



Review article

A critical review of the bioavailability and impacts of heavy metals in municipal solid waste composts compared to sewage sludge

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ABSTRACT

The content, behaviour and significance of heavy metals in composted waste materials is important from two potentially conflicting aspects of environmental legislation in terms of: (a) defining end-of-waste criteria and increasing recycling of composted residuals on land and (b) protecting soil quality by preventing contamination. This review examines the effects of heavy metals in compost and amended soil as a basis for achieving a practical and sustainable balance between these different policy objectives, with particular emphasis on agricultural application.

All types of municipal solid waste (MSW) compost contain more heavy metals than the background concentrations present in soil and will increase their contents in amended soil. Total concentrations of heavy metals in source-segregated and greenwaste compost are typically below UK PAS100 limits and mechanical segregated material can also comply with the metal limits in UK PAS100, although this is likely to be more challenging. Zinc and Pb are numerically the elements present in the largest amounts in MSW-compost. Lead is the most limiting element to use of mechanically-segregated compost in domestic gardens, but concentrations are typically below risk-based thresholds that protect human health.

Composted residuals derived from MSW and greenwaste have a high affinity for binding heavy metals. There is general consensus in the scientific literature that aerobic composting processes increase the complexation of heavy metals in organic waste residuals, and that metals are strongly bound to the compost matrix and organic matter, limiting their solubility and potential bioavailability in soil. Lead is the most strongly bound element and Ni the weakest, with Zn, Cu and Cd showing intermediate sorption characteristics. The strong metal sorption properties of compost produced from MSW or sewage sludge have important benefits for the remediation of metal contaminated industrial and urban soils.

Compost and sewage sludge additions to agricultural and other soils, with background concentrations of heavy metals, raise the soil content and the availability of heavy metals for transfer into crop plants. The availability in soil depends on the nature of the chemical association between a metal with the organic residual and soil matrix, the pH value of the soil, the concentration of the element in the compost and the soil, and the ability of the plant to regulate the uptake of a particular element. There is no evidence of increased metal release into available forms as organic matter degrades in soil once compost applications have ceased.

However, there is good experimental evidence demonstrating the reduced bioavailability and crop uptake of metals from composted biosolids compared to other types of sewage sludge. It may therefore be inferred that composting processes overall are likely to contribute to lowering the availability of metals in amended soil compared to other waste biostabilisation techniques.

The total metal concentration in compost is important in controlling crop uptake of labile elements, like Zn and Cu, which increases with increasing total content of these elements in compost. Therefore, low metal materials, which include source-segregated and greenwaste composts, are likely to have inherently lower metal availabilities overall, at equivalent metal loading rates to soil, compared to composted residuals with larger metal contents. This is explained because the compost matrix modulates metal availability and materials low in metals have stronger sorption capacity compared to high metal composts.

Zinc is the element in sewage sludge-treated agricultural soil identified as the main concern in relation to potential impacts on soil microbial activity and is also the most significant metal in compost with regard to soil fertility and microbial processes. However, with the exception of one study, there is no other tangible evidence demonstrating negative impacts of heavy metals applied to soil in compost on soil microbial processes and only positive effects of compost application on the microbial status and fertility of soil are reported. The negative impacts on soil microorganisms apparent in one long-term field experiment could be explained by the

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exceptionally high concentrations of Cd and other elements in the applied compost, and of Cd in the compost-amended soil, which are unrepresentative of current practice and compost quality.

The metal contents of source-segregated MSW or greenwaste compost are smaller compared to mechanically-sorted MSW-compost and sewage sludge, and low metal materials also have the smallest potential metal availabilities. Composting processes also inherently reduce metal availability compared to other organic waste stabilisation methods. Therefore, risks to the environment, human health, crop quality and yield, and soil fertility, from heavy metals in source-segregated MSW or greenwaste-compost are minimal. Furthermore, composts produced from mechanically-segregated MSW generally contain fewer metals than sewage sludge used as an agricultural soil improver under controlled conditions. Consequently, the metal content of mechanically-segregated MSW-compost does not represent a barrier to end-use of the product. The application of appropriate preprocessing and refinement technologies is recommended to minimise the contamination of mechanically-segregated MSW-compost as far as practicable.

In conclusion, the scientific evidence indicates that conservative, but pragmatic limits on heavy metals in compost may be set to encourage recycling of composted residuals and contaminant reduction measures, which at the same time, also protect the soil and environment from potentially negative impacts caused by long-term accumulation of heavy metals in soil.

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1. Introduction

Heavy metals are naturally present in the environment, soil and food and are widely used in manufacturing processes and in the built environment and, consequently, they transfer to and are present in composted organic residuals (Lineres, 1992). There are many sources of heavy metals in compost and particularly products derived from household municipal solid waste (MSW). For example, these include household dust, batteries, disposable household materials (eg bottle tops), they are present in plastics, paints and inks, bodycare products and medicines and household pesticides (NHHWF, 2000; Bardos, 2004). Consequently, composts derived from source-segregated waste streams or greenwaste are generally reported to contain smaller amounts of heavy metals compared to mechanically-sorted products (Epstein et al., 1992; Sharma et al., 1997; Amlinger et al., 2004). In relation to the application of composted residuals to soil, the main elements generally of concern include: Zn, Cu, Ni, Cd, Pb, Cr and Hg (CA, 2001) because they are potentially present in compost in amounts that may be greater than the background values in the receiving soil. The concentrations of conservative elements like heavy metals increase during the composting process (García et al., 1990; Ciavatta et al., 1993) due to the microbial degradation of part of the organic matter and loss of volatile solids (Smith and Hall, 1991). The long-term accumulation of heavy metals in the soil environment is a concern because they potentially have important consequences for the quality of the human food chain, toxicity to plants and soil microbial processes and once applied they have very long residence times in soil. The principal environmental end-points for heavy metals applied to soil in all organic residual materials (Smith, 1996), including compost (Chaney and Ryan, 1993; Ryan and Chaney, 1993; Déportes et al., 1995), are:

- Reduced plant growth (phytotoxicity) due to phytoaccumulation in plant tissues above tolerable threshold values (Zn, Cu, Ni; Cr may also be listed here, but there is no evidence of crop damage due to Cr in sewage sludge or compost-amended soil);

- Human foodchain *via* crop uptake (Cd);
- Human foodchain *via* direct child ingestion of compost, eg applied to home gardens (Cd, Pb, Hg);
- Human foodchain *via* offal meat from animals ingesting compost-treated soil (Cd and Pb);
- Animal health (Cu, Pb);
- Soil microbial processes (Zn).

A comprehensive receptor and environmental pathway analysis for contaminants in sewage sludge was developed by US EPA (1992) as a basis to regulate the use of sewage sludge on land in the US (US EPA, 1993). As MSW-compost has similar properties to sewage sludge and is treated and utilised by application to land in a similar manner to sludge, the pathway analysis may also provide a technical basis for evaluating the significance of contaminants, including heavy metals, in composted residuals (Chaney and Ryan, 1993; Logan et al., 1999). However, the limit values derived from a quantitative environmental pathway analysis and risk assessment of heavy metals are much larger than the concentrations that are considered acceptable in most European compost standards (Hogg et al., 2002).

The majority of research on the fate and behaviour of heavy metals in MSW-compost amended soil is focussed on mechanically-sorted material (eg Smith, 1992; Gigliotti et al., 1996; Pinamonti et al., 1997), or compost prepared by, for example, manual sorting mixed refuse (eg Pichtel and Anderson, 1997; Baldwin and Shelton, 1999; Jordão et al., 2006). This is because compost derived from these feedstocks is recognised as a potentially important source of heavy metals entering soil and the environment (Epstein et al., 1992). Relatively few studies have specifically examined source-segregated products because these are not considered a priority with regard to metal contamination; those referring to source-segregated compost identified in this review include, for example: Zinati et al. (2001), Liu et al. (2003) and Zheljzakov and Warman (2004a,b). A number of field experiments report long-term effects of MSW-compost additions to soil (eg

Gigliotti et al., 1996; Baldwin and Shelton, 1999; García-Gil et al., 2000; Crecchio et al., 2004). An extensive database of information is available considering the fate and impacts of heavy metals in sewage sludge-amended soil (eg see Smith, 1996).

Quantifying the significance of heavy metals in composted residuals derived from municipal wastes has increased in importance recently, especially in the European context, where, on the one hand environmental policies aim to increase recycling biodegradable wastes and composts to land, as an alternative to landfill disposal, whilst at the same time prevent inputs of contaminants entering the soil. This discussion is therefore relevant to: (1) the expansion of composting as a means of diverting biodegradable waste from landfill disposal (Nikitas et al., 2008), (2) increasing mechanical biological treatment of MSW and production of 'compost-like' outputs (Archer et al., 2005), (3) increased recycling of these materials to land (Nikitas et al., 2008), (4) compost quality standards (BSI, 2005), (5) revision of the European Waste Framework Directive and development of associated 'end-of-waste' criteria for compost and related product protocols (ComEC, 2005a,b; WRAP/EA, 2007), and (6) development of policies to protect soil quality and the environment (ComEC, 2006a,b).

This article gives an overview of the environmental significance and bioavailability of heavy metals in MSW-derived compost and amended soil and in sewage sludge for comparison with a well established soil amendment material, with particular emphasis on agricultural application.

2. Total heavy metal contents in compost derived from different waste streams

In the majority of cases, heavy metal concentrations in compost produced from separately collected organic household waste and greenwaste (eg from parks and gardens, and household waste bring sites) are smaller than in the composted residual organic fraction from mechanically-sorted MSW. For example, following an extensive review of heavy metal concentrations in composted residuals, Amlinger et al. (2004) concluded that the metal content in source-segregated compost was typically reduced by a factor of 2–10 compared to mechanically treated material. This is generally supported by the metal concentration data presented in Table 1, compiled from recent published and unpublished data for source and mechanically-segregated composted MSW, greenwaste and home produced compost. What is also evident from the data in Table 1 is that modern mechanical segregation technologies and process improvements have significantly reduced the metal content of compost produced from mixed MSW compared with 30 years ago.

