Rare earth elements and critical metal content of extracted landfilled material and potential recovery opportunities

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ABSTRACT

Rare earth elements (REEs), Platinum group metals (PGMs) and other critical metals currently attract significant interest due to the high risks of supply shortage and substantial impact on the economy. Their uses in many applications have made them present in municipal solid waste (MSW) and in commercial and industrial waste (C&I), since several industrial processes produce by-products with high content of these metals. With over 4000 landfills in the UK alone, the aim of this study was to assess the existence of these critical metals within landfills. Samples collected from four closed landfills in UK were subjected to a two-step acid digestion to extract 27 metals of interest. Concentrations across the four landfill sites were 58 ± 6 mg kg^{-1} for REEs comprising 44 ± 8 mg kg^{-1} for light REEs, 11 ± 2 mg kg^{-1} for heavy REEs and 3 ± 1 mg kg^{-1} for Scandium (Sc) and 3 ± 1.0 mg kg^{-1} of PGMs. Compared to the typical concentration in ores, these concentrations are too low to achieve a commercially viable extraction. However, content of other highly valuable metals (Al and Cu) was found in concentrations equating to a combined value across the four landfills of around $400 million, which increases the economic viability of landfill mining. Presence of critical metals will mainly depend on the type of waste that was buried but the recovery of these metals through landfill mining is possible and is economically feasible only if additional materials (plastics, paper, metallic items and other) are also recovered for reprocessing.

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1. Introduction

The global demand for metals continues to increase and the costs of extraction are reflected in their prices since sources of high quality ores have reduced and so lower quality mines must be now exploited (DEFRA, 2012; House of Commons Science and Technology Committee, 2011). Availability and sufficiency of certain metals are causing great concerns to governments and industries (Kaartinen et al., 2013a). Several reports (British Geological Survey, 2012; DEFRA, 2010; House of Commons Science and Technology Committee, 2011; U.S. Department of Energy, 2011; European Commission, 2014) investigated the demand and supply risk of several metals including their scarcity, the political stability of the producing countries, the price volatility and the potential of substitution, trying to identify the most critical. Rare earth elements (REEs), Platinum group metals (PGMs), Lithium (Li), Indium (In), Cobalt (Co) and Antimony (Sb) were identified as high risk of supply shortage and increased impact on the economy (Hislop and Hill, 2011).

The most common use of Li, In, Co and Sb is in ceramics and glass materials, but they can also be found in permanent magnets (Co), fluorescent lamps (In and Sb), rechargeable batteries (Li and Co) and in electrical and electronic equipment (Co, In and Sb). Also, In is used in fibre optic telecommunications and alkaline batteries whereas Sb is commonly used as a flame retardant in plastics and textiles (Jorgenson and George, 2005; USGS, 2011; Sadyrbaeva, 2014; Jaskula, 2013; Butterman and Carlin, 2004). PGMs are mainly used in fuel cells, catalysts, glass and glass fibre manufacture, electrical and electronic uses and jewellery. They are essential constituents of automotive catalysts as they significantly decrease the emission of harmful compounds such as carbon monoxide, hydrocarbons and nitrogen oxides (British Geological Survey, 2009; Hagelüken, 2012; Chaston, 1982; Ayres and Peiró, 2013). A typical auto-catalyst has a concentration between 300 and 1000 µg g^{-1} of Pt, 200 and 800 µg g^{-1} of Pd and from 50 to 100 µg g^{-1} of Rh (Jimenez de Aberasturi et al., 2011).

REEs comprise the periodic group of lanthanides plus Scandium (Sc) and Ytrrium (Y) and can be divided in 3 groups; Light (LREEs), Heavy (HREEs) and Sc. LREEs comprise Lanthanum (La), Cerium (Ce), Praseodymium (Pr), Neodymium (Nd) and Samarium (Sm) and the remaining are included in the HREEs. This classification is based on the chemical properties, geological availability, supply

sources, supply demands, market values and end-markets of REEs (European Commission, 2014).

