Environ Sci Pollut Res (2011) 18:918–939
DOI 10.1007/s11356-011-0444-1

RESEARCH ARTICLE

Inventory of heavy metal content in organic waste applied as fertilizer in agriculture: evaluating the risk of transfer into the food chain

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Received: 23 April 2010 / Accepted: 7 January 2011 / Published online: 28 January 2011

Abstract

Background, aim, and scope In this work, an environmental risk assessment of reusing organic waste of differing origins and raw materials as agricultural fertilizers was carried out. An inventory of the heavy metal content in different organic wastes (i.e., compost, sludge, or manure) from more than 80 studies at different locations worldwide is presented.

Materials and methods The risk analysis was developed by considering the heavy metal (primarily Cd, Cu, Ni, Pb, and Zn) concentrations in different organic residues to assess their potential environmental accumulation and biotransfer to the food chain and humans. A multi-compartment model was used to estimate the fate and distribution of metals in different environmental compartments, and a multi-pathway model was used to predict human exposure.

Results The obtained hazard index for each waste was concerning in many cases, especially in the sludge samples that yielded an average value of 0.64. Among the metals, Zn was the main contributor to total risk in all organic wastes due to its high concentration in the residues and high biotransfer potential. Other more toxic metals, like Cd or Pb, represented a negligible contribution.

Conclusions These results suggest that the Zn content in organic waste should be reduced or more heavily regulated to guarantee the safe management and reuse of waste residues according to the current policies promoted by the European Union.

Keywords Risk assessment · Organic waste inventory · Heavy metals · Biotransfer

Responsible editor: Vera Slaveykova

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1 Introduction

Reusing organic waste as a soil fertilizer offers a number of advantages over other management alternatives because it reduces the use of other fertilizers and eliminates the necessity of its subsequent treatment or disposal (Bruun et al. 2006; Hargreaves et al. 2008). Sewage sludge and manure are the most common organic wastes applied either raw or composted (i.e., humification of the organic matter under controlled conditions). The application of such wastes to soil provides nutrients, increases organic matter, improves soil structure, and enhances nutrient absorption by plants (Weber et al. 2007; Singh and Agrawal 2008). Therefore, the use of different types of organic waste in agriculture or farming activities instead of using conventional chemical fertilizers should be preferred in terms of sustainability. These residues can also be used as amendments to regenerate infertile soils and for improving plant cover (Soliva and Paulet 2001).

However, the European legislation has become more restrictive on the content of priority pollutants in residues that are used as raw materials for the production of fertilizers or as fertilizers themselves (European Commission 2004), ultimately limiting waste reuse in agriculture. Currently, there are several types of organic waste and compost, classified according to the origin of its raw materials (European Community 2006): urban residues, agricultural and forest residues, wastewater treatment sludge, residues resulting from terrestrial remediation activities, residues from industrial processes, and mixtures of these. Depending on the raw material, toxicity due to the presence of persistent organic pollutants or heavy metals may become important (Hua et al. 2008; Oleszczuk 2008). The application of organic waste (i.e., compost, sludge or manure) to land, especially agricultural crops, represents a significant input of nutrients (i.e., nitrogen, sulfur, and phosphorus), but also of metals, some of them being toxic like cadmium or lead (Pichtel and Anderson 1997; Pinamonti et al. 1997; Lipoth and Schoenau 2007; Madrid et al. 2007). Thus, organic waste likely to be used as fertilizer must contain metal levels that are suitable for soil application in accordance with Directive 86/278/EEC (European Community 1986), which regulates the use of sewage sludge in agriculture. However, pollutant concentration should be considered a unique criterion for waste reuse. Repeated application over extended periods of time and an increase in application frequency favor metal accumulation and biotransfer. Depending on soil composition and the presence of metals in the reused waste, specific chemical and physical associations can cause the accumulation of these pollutants in soil. This soil build-up might cause severe adverse effects to animal and human health through their incorporation into the food chain, with the intake of food grown in contaminated areas as the most direct route of exposure (Lăcătuşu et al. 1996; Khan et al. 2008; Sridhara Cari et al. 2008; Smith et al. 2009; Zhuang et al. 2009). Environmental risk assessment (ERA) could assist in establishing safety conditions for organic waste application as fertilizer to agricultural crops and pasture production (Franco et al. 2006). In this type of analysis, it is important to consider the proper mechanisms of transfer, accumulation, and exposure for a reliable estimation of human exposure to heavy metals, according to the waste-reuse scenario under consideration.

There are numerous research studies related to the metal contents of different types of organic waste, such as manure (Bolan et al. 2004) and compost (Ciavatta et al. 1993; Ayuso et al. 1996; Ihnat and Fernandes 1996; Goi et al. 2006; Cai et al. 2007; Chen et al. 2008; Farrell and Jones 2009a; Haroun et al. 2009), and the potential biotransfer to soil and crops (Pinamonti et al. 1997; Bazzoffi et al. 1998; Cole et al. 2001; Korboulewsky et al. 2002; Casado-vela et al. 2007; Kidd et al. 2007; Bose and Bhattacharyya 2008; Odlare et al. 2008; Achiba et al. 2009). Many of these authors have stressed both the consequences of the presence of metals for both humans and the environment and the need for controlled agricultural activities.

In this work, a wide inventory of the heavy metal content of different types of organic waste was taken. Data collected in the inventory was used to estimate the possible risk derived from the reuse and application of these residues as fertilizers in agriculture. A multi-compartment fate and exposure model was used. This was the basis of a decision support tool for organic waste management (Río et al. 2011), to evaluate the transfer of heavy metals into the food chain and the possible impacts on human health. The influence of model parameterization on the results obtained was assessed by developing a sensitivity analysis to evaluate the contribution of the different variables considered in the model to uncertainty, especially those related to soil properties. The information and results provided in this work are intended to contribute to the current body of knowledge on the reuse of different types of organic waste as fertilizers within the field of environmental management and safety.

2 Materials and methods

2.1 Data inventory

An exhaustive review of studies presenting the heavy metal content of organic waste was collected from the scientific literature. The resulting inventory included 194 cases of different types of residues, which were classified into three main categories: compost (83 cases, Table 1), sludge and other uncomposted wastes (81 cases, Table 2), and manure (30 cases, Table 3). The inventory focused on residues of domestic origin, assuming a final fate of reuse in agriculture. Special attention was paid to works developed during the last decade, although previous studies were also considered. A higher number of studies involving compost or sludge were considered since, in general, reusing this residue might be more problematic due to its higher metal content compared to other types of organic waste. More cases were included in the inventory to better reflect the effect of possible variations in metal concentration among different sludges (domestic and industrial origin). Even though some studies presented data on several metals, only the five most commonly analyzed (i.e., Cd, Cu, Ni, Pb, and Zn) were considered in the inventory for calculating risk indexes. Another criterion for selecting these metals was to reflect different levels of toxicity in the inventory: high (Cd and Pb), mid (Ni), and low (Cu and Zn).

2.2 Environmental risk assessment model

An ERA was used to estimate the potential adverse effects on human health resulting from the application of organic waste containing heavy metals as fertilizer in the production of forage. The importance of the different metals' distribution mechanisms in the environment varies depending on

Table 1 Metal content inventory, metal hazard quotient (HQ), and hazard index (HI) of composts

Compost source	Origin and feedstock materials	Country	Heavy metal content (mg/kg)					Data reported	HQ					HI
			Cd	Cu	Ni	Pb	Zn		Cd	Cu	Ni	Pb	Zn	
Pinamonti et al. 1997	(MSW) MSW compost from the composting of the organic fraction of unseparated MSW, selected mechanically at the plant	Italy	3.2	437	140	652	1,228	Mean	0.017	0.074	0.147	0.075	0.438	0.751
	(SS+B) Compost produced at the plant through the treatment of a mixture of urban wastewater purification sludge and poplar bark (ratio, 1:2 *v/v*)		1.2	184	25	81	512	Mean	0.012	0.045	0.039	0.019	0.239	0.354
Pichtel and Anderson 1997	(MSW) Composted MSW prepared from municipal wastes that were processed first by manual techniques to remove non-recyclable materials. The compostable fraction included food and yard wastes, paper products, and other organic solids. The solids were exposed to in-vessel biological digesters for pretreatment (3 days), then transferred to piles, where they were composted by the turned-pile method for several weeks	USA	–	236	28.0	210.0	655	Single value	–	0.051	0.041	0.032	0.282	0.406
	(SS) The sludge, derived from primarily domestic wastewater, was anaerobically digested and then composted by the aerated-pile method		–	269	40	340	770	Single value	–	0.055	0.052	0.045	0.314	0.466
Bazzoffi et al. 1998	(MSW) Compost was produced through a pile aerobic maturation process lasting 2 months, starting from urban refuse biomass that was ground after removal of plastics and metals by mechanical sieving and magnetic separators. The composition of the compost was dominated by non-metallic inerts, especially glass and shell fragments	Italy	9.1	248	28	626	540	Single value	0.031	0.053	0.041	0.073	0.248	0.446
Hyun et al. 1998	(SS) The SS compost was obtained from the Joint Water Pollution Control Plant, in one batch, then stored indoors in air-dried conditions	USA	61	475	250	1,100	3,500	Single value	0.132	0.078	0.260	0.118	0.973	1.561
Pascual et al. 1998	(MSW+SS) Compost made by a mixture (ratio, 1:1 in organic matter) of MSW and SS	Spain	3.0	158	221	198	535	Single value	0.017	0.042	0.230	0.031	0.246	0.566
Baldwin and Shelton 1999	(MSW+SS) The co-compost of MSW and SS was produced by an aerobic, in-vessel process	USA	2.9	215	40	203	738	Single value	0.017	0.049	0.052	0.032	0.305	0.455
	(MSW) The compost of MSW was produced in windrows		1.0	53	18	34	96	Single value	0.012	0.027	0.033	0.014	0.099	0.185
	(SS) The SS compost was produced from centrifuged, dewatered SS mixed with wood chips and straw in a ratio of 1:5:1		2.1	173	16	88	499	Single value	0.015	0.044	0.031	0.020	0.235	0.345
Hackett et al. 1999	(SS FA) Combined primary and secondary sludge and power boiler FA from the mill and mixed to yield a 50:50 (*v/v*) mixture of sludge and ash. The pile was left to compost in a static windrow. The compost was produced on an old landfill site with a functional leachate collection system to ensure that all leachate produced was treated at the mill's wastewater treatment plant. This site was wind	Canada	0.06	34.8	17.7	5.5	64.5	Single value	0.009	0.024	0.032	0.011	0.086	0.162