From a review of literature on the chemical characteristics of composted, mechanically-segregated MSW, Bardos (2004) noted that heavy metals are mainly associated with finer particle size classes within the material. Sharma et al. (1997) noted that the highest metal concentrations were present in very fine clusters, of <1 mm. Finely divided components, such as household dust, with large metal contents, tend to adhere to the wet putrescible fraction and small metallic or metal containing objects become embedded in the organic matter and are difficult to remove. Preprocessing strategies are the most effective at removing contaminants and reducing metal concentrations in compost produced by mechanical segregation of MSW (Richard and Woodbury, 1992). Product refinement techniques also have potential to significantly reduce the concentrations of the main metal contaminants in compost, such as Zn and Pb (Zennaro et al., 2005). Furthermore, mechanical screening to remove the fine fraction could increase the overall organic matter content and agricultural value of the final product.

Source segregation of compostable material is widely accepted as the most effective approach to minimise metal concentrations in MSW-compost (Richard and Woodbury, 1992; Amlinger et al., 2004; Bardos, 2004). Veeken and Hamelers (2002) compared the heavy

metal contents of the various constituent fractions (food products, plant material, soil organic matter and soil minerals) in source-segregated biodegradable waste with background values in the constituent materials and showed that biowaste collected separately from households was not contaminated from other sources. Therefore, compost produced from these materials contained only natural background concentrations of heavy metals, which may be very small depending on the dietary content (Liu et al., 2003). Nevertheless, they may exceed the statutory limit values for metals in compost permitted in some European countries, such as The Netherlands, for example. Veeken and Hamelers (2002) therefore argued that the rational basis for compost standards should avoid conflicts between the two government policies of: (1) protecting soil systems, and (2) promoting the recycling and reuse of solid organic waste streams. Amlinger et al. (2004) also referred to the potential problem of complying with existing European limits on metals in composts due to the natural contents in the main source materials in residues from food and prepared foods and given the mineralization of organic matter during the course of aerobic degradation. Similar investigations in Germany by Fricke and Vogtmann (1994) showed that soil is an important source of heavy metals in both greenwaste and source-segregated waste and can be responsible for elevating the metal content of compost above quality limits. For a comprehensive and international overview of quality standards for compost refer to Hogg et al. (2002).

Bardos (2004) pointed out that there was considerable overlap in the metals contents of composts at the upper and lower ranges of concentrations measured in source and mechanically-segregated compost, respectively. This is confirmed by the international data presented in Table 1 for composted residuals produced from these feedstocks, as well as from greenwaste and home compost. Table 1 also shows that Zn and Pb are numerically the elements present in the largest amounts in source and mechanically-segregated MSW and greenwaste composts (Zennaro et al., 2005). Total concentrations of heavy metals in mechanically-segregated compost are usually smaller than in sewage sludge, by approximately 30–50%, except for Pb. The Pb content of compost from mechanical-sorting MSW may be similar to or up to approximately 80% larger than the total Pb concentration measured in sewage sludge.

Composts produced from both source and mechanically-segregated feedstocks can comply with the limits on total metals in the voluntary PAS100 standard adopted in the UK (BSI, 2005), but this is potentially more challenging for mechanical segregation systems. This is also put into perspective by comparing the metal contents of composts from these feedstocks with material generated by home composting for instance. Home composting arguably is an approach that is likely to lead to the smallest degree of contamination with heavy metals and, on average, metal concentrations are well below the PAS100 limits (Table 1). However, maximum metal concentrations measured in home compost may also exceed the PAS100 limit values (Nakou, 2001). This may be explained by the introduction of metals from sources such as printing inks as homeowners are encouraged to add non-recyclable paper and packaging materials, for example, to home compost bins (Smith et al., 2004). In contrast to the recent work of Veeken and Hamelers (2002), Hoffmann et al. (1991) showed that garden waste was the most significant source of heavy metals in compostable waste due to potential contamination by atmospheric deposition. Nevertheless, based on the data reviewed here and presented in Table 1, total metal concentrations in greenwaste compost are consistently below the PAS100 limit values. In all the cases listed in Table 1, total metal concentrations in composts produced from different methods of household waste segregation and greenwaste are less than risk-based limits for agricultural application (Chaney and Ryan, 1993). Following the risk-based approach to evaluating the significance of heavy metals in compost, Pb is identified as the most limiting element to compost use in domestic gardens (Table 1).

Table 1

Heavy metal concentrations (mg kg⁻¹ DS) in mechanically and source segregated MSW-compost, greenwaste compost and home produced compost compared to sewage sludge used for agricultural application, background soil, and UK PAS100 and USEPA limit values (see below for abbreviations)

Type/source ^a	Zn	Cu	Ni	Cd	Pb	Cr	Hg	n	Country	Reference/Source
2006 mean data	194	51	16	0.66	114	23	0.19	285	UK	CA (2006)
2006 90%ile data	251	72	23	0.95	160	33	0.37	285	UK	CA (2006)
2004 data	182	46	17	0.60	96	20	0.20		UK	Defra (2006)
SS (food waste)	38	36	5	0.70	24	<1	nd	1	US	He et al. (1995)
SS	269	57	23	0.98	90	39	0.45	90–140	Italy	cit Sharma et al. (1997)
SS industrial	271	114	30	1.76	62	57	0.39	9–31	Italy	cit Sharma et al. (1997)
SS mean	671	149	649	4.00	200	177	nd	4	SE Spain	Pascual et al. (1997)
SS mean	159	51	17	0.92	131	22	nd	Several	UK	Grimes et al. (1999)
SS	176	82	nd	nd	nd	nd	nd		USA	Zinati et al. (2001)
SS	19	4.2	nd	nd	nd	nd	nd		Japan	Liu et al. (2003)
SS mean	181	50	21	0.42	40	27	0.19	552–582	Austria	cit Amlinger et al. (2004)
SS median	229	45	12	0.82	69	22	0.15	195	Belgium	cit Amlinger et al. (2004)
SS mean	326	110	26	1.07	106	43	0.63	12–27	France	cit Amlinger et al. (2004)
SS mean	219	89	26	1.38	84	33	nd	127	Italy	cit Amlinger et al. (2004)
SS mean	219	39	16	0.41	49	32	0.12	175	Luxembourg	cit Amlinger et al. (2004)
SS median	181	52	13	0.32	47	27	0.18	179–196	Germany	cit Amlinger et al. (2004)
SS mean	233	43	19	0.78	78	34	0.33	490	Germany	cit Amlinger et al. (2004)
SS median	180	43	nd	0.46	43	nd	0.13	60	Germany	cit Amlinger et al. (2004)
SS (food waste+greenwaste, paper waste, sewage sludge, wood wastes, fish process waste)	768	312	21	3.10	224	46	0.60	1	Canada	Zheljzkov and Warman (2004a,b)
SS min	100	32	11	0.47	56	14	0.12	10	UK	Dimambro et al. (2007)
SS max	266	92	20	0.87	162	33	0.34	10	UK	Dimambro et al. (2007)
SS mean	182	49	16	0.63	96	22	0.22	10	UK	Dimambro et al. (2007)
Urban solid waste min	300	150	40	2.00	250	80	nd		Italy	cit Sharma et al. (1997)
Urban solid waste max	600	300	50	4.00	350	100	nd		Italy	cit Sharma et al. (1997)
Manually sorted MSW	655	236	28	nd	210	21	nd		USA	Pichtel and Anderson (1997)
Green, fruit and veg waste	178	55	21	1.00	77	37	0.44	27–78	Italy	cit Sharma et al. (1997)
Waste biomass	451	97	25	<5	134	50	nd	4	Italy	cit Sharma et al. (1997)
GW CA mean	116	40	24	3.90	60	28	<0.5	21–22	UK	EA (2000)
GW CA min	47	20	4	0.70	19	10	<0.5	21–22	UK	EA (2000)
GW CA max	185	69	49	9.10	102	49	<0.5	21–22	UK	EA (2000)
GW CA+GW HC mean	176	48	27	3.72	163	26	<0.5	9	UK	EA (2000)
GW CA+GW HC min	147	30	16	1.50	103	5	<0.5	9	UK	EA (2000)
GW CA+GW HC max	209	72	36	6.10	233	30	<0.5	9	UK	EA (2000)
GW HC+KW mean	130	38	30	3.92	56	27	<0.5	21	UK	EA (2000)
GW HC+KW min	43	19	11	0.60	21	11	<0.5	21	UK	EA (2000)
GW HC+KW max	220	58	64	8.40	109	43	<0.5	21	UK	EA (2000)
GW mean	470	31	13	<LoD	80	nd	0.15		Belgium	Soumaré et al. (2003)
GW mean	168	40	21	0.46	38	24	0.14	65	Austria	cit Amlinger et al. (2004)
GW median	169	32	9	0.70	44	17	0.12	229	Belgium	cit Amlinger et al. (2004)
GW mean	187	51	22	1.40	87	46	0.52	22–123	France	cit Amlinger et al. (2004)
GW mean	166	63	23	0.95	72	33	nd	70	Italy	cit Amlinger et al. (2004)
GW mean	164	32	13	0.34	45	24	0.13	57	Luxembourg	cit Amlinger et al. (2004)
GW median	141	37	13	0.28	31	29	0.12	28–86	Germany	cit Amlinger et al. (2004)
GW mean	168	33	18	0.70	61	27	0.27	490	Germany	cit Amlinger et al. (2004)
GW median	205	42	nd	0.71	56	nd	0.16	12	Germany	cit Amlinger et al. (2004)
GW+KW	347	74	10	1.90	356	23	nd	1	Belgium	Rutten et al. (2006)
MS	1650	630	110	6.00	900	220	5.00		Netherlands	De Haan (1981)
MS	1000	266	nd	3.70	230	nd	2.00		Germany	De Haan (1981)
MS	1000	250	190	7.00	600	270	4.00		France	De Haan (1981)
MS	2330	780	90	12.0	1570	nd	nd		Switzerland	De Haan (1981)
MS mean	583	213	36	3.50	236	72	2.40	5	US	He et al. (1992)
MS	757	312	56	4.88	607	54	nd	1	UK	Smith (1992)
MS mean (6 years)	647	240	52	5.00	750	81	nd		Italy	Perucci (1992), Giusquiani et al. (1994), Gigliotti et al. (1996)
MS min	320	90	15	1.50	79	38	nd	8	US	He et al. (1995)
MS max	1147	424	76	4.70	362	186	nd	8	US	He et al. (1995)
MS mean	655	281	34	3.30	234	76	nd	8	US	He et al. (1995)
MS	1321	451	138	<5.00	687	310	nd	9	Italy	cit Sharma et al. (1997)
MS	650	237	328	2.00	235	365	nd		Spain	Pascual et al. (1999)
MS mean	382	158	22	1.95	210	113	nd		Italy	Crecchio et al. (2001, 2004)
MS	470	110	13	0.69	38	33	0.67		France	Morvan (2004)
MS min	130	25	10	0.22	73	3.7	0.001	16	UK	cit Amlinger et al. (2004)
MS max	560	306	94	1.87	683	51	0.93	16	UK	cit Amlinger et al. (2004)
MS median	286	91	31	0.41	167	16	0.15	16	UK	cit Amlinger et al. (2004)
MS min	235	161	18	0.70	64	24	0.10	9	Austria	cit Amlinger et al. (2004)
MS max	990	500	253	6.10	963	344	4.10	9	Austria	cit Amlinger et al. (2004)
MS median	769	247	149	2.70	224	209	1.30	9	Austria	cit Amlinger et al. (2004)
MS	396	362	nd	1.49	385	nd	nd	3	Spain	Pérez-de-Mora et al. (2006)
MS	411	91	23	1.90	101	nd	nd		Brazil	Jordão et al. (2006)
MS	212	47	21	0.65	37	nd	nd		Brazil	Jordão et al. (2006)
MS (AD)	399	159	21	0.72	470	26	0.27		UK	Rigby (2006)
MS min	390	160	19	<1.0 ^b	120	13	0.33	167	Australia	Lawson (2006)
MS max	1400	690	65	6.30	410	70	6.60	167	Australia	Lawson (2006)