REEs are used in magnets, metallurgy, phosphors, glass and polishing, catalysts, ceramics and for nuclear and defence technologies (Hayes-Labruto et al., 2013; Meyer and Bras, 2011). World-wide, in 2010, 1300 tonnes of REEs were placed in magnets for wind turbines and 8250 tonnes in phosphors applications. Particularly for magnets applications, Nd Dy were used for electrical vehicles (EVs) (4160 tonnes of Nd and 265 tonnes of Dy), in MRI units (450 tonnes of Nd and 35 tonnes of Dy), in electrical and electronic devices (12,450 tonnes of Nd and 830 tonnes of Dy) and in wind turbines (910 tonnes of Nd and 70 tonnes of Dy). While Eu, 50 tonnes were used for phosphors applications; 24.5 tonnes in lighting, 19.5 tonnes in LCDs (Liquid–crystal display) and 6 tonnes in plasma panels (Ayres and Peiró, 2013).

All these products become a great source of critical metals when reaching end-of-life, however, reported recycling rates show poor reuse of these metals; typically between 1% and 25% Sb, <1% for REEs, In, Li and Os, and ⩾50% for Pt, Pd, Rh and Co (Graedel et al., 2011). With the Waste and Electronic Equipment (WEEE) Directive being established in 2002 (EC, 2003), it is presumed that many of the waste products containing these valuable metals were disposed in landfills prior to this date. For decades landfills have been the solution for disposing all types of waste; without any segregation they are a heterogeneous source of materials (Kaartinen et al., 2013a). For example, mobile phones contain several of these critical metals; Co, Nd, and Sb are present in quantities of 52 $\mu g\,g^{-1}$, 1.454 $\mu g\,g^{-1}$, 1 $\mu g\,g^{-1}$, respectively, and between 0.05 and 0.2 $\mu g\,g^{-1}$ of Pd and from 0.005 to 0.1 $\mu g\,g^{-1}$ of Pt (Valero Navazo et al., 2014). The presence of REEs and other critical metals have been reported in municipal solid waste (MSW) (Morf et al., 2013), ashes from the incineration of different types of waste (Zhang et al., 2001, 2008) and in sewage sludge and food industry sludge from wastewater treatment plants (Kawasaki et al., 1998). Furthermore, landfilled industrial wastes such as metallurgical slags, bauxite residue (red mud), phosphogypsum and mine tailings are rich in REEs (Binnemans et al., 2013), although these industrial wastes are more commonly contained in mono-landfills.

Once metal-containing wastes are placed in landfill the mobility of the metals will be limited by a number of attenuating mechanisms (Jain et al., 2005). Metal recovery and extraction from landfill using leachate is hard to achieve as high pH values (>7) of the leachate decreases the mobility of metals and their tendency to leach out (Esakku et al., 2003; Ross et al., 2000; Bozkurt et al., 1999). Also, leachate anions such as sulphides and hydroxides significantly reduce the solubility of metals and induce their precipitation. Where leachate recirculation is applied the concentration of metals in the treated leachate is considerably low due to the reduction of sulphate to sulphide during waste decomposition. Approximately 0.02% of the total heavy metals in a landfill are leached out in 30 years (Reinhart and Al-Yousfi, 1996; Kjeldsen et al., 2002; Esakku et al., 2003; Gould et al., 1990; Ross et al., 2000).

Organic compounds present in landfilled waste greatly influence metals mobility. High organic matter content, as humus material, tends to increase the sorption of metals. After methane production ceases, the remaining organic substrate is harder to degrade and the microbial activity significantly decreases, resulting in the start of the humic phase. Humic substances (humic and fulvic acids) bind metals to hydroxyl and carboxyl groups, and either mobilise metals or delay their release; in aerobic conditions the humic substances lose their binding properties (Bozkurt et al., 1999; Klavinsa et al., 2006; Kochany and Smith, 2001; Leung and Kimaro, 1997). Almost 99.9% of metals are still found at the beginning of the humic phase (Bozkurt et al., 1999). The presence of clay in landfills (liner or daily cover) affects the adsorption of metals. Clay is usually negatively charged and in high

content increases the ion exchange with metals and induces their retention within the clay particles (Bradl et al., 2005; Moldoveanu and Papangelakis, 2012).

There are approximately >4000 open and closed landfills in the UK (Kohler and Perry, 2005) and as these critical metals are currently sourced from overseas (British Geological Survey, 2012), it is appealing to consider excavating closed landfills as they have since evolved into local "mines" of metals (Masi et al., 2014). Landfill mining (LFM) is gaining more attention due to the opportunity to source materials for energy production, recover metals, allow site remediation and land reclamation for development (Hogland et al., 2004; Jain et al., 2014; Van Der Zee et al., 2004). Furthermore, it can yield new business opportunities and decrease dependency on metal exports from the producing countries (e.g. China for REEs and Sb) (Kaartinen et al., 2013a; Wübbeke, 2013; British Geological Survey, 2012).