	exposed, requiring spraying of water on the compost pile during the summer months for dust control and to maintain optimal moisture (50%)													
Wong et al. 1999	(Manure) The manure compost originated from livestock wastes mixed with sawdust followed by a composting period of 60 days	China	1.65	143	–	26.1	475	Mean	0.013	0.039	–	0.013	0.228	0.293
García-Gil et al. 2000	(MSW) MSW compost was obtained from the Valdemingomez Municipal Waste Treatment Plant in Madrid	Spain	<0.2	548	81	681	1,325	Single value	0.009	0.085	0.090	0.078	0.463	0.725
Soliva and Paulet 2001	(SS) Compost obtained from a mixture of SS and GW	Spain	0.4	171	123	16	493	Single value	0.010	0.043	0.130	0.012	0.233	0.428
	(SS) Compost obtained from a mixture of SS and GW		1.5	338	54	110	1,087	Single value	0.013	0.063	0.065	0.022	0.401	0.564
	(SS) Compost obtained from a mixture of SS and GW		1.2	237	26	86	644	Single value	0.012	0.051	0.040	0.020	0.278	0.401
	(SS) Compost obtained from a mixture of SS and GW		0.48	55	33	59	260	Single value	0.010	0.027	0.046	0.017	0.159	0.259
	(SS) Compost obtained from a mixture of SS and GW		5.66	220	62	462	2,886	Single value	0.023	0.049	0.072	0.057	0.836	1.037
	(GW) Compost obtained from GW treatment		0.4	62	13	46	201	Single value	0.010	0.028	0.028	0.016	0.138	0.220
	(GW) Compost obtained from GW treatment		0.1	66	89	39	101	Single value	0.009	0.029	0.097	0.015	0.101	0.251
	(GW) Compost obtained from GW treatment		5.14	97	36	52	1,459	Single value	0.022	0.034	0.048	0.016	0.497	0.617
	(GW) Compost obtained from GW treatment		0.17	42	47	38	76	Single value	0.009	0.025	0.058	0.015	0.091	0.198
	(MSW) Compost obtained from MSW. Selection of organic fraction with GW		0.3	325	82.0	97	197	Single value	0.010	0.062	0.091	0.021	0.137	0.321
	(MSW) Compost obtained from MSW. Selection of organic fraction and GW		0.3	100	81.0	66	247	Single value	0.010	0.034	0.090	0.018	0.154	0.306
	(MSW) Compost obtained from MSW. Organic fraction mechanically separated		0.9	271	192	118	396	Single value	0.011	0.055	0.199	0.023	0.203	0.491
	(MSW) Compost obtained from MSW. Organic fraction mechanically separated		1.35	399	101	324	1,462	Single value	0.013	0.070	0.109	0.044	0.498	0.734
	(MSW) Compost obtained from MSW. Organic fraction mechanically separated		1.06	342	94.0	97	732	Single value	0.012	0.063	0.102	0.021	0.304	0.502
	(MSW) Compost obtained from MSW. Selection of organic fraction and GW from gardens and parks of Barcelona		0.4	42	27.0	38	192	Single value	0.010	0.025	0.041	0.015	0.135	0.226
Greenway and Song 2002	(MSW) The municipal composting site was used for GW (grass and leaves) compost obtained from an open-air windrow-composting system. It was used for composting	UK	1.5	50.2	15	117.2	220.4	Mean	0.013	0.027	0.030	0.023	0.145	0.238
	(MSW) The municipal composting site was used for composting GW mixed with sewage sludge. The compost was obtained from an open-air windrow-composting system		3.2	140.3	16.5	133.5	354.6	Mean	0.017	0.039	0.031	0.025	0.190	0.302
	(MSW) The municipal composting site was used for compost from farmer's vegetable waste. The compost was obtained from an open-air windrow-composting system		0.2	10.8	5.8	13.7	25.9	Mean	0.009	0.020	0.022	0.012	0.070	0.133
	(MSW) The municipal composting site was used for composting of mainly green (woody) waste. The compost was obtained from an open-air windrow-composting system		0.18	10.7	5.7	17.3	35.8	Mean	0.009	0.020	0.022	0.012	0.074	0.137

Table 1 (continued)

Compost source	Origin and feedstock materials	Country	Heavy metal content (mg/kg)					Data reported	HQ					HI
			Cd	Cu	Ni	Pb	Zn		Cd	Cu	Ni	Pb	Zn	
Kaschl et al. 2002	(MSW) MSW compost was obtained from a commercial composting plant. The duration of composting was 100 days	Israel	4.2	756	134	337	743	Single value	0.020	0.106	0.141	0.045	0.307	0.619
Korboulewsky et al. 2002	(SS+B+GW) The SS, a by-product of municipal wastewater treatment, was mixed with pine bark and GW. The mixture was composted for 30 days at 75°C to kill pathogenic microorganisms and decompose phytotoxic substances, then sieved to remove large bark pieces and stored in swathes. The swathes were turned (mixed) several times over 6 months to promote organic matter humification	France	0.8	101	12	34.0	221	Mean	0.011	0.034	0.028	0.014	0.145	0.232
Millares et al. 2002	(SS) Compost obtained from SS of five wastewater treatment plants of Madrid. The compost was subject to aerobic composting for 3 months, with periodic dump, without structuring agent	Spain	5	332	64	371	2,857	Single value	0.022	0.062	0.074	0.048	0.830	1.036
Soumaré et al. 2002	(MSW) Farm compost	Mali	<dl	10.3	6.5	3.4	110	Mean	–	0.020	0.023	0.011	0.104	0.158
	(MSW) Compost from an industrial composter	Belgium	<dl	31	13	80	470	Mean	–	0.023	0.028	0.019	0.226	0.296
Manios et al. 2003	(SS) The SS compost was produced by Thames Water Plc using a Windrow system with SS and straw on a 1:1 basis by volume (*v*/*v*)	Greece	1.5	525	68	189	825	Single value	0.013	0.083	0.078	0.030	0.330	0.534
Millares et al. 2003	(SS) The compost was obtained from SS of five wastewater treatment plants of Madrid	Spain	<3	330	67	140	1,390	Single value	0.017	0.062	0.077	0.025	0.480	0.661
Sebastiaò and Queda 2003	(MSW) The compost was obtained by bio-oxidation process of organic matter, over 60 days, in a locked ward, in trapezoidal aerated piles, with stirring and correction moisture	Portugal	2.4	293	–	247	448	Mean	0.015	0.058	–	0.036	0.220	0.329
Goi et al. 2006	(MSW) Compost originated from the wet fraction of two different MSW and was collected from bags that were to be sold for agricultural purposes. The compost was selected from waste mixtures with poor characteristics	Italy	<2.0	49.9	25.0	127.4	126.8	Mean	0.014	0.027	0.039	0.024	0.111	0.215
	(MSW) Compost originated from the wet fraction of two different MSW and was collected from bags that were to be sold for agricultural purposes. The compost was chosen from a high quality compost product certified by the producer		<2.0	74.2	21.0	92.6	198.4	Mean	0.014	0.030	0.035	0.020	0.137	0.236
Larchevêque et al. 2006	(SS+GW) This compost was elaborated with GW (1/3 volume), pine barks (1/3 volume), and local municipal SS (1/3 volume). The mixture was composted for 30 days at 75°C to kill pathogenic microorganisms and decompose phytotoxic substances, and then sieved to remove large bark pieces and stored in swathes. The swathes were mixed several times in 6 months to promote organic matter humification	France	0.77	122	14.7	65	266	Mean	0.011	0.037	0.030	0.018	0.161	0.257