(continued on next page)

Table 1 (continued)

Type/source ^a	Zn	Cu	Ni	Cd	Pb	Cr	Hg	n	Country	Reference/Source
MS mean	680	309	30	1.15	257	20	1.00	167	Australia	Lawson (2006)
MS 90%ile	834	400	38	1.30	330	24	2.20	167	Australia	Lawson (2006)
MS	583	357	56	4.3	269	56	nd	3	Portugal	Alvarenga et al. (2007)
MS	563	224	197	1.16	414	73	0.37	1	UK	Dimambro et al. (2007)
MS	148	78	14	0.78	108	11	0.18	1	UK	Dimambro et al. (2007)
MS min	100	24	3	<0.1	17	8	<0.02	186	Australia	McCall (2007)
MS max	1500	320	53	14.0	560	340	3.20	186	Australia	McCall (2007)
MS mean	365	108	22	1.18	197	27	0.32	186	Australia	McCall (2007)
Home compost mean	240	52	18	1.77	124	28	nd	64	UK	Nakou (2001)
Home compost max	693	177	107	16.0	745	198	nd	64	UK	Nakou (2001)
UK sewage sludge 1996/97										
Works size weighted mean ^c	792	568	57	3.30	221	157	2.40			Gendebien et al. (1999)
Median UK soil content	97	23	25	0.80	74	41	nd			McGrath and Loveland (1992)
Median worldwide soil content	90	30	50	0.35	35	70	nd			Bowen (1979)
UK PAS 100 limits	400	200	50	1.50	200	100	1.00			BSI (2005)
US EPA 503 risk based limit	2800	1500	420	390	300 ^d	3000	17			US EPA (1992)
US EPA 503 most limiting pathway	Plant tox	Plant tox	Plant tox	Child intake	Child intake	Plant tox	Child intake			US EPA (1992), Logan et al. (1999)

nd - not determined; <LoD - less than analytical limit of detection.

^a Abbreviations: SS - Source-Segregated; MS - Mechanically-Segregated; GW - Greenwaste; CA - Civic Amenity; HC - Household Collected; KW - Kitchen Waste; AD - Anaerobically Digested.

^b 90% of samples less than detection limit of 1.0 mg kg⁻¹ DS.

^c Used in agriculture.

^d Next most limiting pathway for Pb is ingestion by grazing livestock and the risk based limit is 1200 mg Pb kg⁻¹ DS (US EPA, 1992). Note this is equivalent to the limit value for Pb in sewage sludge for application to the surface of grassland in the UK (DoE, 1996).

Nevertheless, the mean Pb concentrations in compost from modern mechanical segregation and composting facilities may be significantly below the risk-based limit value of 300 mg kg⁻¹ dry solids (DS) for this element (Chaney and Ryan, 1993). Thus, mechanical treatment of household waste can also achieve a product suitable for land application that is not constrained by heavy metals (Morvan, 2004).

3. Metal extractability and sorption in compost

One of the main approaches to studying the behaviour of heavy metals in composts has been to use chemical and sequential extraction techniques to remove metals bound in different operationally defined phases (He et al., 1992). Many such studies report reduced chemical extractabilities and water solubility of metals by composting organic residuals and that metal availability decreases with the period of composting and maturation time (Leita and De Nobili, 1991; García et al., 1990, 1995; Chiang et al., 2001; Eneji et al., 2003). Thus, the leaching potential of heavy metals from mature compost is considered to be very low (Leita and De Nobili, 1991; Woodbury, 1992; Tisdell and Breslin, 1995). García et al. (1995) considered that this behaviour was explained by the formation of stable metal-humus complexes during the composting process. Walter et al. (2006) used a sequential extraction technique to identify metal fractions (exchangeable and carbonate, reduced or Fe+Mn oxide bound, oxidisable or organic matter bound and residual) in sewage sludges treated by different methods. Composted sludge contained smaller available fractions of Ni and Zn, but the availability of Cu was larger compared to mechanically dewatered or thermally dried, anaerobically digested material. Cadmium availability was also slightly greater in the composted compared to digested sludge. Eneji et al. (2003) also found the extractability of Zn and Cu was reduced by aerobic composting livestock manure, but Zn availability increased during anaerobic digestion of livestock waste. In general, however, there would appear to be an overall shift to more stable inorganically precipitated forms of metals during anaerobic digestion of sewage sludge compared to raw sludge (Lake, 1987).

Nickel is recognised as a potentially mobile and water-soluble element (Rudd, 1987) and the labile pool in compost is relatively easily solubilized by mild extractants due to the weaker binding to the matrix compared with other key metals (Hernando et al., 1989; García et al., 1995; Tisdell and Breslin, 1995), which is also consistent with its

behaviour in sewage sludge for example (Davis and Carlton-Smith, 1981). However, the majority of Ni in MSW-compost (up to 50%) resides in the residual and low availability fractions (eg carbonate, phosphate and sulphide minerals), probably as metallic or alloy forms, silicate minerals, glass and inert plastics (He et al., 1995; Liu et al., 2007). Chromium is also present predominantly in residual stable forms (He et al., 1995; Liu et al., 2007) and thus has extremely low bioavailability (see later).

Using a five stage sequential procedure, Liu et al. (2007) showed that organic matter decomposition and declining pH value were primarily responsible for the transformation of metals between different pools during composting of sewage sludge. Thus, Pb, Zn and Cd moved from the residual to other extractable forms, whereas the distribution of Cu between the different extractable fractions remained relatively unchanged.

Amir et al. (2005) also used a sequential extraction method to determine the changes in the distributions of heavy metals between different operational fractions during sewage sludge composting. In this case, however, a large proportion of the heavy metals were associated with the residual fraction (70–80%) and more resistant fractions to extraction (NaOH=organically bound, EDTA=organically complexed or carbonates and HNO₃=sulphides) (12–29%). Less than 2% of metals were detected in the potentially available fractions (KNO₃+H₂O). The available fractions of all elements decreased during composting with the exception of Ni, consistent with other studies. Zinc and Cu were mainly associated with organic and carbonate fractions, whereas Pb was preferentially bound to sulphide forms. The observed affinities of heavy metals to the various fractions after treatment by composting were ranked as follows: KNO₃+H₂O: Ni>>Cu>Zn; NaOH: Cu>Zn>Pb>Ni; EDTA: Ni>Cu>Zn>Pb; HNO₃: Pb>Ni>Cu>Zn. The mobility of heavy metals also depended on other physico-chemical properties of the compost besides the total metal content and affinity for various chemical forms. For example, the size of the mobile Zn fraction declined and more resistant forms increased with decreasing organic matter content. Therefore, the stabilisation of raw organic matter during composting transformed Zn from exchangeable to more stable organic forms. For Cu, the mobile fraction declined with increasing content of humic substances and decrease in pH value (7.5–6.7) during composting, consistent with the known affinity of Cu to organic matter. In contrast, mobile Ni increased with declining pH and organic matter content. Amir et al. (2005) considered this behaviour could be attributed to the high proportion of Ni in

the raw material that is present in an organic form that is readily biodegradable therefore releasing Ni in soluble forms during the composting process.

The apparent inconsistencies in the patterns of metal fractionation observed between the studies of Amir et al. (2005) and Liu et al. (2007) may be partly explained because different sludge feedstocks were used. Thus, Liu et al. (2007) composted biological sludge from an activated sludge process, whereas Amir et al. (2005) used a sludge taken from an anaerobic lagoon. It has already been established that the extractabilities and solubilities of metals decrease with time of composting and during maturation. A further explanation for the apparent differences in behaviour may therefore also be attributed to the relatively short processing period of 12 days monitored by Liu et al. (2007), in contrast to 180 days by Amir et al. (2005). Consequently, Liu et al. (2007) were unable to follow the complete transformation dynamics of metals to less mobile forms that occur during composting of biodegradable waste (Leita and De Nobili, 1991).

Grimes et al. (1999) showed that metals in compost produced from source-segregated household and garden waste were tightly bound to the matrix and were not easily removed unless harsh extraction conditions were applied using, for example, concentrated nitric acid. Batch sorption studies using metal salts assessed the binding potential of additional Cd, Cu, Pb and Zn by the compost. The relative binding of the added metals was in the order: Pb > Cd = Cu > Zn. The sorption of metals took place, at least in part, by exchange of Ca bound to the compost and there was evidence that the sorption occurred in both humic and non-humic sites in the compost. In a similar study of heavy metal leachability and retention by compost produced from wood, straw and vegetable wastes, Song and Greenway (2004) also reported the high sorption affinity of compost for heavy metals. This was similar to that reported for humic acid and was in the order: Pb > Cr > Cu > Ni ≥ Cd ≥ Zn, suggesting that humic materials were likely to be the main sites of metal sorption.

A significant proportion of the metals contained in MSW-compost are organically bound. For example, He et al. (1995) showed that >50–80% of Zn was extracted in a relatively labile organic fraction from MSW-composts. Copper, on the other hand, was bound to more humified organic matter (approximately 50%) and was therefore considered to be relatively immobile.