Previous studies (Masi et al., 2014; Kaartinen et al., 2013a; Hogland et al., 2004; Quaghebeur et al., 2013; Kaartinen et al., 2013b; Spooren et al., 2012; Prechthai et al., 2008) have assessed the type of wastes that can be recovered through LFM for reuse. However they mainly focused on raw materials like paper, glass, plastics, textiles, wood, metallic items and the presence of heavy metals. Conventional LFM is the process of recovering materials contained within a landfill mainly with the purpose of remediating the site (Krook et al., 2012), whereas the concept of enhanced landfill mining [ELFM] targets resource recovery by combining integrated and innovative valorisation techniques for material and energy recovery (Jones et al., 2013). Furthermore in-situ, as part of an ELFM approach, is an interesting option for recovering critical metals from specific landfilled waste streams, such as industrial process residues (Binnemans et al., 2013). Therefore, the purpose of this study was to analyse the presence of REEs and critical metals in the landfill mass (involving mixture of cover material, degraded organics and other inorganic materials) within the closed landfills of the UK to assess the availability of these metals and feasibility of recovery regarding the potential economic impact. A possible relationship between the concentration of metals with the organic content of landfilled material and with the depth at which waste was buried in the landfill site was also investigated.

2. Materials and methods

Fifty-five samples were collected from four closed landfill sites (LFS) in the UK. Different cores were drilled vertically across the landfill area and samples were taken at different depths by a specialised private company hired by the LFS operator. An overview of the waste samples collected at the four LFS is provided in Table 1.

The samples were subjected to manual sieving and sorting for removal of plastic, metal, paper, textile, glass and other materials larger than 19 mm. The remaining fraction, the organic-like material, was dried at 105 °C overnight and ground to a size of ⩽1.5 mm (Retsch SM 2000 grinder).

To achieve the highest quantity of the extracted metals, samples were submitted to nitric acid digestion followed by a digestion with aqua regia (HNO_3 + HCl in a ratio of 1:3) (James, 2011), as it was not technically possible to use HF to digest the samples. Briefly, for the nitric acid digestion, 0.5 g of sample was mixed with 8 ml of HNO_3 (70%, 1.42 density, Fisher Scientific) in Teflon tubes and left overnight. The samples were placed in a microwave digester (Multiwave 3000, Anton Paar) for 45 min at 800 W. The mixtures were then filtered (150 mm, hardened low ash paper, Fisher Scientific) and the remaining liquid was diluted to 100 ml with distilled water and retained for analysis. The filter paper and residue was then dried for 2 h at 105 °C and placed into a digestion tube with 6 ml of HCl (37%, 1.18 density, Fisher

Table 1
Landfill sites, waste type and number of samples collected.

Landfill site and location	Operational time and type of waste	Depth of sample/m (core numbers shown in brackets)
Site A	2007–2013: 75% MSW and 25% commercial and industrial waste (C&I)	2 (1–5)
		4 (1–5)
West central lowlands of Scotland	2013–2014: 75% C&I and 25% MSW	5 (4)
		6 (1–3, 5)
	Approximately 2.25 Mt of waste on site	8 (1–3, 5)
		10 (1–3, 5)
		12 (1–3)
		13 (1)
Site B	1998–2005: MSW and C&I	5 (4–7)
		6 (1)
South-west coast of England	Approximately 1 Mt of waste on site	6.5 (2)
		7 (3)
		12.5 (1)
		13.5 (3)
Site C	1992–2011: MSW and C&I	10 (2, 3, 5)
		15 (1–3)
South coast of England	Approximately 5 Mt of waste on site	20 (2, 4, 5)
		25 (1)
Site D	1980–1999: C&I	7.5 (1)
		8 (2, 5)
South-west coast of England	2000–2004: no activity	10 (3)
		13 (1)
	2004–2008: MSW	15 (3)
		16 (5)
	Approximately 1.5 Mt of waste on site	17.5 (2)
		19 (4)

Scientific) and 2 ml of HNO_3. The mixture was subjected to microwave irradiation at 800 W for 45 min. The solution was filtered and the liquid diluted to 100 ml and retained for analysis.