Ramos 2006	(Manure) Composted cattle manure	Spain	0.8	35	–	9.8	142	Mean	0.011	0.024	–	0.012	0.117	0.164
Walter et al. 2006	(SS) The composted sludge was obtained from an anaerobically digested sludge mixed with pine bark at an initial sludge/wood ratio of 1:1.5 *v/v*. Composting was performed in the open air at a private facility, turning the piles periodically twice during the first month and then monthly until the end of the process. The final solid content was approximately 65–67%	Spain	3.5	220	42.5	179	820	Mean	0.018	0.049	0.054	0.029	0.328	0.478
Zheljazkov et al. 2006	MSW+SS	Canada	–	114	–	75.0	280	Single value	–	0.036	–	0.019	0.165	0.220
Casado-Vela et al. 2007	(SS) Aerobically composted SS from a wastewater treatment facility was used. It was composted in the plant using a three-step process involving: firstly, air drying of sewage sludge and addition of sawdust; secondly, turning of the feedstock every 7 days to promote aeration; and finally, mechanical mixing of the feedstock and collection after 3 months of stabilization	Spain	1.6	157	–	40.8	470	Single value	0.013	0.042	–	0.015	0.226	0.296
Madrid et al. 2007	(MSW) Compost obtained from the MSW treatment plant of Villarrasa (SW Spain)	Spain	–	128	23	98	261	Mean	–	0.038	0.037	0.021	0.159	0.255
	(MSW) Compost was obtained from the MSW treatment plant of Villarrasa (SW Spain)		–	312	54	172	494	Mean	–	0.060	0.065	0.028	0.234	0.387
	(MSW) Compost obtained from the MSW treatment plant of Villarrasa (SW Spain)		–	244	39	203	512	Mean	–	0.052	0.051	0.032	0.239	0.374
Paradelo Núñez et al. 2007	(MSW) MSW compost obtained by anaerobic fermentation of the biodegradable fraction of MSW, separated before collection, followed by an aerobic composting step	Spain	3.5	325	57	188	608	Mean	0.018	0.062	0.067	0.030	0.268	0.445
	(MSW) Aerobic MSW compost obtained from the source separated organic fraction of MSW		3.1	829	75	223	1,149	Mean	0.017	0.114	0.084	0.034	0.417	0.666
	(MSW+GW) Commercial compost obtained from source separated MSW mixed with GW		2.1	52	25	62	200	Mean	0.015	0.027	0.039	0.017	0.138	0.236
	(SS+GW) Compost obtained from municipal garden trimmings mixed with SS		2.7	688	71	180	896	Mean	0.016	0.100	0.080	0.029	0.349	0.574
Rosal et al. 2007	(MSW) A compost pile, with 20 t, was periodically turned and moistened as necessary for 140 days to ensure biological stability. Compost obtained during first year of the experiment	Spain	3.0	276	50	165	415	Single value	0.017	0.056	0.061	0.028	0.209	0.371
	(MSW) A compost pile, with 20 t, was periodically turned and moistened as necessary for 140 days to ensure biological stability. Compost obtained during second year of the experiment		3.0	252	57	120	579	Single value	0.017	0.053	0.067	0.023	0.259	0.419
	(MSW) A compost pile, with 20 t, was periodically turned and moistened as necessary for 140 days to ensure biological stability. Compost obtained during third year of the experiment		2.0	373	64	144	603	Single value	0.014	0.067	0.074	0.026	0.266	0.447
Sager 2007	GW	Austria	0.43	100	25.7	43.4	267	Median	0.010	0.034	0.039	0.015	0.161	0.259
Weber et al. 2007	(MSW) Commercial compost from Katowice produced by the MUT-DANO system represents MSWs originating from a highly industrialized region	Poland	11.7	366	168	972	1,825	Single value	0.037	0.066	0.175	0.106	0.588	0.972

Table 1 (continued)

Compost source	Origin and feedstock materials	Country	Heavy metal content (mg/kg)					Data reported	HQ					HI
			Cd	Cu	Ni	Pb	Zn		Cd	Cu	Ni	Pb	Zn	
	(MSW) Commercial compost from Zywiec produced by the HERHOFF system, utilized selectively collected MSWs rich in organic carbon		3.3	34	41	65.0	228	Single value	0.018	0.024	0.053	0.018	0.148	0.261
Alvarenga et al. 2008	(MSW) Compost from the organic fraction of unsorted MSW, obtained in a composting plant near Setúbal (Portugal)	Portugal	4.3	357	56	269	583	Mean	0.020	0.065	0.067	0.038	0.260	0.450
	(GW) Garden waste compost from a composting plant in Tavira (Portugal), which receives source separated garden residues (namely grass clippings, leaves and brush), were used		1.4	14	16	34	35	Mean	0.013	0.020	0.031	0.014	0.074	0.152
Jordan et al. 2008	SM	Ireland	6.2	54	5.8	10.4	143	Mean (63 samples of SM)	0.025	0.027	0.022	0.012	0.117	0.203
Ko et al. 2008	(Manure) Compost consisted of sawdust as the bulking agent and animal manures at 10:90 *v/v* ratios. Animal manures were composed of 50% dairy manure (collected on an open feedlot using a wheel loader), 30% beef manures (collected in a sawdust bed barn using a wheel loader) and 20% swine manure (collected at a mechanical manure separator) collected from an integrated livestock experimental building	Korea	1.1	466	11	38.2	566	Mean	0.012	0.077	0.027	0.015	0.255	0.386
Lakhdar et al. 2008	(MSW) The compost was mechanically produced by mixing weekly the waste heap under aerobic conditions by fast fermentation	Tunisia	3.37	91.63	–	251.63	290.19	Mean	0.018	0.033	–	0.036	0.169	0.256
Mbarki et al. 2008	MSW	Tunisia	2.56	278	–	668	649	Single value	0.016	0.056	–	0.077	0.280	0.429
Oleszczuk 2008	(SS) SS was composted during 76 days. Ventilation was provided through air distribution tubes. In order to increase oxygen inflow, the composted material was additionally mixed once a fortnight	Poland	2.25	192	22	52.5	1,490	Mean	0.015	0.046	0.036	0.016	0.505	0.618
	(SS) SS was composted during 76 days. Ventilation was provided through air distribution tubes. In order to increase oxygen inflow, the composted material was additionally mixed once a fortnight.		76	236	177.5	37.5	1,270	Mean	0.160	0.051	0.185	0.015	0.449	0.860
	(SS) SS was composted during 76 days. Ventilation was provided through air distribution tubes. In order to increase oxygen inflow, the composted material was additionally mixed once a fortnight		1.95	314	17.7	35.2	1,125	Mean	0.014	0.060	0.032	0.014	0.411	0.531
	(SS) SS was composted during 76 days. Ventilation was provided through air distribution tubes. In order to increase oxygen inflow, the composted material was additionally mixed once a fortnight		2.75	155	58	37.8	935	Mean	0.016	0.041	0.068	0.015	0.360	0.500
Pengcheng et al. 2008	SS+GW	China	3.72	156	–	61.9	1,105	Single value	0.019	0.041	–	0.017	0.406	0.483
Zubillaga et al. 2008	MSW	Argentina	<4.0	727	109	383	1,183	Single value	0.019	0.104	0.117	0.049	0.426	0.715
Achiba et al. 2009	(MSW) The MSW was prepared from a mixture of the separated and shredded organic fraction of house-	Tunisia	3.3	278	44	325	410	Mean	0.018	0.056	0.056	0.044	0.208	0.382

	hold rubbish and garden waste by aerobic fermentation													
Businelli et al. 2009	MSW	Italy	5.0	240	52	750	647	Mean	0.022	0.052	0.063	0.085	0.279	0.501
Cherif et al. 2009	(MSW) MSW compost obtained from sorted MSW by aerobic composting process for 120 days	Tunisia	2.3	337	90.8	80.1	290	Mean (the values reported are the means of four replicates)	0.015	0.063	0.099	0.019	0.169	0.365
Farrell and Jones 2009b	(MSW) MSW compost was produced in the EcoPOD® experiment	UK	0.69	261	46	614	249	Mean	0.011	0.054	0.057	0.072	0.155	0.349
	(MSW+GW) MSW compost was produced in the EcoPOD® experiment		0.49	276	37	232	213	Mean	0.010	0.056	0.049	0.034	0.142	0.291
	(GW) GW compost derived from source separated municipal GW waste was obtained from Flintshire County Council's open windrow-composting facility at Greenfields, Flintshire, UK		1.30	63	20	198	369	Mean	0.013	0.029	0.034	0.031	0.195	0.302
Haroun et al. 2009	(TSS) The sludge (100 kg) was mixed with sawdust (50 kg), chicken manure (30 kg), beneficial organisms (1 l) and rice bran (20 kg) in a pile on a composting windrow type. With the aim of maintaining aerobic conditions during the process, the pile was turned manually every 10 days. The mature compost was obtained at the end of 60 days of composting	Malaysia	1.6	54.0		2.2	148	Single value	0.013	0.027	–	0.011	0.119	0.170
Qazi et al. 2009	(MSW) The compost was originated from recycled mixed MSW. Windrow composting is applied to generate the compost	Pakistan	34	480	49	73	1,622	Single value	0.082	0.078	0.060	0.018	0.538	0.776
Roca-Pérez et al. 2009	(SS+GW) The compost included SS and rice straw and the composting during 90 days	Spain	1.2	170	36	94	700	Mean	0.012	0.043	0.048	0.021	0.295	0.419
Tejada et al. 2009	(GW) The vermicompost was obtained using green forages (constituted basically by grasses, green vegetable leaves, herbs and plant materials) as substrate	Spain	<0.1	1.4	<0.1	<0.1	3.2	Mean (data are the means of five samples)	0.009	0.018	0.018	0.011	0.059	0.115
	(GW+BV) The compost was obtained by the co-composting of the beet vinasse and the vermicompost at a 1:1 rate (weight/weight)		<0.1	2.5	<0.1	<0.1	12.8	Mean (data are the means of five samples)	0.009	0.018	0.018	0.011	0.064	0.120
Mean			4.4	222.7	55.0	181.3	644.0		0.019	0.048	0.067	0.029	0.266	0.420
Min			0.06	1.4	0.1	0.1	3.2		0.009	0.018	0.018	0.011	0.059	0.115
Max			76	829	250	1,100	3,500		0.160	0.114	0.260	0.118	0.973	1.561