Interestingly, Soumaré et al. (2003) measured larger amounts of water soluble and exchangeable Zn in a peat-based substrate compared to composted greenwaste, despite a smaller total Zn content in the peat material (104 mg kg⁻¹ DS) relative to the compost (470 mg kg⁻¹ DS). This behaviour was probably explained by the lower pH value of the peat substrate (pH 5.5) compared to the compost (pH 7.2), thus emphasising the importance of chemical properties of organic residuals other than their total metal contents for the potential bioavailability of heavy metals. Similar amounts of Cu (10%) were organically bound in both substrate types, but only a small proportion of the total Zn content (<3%) was found in the organic fraction, illustrating the relatively higher affinity of Cu for organic matter. The majority of metals (<70%), however, were bound in stable residual forms indicating they were unavailable for plant uptake.

The strong sorption of Pb to compost emerges as a consistent characteristic from the scientific literature on metal behaviour in composted organic materials. For example, Zheng et al. (2004) measured the effects of composting sewage sludge on the chemical speciation, distribution and bioavailability of Pb during the treatment process. A sequential extraction method was adopted to determine the operationally defined speciation of Pb into exchangeable, carbonate, Fe–Mn oxide bound, organic, sulphide and residual fractions. The results showed a shift in the speciation of Pb from the more soluble fractions (exchangeable, carbonate and Fe–Mn oxides bound) into less soluble fractions (organic, sulphide and residual) and Zheng et al. (2004) concluded that the bioavailability of Pb would therefore be potentially reduced by composting treatment.

4. Metal extractability and sorption in MSW-compost amended soil

Compost and sewage sludge contain larger concentrations of heavy metals than the background values found in soil (Table 1) and, consequently, their regular application to land gradually raises the total metal content of soil in the long-term. A shift in the distribution of metals occurs in sewage sludge-amended soil generally increasing the amounts of the potentially labile and extractable fractions (for example, see Carlton-Smith, 1987). Compost inputs to contaminated soils containing large fractions of labile elements reduce the overall bioavailabilities of metals due to sorption processes and thus provide an effective soil remediation technique (Brown et al., 2003, 2004). The extractability of heavy metals (eg Zn and Cu) may also apparently decline following compost application to soil, previously containing only background concentrations of metals, compared to the untreated control when examined relative to the total metal contents in amended and unamended control soils (Zheljzkov and Warman, 2004b). However, absolute concentrations of extractable metals increase with compost amendment compared to unamended control soil containing background metal values, including for source-segregated material (Pichtel and Anderson, 1997; Zheljzkov and Warman, 2004a). This is because, although metals are present in compost in relatively resistant forms, these are more labile in the environment than the very stable, residual pool of metals of geological origin present in uncontaminated soils. Therefore, a new equilibrium is established following compost application causing a shift to a moderately higher proportion of labile and extractable fractions compared to untreated soil, in a similar manner to sewage sludge. However, as will be discussed later, at equivalent loading rates of metals to soil, the availability of metals applied to soil in compost are likely to be smaller than for sewage sludge and are also expected to decline with decreasing metal content in compost.

Zinc is frequently the element where the largest increase in labile forms is reported when composted residuals or sewage sludge are applied to soil, particularly under acidic soil conditions (Planquart et al., 1999). In contrast, Vaca-Paulín et al. (2006) found the application of sewage sludge and composted sludge at a rate equivalent to 200 t ha⁻¹ on a DS basis to clay loam soil (pH 6.0) increased the sorption of Cu and Cd during a short-term laboratory incubation and sequential extraction study. Pichtel and Anderson (1997) measured similar increases in the total concentrations of Zn, Cu, Ni, Cr and Pb in silty loam soil (pH 6.8) amended with composted MSW (from manually-sorted mixed MSW) or composted sewage sludge, but the increases were generally in the most resistant NaOH (organic), EDTA (carbonate) and HNO₃ (sulphide) fractions. They concluded that compost metals added to soil in the neutral pH range would be transformed mostly to insoluble or only partially soluble forms upon incorporation. Thus, different types of composted organic residuals, with comparable total metal concentrations, appear to have similar effects on metal behaviour in soil, irrespective of the origin or nature of the initial feedstock materials.

The soil pH value has a critical role in controlling metal availability and declining soil pH increases the mobility of the most labile elements in soil: Zn, Ni and Cd (Smith, 1994a,b). MSW-compost application may not only reduce metal bioavailability by increasing the soil pH value (Chu and Wong 1987; Hernando et al., 1989; Zheljzkov and Warman, 2004a; Jordão et al., 2006), but may further protect soil from potential degradation due to contamination with heavy metals by improving the overall pH buffering capacity of soil due to short and long-term effects of compost application on soil humic acid properties (García-Gil et al., 2004). Zinc, for example, the most abundant element in MSW-compost and amended soil, has a high affinity for sorption on the surfaces of Fe and Mn oxides, particularly as the soil pH is raised (Zheljzkov and Warman, 2004a,b).

Whereas the bioavailability of most elements declines with increasing pH, the Mo anion becomes more mobile with increasing

pH in MSW-amended soil (De Haan, 1981). However, no studies were found suggesting Mo presented a specific problem for the use of MSW-compost and this is probably because the concentration of Mo in MSW-compost is relatively small compared to other important phytotoxic elements. For example, He et al. (1995) found the mean content of Mo in a representative range of MSW-composts in the US was 7.5 mg kg^{-1} DS.

Jordão et al. (2006) observed increases in the extractability of heavy metals by DTPA (diethylenetriaminepentaacetic acid) with increasing metal content of municipal waste composts (probably of manually-sorted mixed refuse origin) added to clay soil in a pot experiment, and also in relation to time from incorporation in response to a decline in soil pH value. In a calcareous tropical soil, where trace element deficiencies are a problem, Zinati et al. (2001) considered that MSW-compost had the potential to supply larger amounts of plant-available Cu and Zn compared to composted sewage sludge or a material produced by cocomposting MSW and sewage sludge together in the ratio 75%:25%, respectively. This was because the dominant forms of these elements were organically bound in soil amended with MSW-compost, whereas Cu and Zn accumulated mainly in the residual and Fe–Mn oxide fractions in soil receiving the composted sludge or cocompost.

Baldwin and Shelton (1999) also found that the metal concentration in MSW-compost (no details were given relating to the nature of the feedstock except the compost was produced in windrows and therefore probably received some manual pre-sorting) had an important influence on metal extractability in soil and may therefore affect the availability of metals in amended soils. Three composts of varying metal contents were applied in this field experiment to a clay soil, including: MSW-compost, sewage sludge compost and MSW cocomposted with sludge. DTPA-extractable metal concentrations (Cd, Cu and Pb) increased in soil receiving high metal compost compared with a low metal material, at equivalent rates of metal addition. Interestingly, however, there was no difference apparent in DTPA extractable Zn or Ni between high and low metal composts, which are potentially the most labile elements in compost and soil. Two soil pH treatments (5.8 and 6.5) were also included in the field experiments. In general, there was no effect of pH value on DTPA-extractable metals, which may indicate the importance of other binding mechanisms operating in clay soil types, which usually have a high affinity for immobilising heavy metals. However, the extractable concentrations of Zn, Cu, Cd and Pb were raised with increasing soil pH value at the highest rate of addition (100 t DS ha^{-1}) of the co-composted material, in contrast to the usual response to increasing soil pH, which generally reduces the availability of heavy metals in soil. In this case, the increase in extractability at the higher pH value was probably explained because the co-composted material contained large amounts of Na (6126 mg kg^{-1}) and increasing ionic strength and electrical conductivity in soil leads to preferential adsorption of monovalent cations. Competition between Zn, Cu, Ca and Mn in solution, and Cd–Cl ion pair formation due to high Cl inputs to soil in this compost may have also contributed to the greater extractability of Cd as pH increased. Thus, at high application rates of the co-compost, preference for metal adsorption decreased and extractable concentrations generally increased in soil with increasing soil pH value. Overall, however, the extractability of heavy metals did not vary greatly with pH even at the highest rates of compost addition.

Organic matter is frequently reported to have a dominant role in controlling the behaviour of Cu in soil (Karapanagiotis et al., 1991), which is also a potentially important binding site for this element in compost-amended soil (Zheljazkov and Warman, 2004b). However, organic matter may not necessarily represent the main binding site for Cu in soil, which may depend to an extent on the soil type. For example, Vaca-Paulin et al. (2006) showed that significant amounts of Cu in a clay loam soil (pH 6.0), amended with both sludge and composted sludge at a rate equivalent to 200 t DS ha^{-1} , were in tightly

bound forms possibly corresponding to Cu held to mineral lattices or sorbed to clays. Hernando et al. (1989) measured an increase in water soluble Cu from MSW-compost amended soil with time due to the release of organically bound Cu, whereas the opposite trend was observed for Zn.

Herencia et al. (2008) showed that the addition of compost produced from source-segregated vegetable waste, containing 55 and 18.5 mg kg^{-1} DS of Zn and Cu, respectively, and applied at a rate of 220 t ha^{-1} over six years to a loam soil with a pH value of 7.6, had no significant influence on the total metal content of the soil. However, this work is important because it showed that, even where there is no overall effect on the total soil metal content, the addition of organic matter to soil in a composted residual can increase the extractable concentrations of metals compared to unamended soil receiving mineral fertilisers. The amount of soluble Cu in the soil was small and the organic amendment redistributed Cu from exchangeable to less mobile oxidizable fractions presumably bound to organic matter. On the other hand, compost application increased the size of the most labile fractions of Zn in the soil. The addition of organic matter can supply soluble organic ligands to the soil thus increasing the potential mobility of metals (Almås et al., 2000). Nevertheless, the majority of the metal remained bound to the residual fraction, which accounted for >80% of the Cu and approximately 70% of the Zn in both compost-amended and mineral fertilized soils. In contrast to unstabilised organic residues, however, such as livestock wastes, water soluble, including soluble organic metal fractions, are greatly diminished in mature aerobically composted biowastes, and this mitigates the potential leaching risk of metals from compost-amended soil (Leita and De Nobili, 1991; Tisdell and Breslin, 1995).

