the results may have been influenced by the limited number of studies investigating inorganically bound Cd and the higher doses of Cd involved in the tests with inorganic inputs.

Phillips and Tudoreanu (2011) integrated the model of Prankel et al. (2004) with models for accumulation of Cd in fodder crops derived from soil characteristics, to produce a combined function relating liver and kidney concentrations in sheep to soil Cd. The model assumed that sheep were fed on ryegrass and/or a supplement composed of maize and soybean, that feed Cd was organically bound and that water and soil Cd were inorganically bound. The model assumed inorganically bound Cd in soil accumulated at a slower rate compared to organically bound forms. Sensitivity analysis demonstrated that kidney accumulated Cd at a faster rate than liver and that the total Cd content in the soil in which fodder was grown exerted the largest overall influence on the tissue accumulation rates, which is verified by experimental data. However, other model predictions are less convincing and suggest there were limitations to some of the fundamental assumptions. For instance, soil pH value had relatively minor effects on accumulated Cd, but soil pH value is one of the main factors controlling Cd uptake by fodder crops (Smith, 1994). The model also suggested that Cd accumulation in liver and kidney was independent of the Cd concentration in ingested soil during grazing (at soil ingestion rates between 0 and 15%). However, direct feeding studies confirm that the Cd concentration of ingested soil is an important factor influencing the accumulation rate in liver and kidney, which is also modulated by the soil type.

Rodrigues et al. (2012) also developed a soil–plant-animal transfer model to back-calculate soil threshold PTE concentrations to avoid exceeding the limits for PTEs in animal fodder and EC, 2006 regulations controlling the concentrations of PTEs in muscle, liver and kidney tissue. Using modelling tools to estimate potential transfers of PTEs to animal tissues from biosolids and soil has considerable potential for evaluating appropriate environmental quality standards to protect food quality and the human foodchain. However, a critical weakness of this model was the assumption that common bioaccumulation factors apply to the transfer of PTEs to animal tissues. Experimental evidence clearly shows this is not the case and that many soil and biosolids properties, and husbandry and environmental conditions, can influence this complex behaviour.

Thus, current attempts to model PTE accumulation by animals from different exposure routes and dietary sources, including biosolids-amended soils and pasture crops, can determine some overall patterns in Cd concentrations in liver and kidney. However, current models fall short of being able to discern the effects of different sources of Cd and the Cd content in some of the dietary pools on the accumulation in animal tissues.

7. Implications of research on the animal ingestion of cadmium from biosolids-amended soil for the human food chain

Stark et al. (1998) concluded that the MAFF/DoE indoor feeding trials alone did not indicate a need to tighten soil limits for Cd. However, these trials used grass with low Cd concentrations as a basal diet (< 0.1–0.131 mg kg^{-1} DM). It is possible that combined intakes of Cd from biosolids-amended soil and herbage under extreme circumstances could further contribute to Cd concentrations in liver and kidneys, and particularly the kidneys of aged livestock, such as breeding ewes. Stark et al. (1998) recommended, therefore, that livestock diets should not exceed the NRC, 1980 limit of 0.5 mg Cd kg^{-1} DM in feeding stuffs to protect human health, although, this limit was subsequently raised to 10 mg Cd kg^{-1} DM (Table S2, Supplementary Material) in 2005 as the concern about the risk of Cd to animal health had apparently diminished (NRC, 2005). By contrast, the EC (2002) Directive on undesirable substances in animal feed set a limit of 1 mg kg^{-1} for Cd in complete feed to provide comprehensive protection against potentially harmfull effects of Cd based on animal, human health and environmental considerations. Nevertheless, risk assessment of the dietary impact (Stark et al., 1998) demonstrates that, assuming livestock consume 10% soil in the total diet containing 3.9 mg Cd kg^{-1} ds (3.9 times the current EU limit for feed and approximately 30% larger than the soil limit), and that Cd concentrations in grass are 10% of the statutory/advisory limit soil concentration (Stark et al., 1998), the dietary Cd concentration would be higher than the earlier maximum limit recommended by NRC (1980) at the time of 0.5 mg Cd kg^{-1} DM, at 0.7 mg Cd kg^{-1} DM. However, this value falls below the current EC (2002) limit for Cd in complete feeding stuffs.