MSW municipal solid waste, *SS* sewage sludge, *GW* green waste, *FA* fly ash, *B* bark, *SM* spent mushroom, *TSS* tannery sewage sludge, *BV* beet vinasse

Table 2 Metal content inventory, metal hazard quotient (HQ), and hazard index (HI) of sludge and other wastes

Sludge source	Origin and feedstock materials	Country	Heavy metal content (mg/kg)					Data reported	HQ					HI
			Cd	Cu	Ni	Pb	Zn		Cd	Cu	Ni	Pb	Zn	
Moreno et al. 1997	(MSW+SS) The SS base originated from an aerobic sewage treatment plant receiving municipal and food industry effluents. In this treatment plant, sewage is submitted to a biological-type depuration process		2.0	275	105	–	776	Single value	0.014	0.056	0.113	–	0.316	0.499
Pascual et al. 1998	(SS) The SS was obtained from an aerobic-treatment	Spain	6.0	151	228	85	415	Single value	0.024	0.041	0.237	0.020	0.209	0.531
	(MSW) Organic fraction of MSW		2.0	77	178	77	281	Single value	0.014	0.031	0.185	0.019	0.166	0.415
Fang and Wong 1999	(SS) Dewatered anaerobically digested SS was collected from the Tai Po sewage treatment plant	China	–	785	72.5	–	2,786	Mean (the values reported are the means of triplicates)	–	0.109	0.082	–	0.813	1.004
Saviozzi et al. 1999	SS	Italy	4.0	236	40	60	1,640	Mean (the values reported are the means of triplicates)	0.019	0.051	0.052	0.017	0.542	0.681
López Fernández et al. 2000	(SS) SS obtained from wastewater treatment plant of Burgos	Spain	4.84	148.27	46.91	158.52	1,023.37	Single value	0.022	0.040	0.058	0.027	0.384	0.531
	(MSW) Urban wastes obtained from municipal landfill of Burgos		5.48	251.80	87.81	626.56	716.65	Single value	0.023	0.053	0.096	0.073	0.299	0.544
Cole et al. 2001	(SS) SS were derived from uncontaminated sludge	UK	1.94	722	45	161	725	Mean	0.014	0.103	0.057	0.027	0.302	0.503
	(SS) SS were derived from Zn-rich sludge		17.2	1,438	629	1,075	6,619	Mean	0.049	0.171	0.691	0.115	1.630	2.656
	(SS) SS derived from Cd-rich sludge		48.9	617	188	494	1,244	Mean	0.110	0.093	0.195	0.060	0.442	0.900
Illera et al. 2001	(SS) The SS was obtained from wastewater treatment plant of Madrid, mainly urban origin. It was obtained from anaerobic digestion	Spain	0.6	174	15.3	252	445	Single value	0.011	0.044	0.030	0.036	0.219	0.340
	(MSW) The MSW was obtained from waste treatment plant of Valdemingómez (Madrid) and correspond to organic fraction composted of domestic wastes		1.5	203	21.6	191	335	Single value	0.013	0.047	0.036	0.030	0.184	0.310
Soliva and Paulet 2001	IS	Spain	0.20	166	59	15	521	Single value	0.009	0.043	0.069	0.012	0.242	0.375
	IS		0.30	110	6	16	683	Single value	0.010	0.035	0.023	0.012	0.290	0.370
	IS		0.50	49	63	15	87	Single value	0.010	0.026	0.073	0.012	0.095	0.216
	IS		2.5	1,140	38	30	2,993	Single value	0.016	0.143	0.050	0.014	0.860	1.083
	(MSW) Organic fraction of MSW		2.0	156	53	190	569	Single value	0.014	0.041	0.064	0.030	0.256	0.405
	(MSW) Organic fraction of MSW		0.12	14	16	6	43	Single value	0.009	0.020	0.031	0.011	0.077	0.148
Millares et al. 2002	(SS) Fresh SS obtained from wastewater treatment plant of Viveros	Spain	1.0	197	15	197	577	Single value	0.012	0.047	0.030	0.031	0.259	0.379

Acosta et al. 2003	(SS) SS obtained from wastewater treatment plant of Punta Cardón	Venezuela	3.7	206.6	28.1	253	878.6	Mean	0.019	0.048	0.042	0.037	0.345	0.491
Chicón Reina 2003	(SS) SS obtained from urban wastewater treatment plant	Spain	3.3	250	125	365.7	864.9	Single value	0.018	0.053	0.132	0.048	0.341	0.592
Manios et al. 2003	SS	UK	1.2	599	99	191	728	Single value	0.012	0.091	0.107	0.030	0.303	0.543
Millares et al. 2003	(SS) Mixture of SS obtained from 5 wastewater treatment plants of Madrid	Spain	1.2	339	70	64	1,650	Single value	0.012	0.063	0.079	0.017	0.545	0.716
Fuentes et al. 2004	(SS) SS came from wastewater treatment plant in the Region of Murcia. SS was obtained from aerobic digestion	Spain	1.10	204	17	58	487	Mean	0.012	0.047	0.032	0.017	0.232	0.340
	(SS) SS came from wastewater treatment plant in the Region of Murcia. SS was obtained anaerobically		18.3	337	29	167	871	Mean	0.051	0.063	0.042	0.028	0.343	0.527
	(SS) SS came from wastewater treatment plant in the Region of Murcia. It was stabilized in a waste stabilization pond		11.4	167	15	250	697	Mean	0.036	0.043	0.030	0.036	0.294	0.439
	(SS) SS came from wastewater treatment plant in the Region of Murcia. Non-stabilized SS		1.14	146	25	87	458	Mean	0.012	0.040	0.039	0.020	0.223	0.334
Kandpal et al. 2004	(SS) Bulk sample of SS was collected in plastic bags from Karula drain of Moradabad, UP, India, a city having brass plating and policing industrial units. The sample was processed to remove the non-recyclable materials	India	16	1,434.50	168	340.5	2,164	Mean (the values reported are the means of triplicate samples)	0.046	0.171	0.175	0.045	0.669	1.106
Ahlberg et al. 2006	(SS) SS was collected directly from Ryaverken, the sewage works of Gothenburg, Sweden. The sludge produced is digested anaerobically and had 29.2% (by weight) dry solids (DS) content. The organic content of DS was 54%	Sweden	1.64	501.9	24.7	43.79	748.7	Mean	0.013	0.081	0.039	0.015	0.308	0.456
García et al. 2006	(SS) SS obtained from closed digestion	Venezuela	6.8	226.01	76.46	304.29	1,474.79	Mean	0.026	0.050	0.086	0.042	0.501	0.705
Goi et al. 2006	(SS) Sludge sample is representative of 1 month of sludge production and come from MWW treatment plants treating mainly domestic wastewaters	Italy	<2.0	20.1	11.0	13.4	152.8	Mean	0.014	0.022	0.027	0.012	0.121	0.196
	(SS) Sludge sample is representative of 1 month of sludge production and come from MWW treatment plants treating mainly domestic wastewaters		<2.0	69.5	4.3	58.7	410.1	Mean	0.014	0.030	0.021	0.017	0.208	0.290
	(SS) Sludge sample is representative of 1 month of sludge production and come from MWW treatment plants treating mainly domestic wastewaters		<2.0	71.7	16.2	27.0	355.1	Mean	0.014	0.030	0.031	0.014	0.190	0.279
	(SS) Sludge sample is representative of		<2.0	73.5	12.5	27.0	254.6	Mean	0.014	0.030	0.028	0.014	0.157	0.243

Table 2 (continued)