A long-term study of metal behaviour in sewage sludge and composted sludge-amended soil is reported by McGrath (1984). In this work, 25 applications of air-dried, anaerobically digested sludge and compost produced with the same sludge by mixing with straw, were applied to a neutral sandy loam soil in a field experiment (known as the Woburn Market Garden Experiment) over the period 1942–1961. The sludge compost was prepared by mixing the sludge with straw in piles in the ratio 1:2 by fresh weight. The material was composted for four months with periodic turning and water addition (Johnston and Wedderburn, 1974). McGrath (1984) measured the extractability of the metals with EDTA in archived soil samples taken from the field plots in 1960. After 20 years of additions the extractable fractions were similar regardless of the type of organic material and on average were equivalent to 50% of the total metal concentration in the soil. The apparent similarity between the soil amendments could be explained because the metals were present in the same chemical forms, or the speciation changed with time and became identical as the manures degraded to soil organic matter by biological activity (McGrath, 1984). Alternatively, the extractant used did not detect potentially important differences in the types of chemically bound forms of metals in the soil. Indeed, EDTA is a relatively aggressive soil extractant and removes most of the major metal pools (including soluble, exchangeable, sorbed and organically bound, and metals bound in oxide form and to clay minerals), except for the residual fraction (Ure, 1995). The fractionation of soil metals requires sequential extraction or milder extractants (eg NH_4NO_3 or CaCl_2) to determine the potentially bioavailable pool (Pinamonti et al., 1997).

More recently, McGrath et al. (2000) reported the 0.1 M CaCl_2 extractable concentrations of Cd and Zn in Woburn soil sampled in the years: 1960, 1967, 1972, 1980, 1983 and 1984. Extractability of Zn, as a proportion of the total soil concentration, ranged from 0.5 to 3.0% and that of Cd from 4 to 18% and was higher in soil amended with sewage sludge and sludge-straw compost than for farmyard manure and fertilizer-amended soils. No differences in the extractabilities of the elements were noted between the composted and uncomposted sludge. The potential for heavy metals to be redistributed, with the possibility that their bioavailability may increase, has been suggested in response to the degradation of organic matter in soil receiving long-

term compost applications (eg see Zheljzkov and Warman, 2004b; Hargreaves et al., 2008). However, over a 23-year period after 1961, when sludge was last applied at Woburn, the 0.1 M CaCl₂ extractability of both metals fluctuated, but neither increased or decreased (McGrath et al., 2000). Considerable mineralization of the applied organic matter had taken place over this period and only 10–20% of the compost organic matter remained in the soil 23 years after the last application. Consequently, there was no evidence that metal availability increased with time as a result of the mineralization of applied organic matter in compost-amended soil. The significance of the apparent metal behaviour in sludge and compost-amended soil at Woburn is discussed further in the next section, which considers the effects of compost on the bioavailability of heavy metals to plants.

5. Bioavailability to crops

Increasing metal concentrations and changes in the distribution of metals in soil amended with compost in the long-term are generally reported to increase the concentrations of heavy metals in the tissues of plants growing in the soil (Gigliotti et al., 1996; Zhao et al., 1997; Zheljzkov and Warman, 2004b; Wei and Liu, 2005). This depends on the amount of metal in the soil, soil physico-chemical properties (eg pH value), the strength of binding of the element in soil and the ability of the plant to regulate uptake of the element. Thus, Zn is relatively labile and is readily transferred to plant tissues, as predicted from its susceptibility to chemical extraction (Section 3), and it is also usually present in larger concentrations in sludge and compost-amended soil compared with the other elements of interest (Table 1; Speir et al., 2004). Copper, on the other hand, tends to be more strongly sorbed in soil and plants regulate the uptake of this element more effectively than they do Zn (Kabata-Pendias and Pendias, 1992). Therefore, plant tissue concentrations and availability of Cu are usually much lower and less sensitive to inputs of this element to soil in compost or sludge compared with more mobile elements such as Zn (Planquart et al., 1999; Zheljzkov and Warman, 2004b).

Zheljzkov and Warman (2004b) found that increases in plant tissue metals were not proportional to the total metal concentrations measured in soil amended with source-segregated MSW-compost. Both the Bioavailability Factor (= ratio of the exchangeable relative to the total metal content in the soil) and Transfer Factor (= the ratio of metal content in plant tissue relative to the total concentration in the soil) for Zn and Cu in Swiss chard and basil, grown in sandy loam soil in a pot experiment, declined in compost-amended soil (up to 60% by volume, equivalent to a soil mixture containing approximately 17% compost w/w, or an application of 600 t ha⁻¹ DS in the field). In this pot trial, therefore, compost addition reduced the *relative* bioavailability of both elements compared to the unamended control soil. The decline in the relative availability in amended soil was explained due to the influence of the compost matrix on the sorption of metals compared to the unamended control treatment. The actual concentrations of Zn and Cu measured in plant tissues were raised overall compared to the unamended control (see earlier). However, although bioavailable forms of Cu and Zn in compost-amended soil, determined by sequential extraction of the compost-soil mixtures, were increased, as were tissue concentrations of the elements in both crop species, these were within the normal ranges in plants, and below the critical phytotoxic amounts. Thus, Zheljzkov and Warman (2004a) concluded that source-separated MSW-compost containing relatively large amounts of metals (Table 1) may be used as a soil amendment on agricultural crops without a risk of phytotoxicity, or risk of increasing crop tissue concentrations of Cu, Pb and Zn above the normal range. However, long-term effects of compost application on metal availability and effects on crops needed to be considered.

Sequential extraction tests often show a shift in metals from immobile to more mobile forms in MSW-compost amended soil

including, for example, an increase in operationally defined exchangeable or relatively labile organic pools, which are considered to be potentially bioavailable. However, the bioavailability classification assumed for particular metal fractions in sequential extraction schemes (eg He et al., 1995) provides poor predictions of actual potential transfers of metals to crop tissues from compost-amended soil (Zheljzkov and Warman, 2004a; Warman et al., 1995).

As expected from the discussion on the effects of soil pH on the chemical extractability of heavy metals in soil, this soil parameter has a major impact on crop uptake of metals. Indeed, the pH value of soil can be more important in controlling phytoavailability than the heavy metal concentration in compost (Lutz, 1981). For example, Chu and Wong (1987) applied MSW-compost (the source was not specified) and sewage sludge (activated sludge) to a loamy sand soil in a pot experiment with a variety of vegetable crops. Crops grown on compost-treated soil accumulated smaller concentrations of heavy metals compared with sludge, despite its larger metal content. This behaviour was attributed to the higher pH and liming effect observed with MSW-compost and larger organic matter content, which also reduced metal availability, compared to sludge (Chu and Wong, 1987).

Very low transfers of heavy metals to plant tissues occur in high pH and calcareous soils. For example, Gigliotti et al. (1996) reported that soil-plant transfer coefficients were within the normal lower range for maize after application of 90 t ha⁻¹ y⁻¹ of MSW-compost (at 70.5% DS) to calcareous clay loam soil (pH 8.3) for 6 years (total application = 540 t ha⁻¹). This demonstrated the very limited uptake of heavy metals from this soil type and the absence of risks to plant, animal or human health. The heavy metal concentrations in the applied compost are presented in Table 1 and were generally representative of contemporary values in compost sourced from mechanically-segregated MSW. Ozores-Hampton et al. (1997) applied biosolids and MSW-composts, containing up to 607, 650, 34 and 4.1 mg kg⁻¹ DS of Zn, Cu, Ni and Cd, respectively, at a rate up to 48 t ha⁻¹ y⁻¹ to a calcareous soil for two growing seasons and similarly reported no significant change in fruit concentrations of metals in tomato or squash. Although in this case the results could also be explained because transfers of metals to fruit tissues tend to be regulated and controlled physiologically by the plant, nevertheless, high soil pH was probably a major factor reducing metal uptake.

Soil types with a strong affinity for metal sorption, eg soils with high clay content, may be less responsive to the effects of pH (Baldwin and Shelton, 1999) on metal availability to plants and other mechanisms may have a more dominant role in controlling metal behaviour in this case. Differences in the availability of metals for crop uptake between sewage sludge and compost may be explained to some extent by the effects of these materials on the soil pH value, as well as by intrinsic properties of the materials themselves. For example, Simeoni et al. (1984) concluded that the increased uptake of Zn and Cd by lettuce and oats measured for an acid soil amended with digested sludge was largely explained due to a decrease in soil pH value, compared to compost-treated soil. The complexity of the interactions between soil and compost properties that determine the behaviour and bioavailability of heavy metals, emphasise the need for the careful management and control of experimental conditions designed to elucidate the specific factors governing metal uptake in compost and sludge-amended systems.

Pichtel and Anderson (1997) detected increased uptake and potentially phytotoxic concentrations of Zn, Cu and Ni in plant tissues of oats grown in a fine-textured soil (pH 6.8) in a pot experiment amended with MSW (manually sorted) and sewage sludge composts and this was associated with yield decreases at the highest rates of compost application up to 240 t ha⁻¹ DS. However, pot experiments need to be viewed with caution as they tend to accentuate the uptake of metals compared to crops grown in field soil (Logan and Chaney, 1983). In contrast, Smith (1992) measured very low apparent bioavailabilities of metals in compost in a pot experiment with petunias grown in acidic sandy soil-compost mixtures (initial soil pH=5.0) and

in 100% of sludge cake, liquid sludge and mechanically-segregated MSW-composts, containing up to 1100 mg kg⁻¹ DS and 1120 mg kg⁻¹ DS of Zn and Cu, respectively, the main phytotoxic elements in compost. No phytotoxicity was observed due to metals and plant tissue concentrations were below the upper critical toxicity values. Indeed, a positive relation was found between the Cu content in the leaf tissues and flower productivity. Warman et al. (1995) also reported only small increases in the metal content of Swiss chard, a recognized accumulator crop, grown in biosolids–compost soil mixtures and 100% compost in containers. In this experiment, the soil type was a loamy sand with pH value of 5.7 and the compost contained 831, 323, 56 and 8 mg kg⁻¹ DS of Zn, Cu, Ni and Cd, respectively. These studies therefore demonstrated the very low bioavailability and generally recalcitrant nature of heavy metals in composted organic residuals.

