Waste Management 48 (2016) 404–408

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Metal concentrations in lime stabilised, thermally dried and anaerobically digested sewage sludges

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ARTICLE INFO

Article history:
Received 12 May 2015
Revised 20 October 2015
Accepted 14 November 2015
Available online 25 November 2015

Keywords:
Treated sludge
Biosolids
Metals
Land application

ABSTRACT

Cognisant of the negative debate and public sentiment about the land application of treated sewage sludges ('biosolids'), it is important to characterise such wastes beyond current regulated parameters. Concerns may be warranted, as many priority metal pollutants may be present in biosolids. This study represents the first time that extensive use was made of a handheld X-ray fluorescence (XRF) analyser to characterise metals in sludges, having undergone treatment by thermal drying, lime stabilisation, or anaerobic digestion, in 16 wastewater treatment plants (WWTPs) in Ireland. The concentrations of metals, expressed as mg kg^{-1} dry solids (DS), which are currently regulated in the European Union, ranged from 11 (cadmium, anaerobically digested (AD) biosolids) to 1273 mg kg^{-1} (zinc, AD biosolids), and with the exception of lead in one WWTP (which had a concentration of 3696 mg kg^{-1}), all metals were within EU regulatory limits. Two potentially hazardous metals, antimony (Sb) and tin (Sn), for which no legislation currently exists, were much higher than their baseline concentrations in soils (17–20 mg Sb kg^{-1} and 23–55 mg Sn kg^{-1}), meaning that potentially large amounts of these elements may be applied to the soil without regulation. This study recommends that the regulations governing the values for metal concentrations in sludges for reuse in agriculture are extended to include Sb and Sn.

1. Introduction

More than 10 million tonnes of sewage sludges were produced in the European Union (EU) in 2010 (Eurostat, 2014). Legislation such as the Landfill Directive, 1999/31/EC (EC, 1999), the Urban Wastewater Treatment Directive 91/271/EEC (EC, 1991), the Waste Framework Directive (2008/98/EC; EC, 2008) and the Renewable Energy Directive (2009/28/EC; EC, 2009), means that rather than incinerating it or sending it to landfill, there is an increased emphasis on its reuse as a 'product'. Consequently, it is used in the production of energy (Gikas, 2014), bio-plastics (Yan et al., 2008), construction materials (Jiang et al., 2011) and, when appropriate treatment is applied, as an agricultural fertiliser (Koutroubas et al., 2014).

There are considerable public acceptance issues around the reuse of treated municipal sludge ('biosolids') as fertiliser (LeBlanc et al., 2008) and, depending on the part of the world, legislation regarding its reuse as such, differs (Milieu et al., 2013a,b,c). Moreover, in some countries such as Belgium (Brussels and Flanders), Switzerland and Romania, the use of biosolids in agriculture is prohibited (Milieu et al., 2013a,b,c). While concerns over the presence of persistent organic pollutants and emerging contaminants, such as pharmaceuticals, have been expressed (Clarke and Cummins, 2014), the presence of toxic metals in sludge, due to the mixing of industrial wastewater with sewage, means that the application of metal-contaminated sludge may cause the contamination of soil and water (Cornu et al., 2001) and accumulation of metals in the food chain (Kidd et al., 2007; Latare et al., 2014). In an attempt to address these concerns, guidance values concerning the maximum allowable concentration of certain metals in biosolids (Table 1) are in place in countries where the reuse of biosolids on land is permitted. The level of exceedance in wastewater treatment plants (WWTPs) is therefore of interest.

The application of biosolids to agricultural land is governed by various legislation (e.g. in Europe by EU Directive 86/278/EEC (EEC, 1986); in the US by 40 CFR Part 503 (US EPA, 1993)). These require that sewage sludge undergoes biological, chemical or heat treatment, long-term storage, or any other process to reduce the potential for health hazards associated with its use. In the EU, land

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http://dx.doi.org/10.1016/j.wasman.2015.11.028
0956-053X/

Table 1
Limit values for metal concentrations in sludge for use in agriculture.