Sludge source	Origin and feedstock materials	Country	Heavy metal content (mg/kg)					Data reported	HQ					HI
			Cd	Cu	Ni	Pb	Zn		Cd	Cu	Ni	Pb	Zn	
	1 month of sludge production and come from MWW treatment plants treating mainly domestic wastewaters													
	(SS) Sludge sample is representative of 1 month of sludge production and come from MWW treatment plants treating mainly domestic wastewaters		<2.0	55.6	10.4	18.9	195.8	Mean	0.014	0.027	0.026	0.013	0.136	0.216
	(SS) Sludge sample is representative of 1 month of sludge production and come from MWW treatment plants treating mainly urban wastewaters		<2.0	105.8	26.2	48.4	404.1	Mean	0.014	0.035	0.040	0.016	0.206	0.311
	(SS) Sludge sample is representative of 1 month of sludge production and come from MWW treatment plants treating mainly urban wastewaters		<2.0	12.5	24.5	3.7	30.4	Mean	0.014	0.020	0.038	0.011	0.072	0.155
	(SS) Sludge sample is representative of 1 month of sludge production and come from MWW treatment plants treating mainly urban wastewaters		<2.0	20.2	35.9	17.3	134.1	Mean	0.014	0.022	0.048	0.012	0.114	0.210
	(SS) Sludge sample is representative of 1 month of sludge production and come from MWW treatment plants treating mainly urban wastewaters		3.6	61.4	21.4	17.0	275.0	Mean	0.018	0.028	0.036	0.012	0.164	0.258
	(SS) Sludge sample is representative of 1 month of sludge production and come from MWW treatment plants treating mainly urban wastewaters		2.8	50.8	19.8	16.4	236.8	Mean	0.016	0.027	0.034	0.012	0.151	0.240
Walter et al. 2006	(SS) Anaerobilcally digested sludge produced at a wastewater treatment facility in Madrid, Spain	Spain	2.5	202	20.5	164	497	Mean	0.016	0.047	0.035	0.028	0.235	0.361
	(SS) Heat-dried sludge produced from a mixture of anaerobic SS produced by the 7 municipal wastewater treatment facilities in Madrid		2.7	242	37.5	197.2	689	Mean	0.016	0.052	0.050	0.031	0.291	0.440
Cai et al. 2007	(SS) Secondary dewatered sludge was taken from Datansha wastewater treatment plant in Guangzhou city	China	0.54	396	–	57	1,213	Single value	0.010	0.069	–	0.017	0.434	0.530
	(SS) Secondary dewatered sludge was taken from Zhen'an wastewater treatment plant in Foshan city		1.74	357	–	134	1,190	Single value	0.014	0.065	–	0.025	0.428	0.532
Fuentes et al. 2007	(SS) Anaerobically digested SS from a domestic wastewater treatment plant (Pinedo I, located at the city of Valencia)	Spain	3.3	406	47	182	1,036	Single value	0.018	0.071	0.058	0.029	0.387	0.563
Kidd et al. 2007	(SS) Digested SS	Spain	<5	230	35.0	69.0	500.0	Single value	0.022	0.051	0.048	0.018	0.236	0.375

Sager 2007	SS	Austria	0.82	166	25.6	38.3	683	Median	0.011	0.043	0.039	0.015	0.290	0.398
Salcedo-Pérez et al. 2007	(SS) SS collected from a wastewater treatment plant of electronics manufacturing company of the central region of Jalisco, México	México	1.08	383.4	9.69	117.22	539.9	Single value	0.012	0.068	0.026	0.023	0.248	0.377
Bose and Bhattacharyya 2008	(IS) Roadside sludge collected from pickling–rolling and electroplating industrial area	India	30.16	1,290	1,807	440	410	Mean	0.074	0.157	2.240	0.055	0.208	2.734
Chen et al. 2008	SS	China	7.2	111	–	152	424.8	Single value	0.027	0.036	–	0.026	0.212	0.301
	SS		10.7	130.4	–	53.6	450.9	Single value	0.035	0.038	–	0.016	0.220	0.309
	SS		15.7	159.6	–	71.8	444.6	Single value	0.045	0.042	–	0.018	0.219	0.324
	SS		7.9	67	–	98.4	361	Single value	0.029	0.029	–	0.021	0.192	0.271
	(IS+SS) The SS was collected from Qingshuitang area in Zhuzhou, where many chemical plants were centralized		903.8	659	–	1,270.2	1,105.9	Single value	1.536	0.097	–	0.134	0.406	2.173
Hua et al. 2008	(SS) The SS was collected from the wastewater treatment plant in Ningbo	China	10.86	311.0	25.6	58.9	1,652.4	Single value	0.035	0.060	0.039	0.017	0.546	0.697
	(SS) The SS was collected from the wastewater treatment plant in Fuyang		13.0	240.2	25.1	47.0	1,406.2	Single value	0.040	0.052	0.039	0.016	0.484	0.631
	(SS) The SS was collected from the wastewater treatment plant in Lin'an		23.4	227.7	38.9	123.1	2,445.3	Single value	0.061	0.050	0.051	0.024	0.735	0.921
	(SS) The SS was collected from the wastewater treatment plant in Shaoxing		13.3	452.3	54.2	72.8	2,231.3	Single value	0.040	0.075	0.065	0.018	0.685	0.883
	(SS) The SS was collected from the wastewater treatment plant in Huzhou		2.1	220.1	42.7	93.7	1,521.4	Single value	0.015	0.049	0.054	0.021	0.513	0.652
	(SS) The SS was collected from the wastewater treatment plant in JH		8.0	382.2	67.7	123.3	2,037.9	Single value	0.029	0.068	0.077	0.024	0.639	0.837
	(SS) The SS was collected from the wastewater treatment plant in Lishui		3.7	1,191.3	31.1	41.2	3,066.7	Single value	0.019	0.148	0.044	0.015	0.877	1.103
	(SS) The SS was collected from the wastewater treatment plant in XS		16.8	861.5	106.6	162.7	2,678.6	Single value	0.048	0.117	0.114	0.028	0.789	1.096
	(SS) The SS was collected from the wastewater treatment plant in Qige		19.4	266.2	102.3	195.1	2,431.6	Single value	0.053	0.055	0.110	0.031	0.732	0.981
	(SS) The SS was collected from the wastewater treatment plant in Sibao		9.0	210.6	28.5	260.8	2,008.5	Single value	0.031	0.048	0.042	0.037	0.632	0.790
	(SS) The SS was collected from the wastewater treatment plant in JJ		4.9	393.1	90.1	327.2	1,950.9	Single value	0.022	0.069	0.098	0.044	0.618	0.851
	(SS) The SS was collected from the wastewater treatment plant in Huangyan		2.9	753.7	77.4	452.2	3,699.2	Single value	0.017	0.106	0.086	0.056	1.020	1.285
Oleszczuk 2008	(SS) Dewatered SS were collected from wastewater treatment plant	Poland	1.9	201	21.7	59.5	1,385	Mean	0.014	0.047	0.036	0.017	0.478	0.592
	(SS) Dewatered SS were collected from wastewater treatment plant		76	214	155	39.3	1,220	Mean	0.160	0.049	0.162	0.015	0.436	0.822
	(SS) Dewatered SS were collected from wastewater treatment plant		1.95	335	43.4	37.9	1,220	Mean	0.014	0.063	0.055	0.015	0.436	0.583

Table 2 (continued)

Sludge source	Origin and feedstock materials	Country	Heavy metal content (mg/kg)					Data reported	HQ					HI
			Cd	Cu	Ni	Pb	Zn		Cd	Cu	Ni	Pb	Zn	
	(SS) Dewatered SS were collected from wastewater treatment plant		2.8	156	22.3	46.8	1,015	Mean	0.016	0.041	0.036	0.016	0.382	0.491
Stylianou et al. 2008	(SS) SS samples were collected from wastewater treatment plant in Psittalia and stored at 4°C	Greece	–	429	149	7.8	851	Mean (the values reported are the means of triplicates)	–	0.073	0.156	0.011	0.337	0.577
Zorpas et al. 2008	(SS) Dewatered anaerobically stabilized primary SS, as result of primary treatment of municipal wastewater along with industrial wastes	Greece	2.0	258	41	326.0	1,739	Single value	0.014	0.054	0.053	0.044	0.567	0.732
Egiarte et al. 2009	(SS) The anaerobic SS was obtained from the Durango wastewater treatment plant	Spain	5.7	456	208	151	10,924	Single value	0.024	0.076	0.216	0.026	2.470	2.812
Haroun et al. 2009	(TS) The TS was collected from Kenny Leather Sdn Bhd (Melaka, Malaysia)	Malaysia	8.0	80	–	10.0	200	Single value	0.029	0.031	–	0.012	0.138	0.210
	(GW) Rice bran waste		0.2	24.33	–	1.2	127	Single value	0.009	0.022	–	0.011	0.111	0.153
Lasheen and Ammar 2009	IS+SS	Egypt	3.02	197.70	39	–	1,770.34	Mean (the values reported are the means of triplicates)	0.017	0.047	0.051	–	0.575	0.690
	IS+SS		2.56	311.23	55.80	–	515.40	Mean (the values reported are the means of triplicates)	0.016	0.060	0.066	–	0.240	0.382
	IS		3.42	1,391.42	291.53	–	3,237.52	Mean (the values reported are the means of triplicates)	0.018	0.167	0.305	–	0.915	1.405
	IS+SS		3.56	200.20	56.30	–	1,181.62	Mean (the values reported are the means of triplicates)	0.018	0.047	0.067	–	0.426	0.558
	IS+SS		2.16	184.88	36.79	–	684.95	Mean (the values reported are the means of triplicates)	0.015	0.045	0.049	–	0.290	0.399
Roca-Pérez et al. 2009	(SS) Dewatered digested SS was collected from the Metropolitan sewage industry (EMARSA)	Spain	2.55	230	53	50	1,100	Mean (the values reported are the means of triplicates)	0.016	0.051	0.064	0.016	0.404	0.551
Mean			18.0	331.4	91.8	158.8	1,232.0		0.044	0.060	0.110	0.027	0.416	0.641
Min			0.12	12.5	4.3	1.2	30.4		0.009	0.020	0.021	0.011	0.072	0.148
Max			903.8	1,438	1,807	1,270.2	10,924		1.536	0.171	2.240	0.134	2.470	2.812