The retained solutions were analysed for metal content using inductively coupled plasma mass spectrometry (ICP-MS) (Elan 9000 Perkin-Elmer SCIEX) using several certified standard solutions for calibration: rare earth ICP-MS standard (CMS-1, Inorganic Ventures), precious metals ICP-MS standard (CMS-2, Inorganic Ventures) and single ICP standards (analyte concentration of 1000 µg ml^{-1}, SCP SCIENCE) for the rest of critical metals. A total of 27 metals were analysed, Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu from the REEs; Pt, Pd and Ru from the PGMs and Li, In, Co, Sb, Ag, Au, Cu and Al.

Both subsamples form the 2-step acid digestion were analysed by ICP-MS and the metal concentration of the original landfill sample was calculated using the following equation:

$$C \ (\mu g \ L^{-1}) = 100 \times [(M_1 - M_b) + (M_2 - M_b)]/m \qquad (2\text{-}1)$$

where M_1 and M_2 refers to the concentration in the sample extract (µg L^{-1}) from nitric acid and aqua regia digestion, respectively. M_b is the analyte metal concentration in the blank extract (µg L^{-1}) and m is the total weight of the landfill sample (g) being digested.

Relative standard deviations of the concentrations of each metal in samples collected from each and across all four landfill sites were calculated to assess concentration variation.

Total solids (TS) and volatile solids (VS) were determined on all samples according to the Method 1648 of the U.S. Environmental Protection Agency (U.S. Environmental Protection Agency, 2001).

Analysis of the possible relation between volatile solids content and depth of waste with metal concentration was made through visual assessment of scatter plots and by calculating the correlation coefficient (or Pearson coefficient).

3. Results and discussions

3.1. Critical metals concentration across the four landfill sites

Fig. 1 shows the average concentrations of the extracted metals from the four LFS. Ce, Nd, Li, Sb and Co were the most abundant metals retrieved across the landfill sites. The two step extraction process further showed that for most of the metals investigated more than 90% of the content was extracted by the first digestion (HNO_3), except for Ce and Sb. Sb is relatively stable in dissolved HCl making it soluble in contact with this acid (Hashimoto et al., 2003), whereas in contact with HNO_3 Sb is less stable and oxidizes avoiding solubilisation (Ahrland and Bovin, 1974).

Average of metal concentrations at each LFS along with their RSD are summarised in Table 2. REEs concentrations in each landfill site were similar and did not present significant variations.

PGMs concentrations were low in all four landfills, with the exception of Pd in Site A yielded an average concentration of 0.8 mg kg^{-1}, which was twice as much as any of the other analysed landfills. Additionally, it is observed that concentrations of Pt and Ru vary most significantly across all samples, indicated by large RSD. This variability of results is likely due to limitation of the analytical instrument, as the levels of metal concentration are close to the detection limit of ICP-MS.

For the other critical metals, Li had the highest concentration with 20 ± 11 mg kg^{-1}, followed by Co, Sb and In. The concentrations presented high variability within the LFSs and between each other providing wide ranges of possible concentrations. Despite the wide intervals found for Li, In, Sb and Co concentrations, their ranges at each LFS intersect with each other providing confidence of the presence of these metals in the landfills.

Other metals including Cu, Ag and Au their concentrations were highly variable across the LFS which can be attributed to specific solid waste input rich in those metals, like jewellery or cutlery (Muchova et al., 2011; Morf et al., 2013), which would significantly increase their concentration.

Site D, the oldest site, was expected to have higher concentrations of critical metals since it has stored MSW and commercial and industrial (C&I) waste for several periods where less segregation existed. Also, for almost 20 years it received mainly C&I wastes which in large quantities are a great source of REEs (Binnemans et al., 2013). However, it had a period without receiving any type
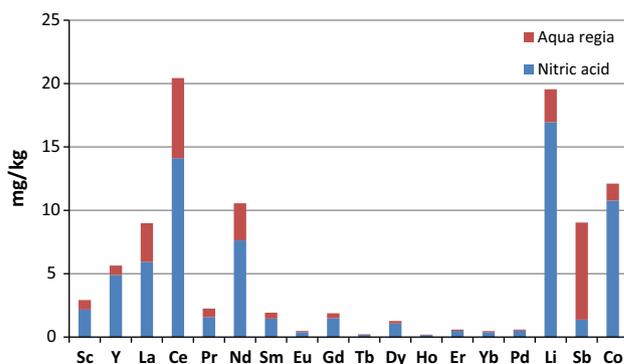

Fig. 1. Average concentration of metals extracted from the four sampled landfills. Lu, Tm, Pt, Ru and In were not included since their total extractions were below 0.06 mg kg^{-1}.