	Selenium (Se)	Molybdenum (Mo)	Arsenic (As)	Copper (Cu)	Nickel (Ni)	Lead (Pb)	Zinc (Zn)	Cadmium (Cd)	Chromium (Cr)	Mercury (Hg)	Reference
	mg kg^{-1} dry weight (=ppm)										
Brazil	100	50	41	1500	40	300	2800	39	1000	17	LeBlanc et al. (2008)
China			75	800–1500	100–200	300–1000	2000–3000	5–20		5–15	LeBlanc et al. (2008)
EU				1000–1750	300–400	750–1200	2500–4000	20–40	–	16–25	EEC (1986)
Japan			50		300	100		5	500	2	LeBlanc et al. (2008)
Jordan	100	75	41	1500	300	300	2800	40	900	17	LeBlanc et al. (2008)
Russian Fed.			10	750	200	250	1750	15	500	7.5	LeBlanc et al. (2008)
USA	100	75	41–75	1500–4300	420	300–840	2800–7500	39–85		17–57	US EPA (1993)

application of biosolids is typically based on its nutrient and metal content, although individual member states often have more stringent limits than governing directives (LeBlanc et al., 2008; EC, 2010; Milieu et al., 2013a,b,c). Guidelines govern the maximum rate of nutrients and metals (e.g. Fehily Timoney and Company, 1999), although as the metal content is normally low relative to the nutrient content of biosolids, application rates are frequently determined by the nutrient content of the biosolids and not their metal content (Lucid et al., 2013). As soil acidification may increase the solubility of metals (Antoniadis et al., 2008), there is a potential risk of metal accumulation in the soil (Álvarez et al., 2002; Mamindy-Pajany et al., 2014), in plants (Latare et al., 2014), or of transport to groundwater, particularly if added in excess (McBride et al., 1999). In countries such as the USA, where in the majority of states biosolids are applied to land based on the nitrogen (N) requirement of the crop being grown and not on a soil-based test (McDonald and Wall, 2011), excessive metal accumulation in soil and plants (Wen et al., 2014), or losses in surface and subsurface waters (Oun et al., 2014), may potentially occur.

Laboratory and field studies have demonstrated that the addition of biosolids to land as a fertiliser replacement has several beneficial effects (Monera et al., 2002; Latare et al., 2014). They provide nutrients and micronutrients (e.g. zinc (Zn), copper (Cu), cobalt (Co)) required for plant and crop growth, and can be used as an aid in the development of a soil's physical and chemical characteristics. Latare et al. (2014) found that applications of biosolids to land at rates ranging from 10 to 40 tonnes ha^{-1} increased the grain yield of rice by up to 40% and increased the available nutrient content of the soil in comparison to equivalent doses of fertilizers. However, the metal content of both the plants (cadmium (Cd)) and soil (Zn) also increased in comparison to the regular fertiliser. Similar results have been found by other researchers (McBride et al., 1999; Stietiya and Wang, 2011).

Due to the increasing awareness regarding potential risks to the environment and human health, the application of sewage sludge, following treatment, to land as a fertilizer in agricultural systems has come under increased scrutiny. This is mainly a perception issue by the food production sector, which is driven by the belief that best practices for sludge treatment are not being followed (EPA, 2014b). As metals are likely to remain in the soil indefinitely, the characterisation of biosolids prior to land application is important. The aim of this study was to: (1) examine if the metal content of biosolids from high population equivalent (PE) WWTPs in Ireland exceeded permitted limit values and (2) establish a baseline for unregulated metals – potential pollutants of which little is known and from which other global studies may be compared. To our knowledge, this is the first time that extensive use was made of a handheld X-ray fluorescence (XRF) analyser to carry out analysis on biosolids.

1.1. Study context in Ireland

In Ireland there were 541 urban areas, with PEs ranging up to 2.3 million, that received either preliminary, primary, secondary, or secondary treatment and nutrient reduction in 2012 (EPA, 2014a). In 2012, approximately 94% of the national wastewater load received at least secondary treatment, and the WWTPs produced sewage sludge with a total load of 72,429 tonnes (dry solids, DS), of which 94.3% was diverted to agriculture, 5.7% was diverted to composting and other uses, and <0.01% was sent to landfill (EPA, 2014a). Of the treatment processes currently in use in Ireland (anaerobic and aerobic digestion, composting, thermal drying), lime stabilisation remains the most popular, due to the relatively small amount of costs involved (EPA, 2014b).