SS sewage sludge; *IS* industrial sludge; *TS* tannery sludge; *MWW* municipal wastewater; *MSW* municipal solid waste; *GW* green waste

Table 3 Metal content inventory, metal Hazard Quotient (HQ) and hazard index (HI) of manure

Manure Source	Origin and feedstock materials	Country	Heavy metal content (mg/kg)					Data reported	HQ					HI
			Cd	Cu	Ni	Pb	Zn		Cd	Cu	Ni	Pb	Zn	
Ayuso et al. 1996	(Sheep) Manure (fresh organic material) from sheep kept indoors	Spain	ND	14	37	18	94	Single value	–	0.020	0.049	0.013	0.098	0.180
Ihnat and Fernandes 1996	(Poultry) The materials used were from a poultry manure aeration composting study conducted with poultry manure slurry	Canada	0.48	54.3	7	2.3	550	Mean (2 samples were analyzed)	0.010	0.027	0.023	0.011	0.251	0.322
Pinamonti et al. 1997	(Cattle) Uncomposted cattle manure produced by dairy-cows in sheds with straw bedding	Italy	0.7	56	12	31	253	Mean	0.011	0.028	0.028	0.014	0.156	0.237
Nicholson et al. 1999	Dairy cattle farmyard	UK	0.38	37.5	3.7	3.61	153	Mean (6 samples were collected)	0.010	0.025	0.021	0.011	0.121	0.188
	Dairy cattle slurry		0.33	62.3	5.4	5.87	209	Mean (20 samples were collected)	0.010	0.028	0.022	0.011	0.141	0.212
	Beef cattle farmyard		0.13	16.4	2.0	1.95	81	Mean (12 samples were collected)	0.009	0.021	0.019	0.011	0.093	0.153
	Beef cattle slurry		0.26	33.2	6.4	7.07	133	Mean (8 samples were collected)	0.010	0.024	0.023	0.011	0.113	0.181
	Pig farmyard		0.37	374	7.5	2.94	431	Mean (7 samples were collected)	0.010	0.067	0.024	0.011	0.214	0.326
	Pig slurry		0.30	351	10.4	2.48	575	Mean (12 samples were collected)	0.010	0.064	0.026	0.011	0.258	0.369
	Turkey litter		0.42	96.8	5.4	3.62	378	Mean (12 samples were collected)	0.010	0.034	0.022	0.011	0.198	0.275
	Layer manure		1.06	64.8	7.1	8.37	459	Mean (8 samples were collected)	0.012	0.029	0.023	0.012	0.223	0.299
Saviozzi et al. 1999	Farmyard	Italy	6.0	66	14	60	340	Mean (the values reported are the means of triplicates)	0.024	0.029	0.029	0.017	0.185	0.284
García-Gil et al. 2000	Cow	Spain	<0.2	<3	3	<3	28	Single value	0.009	0.018	0.020	0.011	0.071	0.129
Soliva and Paulet 2001	Cow	Spain	0.24	59	46	8	219	Single value	0.009	0.028	0.057	0.011	0.144	0.249
Charest and Beauchamp 2002	(Poultry) Poultry manure came from a poultry farm near St-Henri-de-Lévis	Canada	<1	160	12	<20	550	Mean (chemical analyses were done in triplicate)	0.012	0.042	0.028	0.013	0.251	0.346
	(Broiler litter) Poultry broiler floor litter came from a poultry farm near St-Henri-de-Lévis		<1	47	<10	<20	280	Mean (chemical analyses were done in triplicate)	0.012	0.026	0.026	0.013	0.165	0.242
Acosta et al. 2003	(Goat) Goat manure collected from local breeding "El Taparo"	Venezuela	1.0	13	4.4	3.7	71	Mean	0.012	0.020	0.021	0.011	0.089	0.153
Zheljazkov et al. 2006	(Mixture) The solid manure represents a mixture of mostly cattle, sheep, and chicken manures, plus some mink and fox manure	Canada	–	8.3	–	–	91	Single value	–	0.019	–	–	0.097	0.116
Clemente et al. 2007	(Cow) Fresh cow manure was collected from a cattle farm in Santomera (Murcia)	Spain	<0.5	26	–	9	12	Single value	0.010	0.023	–	0.012	0.064	0.109
Sager 2007	Cattle	Austria	0.27	51	6.3	4.1	164	Median	0.010	0.027	0.023	0.011	0.125	0.196
	Pig		0.46	282	12.5	1.9	1,156	Median	0.010	0.057	0.028	0.011	0.419	0.525
	Biogas		0.56	94	14.1	7.7	349	Median	0.010	0.033	0.029	0.011	0.188	0.271
Salazar and Saldana 2007	(Trout) Trout manures collected from raceways	Chile	1.13	33.4	4.94	5.54	605	Mean	0.012	0.024	0.022	0.011	0.267	0.336
Odlare et al. 2008	Pig+mineral N	Sweden	0.3	140	4.0	1.0	631	Mean (the values represent mean values for 4 years)	0.010	0.039	0.021	0.011	0.275	0.356

Table 3 (continued)

Manure Source	Origin and feedstock materials	Country	Heavy metal content (mg/kg)					Data reported	HQ					HI
			Cd	Cu	Ni	Pb	Zn		Cd	Cu	Ni	Pb	Zn	
	Cow+mineral N		0.4	76	7.0	4.0	415	Mean (the values represent mean values for 4 years)	0.010	0.031	0.023	0.011	0.209	0.284
Tripathy et al. 2008	(Cow) Decomposed cow manure	South Korea	0.5	10	4	21	21	Single value	0.010	0.020	0.021	0.013	0.068	0.132
Achiba et al. 2009	(Cow) The manure was taken from the cow-shed of the experimental farm of the Agronomic National Institute of Tunisia	Tunisia	0.7	26	22	10.0	120	Mean	0.011	0.023	0.036	0.012	0.108	0.190
Cherif et al. 2009	Farmyard	Tunisia	2.10	25.50	22.40	8.90	117	Mean (the values reported are the means of determinations made on 4 replicates)	0.015	0.023	0.037	0.012	0.107	0.194
Hachicha et al. 2009	(Poultry) The poultry manure was collected from an industrialized farm in the city of Sfax (Tunisia)	Tunisia	<4	34	<88	<41	75	Mean	0.019	0.024	0.096	0.015	0.091	0.245
Haroun et al. 2009	Chicken	Malaysia	0.5	330	–	1.3	635	Single value	0.010	0.062	–	0.011	0.276	0.359
Mean			0.90	88.2	14.0	10.9	306.5		0.011	0.031	0.030	0.012	0.169	0.249
Min			0.13	3	2	1	12		0.009	0.018	0.019	0.011	0.064	0.109
Max			6	374	88	60	1,156		0.024	0.067	0.096	0.017	0.419	0.525

soil characteristics (e.g., pH, organic matter, and texture), climatic conditions (e.g., rainfall), and agricultural practices (e.g., intensity and frequency).

The accumulation of heavy metals in soil was assessed by establishing a dynamic mass balance between input and output fluxes according to Boekhold and van der Zee (1991) and Moolenaar et al. (1997). The input of metals to the agricultural soil surface may have several contributors: addition of organic waste (i.e., sewage sludge, manure, or compost), irrigation with wastewater, application of commercial fertilizers, or atmospheric deposition. Considering the scope of this work, only the application of organic waste was considered as an input to the model. Output fluxes from soil included leaching from plough to deeper soil layers by precipitation and plant uptake. Data corresponded to areas with different soil types/characteristics, climatology, and precipitation rates. Since metal concentration in solution is usually correlated with soil properties (e.g., pH, metal soil concentration, metal transfer by soil erosion, organic matter, cation exchange capacity, and fulvic and humic acid concentration) and climatology characteristics (e.g., precipitation rate), the leaching of heavy metals into groundwater may be more important in some areas than in others (Sauvé et al. 1997, 2000; Krishnamurti and Naidu 2002; Keller and Schulin 2003; Carlon et al. 2004). Plant absorption rate is related to metal concentration in solution and, therefore, is also dependent on soil type. With the aim of analyzing the effect of organic waste metal content on total risk regardless of soil location, the parameterization of the fate model (i.e., initial soil concentrations, waste application rates, and soil characteristics) was the same for all cases included in the inventory (Table 4). This criterion was also adopted due to the lack of data for these parameters in the majority (>60%) of studies.

Human exposure was estimated by taking into account five exposure pathways according to the scenario evaluated: (1) intake of meat from cattle grazing in the area, (2) ingestion of milk from cattle grazing in the area, (3) dermal absorption from soil, (4) ingestion of soil, and (5) inhalation of resuspended soil particles. Some of the exposure routes were selected based on the primary activities of the population inhabiting in the study area (e.g., farming). Minor contributions from pathways with a soil exposure source were also expected.

Cattle are exposed to metals through ingestion of contaminated food (i.e., soil, vegetation, and water), by inhalation of resuspended soil particles, or by absorption through the skin. However, only the ingestion pathways were considered to evaluate cattle exposure because dermal contact and inhalation are generally not as significant (ORNL 2004). The equations and empirical multicorrelation models used to estimate metal concentrations in solution (Sauvé et al. 2000), plants (Efroymson et al.