Nickel is identified as a potentially toxic element in soil (eg Rooney et al., 2007) and is included in the routinely monitored suite of elements in sludge and composted organic residuals. However, Ni ecotoxicity is rarely reported in practice, other than under exceptional circumstances (eg Frank et al., 1982), and Ni attracts a very low hazard ranking to the human foodchain or terrestrial ecosystems relative to other principal heavy metals (Smolders et al., 2004). Consequently, the concentrations of Ni measured in contemporary sludge or compost (Table 1) have no identified or reported environmental significance (for a review of Ni behaviour and significance in soil, see Carrington et al., 1998); also Chang et al., 1992; Gigliotti et al., 1996). Nickel concentrations are usually relatively low in MSW-compost. Woodbury (1992) noted the mean Ni content in mixed MSW-composts in N America was 31 mg kg⁻¹, whereas soils contain 22 mg Ni kg⁻¹ on average worldwide and upper critical foliar concentrations are 10–50 mg Ni kg⁻¹. Bowen (1979) gives a median value for Ni for normal soils worldwide of 50 mg kg⁻¹ (Table 1). Thus, MSW-compost is unlikely to cause phytotoxicity due to the addition of Ni to soil.

Wen et al. (2002a) describe a series of field experiments to examine the effects of dewatered digested, digested-irradiated and composted sludge, and composted livestock manure applied to a loamy sand soil (pH 6.6) on plant availability of Zn. The soil amendments were applied up to a maximum rate of 40 t DS ha⁻¹ y⁻¹ during two growing seasons and the crops grown included lettuce and snap bean and an ornamental plant, petunia. Composting significantly reduced the availability and crop uptake of Zn compared to the uncomposted sludges. Zinc applied in composted sludge did not increase the crop Zn concentration or soil availability index (estimated from the DTPA extractable concentration and pH value of the soil).

Simeoni et al. (1984) detected similar Cu concentrations in lettuce and oats grown in a glasshouse pot study in two soils (acid loamy sand – pH 5.4 and neutral clay loam – pH 7.6) amended with either anaerobically digested sludge or compost produced from the sludge by mixing with wood chips. However, the sludge contained almost 70% more Cu than the compost (due to dilution with the bulking agent and possible loss of soluble Cu complexed during composting), suggesting potentially increased Cu availability after composting. In general, however, the majority of published research indicates that metal transfers to crops in the short-to-medium term are reduced by composting, or may be similar in the long-term compared to sewage sludge. For example, with regard to Cu, in the field experiments of Wen et al. (2002b), no increase in the Cu concentration of plant tissues was measured with composted sludge and crop Cu values were generally lower compared with dewatered digested sludge or digested-irradiated sludge. Thus, composting sludge apparently reduced the phytoavailability of Cu. Baldwin and Shelton (1999) noted that the leaf Cu concentration in tobacco grown in soil amended with MSW and sewage sludge composts were low and not influenced by pH, and concluded that the adsorptive capacity of compost controlled the availability of this element in soil. Nevertheless, leaf Cu concentrations were influenced by the amount of Cu in the com-

post and were raised with increasing compost Cu content (from 173 to 215 mg Cu kg⁻¹ DS). However, the apparent relationship between the plant and compost concentrations of Cu (and Zn) could have also been influenced by other compost parameters (eg salinity) that potentially influenced plant growth and the dilution of metals in the tissues, although the plant transfer results were consistent with DTPA-extractable Cu data (Baldwin and Shelton, 1999).

Lead has exceptionally low plant availability in sludge and compost-amended soil (Carlton-Smith, 1987; Warman et al., 1995; Planquart et al., 1999; Zheljzakov and Warman, 2004a) and uptake of this element by crop plants is not a concern with regard to the human foodchain (Table 1), which is protected by the soil-plant barrier (Logan and Chaney, 1983). Compost is reported to have a high affinity for binding Pb into stable forms, for example, in sulphide and residual fractions that have very low availability (Grimes et al., 1999; Song and Greenway, 2004; Zheljzakov and Warman, 2004a; Zheng et al., 2004; Amir et al., 2005). Gigliotti et al. (1996) observed very limited mobility of Pb to corn plants grown in calcareous soil (pH 8.3) amended with mechanically-segregated MSW-compost at a rate of 90 t ha⁻¹ y⁻¹ for 6 years. Chromium is also an element with extremely low bioavailability in soil, sewage sludge and composted sludge and MSW residuals (Hernando et al., 1989; Smith, 1992, 1993; Warman et al., 1995).

Chang et al. (1984) report data on metal accumulation in soil from a long-term field trial with composted sewage sludge and two liquid anaerobically digested sludge types applied annually for six years. The data were presented as a recovery mass balance and included the total metal additions to the plots in the different materials and the total removals of metals in the crops for the six year period. The data have been used here to compare the bioavailabilities of Zn and Cd, two of the most potentially labile elements in sludge and compost, in the sludges treated by composting and digestion processes (Table 2). The values presented in Table 2 are the mean metal recoveries in the harvested plant material as a proportion (%) of the total amounts of metals added at three rates of application of each material on two soil types (sandy loam and fine loam), excluding the background uptake by the unamended controls. The results show that approximately 0.5% of the added Zn and Cd in liquid digested sludge types were removed in crop tissue, and were therefore bioavailable. By comparison, the metal recoveries from composted sludge were reduced by approximately 70% to a value of about 0.15%. All the materials examined by Chang et al. (1984) were derived from wastewater treatment residuals. However, this comprehensive and long-term work provides clear evidence for the lower bioavailability of metals in composted organic residuals compared to anaerobically treated products.

Composted sludge significantly decreased the bioavailability and uptake by maize of Cd added as a soluble salt to a relatively acid silty loam soil (pH 5.5) in a pot experiment reported by Shuman et al. (2002). The decrease in plant Cd was explained by the redistribution of Cd from the water soluble and exchangeable fractions in soil, without compost addition, to the organic fraction with compost amendment.

Table 2

Mean total removal of Zn and Cd in crop tissues in a long-term field experiment from a 6 year crop rotation on two soils types (sandy loam and loam) amended annually with three rates of composted and two liquid digested sewage sludges (adapted from Chang et al., 1984)

Sludge type	Mean metal recovery in crop tissues (% total addition) ^a	
	Zn	Cd
Composted	0.16	0.14
Liquid digested 1	0.43	0.42
Liquid digested 2	0.62	0.54

^a Excludes the background offtakes of metals in unamended controls.

Shuman et al. (2002) concluded, therefore, that composted sludge would be potentially useful and effective in decreasing Cd uptake by plants from soil contaminated with this element.

In the long-term study reported by McGrath et al. (2000), however, no differences in the bioavailabilities of Zn and Cd to red beet, sugar beet or barley were found between sewage sludge or composted-sludge amended soil from the Woburn Market Garden Experiment, which were also higher than for the unamended control plots. This would suggest that, in the long-term, the type of organic amendment and stabilisation treatment process may have no overall significance for the availability of heavy metals in amended soil. However, the sludge and compost applied at Woburn had exceptionally high metal concentrations (McGrath, 1984) compared to contemporary amounts (Table 1), and short-term studies suggest this may significantly increase the availability of metals in compost-amended soil (Baldwin and Shelton, 1999). Although a significant amount of the organic matter had decomposed following the last application of sludge and compost to Woburn soil, and important elements including Zn (and Cu) may be present in organically bound fractions in compost-amended soil, there was no evidence of an increase in bioavailability of Zn and Cd to the crop plants with time (McGrath et al., 2000), consistent with the CaCl_2 extractable metal concentrations measured in the soil.

From an early critical review of long-term field studies, Woodbury (1992) concluded that heavy metals in MSW-compost were unlikely to pose a risk of phytotoxicity to crops or accumulate in crop tissues to levels of concern to the foodchain. Zinc is the most mobile element with the largest concentration in MSW-compost and the greatest potential to transfer to crop tissues (De Haan, 1981), but even so, Zn phytotoxicity is rarely reported (Woodbury, 1992; He et al., 1995). The evidence reviewed here on metal bioavailability in MSW-compost and amended soil confirms that MSW-composts present little risk to crops or to the foodchain through the soil-crop-human exposure pathway.

6. Soil microbial activity and fertility

There is increasing interest in quantifying the ecotoxicological risk of composted residuals. The extensive array of ecotoxicity test protocols for compost, employing microorganisms, enzymes, soil fauna and plants, are discussed in the review by Kapanen and Itävaara (2001).

Gallardo-Lara and Nogales (1987) give an early overview of the effects of MSW-compost incorporation on soil biological properties. These generally included increases in the fungal and bacteria populations in amended soil, and an increase in CO_2 production. As may be expected, the effect of compost application on the total and fungal microflora decreased with increasing maturity and stability of the material, presumably in response to the lower substrate availability, whereas the number of actinomycetes increased. Populations of rhizosphere microorganisms were also reported to increase in relation to increasing inputs of composted organic matter to soil. Woodbury (1992) also noted that compost application often had beneficial effects on soil microorganisms and concluded that evidence relating to potential impacts of heavy metals applied to soil in MSW-compost on the soil microbiota was contradictory.

However, medium to relatively long-term field studies conducted in Italy (Perucci, 1992; Giusquiani et al., 1994; Crecchio et al., 2001) and Spain (Pascual et al., 1999) have consistently shown positive effects of MSW composts (see Table 1 for heavy metal concentrations) on soil enzyme activities, respiration rate and microbial biomass C concentration, and there was no effect on soil bacterial community biodiversity. Some of the conditions examined represented extreme doses of MSW-compost application, equivalent to 10–20 years of inputs under normal agronomic practice, up to a maximum of 270 t ha^{-1} in one case (Giusquiani et al., 1994), and these positive effects could be detected up to eight years after incorporating MSW-compost into the soil (Pascual

et al., 1999). Similarly, no negative impacts on soil microbial biomass, substrate induced respiration or soil enzyme activities were detected in the soil incubation study reported by Bhattacharyya et al. (2003) for an acidic soil (pH 5.5) supplied with MSW-compost at a rate equivalent of up to 40 t ha^{-1} and containing 691 and 149 mg kg^{-1} of Zn and Cu, respectively. Whilst increases in soil metals (eg see Giusquiani et al., 1994) were below European maximum permissible limits for sewage sludge-treated agricultural soil (CEC, 1986), these studies indicate MSW-compost application had no direct toxic action on the soil microbial biota.