2. Materials and methods

2.1. Sample collection and preparation

Biosolids were collected from 16 WWTPs or agglomerations, with PEs ranging up to approximately 2.3 million (Table 2). Selection of the WWTPs was predicated on willingness to participate in this monitoring study and geographical location (a good geographical spread was desirable). None of the plants selected had a history of persistent failures in meeting water discharge standards (EPA, 2014a). Of the WWTPs examined, most received landfill leachate in low quantities (no greater than 2% of the total BOD loading on the WWTP), while others received industrial, commercial and domestic/septic tank sludge comprising up to 30% of the total influent BOD loading on the WWTP (Table 2). Eight discrete samples (n = 8) of 100 g were collected in clean LDPE containers (Fisher, UK) from each WWTP and stored at −20 °C prior to analysis. The biosolids samples were freeze dried (Freezone 12, Labconco, Kansas City, USA) at −50 °C and pulverised in an agate ball mill (Fritsch™ Pulverisette 6 Panetary Mono Mill) with a rotational speed of 500 rpm for 5 min (repeated three times) using an 80 ml agate vial and balls (Ø 10 mm).

2.2. Elemental determination

A handheld X-ray fluorescence (XRF) analyser (DELTA Series 4000, Olympus INNOV-X, Woburn, MA, USA) in the laboratory

Table 2
Site agglomerations and type of treatment conducted in each location.

Site no.	WWTP/ agglomeration size (PEs)	Leachate as % of influent BOD load	Industrial/commercial and domestic/septic tank sludge[a] as % of influent BOD load	Type of treatment
1	2,362,329	<0.01	<0.01	Thermal drying, anaerobic digestion
2	284,696	0.3	24	Thermal drying
3	179,000	Unknown	30	Anaerobic digestion
4	130,000	Unknown	0.008	Thermal drying
5	101,000	2.0	Unknown	Lime stabilisation
6	86,408	0.2	2.1	Anaerobic digestion
7	76,456	0	0	Anaerobic digestion
8	46,428	0.1	25	Lime stabilisation
9	42,000	<0.01	15	Thermal drying
10	31,788	0.25	Unknown	Lime stabilisation
11	30,000	0.081	0	Thermal drying
12	27,731	0	2.8	Anaerobic digestion
13	27,000	0.2	0	Thermal drying
14	25,000	0.7	0	Thermal drying
15	22,440	0	0	Lime stabilisation
16	6500	Unknown	Unknown	Thermal drying

[a] Most recent available figures in all WWTPs (2013).

(mounted in an integrated bench-top workstation and interfaced with a PC) in soil environmental mode was employed to determine metal (Cd, chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), molybdenum (Mo), nickel (Ni), lead (Pb), antimony (Sb), selenium (Se), tin (Sn), and Zn) concentrations. This portable XRF system consists of a powerful X-ray tube (4 W, Au anode) and a 30 cm^2 Silicon Drift Detector (SDD). An internal instrument standardisation was performed using an alloy chip (aligns the Fe and Mo peaks on the spectrum to compensate for temperature drift) and sewage sludge certified reference materials (Trace Metals – Sewage Sludge 2 CRM029, Sewage Sludge 3 CRM031 and Sewage Sludge 4 CRM055, Sigma–Aldrich RTC, Inc., USA) were used for calibration/verification of the P-XRF to matrix match the 'unknown sewage sludge samples' as closely as possible in order to eliminate matrix effect from the P-XRF analysis. Calibration using the Certified Reference Materials (CRMs) was achieved by plotting the XRF data against certified data and inserting a linear trend line to determine the linearity of the calibration (which is used to calculate the factor and offset required to correct the data within the instrument). An aliquot of the homogenised biosolids (approximately 5 g) was packed into polyethylene XRF sample cups and covered with a 4 μm Prolene sample support window (Chemplex® Industries Inc., USA). Metal concentrations were detected simultaneously and the operating parameters included a measurement time of 180 s at beam currents of up to 200 μA (maximum voltage of 40 kv and energy resolution of 150 eV). The software uses a compton normalisation algorithm to determine mg kg^{-1} concentrations of elements by correlation of the X-ray tube parameters and the intensity and energy seen by the detector.

2.3. Quality control

Quality control included the use of instrumental blanks (SiO_2), analysis of duplicate samples, and the performance of the method and stability of the instrument was evaluated by using CRMs of sewage sludge (Trace Metals – Sewage Sludge 2 CRM029, Sewage Sludge 3 CRM031 and Sewage Sludge 4 CRM055, Sigma–Aldrich RTC, Inc., USA), sediments (LKSD-4, lake sediment and PACS-1 marine harbour sediment, National Resources Canada) and soils (SRM 2709a San Joaquin Soil and SRM 2710a Montana Soil I, National Institute of Standards and Technology (NIST), USA). The results of the analysis of the CRMs were in good agreement with their respective certified and reference ranges (Tables S1 and S2). Further confirmation of the validity of the P-XRF technique was provided by the analysis of 15% of the sewage sludge samples (taken systematically, representing elemental concentrations across the entire range, as determined by P-XRF) using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (Agilent 7700) after digestion with *aqua-regia* (Trace SELECT®, Sigma Aldrich) in a graphite heating block. For the elements that were above the limit of detection (LOD) of the P-XRF technique (Fe, Cu, Zn, Pb, Se, Mo, Ni, Sn and Cr) in this portion of the sewage sludge samples, a comparison was made between the results obtained from the P-XRF and the concentrations determined by ICP-MS. Correlation coefficients (Pearson Product Moment Correlation for normal distributions and Spearman's Rank Order Correlation for non-normal data) between the P-XRF and ICP-MS results were also determined (SigmaPlot 12, Systat Software Inc, San Jose, CA).