Table 4 Parameter values for the distribution model

Parameter	Units	Value
Application rate	$t{\cdot}ha^{-1}{\cdot}year^{-1}$	10
Cd (initial)in soil	$mg{\cdot}kg^{-1}$	1.0
Cu (initial)in soil	$mg{\cdot}kg^{-1}$	19.3
Ni (initial)in soil	$mg{\cdot}kg^{-1}$	11.1
Pb (initial) in soil	$mg{\cdot}kg^{-1}$	33.0
Zn (initial) in soil	$mg{\cdot}kg^{-1}$	42.4
Average pasture production	$kg{\cdot}ha^{-1}{\cdot}year^{-1}$	12,000
Soil pH	Unitless	5.49
Soil organic matter	% C	11.69
Precipitation	$m{\cdot}year^{-1}$	0.9
Infiltration factor	Unitless	0.44
Soil bulk density	$kg{\cdot}m^{-3}$	1,300
Depth plough layer	m	0.2
Time	year	100

Data references in Franco et al. (2006)

2001), and soil can be found in a previous work (Franco et al. 2006), as along with the exposure model equations and their parameterization.

Quantification of the potential non-carcinogenic risk was determined by a hazard quotient (HQ), which was calculated by dividing the individual doses (milligrams contaminant per kilogram of body weight per day) of each metal by the corresponding reference dose (RfD, milligrams contaminant per kilogram of body weight per day) as shown in Eq. 1.

$$HQ = \frac{\text{Individual dose}}{\text{RfD}} \tag{1}$$

Route-to-route extrapolations were needed when no specific dose–response data were available (IRIS database, US EPA 2010). A hazard index (HI) was obtained for each case in the inventory by aggregating the HQs corresponding to the different metals contained in each of the organic wastes considered, reflecting the global risk (Eq. 2).

$$HI = \sum HQ_{\text{metal}} \tag{2}$$

A HI higher than 1.0 indicates that adverse human health effects are expected to occur.

2.3 Sensitivity analysis

A Monte Carlo simulation of 10,000 iterations was developed using the commercial software, Crystal Ball, Version 7 (Decisioneering). This numerical technique propagates parameter uncertainty through the model equations. In this particular case, the sensitivity analysis was only performed on the fate model's parameters to evaluate the influence that different locations with different soil characteristics and climatology might have on both the HQ and HI. Probability distributions with a standard deviation of 50% around the nominal value were assigned to average production, soil organic matter, and soil infiltration (Table 4). A standard deviation of 100% was assigned to the precipitation rate to observe the effect of precipitation absence in arid locations. Finally, soil pH was allowed to vary between 5.0 and 7.5.

3 Results and discussion

3.1 Risk indexes

The data compiled on heavy metals content in compost, sludge, and manure are shown in Tables 1, 2, and 3 (inventory tables), respectively. It can be seen that sludge contained the highest values of average heavy metal concentration, 50–90% higher than in compost (depending on the metal) and considerably higher than in manure (almost 20 times higher for toxic metals like Cd or Pb). Sludge composition primarily depends on the origin of the effluent treated in the biological reactor. Metal concentrations of concern are typically found in sludge (or compost) coming from a wastewater treatment plant that collects industrial effluents (Soliva and Paulet 2001; Bose and Bhattacharyya 2008), although high concentrations can also be found in domestic sewage depending on the country of origin (Kandpal et al. 2004; Chen et al. 2008; Hua et al. 2008; Egiarte et al. 2009; Lasheen and Ammar 2009).

In general, our metal content values in sludge are within the ranges of those compiled in other works (Pathak et al. 2009). More specifically, average contents of Cu, Pb, and Zn in Table 2 agreed well with sludge values proposed by the EU, while mean values for Ni and Cd were in accordance with those reported by the USA (Stylianou et al. 2008). In Table 2, it should be highlighted that other uncomposted wastes like municipal solid waste or green waste were considered in addition to sludge. Although composting can effectively reduce the availability of metals (García et al. 1995; Smith 2009), it has proved difficult to significantly reduce the total metal content of the initial residue (Manios et al. 2003; Nomeda et al. 2008; Oleszczuk 2008). In fact, this content can be even higher in compost than in the initial waste for certain metals due to the weight loss suffered through mineralization (García et al. 1995). Intermediate metal levels between sludge and manure can be found in compost because composted waste can be either sludge or manure.

On the other hand, the presence of metals in manure is due to animal (e.g., cattle, pig, and poultry) excretion of trace elements contained in their diet or other health supplements (Petersen et al. 2007; European Commission (2003)). Thus, the concentration of metals in manure is generally moderate, especially for toxic Cd and Pb. Micronutrients like Cu and Zn can reach substantial levels because the animal is usually overdosed with these oligoelements to increase productivity and disease resistance (Nicholson et al. 1999).

The metal HQ and HI were calculated for each of the 194 cases in the inventory tables using the multi-compartment risk assessment model described in the previous section. It can be seen in Tables 1, 2, and 3 that the HI value exceeded the recommended ERA safety limit of 1.0 in 14% of sludge cases, with an average value of 0.64. The percentage of cases above 1.0 was lower for compost (4%), with an average value 0.42. However, it is important to note that the risk estimated is incremental in that it only reflects one of the possible routes of metal exposure for humans, and the obtained HI values for sludge and compost become of greater concern within this context despite being lower than 1.0 in most cases. Regarding manure, its reuse as agricultural fertilizer could be considered a safer practice (0.25 average HI). Note that only total metal contents in waste were used to calculate HQs and the HI, and aspects like bioavailability were not assessed in this work. This fact could reduce the final value of the HI because some metals may be strongly complexed with organic matter (García et al. 1995; Zheng et al. 2004; Nomeda et al. 2008). Hence, it is possible that taking bioavailability into account would result in the reduction of the HI for organic wastes. However, metal bioavailability depends not only on metal content, but also on the chemical properties of organic waste (Smith 2009).

Average metal-specific HQs and an average HI were calculated for each type of waste (Fig. 1). The highest contribution to the HI was the essential trace element Zn, and typical toxic elements like Cd and Pb posed a minor

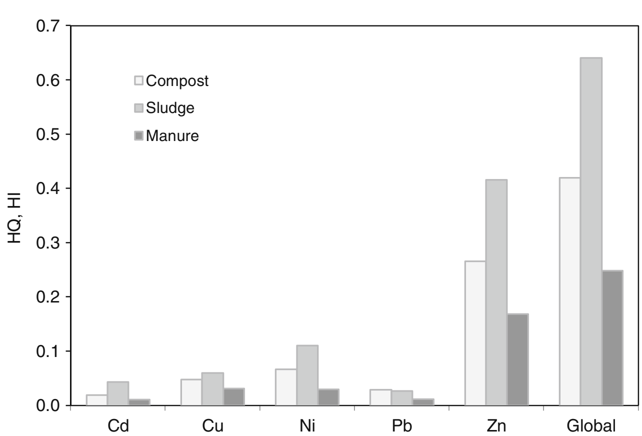

Fig. 1 Influence of metal and organic waste type on risk indexes, hazard quotient (*HQ*), and hazard index (*HI*)

contribution to total risk. Although a very low dose (RfD) of these metals can result in severe adverse effects to human health, it is necessary to take into account each evaluated case. From the original organic waste applied on land, metals have to be transferred to vegetation and cattle, then to humans. Thus, the biotransfer potential, rather than the toxicity potential, would be the best indicator of the magnitude of risk in this particular scenario. According to the Risk Assessment Information System (ORNL 2010), biotransfer factors (BTFs) to meat and milk for Cd, Cu, and Pb ranged between $1{\cdot}10^{-03}$ and $1{\cdot}10^{-04}$ in magnitude, while for Zn, the values were $1{\cdot}10^{-01}$, and $1{\cdot}10^{-02}$ for meat and milk, respectively. Thus, although the ingestion RfDs of Zn was significantly higher in comparison with the other metals (i.e., the dose a human ingests must be high to produce any adverse effect on health), significant concentrations of Zn in either type of organic waste and high BTFs resulted in large HQs, exceeding the safety limit for several cases of compost and sludge. Ni also contributed significantly to the HI because of its high BTF to milk ($1.6{\cdot}10^{-01}$). An analysis of the exposure pathways considered in the scenario revealed that ingestion of meat, followed by milk ingestion, represented between 75% and 90% of the total risk on average in all cases inventoried. As expected, pathways involving direct absorption from soil contact and inhalation had a minor effect on the risk index, and both the Cd and Pb HQ were low.

The HQs of metals for each type of organic waste were proportional to their concentration. The contribution of Ni to the HI was approximately 10–12% for compost and sludge and 6% in manure. In the case of Zn, the opposite trend occurred, with a contribution to manure of 68% and to compost and sludge of 64%. So, although some authors have indicated that levels of Zn in manure are generally lower than in other types of organic waste (Soliva and Paulet 2001; Achiba et al. 2009), we found similar levels in manure, compost, and sludge for the cases included in the inventory. Together with Cu, Zn content was higher than that of other metals in manure due to excretion of these oligoelements after supplementation in cattle. Zn concentration was also highest in compost and sludge, but a more significant presence of the other metals was also found, especially for the toxic Cd and Pb. The average level of Zn in sludge calculated from the studies in the inventory was 1,200 $mg{\cdot}kg^{-1}$, while in manure it was 300 $mg{\cdot}kg^{-1}$.