Table 2
Average content and relative standard deviation (RSD) of REEs, PGMs, critical metals and others found in the four UK closed landfills.

Metal	Site A		Site B		Site C		Site D	
	mg/kg	RSD (%)	mg/kg	RSD (%)	mg/kg	RSD (%)	mg/kg	RSD (%)
REEs								
Sc	3.46	36	2.02	23	3.27	57	1.81	25
Y	6.42	26	5.02	24	5.78	59	3.78	28
La	9.36	22	8.14	20	10.77	58	6.74	22
Ce	21.38	26	18.90	26	25.20	57	13.85	31
Pr	2.39	24	2.08	22	2.49	54	1.73	25
Nd	11.75	27	9.64	18	10.14	50	8,34	23
Sm	2.06	26	1.90	19	2.07	56	1.38	32
Eu	0.59	28	0.42	17	0.43	49	0.31	36
Gd	2.07	29	1.77	24	1.98	57	1.27	29
Tb	0.24	27	0.21	25	0.23	56	0.15	27
Dy	1.44	29	1.15	25	1.26	58	0.87	26
Ho	0.21	28	0.19	25	0.21	59	0.13	29
Er	0.65	27	0.55	24	0.62	59	0.41	27
Tm	0.08	27	0.06	23	0.07	59	0.05	29
Yb	0.52	27	0.42	23	0.52	59	0.33	27
Lu	0.07	29	0.056	21	0.06	58	0.04	30
PGMs								
Pt	0.02	71	0.03	114	0.01	52	0.02	68
Pd	0.77	81	0.41	46	0.45	25	0.41	38
Ru	0.003	145	0.0005	152	0.0013	71	0.002	88
Other critical								
Li	21.90	54	11.17	29	27.66	54	11.86	34
In	0.10	69	0.05	84	0.04	36	0.06	61
Sb	7.71	53	6.40	118	9.51	39	15.15	77
Co	14.14	32	13.05	105	8.83	28	8.72	35
Others								
Cu	1076	131	1027	103	2595	218	1830	67
Ag	2.26	68	2.77	71	3.63	179	5.02	158
Au	0.18	193	0.13	118	0.16	111	0.05	65
Al	17,274	48	12,357	43	12,594	63	12,079	42

of waste (Table 2) and only for 4 years it mainly received MSW, which could have an effect on the retention of metals.

Site B mostly contains MSW and Site C accepted both MSW and C&I, while Site A, which had the higher concentrations of critical metals, only for one year received mainly C&I waste (2013–2014).

Based on the types of waste each landfill received, it appears that a specific mixture of organic and inorganic materials must be achieved so the metals of interest can be retained in the organic-like component (soil) of the landfill. However further analysis is required to consider the specifications of where the samples were taken (sampled cell, type of waste mainly buried in place, age of waste, etc.) to assess how waste composition of a landfill site affects critical metal content and better understand the major source of these metals.

Other studies where critical metal analysis was carried out in waste samples from landfill sites could not be found. However Morf et al. (2013) analysed the presence of some REEs, PGMs and other metals in MSW used as fuel in an incinerator in Switzerland. Furthermore, James (2011) assessed the quantity of REEs and PGMs in landfill leachate and its sediment. Fig. 2 shows the concentrations of selected metals (REEs, PGMs and precious metals) found in the previous mentioned studies and compare them against the total average values obtained from the four UK landfill sites.

Concentrations of Sc, Y, Pr, Nd and Gd were consistent across MSW and landfilled material (Fig. 2). Values ranged between 1 and 3 mg kg^{-1} and could imply that these metals are not lost within the reactions of the landfill (e.g. mobilised by leachate) (Gould et al., 1990). However, even though no significant concentrations were found in landfill leachate for REEs and PGMs (James, 2011), the sediment of the leachate showed presence of

Y, La, Ce, Nd and Pd. La and Ce content was more than twice the content in landfilled waste with 4.8 mg kg^{-1} and 11.5 mg kg^{-1}, respectively, while for Nd and Y, its solubility in leachate sediment is lower (significantly for Y). This suggests that La, Ce and Nd concentrations can be reduced by almost half of their content in the landfill by leachate. The use of leachate to recover materials of value from landfill sites is an example of in-situ landfill mining, as mentioned previously. However further studies are required where the average content of critical metals in the landfilled waste and in the leachate (both liquid and sediment) are analysed to assess the possibility of leachate to dissolve and transport critical metals within the landfill.