In a long-term field experiment, García-Gil et al. (2000) applied up to $80 \text{ t ha}^{-1} \text{ y}^{-1}$ of MSW-compost to a sandy soil (pH 6.4 with low organic matter) for a period of 5 years over a total duration of nine years, when the plots were sampled to determine enzyme activities and microbial biomass content. The metal contents measured in the compost (Zn = 1325 mg kg^{-1} DS, Cu = 548 mg kg^{-1} DS and Pb = 681 mg kg^{-1} DS) suggested this was produced from mechanically-sorted MSW, although no details are provided about the source. At the highest accumulative rate of addition, the soil contained: $159 \text{ mg Zn kg}^{-1}$, 61 mg Cu kg^{-1} , 13 mg Ni kg^{-1} . The background soil concentrations were: 18 mg Zn kg^{-1} , 6 mg Cu kg^{-1} , 4 mg Ni kg^{-1} ; there was no evidence of Cd accumulation and the concentration of this element in the unamended control and amended treatment plots was 0.2 mg kg^{-1} . MSW-compost increased the microbial biomass content of the soil compared to the control due to substrate addition, although the lowest overall biomass C:soil organic C ratio was measured at the high rate of compost application. This could imply a toxic response to heavy metals in the soil, however, García-Gil et al. (2000) suggested that the most likely explanation was the high stability and humification of organic matter in compost, which increased its resistance to microbial attack, compared to cow manure treatments that give higher microbial biomass and ratio values. The activities of the intracellular enzymes: dehydrogenase, catalase and β -glucosidase, increased with the rate of compost addition, compared to the control, indicating an increase in microbial metabolism in the soil due to the mineralization of biodegradable C inputs, although the response was smaller compared to cow manure, again reflecting the differences in the intrinsic biodegradabilities of the two organic materials. These enzymes were therefore not affected by the concentrations of heavy metals measured in the high rate accumulation plots. In contrast, urease and phosphatase activities were reduced in compost-amended soil compared to control and mineral fertiliser plots. Heavy metal toxicity was indicated as one possible cause of the inhibition of these enzymes, however, urease activity is also reduced in the presence of NH_4^+ released by mineralization processes in the soil and phosphatase is inhibited by the presence of inorganic P. Compost application increases both of these major nutrients in soil (Speir et al., 2004), which may offer a more plausible explanation for the apparent changes observed in the activities of these enzymes in the amended soil.

More recently, Crecchio et al. (2004) reported a 6 year field investigation on a clay soil (pH 8.3) receiving annual dressings of MSW-compost, also probably supplied from mechanically-treated MSW (see Table 1 for the heavy metal contents), at a rate of up to $24 \text{ t ha}^{-1} \text{ y}^{-1}$. The crops grown were a sugar beet and durum wheat rotation. Soil samples were collected for biochemical and microbiological examination at the end of the 6 year experimental period. Unfortunately, however, the authors omitted to include basic heavy metal concentration data for the soil and therefore it is not possible to draw comparisons between these results and other experimental work with regard to the potential effects of heavy metals in the compost-amended soil. Dehydrogenase, β -glucosidase, urease, nitrate reductase and phosphatase enzyme activities were slightly increased in compost-amended soil at the 12 t ha^{-1} rate, albeit not significantly, and there was no further increase at the higher rate of compost addition compared to the control, whereas protease activity was significantly reduced by compost application at 24 t ha^{-1} . Crecchio et al. (2004) speculated that the absence of an increase in microbial activity at the higher rate of organic matter input in compost could be

attributed to heavy metal toxicity and protease was possibly more sensitive to metals in soil compared to the other enzymes assayed, but there was no direct evidence of this. Indeed, this seems unlikely as García-Gil et al. (2000) found that protease activity was stimulated in a sandy soil amended with up to 80 t ha⁻¹ y⁻¹ of MSW-compost for five years under field conditions compared to the unamended control, and the applied compost contained significantly larger concentrations of heavy metals than that used by Creccchio et al. (2004). Giusquiani et al. (1994) suggest extremely high rates of MSW-compost addition may not stimulate soil enzyme activities further because enzyme activities may reach their maximum value for soil (Martens et al., 1992). Furthermore, Creccchio et al. (2004) reported that compost application had no effect on the bacterial genetic diversity of the soil, or on the variation in specific taxonomic groups regarded as essential for soil fertility. Therefore, Creccchio et al. (2004) concluded that these biochemical and molecular results demonstrated that MSW-compost application had a beneficial effect overall and no significant negative effects on important biological components of soil were detected. MSW-compost also enhanced soil fertility in terms of crop production and quality.

Speir et al. (2004) provided annual applications of composted sewage sludge for 4 years up to a maximum rate of 200 t ha⁻¹ y⁻¹ DS to a sandy loam soil at two pH ranges (5.0–5.5 and 6.0–6.5) and measured various soil biological properties after the last application. The maximum total metal concentrations in the soil were: 184 mg Zn kg⁻¹, 101 mg Cu kg⁻¹, 19 mg Ni kg⁻¹ and 0.63 mg Cd kg⁻¹ dry soil. The mean background concentrations in the unamended control soil were: 45 mg Zn kg⁻¹, 14.5 mg Cu kg⁻¹, 12 mg Ni kg⁻¹, 0.18 mg Cd kg⁻¹ dry soil. Soil basal respiration, microbial biomass C and anaerobically mineralisable N were significantly increased in the compost-amended plots relative to the control. Sulphatase and phosphatase activities also increased, although not significantly. There were no effects of compost addition on the numbers of the N-fixing bacteria, *Rhizobium*, free-living in the soil, or on sensitive microbial biosensors (*Rhizotox C* and *lux*-marked *Escherichia coli*). The application of sewage sludge compost was shown to enhance soil fertility, productivity and microbial biomass and activity, with no apparent adverse effects attributable to heavy metals. Importantly, the maximum Zn concentration in the soil was very close to the maximum advisory limit recommended for this element (200 mg kg⁻¹) in sludge-treated agricultural soil in the UK (DoE, 1996).

The potential long-term implications of heavy metal accumulations in soil from the agricultural use of sewage sludge have received considerable debate in recent years (McGrath and Chaudri, 1999; Smith, 2000) and are also relevant to the use of composted organic residuals as soil improvers. An extensive amount of research has been completed in this area and a programme of multi-site, long-term experiments is ongoing in the UK to quantify the effects of heavy metals on the long-term fertility of sewage sludge-amended agricultural soil, although the field experiments do not include compost treatments (Chambers, 2006; Gibbs et al., 2006a,b). Results emerging from the research are equivocal at a number of the sites as negative effects are apparent on high-metal cake sludge-amended plots, but not where Zn metal salt treatments, with higher overall bioavailability, have been applied (Chambers, 2006; UKWIR, 2007). It is widely accepted that the bioavailability and toxicity of metal salts in soil is usually greater than for metals present in sludge-amended soil due to the complexation mechanisms operating within the sludge matrix (Basta et al., 2005). Therefore, further careful interpretation and data analysis, experimental investigations on metal speciation and comparisons with other international published literature are necessary to establish the significance and implications of the results before they should be adopted as a basis for developing soil protection policies and metal limits. These apparent anomalies may be explained for instance, because microorganisms were exposed to localised particles of high-metal sludge cake containing toxic amounts of Zn, compared to the more representative averaging of the metal concentration in soil receiving the metal salt. De Brouwere and Smolders (2006) observed

similar effects of Zn-rich sludge, compared to low Zn sludge, on the phytotoxicity of this element to plants.

Only a brief overview of the other main scientific literature can be presented here. Early data from the Woburn Market Garden Experiment showed a significant reduction in the microbial biomass content of sludge-treated soil compared to farmyard manure and control plots due to an apparent toxic effect of increased heavy metal concentrations on the soil microbial community (Brookes and McGrath, 1984). Subsequent work measured the effects of heavy metals on N-fixation and the populations of the symbiotic N-fixing bacteria *Rhizobium leguminosarum* biovar *trifolii* in Woburn soil (McGrath et al., 1988; Giller et al., 1989). However, the high Cd content of the sludge-amended soil at Woburn may be a significant contributory factor to the apparent negative impacts on soil microorganisms observed at this site compared to soils supplied with contemporary sewage sludges with low metals contents, including Cd (McGrath, 1984; MAFF/DoE, 1993; Gendebien et al., 1999; also see: Chaudri et al., 1992; Giller et al., 1993; Chander et al., 1995). For example, McGrath et al. (1988) reported a Cd concentration in the soil receiving sewage sludge of 15.4 mg kg⁻¹ dry soil, more than 5 times the UK legal limit in sludge-treated agricultural soil (SI, 1989). Irrespective of the source, MSW-compost contains relatively small amounts of Cd, similar to or slightly higher than the median background concentration in soil, but well below concentrations that could pose a threat to soil ecology. By contrast, the maximum mean Cd concentration in the sludge applied to the Woburn field experiment recorded for the period 1957–61 was 137 mg kg⁻¹ DS (McGrath, 1984). Overall, the mean Cd concentration in sludge-compost was approximately 80% of the mean value in the sludge resulting from dilution with the straw bulking agent. Consequently, because there were a range of metals in the sludge-treated plots at Woburn, it was not possible to determine which metals were toxic to rhizobia (Chaudri et al., 1993a). However, investigations at another sludge-amended field trial site at Braunschweig, Germany (Chaudri et al., 1993a), identified Zn as the critical element that may be potentially toxic to *Rhizobium* survival in relation to the European and UK maximum permissible limit values for heavy metals in sewage sludge-treated agricultural soil (CEC, 1986; SI, 1989; DoE, 1996). Certain sludge treatments at Braunschweig were incubated with metal salts and the sludge was applied to the experimental plots as digested liquid. No compost was used in this experiment so it is not possible to compare the effects of sewage sludge and compost on metal behaviour or soil microbiology. However, other medium-term, field based research (Chang et al., 1984) has shown that metal availabilities in compost-amended soil are significantly smaller than for liquid anaerobically digested sludge (Table 2).

Other research on long-term, operationally sludge-treated sites, however, did not support the link between metal contamination of soil and toxicity to *Rhizobium* (Smith, 1997). In this work, other factors (eg soil pH and the presence of the host crop) were found to be much more significant in controlling the presence of the bacteria in the free-living state in soil than the soil heavy metal content. Investigations on the microbial biomass content of historically sludge-treated soils indicated the upper critical toxic Zn concentration in soil was 580 mg kg⁻¹ dry soil (Smith, 1998). Furthermore, no effect of Zn on the microbial biomass was detected in a field experiment in sandy loam soil containing 450 mg Zn kg⁻¹ dry soil across the pH range 5.4–7.0 (Smith et al., 1999).