3. Results and discussion

3.1. Validation of the P-XRF technique

Correlation coefficients between P-XRF and ICP-MS results indicated the suitability and satisfactory use of the P-XRF technique for the quantification of these elements in sewage sludges (Fe: $r = 0.99$, $P < 0.001$; Cu: $r = 0.95$, $P < 0.0001$; Zn: $r = 0.98$, $P < 0.0001$; Se: $r = 0.95$, $P < 0.0001$; Mo: $r = 0.79$, $P < 0.0001$; Sn: $r = 0.63$, $P < 0.01$; Ni: $r = 0.85$, $P < 0.001$; Cr: $r = 0.82$, $P < 0.01$; Pb: $r = 0.99$, $P < 0.0001$). Results of the ICP-MS analysis also confirmed that the levels of Sb and Hg were below the LOD of the P-XRF technique for this portion of comparative samples.

3.2. Overview of metal concentrations in sewage sludge

The mean concentrations of the metals in the sewage sludge following treatment in the 16 WWTPs are given in Table 3. The concentrations of the metals, which are regulated in the EU, and all expressed as mg kg^{-1} DS, ranged from 11 (Cd, anaerobically digested (AD) biosolids) to 1273 mg kg^{-1} (Zn, AD biosolids), and were well under EU regulatory limits. Of the parameters not regulated in the EU, but regulated elsewhere (Table 1), As, Se, Mo and Cr (Table 3) were well below the upper limits of 75, 100, 75 and 1000 mg kg^{-1}, respectively. Of the elements considered bio-essential micro-nutrients measured in this study (Se, Fe, Cu and Zn), all were within either EU or international limits (Table 1) (no limits govern Fe).

The biosolids from one WWTP, in which anaerobic digestion was carried out, had an average Pb concentration of 3696 mg kg^{-1}, well in excess of the threshold value of 1200 mg kg^{-1}. The average concentrations (across all treatments) of Cu, Pb and Zn were also well above the median values of internationally published results (Table 4). Lead is amongst the most hazardous metals, which are potentially harmful to human health (Johnson and Bretsch, 2002). Other metals measured in this study, which are also

Table 3
Mean (±standard deviation, SD) metal concentration (mg kg^{-1} dry weight) in sludge following anaerobic digestion, lime stabilisation, or thermal drying. *n* refers to the number of treatments.

Metal	Anaerobic digestion ($n = 5$)		Lime stabilisation ($n = 4$)		Thermal drying ($n = 8$)		EU regularity upper limits EEC (1986)
	Mean	SD	Mean	SD	Mean	SD	
Regulated parameters in EU							
Cu	640	411	491	452	464	205	1750
Ni	25	5	13	2.5	15	7	400
Pb	791	1625	33	25	54	30	1200
Cd	11	1	13	1	10	3	40
Zn	1273	749	526	388	869	400	4000
Hg[a]	<LOD		<LOD		<LOD		25
Non-regulated parameters in EU							
As[b]	<LOD		<LOD		<LOD		
Se	3	2	3	1	2	1	
Sr	162	61	183	75	114	36	
Mo	5	2	4	1	5	1	
Ag	11	2	11	3	8	3	
Sn	55	57	23	4	23	5	
Sb	20	5	17	3	17	4	
Cr	51	43	25	15	16	12	
Fe	32,135	41,717	9654	7264	33,087	43,373	

[a] Limit of detection (LOD) = 10 ppm.
[b] LOD = 100 ppm.