Morf et al. were not able to determine the concentration of Pd in MSW due to several interferences within the analysis (Morf et al., 2013) but from the values obtained in this work and from James (2011) it can be established that Pd content will not be high enough to identify landfills as a source for recovery. PGMs concentration of 0.61 mg kg^{-1} was found in landfilled material of which 97% was Pd. In contrast the concentration of PGMs in leachate was 0.72 mg kg^{-1} and the concentration of Pd was 1.2 mg kg^{-1} in the leachate sediment. This finding suggests that Pd is mobilised and washed out from the waste material by the leachate then accumulate/adsorb in its sediment phase.

Furthermore, James (2011) found only Ag and Au in relatively high concentrations in the leachate compared to the rest of the analysed metals. It is worth noting that almost half of the concentrations of these metals found in MSW disposed seem to leach out of the landfill.

3.2. Distribution of critical metals within the landfill

A relationship between metal content and the depth of the waste was investigated. Fig. 3 shows the presence of REEs and other critical metals found in relatively high quantities at the different depths.

For all metals the correlation coefficients were low ($R \leqslant 0.2$) indicating that there is no relationship between concentration of metals and the depth.

Previous studies have assessed the capacity of leachate to mobilise metals out of the landfilled waste. Although these studies only focus on heavy metals (Cr, Hg, Ni, Cd and Zn) and on the possibility of leachate to dissolve high concentrations of these metals, they found that metals mobility increases with pH below 6 values and mostly they were retained in the organic compounds (Gould et al., 1990; Bozkurt et al., 1999; Esakku et al., 2003). Also, it is known that acid or alkaline solutions are used in practice to solubilise REEs, PGMs and other metals from minerals due to their very high or low pH values (Xie et al., 2014; Crundwell, 2014a,b). The fact that no direct relationship between depth and concentration of critical metals was found could imply that leachate (which presents neutral pH of around 7 units) is not vertically mobilising these metals through the landfill and that metals are being retained between soil layers.