The chemical and ecotoxicological characteristics of anaerobically digested sewage sludge and unsorted MSW-compost and green waste-compost were compared by Alvarenga et al. (2007). This study measured the direct ecotoxicity of these organic residuals using plant growth tests (cress and barley) and earthworm mortality. Indirect exposure assays with leachate from the residues were based on luminescent bacteria, seed germination of the same plant species and *Daphnia magna* immobilization. Sewage sludge gave the highest overall ecotoxicity response measured by the indirect assays and to earthworms compared to the other materials examined. The direct

exposure assays and luminescent bacteria test showed significant ecotoxic effects of the MSW-compost although the response was smaller compared to sludge. Only luminescent bacteria responded negatively to the green waste compost. However, the relevance of such direct and leachate toxicity testing of organic residuals to the behaviour of these materials in soil is questionable. For example, digested sludge contains appreciable amounts of ammonia-N, which is potentially biocidal, but is an important plant nutrient and substrate for nitrification in soil. Also, the Zn content of the sludge used by Alvarenga et al. (2007) would not be acceptable for use on agricultural land (CEC, 1986) being almost 10 times the typical average value measured in sludge (Table 1). The relatively high electrical conductivity of MSW-compost (5.7 mS cm^{-1}), compared to the other substrates examined, could also explain the detrimental effects of this material to some of the assayed organisms. For instance, salt content and toxicity is much more of a concern than compost metal content especially in arid and semi-arid conditions (Maftoun et al., 2004).

In artificial laboratory culture, soil bacteria show extreme sensitivity to Cu and, because of this, Cu has been suggested as the most environmentally relevant element found in organic wastes (Keeling and Cater, 1998). However, whilst demonstrating the potential antimicrobial potency of Cu, these experimental systems do not provide representative information on the actual behaviour of metals in soil, due to the high solubility of metal salts in the growth media (Chaudri et al., 1993b). Copper has not emerged as an element of concern for recycling organic residuals on land or in relation to the European soil metal limits for agricultural use of sewage sludge (CEC, 1986; MAFF/DoE, 1993). This is because Cu availability is relatively limited in sludge and compost-amended soil due to the extent of its complexation, particularly with organic matter, in soil and in sludge and composted matrices.

In a detailed study of the microbial community structure and function of a metal contaminated soil, Pérez-de-Mora et al. (2006) showed that the addition of compost produced from MSW or sewage sludge raised the soil pH value and reduced the solubilities of metals determined by chemical extraction with CaCl_2 . Compost application was one of the most effective of a range of soil amendments examined and increased the soil microbial biomass C (MBC), the ratio of MBC to total soil organic C (TOC) and certain enzyme activities. Interestingly, however, there was increased microbial diversity in the control soils, with the highest metal availabilities and the lowest MBC concentrations, compared to compost-amended soil. This may be explained because elevated metal concentrations can exert a selective pressure on the microbial community increasing the numbers of metal-tolerant or resistant strains, compared to soil ameliorated by compost application with reduced metal bioavailability (Pérez-de-Mora et al., 2006).

7. Conclusions

- International data on the total concentrations of heavy metals in MSW and greenwaste compost are compiled and show that all types of compost contain larger concentrations of heavy metals than the background values present in soil, irrespective of the source. Therefore, heavy metals will slowly accumulate in soil following the long-term application of all types of composted biodegradable waste materials.
- The metal concentrations measured in source-segregated and greenwaste composts are determined by the natural background contents of metals in food and plant material wastes.
- Source-segregated MSW and greenwaste generally result in compost with the smallest amounts of heavy metals and total metal concentrations in these composted materials are typically below UK PAS100 limits. Compost produced from mechanically-segregated MSW generally contains larger metal contents than from source-segregated MSW or greenwaste. However, the review of international metal content data has shown that mechanical treatment can achieve a final product compliant with UK PAS100 indicating that the main sources of contaminants can be removed relatively effectively by modern mechanical screening processes. Total concentrations of heavy metals in mechanically-segregated compost are usually smaller than in sewage sludge, by approximately 30–50% except for Pb. The Pb content of compost from mechanically-segregated MSW may be similar or up to approximately 80% larger than the total Pb concentration measured in sewage sludge.
- Zinc and Pb are numerically the elements present in the largest amounts in source and mechanically-segregated MSW and greenwaste composts. Lead is identified as the most limiting element overall to use of mechanically-segregated MSW compost in domestic gardens. Although compost from mechanically-sorted waste generally contains more Pb than from source segregation, the mean Pb concentration in compost from modern mechanical segregation and treatment facilities is significantly below the risk-based limit value of 300 mg kg^{-1} for this element in compost.
- Metal contamination in mechanically-segregated compost is generally associated with fine particle size material and preprocessing and product refinement techniques to remove this fraction significantly reduce the concentration of heavy metals in the final product. This may also increase the overall organic matter content and agricultural value of the screened compost.
- The literature relating to the behaviour and availability of heavy metals applied to soil in compost and sewage sludge is extensive and includes short and long-term investigations, chemical extraction and plant uptake studies and pot and field experimental work. However, the amount of information specific to source-segregated compost is relatively limited. In summary, compost and sewage sludge additions to agricultural and other non-metal contaminated soils raise the soil content and the availability of heavy metals for transfer to crop plants. The availability in the soil depends on the nature of the chemical association between a metal with the organic residual and soil matrix, the pH value of the soil, the concentration of the element in the compost and the soil, and the ability of the plant to regulate uptake of a particular element.
- Research investigating the nature of metal binding mechanisms to compost show that composted residuals derived from MSW and greenwaste have a high affinity for metal sorption. There is general consensus in the scientific literature that aerobic composting processes increase the complexation of heavy metals in organic waste residuals, and that metals are strongly bound to the compost matrix and organic matter, limiting their solubility and potential bioavailability in soil. Lead is the most strongly bound element and Ni the weakest, with Zn, Cu and Cd showing intermediate sorption characteristics.
- Consequently, there is little evidence of phytotoxic effects, or accumulations of metals in crop tissues that may pose a risk to human health, from application of MSW-composts to soil.
- Assumptions about the relatively bioavailabilities of different sequentially extracted metal fractions, such as exchangeable or organically bound forms, appear to have poor associations with actual transfers of metals to crop tissues from compost or compost-amended soil.
- Certain elements such as Cu and Zn may have a strong association with organic fractions in MSW-composts; however, there is no evidence from long-term field studies that the bioavailability of heavy metals in compost-amended soil increases with organic matter decomposition.
- There is good experimental evidence demonstrating the reduced bioavailability and crop uptake of metals from composted biosolids compared to other types of sewage sludge. It may therefore be inferred from this that composting processes overall are likely to contribute to lowering the availability of metals in amended soil compared to other waste biostabilisation techniques.
- The metal concentration in compost also emerges as an important factor controlling crop uptake of labile elements, like Zn and Cu, which

increase with increasing total content of these elements in compost. Therefore, low metal materials, which include source-segregated and greenwaste compost, are likely to have lower metal availabilities overall, at equivalent metal loading rates to soil, compared to composted residuals with larger metal contents. This is explained because the compost matrix modulates metal availability and materials low in metals have stronger sorption capacity compared to high metal composts. Composts are divided into different categories for operational reasons depending on the nature of the feedstock and whether it is derived from source or mechanically-segregated MSW, or greenwaste. The input metal content of the feedstock affects the final concentration in the compost, and therefore, the availability of metals. However, it is emphasised that it is the concentration of metals in the feedstock used for composting and not the type of feedstock *per se* that is important in controlling metal availability. Thus, compost produced from source-segregated or mechanically-segregated material, for example, would be expected to have broadly similar metal availabilities if they contained similar total amounts of metals.

- Data from one long-term, historical field experiment in the UK suggest that metal availabilities in soil amended with sewage sludge or sludge-compost may tend towards equivalency over time. However, in this case, both sludge and compost were exceptionally contaminated with metals and this may have had a profound and over-riding influence on metal behaviour in the soil compared to contemporary sludge and composted residuals applied to land.
- Soil pH is a critical factor influencing plant uptake of labile elements (eg Zn, Ni and Cd). The effect of compost or sludge addition on this soil parameter, in the short to medium-term, is likely to have an important influence on metal availability and potential transfer to crop plants. In most situations, metal extractability and uptake declines with increasing soil pH value; compost application generally increases soil pH, therefore reducing crop uptake, whereas digested sewage sludge tends to lower the soil pH value, thus potentially increasing metal transfers to plant tissues. Very low transfers of heavy metals are reported from compost additions to neutral pH and calcareous soils. Soil pH may be less important for controlling metal availability in soil types with a strong affinity for metal sorption, such as soils containing a large proportion of clay. Compost application also improves the pH-buffering capacity of soil due its influence on soil humic acid characteristics.
- Zinc is the element in sewage sludge-treated agricultural soil identified as the main concern in relation to potential impacts on soil microbial activity and is, therefore, also the most significant metal in compost with regard to long-term soil fertility. With the exception of one long-term field experiment in the UK, there appears to be no other tangible evidence demonstrating negative impacts of heavy metals applied to soil in compost on soil microbial processes or fertility and only positive effects of compost application on the soil microbial status and fertility are reported. In one study, the Zn concentration of the soil was close to the UK maximum advisory limit recommended for this element (200 mg kg⁻¹) in sludge-treated agricultural soil, nevertheless, only positive effects on soil microbial processes were observed. The apparent negative impacts on soil microorganisms reported in the long-term experiment referred to above could be explained by the exceptionally high concentrations of Cd in the compost-amended soil and of metals generally in the applied compost, which are unrepresentative of current practice and compost quality.
- The review of scientific literature has indicated that the risks to the environment, human health, crop quality and yield, and soil fertility, from heavy metals in MSW-compost are minimal. The smallest metal contents are generally reported for source-segregated and greenwaste compost, compared to mechanically-sorted compost. However, the total metal concentrations measured in all these compost types are smaller than for sewage sludge. Preprocessing and refinement technologies are necessary to minimise the

contamination of mechanically-segregated MSW-compost as far as practicable and this could also improve other agronomic properties of the compost.

- The scientific evidence indicates that conservative, but pragmatic limits on heavy metals in compost may be set to encourage recycling of composted residuals and contaminant reduction measures, which at the same time, also protect the soil and environment from potentially negative impacts caused by long-term accumulation of heavy metals in soil.

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