The calculated dietary concentration of 0.7 mg Cd kg^{-1} DM is slightly in excess of the maximum concentration fed to sheep in the MAFF/DoE trials of 0.6 mg kg^{-1} DM. However, the data in Table 2 show that, at a 10% rate of DM ingestion of biosolids-treated soil with total Cd concentrations of 3 to almost 9 mg kg^{-1} ds (i.e. equivalent to the soil limit and up to 3 times the maximum permissible value in biosolids-amended soil – Table S1, Supplementary Material), Cd concentrations in liver and kidney of sheep were significantly below the relevant food standards for Cd in offal meat (Table 1).

The UK grazing trials (Wilkinson et al., 2001) under practical field husbandry conditions, which arguably provide a representative worst-case, confirmed that Cd accumulation in offal meats was below the upper limit permitted by current food standards (Section 5.3). Therefore, the available experimental evidence from controlled dietary investigations and field grazing trials with sheep indicates that the current UK maximum permissible limit value for Cd in biosolids-treated agricultural soil protects the human food chain from intakes of this element via the animal ingestion pathway. Furthermore, offal is not consumed in significant amounts in the UK diet and so there is little risk of liver and kidney being a significant source of Cd to consumers. As an added precaution, removing the livers and kidneys of mature livestock that is potentially at risk from the continuous ingestion of a diet at the maximum permissible feed limit value for Cd has been suggested as a measure to protect the human food chain from Cd transfer (Wilkinson et al., 2003; Prankel et al., 2004).

8. Conclusions

Controlled indoor feeding and outdoor sheep grazing trials to quantify the transfer of PTEs to livestock, funded by the UK Government (MAFF/DoE) in the 1990s, represent one of the most extensive programmes of research to be conducted internationally to investigate the potential impacts of PTEs in biosolids-treated soil on animal welfare and the human foodchain through the animal ingestion route. The results of this work have been integrated and interpreted with other and more recent investigations to assess the overall significance of Cd ingestion by grazing livestock from biosolids-amended soil in relation to current food quality standards in the EU and UK, and the following major conclusions were reached:

- The most significant potential route identified for transfer of Cd from biosolids-amended pasture to livestock is through ingestion of biosolids/biosolids-amended soil.
- The international research evidence base demonstrates there is little or no accumulation of Cd in the muscle tissue of animals grazing biosolids-amended soil containing PTE concentrations at or above UK maximum permissible limit values. Therefore, Cd in animal muscle meat is unlikely to represent a risk to the human foodchain in general or from biosolids-amended agricultural land in particular.
- By contrast, Cd accumulates in the liver and kidney of grazing animals depending on the dietary source and concentration. However, Cd in sheep offal was below the maximum concentrations permitted by current food quality standards in the EU and UK (CEC, 2006) at the UK maximum permissible limit value for Cd in biosolids-amended soil. Risk assessments and field grazing trials, representing reasonable worst-case grazing and exposure conditions, also show Cd accumulation by livestock, from combined crop and soil sources, was below food safety standards for Cd in offal at the Cd soil limit.
- The rate of Cd accumulation by liver reached a maximum value after 4 months in sheep ingesting diets containing biosolids-amended soil, however kidney tissues accumulated Cd continuously. Therefore, removing the kidney of older livestock exposed to high Cd soils from the human food chain has been suggested as a preventative measure to reduce human dietary intake of Cd from this source.
- The limited accumulation of Cd below food safety standards in animal offal tissues at the UK limit value in biosolids-amended agricultural soil under worst-case grazing conditions, combined with the decline in Cd concentrations in contemporary biosolids sources to less than the soil limit value, and changing agricultural biosolids application practices, which limit biosolids ingestion by livestock, effectively remove the risk to the human food chain of Cd from biosolids-treated pasture by livestock ingestion in the practical field grazing situation.

Declaration of Competing Interest

None.

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Appendix A. Supplementary material

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