Zn can end up in wastewater and sludge from several different sources: excretion by humans from ingested food or water, use of galvanized materials, car emissions, car washes, metallurgy, mining, painting, and any applications that involve high levels of Zn in domestic and industrial wastewaters (Sörme and Lagerkvist 2002). Zn is an essential element for humans, with a recommended dietary intake of approximately 0.16 $mg{\cdot}kg^{-1}{\cdot}day^{-1}$ for men and

Table 5 Limit values of heavy metals content in compost according to Legislation and its correspondent HQ and HI

Source	Country	HQ (Heavy metal content mg·kg^{-1})					HI
		Cd	Cu	Ni	Pb	Zn	
Spanish Government (2005)	Spain-class A	0.011 (0.7)	0.029 (70)	0.039 (25)	0.015 (45)	0.138 (200)	0.232
	Spain-class B	0.014 (2)	0.059 (300)	0.098 (90)	0.026 (150)	0.236 (500)	0.433
	Spain-class C[a]	0.013 (3)	0.047 (400)	0.061 (100)	0.021 (200)	0.236 (1,000)	0.378
Cai et al. (2007)	Netherlands (clean compost)	0.011 (0.7)	0.022 (25)	–	0.018 (65)	0.091 (75)	0.142
	Netherlands	0.012 (1)	0.028 (60)	–	0.021 (100)	0.138 (200)	0.199
	Canada Class A	0.017 (3)	0.034 (100)	–	0.026 (150)	0.236 (500)	0.313
	Poland	0.022 (5)	0.059 (300)	–	0.046 (350)	0.508 (1,500)	0.635
	UK	0.013 (1.5)	0.047 (200)	–	0.026 (150)	0.205 (400)	0.291
	Australia	0.017 (3)	0.047 (200)	–	0.031 (200)	0.155 (250)	0.250
	USA	0.019 (4)	0.059 (300)	–	0.026 (150)	0.205 (400)	0.309

Limit values for heavy metal content are indicated in parentheses

[a] Application rate <5 t ha^{-1} year^{-1} in agriculture

0.13 mg kg^{-1} day^{-1} for women (ATSDR (Agency for Toxic Substances and Disease Registry) 2005). However, prolonged oral exposure to zinc at high levels (~2 mg kg^{-1} day^{-1} Zn) may cause severe symptoms of copper deficiency, including anemia and neutropenia (Ramadurai et al. 1993).

3.2 Legislative limits

Proposed limits for heavy metals in organic soil fertilizer amendments are given in Table 5, and HIs for each specified-use class (A, B, and C) have been calculated. Considering metal content, class A was the most appropriate for cultivating crops intended for direct human consumption. The resulting HI after 100 years of applications of this type of organic waste was 0.23, but a low percentage of compost (20%) and sludge (10%) considered in the inventory can be classified within this category. This percentage increased to 45% of cases adequate to be applied according to class A guidelines in manure. Sixty percent of compost and 40% of sludge fell into the type B classification, which is more adequate to fertilize land for forage or fruit production. Finally, despite its higher metal content, fertilizers classified under type C had HQs and a global HI that were similar to type B because of its limited application rate, which must be lower than 5 t ha^{-1} year^{-1}.

In general, countries presented similar values of maximum permissible contents in compost for each metal, providing, an acceptable HI as a first approximation. However, different soil properties and climate could influence the final value of the risk index, which was evaluated with a sensitivity analysis. Finally, although legislation allows the use of sludge containing much higher concentrations of heavy metals (Goi et al. 2006; Stylianou et al. 2008), its application in agriculture is usually strongly constrained to low application rates and frequencies, as well as to specific times of the year. These restrictions were not considered in the estimation of sludge HI, although they could result in a decrease of metal risk indexes. Despite this worst-case scenario, incremental risk cannot be considered negligible, and metal limits in organic waste should be decreased, as stated previously in literature (Madrid et al. 2007).

3.3 Sensitivity analysis

Figure 2 illustrates the influence of soil properties and climate in the HQ of each metal and in the total HI. Soil pH

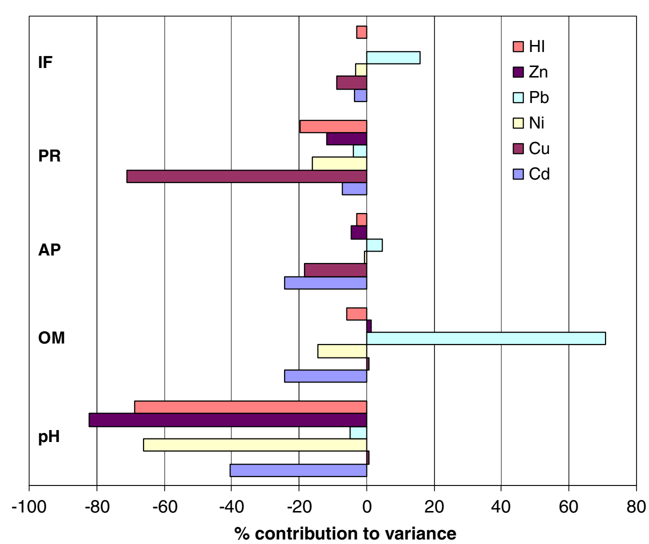

Fig. 2 Influence of soil and climate characteristics (*pH*), organic matter (*OM*), average production (*AP*), precipitation rate (*PR*), and infiltration factor (*IF*) on metal hazard quotient (*HQ*), and hazard index (*HI*)

played a key role in the magnitude of total risk for Cd, Ni, and Zn because an increase in the value of this parameter provoked a significant reduction in HQ and HI. Low pH values enhance metal solubility, mobility, and bioavailability in soil (Smith 1994; Planquart et al. 1999), as reflected in certain countries' legislation that establishes a different organic waste application rate depending on the pH value (i.e., lower or higher than 7).

Soil organic matter only influenced the HQ of Pb significantly (70.9% of variance). It had a lower effect on Cd and Ni and was negligible for Cu and Zn. Figure 2 shows that an increase in soil organic matter resulted in an increase in the Pb HQ (i.e., positive effect). Pb is one of the most strongly adsorbed metals by organic matter and, thus, may be effectively retained and accumulated in the soil matrix (Schroth et al. 2008). Lead's low biotransfer potential implies that the direct soil exposure pathways contributed more to its HQ. Organic matter can fix and increase the Pb concentration in soil and increase its HQ accordingly, although this value was very low compared with the total HI. Therefore, the influence of organic matter could be significant in scenarios where direct and prolonged contact with Pb-contaminated soil is expected.

Finally, the HQ of Cu was primarily affected by climatic conditions (i.e., precipitation rate) and was less sensitive to pH changes (Smith 1994). In contrast to the behavior of the other metals, an increase in precipitation would result in a decrease in risk due to Cu according to the sensitivity analysis. Enhanced leaching of Cu through the soil matrix (Kidd et al. 2007) escapes metal biotransfer from soil solution to vegetation and cattle, and subsequently to humans, leading to a low HQ.

The high influence of pH on the global HI can also be seen in Fig. 2. This influence is due to the high contribution of Zn, followed by Ni, because both metals significantly depend on pH. Precipitation rate is the second most influential variable at 20%, due to the contribution of Cu (after Zn and Ni). Thus, soil and climate properties (i.e., location) can significantly vary the magnitude of risk depending on the metal. For example, the sensitivity analysis revealed that in the case of organic waste reuse, locations with acidic soils and high precipitation rates would be more affected by Zn exposure. These two scenarios can be found within the same country, Spain, where the Mediterranean area has basic soils and low precipitation rates, but the Atlantic area (NW) has acidic soils and high precipitation rates.

4 Conclusions

In this study, a wide inventory of the heavy metal content in three types of organic wastes (i.e., compost, sludge, and manure) was taken. Health risks due to the reuse of these residues as agricultural fertilizers were determined by an ERA. The results indicated that sludge contained the highest concentrations of metals, and the presence of toxic metals like Cd and Pb was more significant than in compost and manure. As expected, sludge reuse in the proposed scenario resulted in the highest incremental risk. Surprisingly, the metal with the greatest risk contribution to the three types of organic waste was Zn, making the presence of toxic Cd and Pb almost negligible in terms of risk. Although Zn presents a very low level of toxicity as an essential element to life, its high biotransfer potential may create in significant concentrations that exceed the recommended doses in organic matrices like plants, cattle, and humans. Therefore, specific measures should be taken to regulate the Zn content of organic waste depending on its final management solution. The origin of the Zn should also be established for proper reduction measurements in emissions, especially in sludge. However, a worst-case scenario approach was selected, and the risk may be overestimated because legislation restrictions on the application of sludge were not considered. Another key aspect, bioavailability, was not addressed in the present work. Future efforts should be focused on assessing metal speciation in the soil solution, either as inorganic complexes or bound to humic and fulvic acids.

Acknowledgments This work was supported by the Spanish Government (Science and Innovation Ministry) through the Project INDIE (CTM2010-18893). Marta Herva wishes to thank the University of Santiago de Compostela for her pre-doctoral contract. Dr. Amaya Franco-Uría would like to thank MICINN for the support provided by the "Juan de la Cierva" Subprogram.

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