potentially harmful, are: Cr, Cd, Sn and Sb. Of these parameters, to date no international standards exist for Sb or Sn in biosolids for reuse in agriculture. In the present study, the average concentration of Sb ranged from 17 to 20 mg kg^{-1} (Table 3), which was substantially higher than recorded elsewhere, e.g. <0.01–0.06 mg kg^{-1} (LeBlanc et al., 2008), 3.4 mg kg^{-1} (Eriksson, 2001). As the average concentration of Sb in non-polluted soils is around 0.53 mg kg^{-1} (Fay et al., 2007) and elevated concentrations in the soil inhibit the early growth of crop plants (Fjällborg and Dave, 2004; Baek et al., 2014), the possibility exists that potentially large applications of this parameter are being land applied without regulation. Tin, in inorganic form, is non-toxic, but a significant portion of sewage sludges may be in a highly toxic, organic form and include compounds such as tributyltin (McBride, 2003). The concentrations of Sn measured in this study ranged from 23 to 55 mg kg^{-1} (Table 3), which was of the same order as other studies (26 mg kg^{-1} – Eriksson, 2001). Normal ranges of Sn in non-polluting Irish soils are around 1.68 mg kg^{-1} (Fay et al., 2007). Both parameters, Sb and Sn, however, are not considered to be of risk to animals or humans (US EPA, 1995).

3.3. Environmental policy and management implications

Land application of biosolids is, in the main, determined by the nutrient content of biosolids and not by the metal content (Lucid et al., 2013). Therefore, the metal content, even if present in relatively high concentrations in the biosolids, may not have any significant impact on soil quality in the short term. However, accumulation of metals in soil following repeated applications of biosolids, may be problematic – particularly for those elements that are not regulated and are harmful to human health. Guidelines should aim to govern the maximum allowable concentrations of these elements in biosolids, as well as the land to which they are applied. Handheld XRF analysis is a useful, quick and relatively inexpensive method for determining the metal content of biosolids, and should be used frequently to characterise it.

4. Conclusions

The metals from 16 WWTPs in Ireland were below the maximum allowable concentrations of metals for use in agriculture in the EU. In addition, they were also within the median levels for biosolids globally. While current EU and international regulations govern certain priority metal pollutants and bio-essential elements, other metals that are potentially harmful to human health, such as Se and Sn, are omitted from the regulations. This means that a number of toxic metals, which are much higher than their baseline concentrations in soils, are being applied without regulation. It is recommended that the regulations governing the values for metal concentrations in biosolids for reuse in agriculture are extended to cover Sn and Sb. A handheld XRF analyser is a cost-effective and rapid method for the analysis of biosolids, and may be easily applied in WWTPs. Its frequent use would mean that

Table 4
Measured values for metal concentrations in sludge for use in agriculture (adapted from LeBlanc et al. (2008)) compared with average concentrations (across all treatments) measured in the current study.

	Selenium (Se)	Molybdenum (Mo)	Arsenic (As)[a]	Copper (Cu)	Nickel (Ni)	Lead (Pb)	Zinc (Zn)	Cadmium (Cd)	Chromium (Cr)	Mercury (Hg)[b]
	mg kg^{-1} dry weight (=ppm)									
Brazil	27	113	15	255	42	80	689	11	144	2
Bogota, Columbia	24		19	163	43	88	1014	76	73	8
Denver, USA	15	20	3	670	16	39	714	2		1
Los Angeles, USA	15	18	6	1060	51	39	1180	10	84	2
Milwaukee, USA	4	11	8	266	32	57	534	4	289	0.3
Ottawa, Canada			1	460	16	51	593	1	50	1
British Columbia, Canada	4	8	5	888	26	56	588	3	51	3
Finland				244	30	9	332	1	18	0.4
Germany				380	32	62	956	2	61	1
Italy				261	16	76	577	2	22	0.2
Slovenia			2	200	35	150	600	1	90	2
Turkey				70	62	34	300	1	34	
Sapporo, Japan			7	140	35	10	300	<1	29	0.2
Suzu, Japan			8		32	5		2	20	1
Moscow, Russ Fed.			0–24	0.9–1200	1.4–306	0.8–1070	3–3820	0–300	18–1280	0–11
Current study	3	5	<LOD	520	18	252	886	12	35	<LOD

[a] LOD = 100 ppm.
[b] Limit of detection (LOD) = 10 ppm.

plant managers may determine, with relative ease, the suitability of biosolids for reuse in agriculture.

Acknowledgements

The authors wish to acknowledge funding from the Irish EPA (Project reference number 2012-EH-MS-13) and the Department of Communications, Energy and Natural Resources under the National Geoscience Programme 2007–2013 (Griffiths Award). The views expressed in this study are the authors' own and do not necessarily reflect the views and opinions of the Minister for Communications, Energy and Natural Resources.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.wasman.2015.11.028.

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