Furthermore, measurements of the TS and VS were carried out to establish a general composition of the samples and to assess whether there is a correlation between metal content and volatile solids. Results from each LFS were similar between each other and have no major variations. Average TS content in each LFS was 58 ± 7% for Site A, 79 ± 9% for Site B, 63 ± 6% for Site C and 66 ± 9% for site (Fig. 4). From all samples, the TS content was above 44% and below 99% resulting in average moisture content of 36 ± 10%. Results are within the range of what has been reported in the literature; studies where waste extracted from several landfills was analysed for possible material recovery had in average between 50% and 84% TS content (Kaartinen et al., 2013a,b; Prechthai et al., 2008).
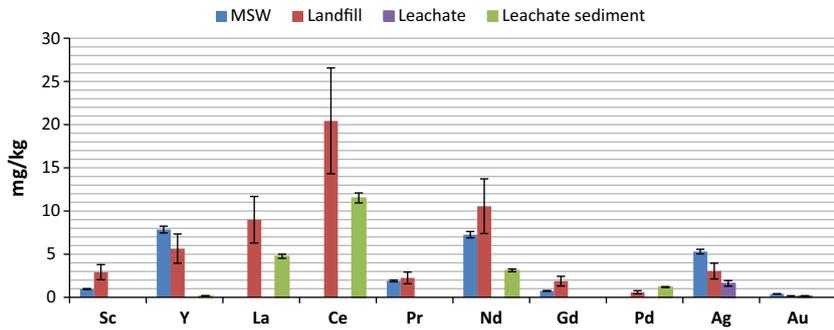
Fig. 2. Average concentration of selected metals present in MSW (Morf et al., 2013), landfill, landfill leachate and sediment of landfill leachate (James, 2011). Landfill category refers to values obtained in this work.
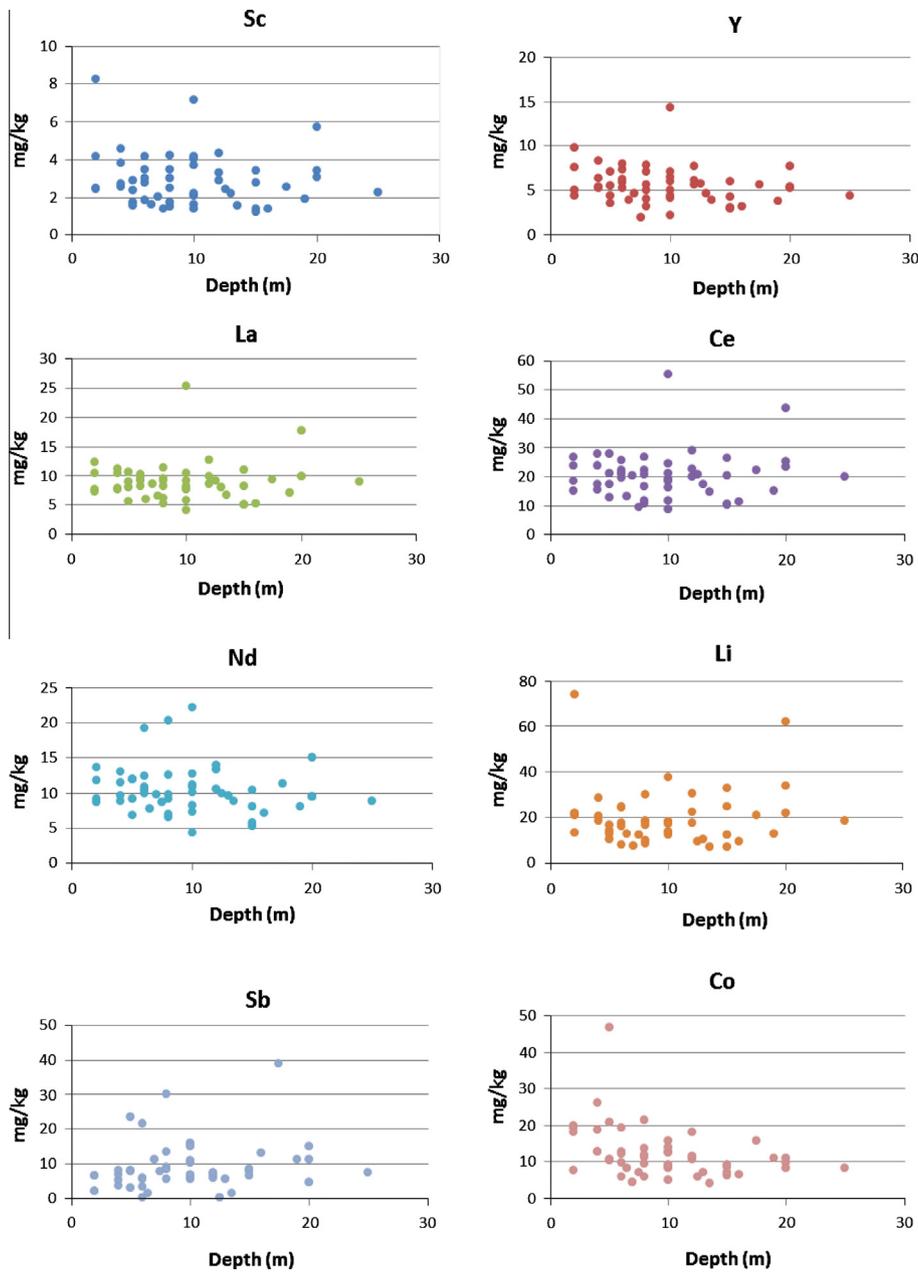

Fig. 3. Concentrations of selected critical metals at different depths in a LFS.
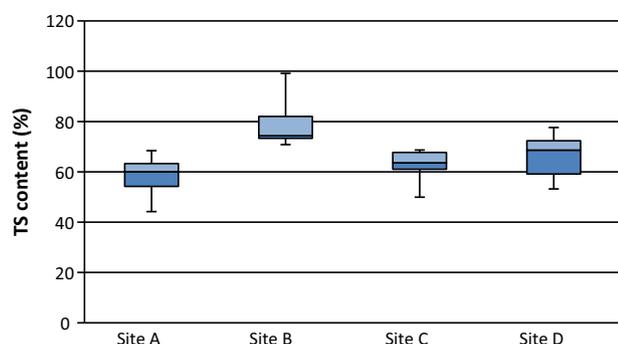
Fig. 4. Total solids content of the landfilled material from each landfill site.

VS content resulted in 41 ± 9%, 25 ± 10%, 32 ± 11% and 34 ± 12% for Site A, Site B, Site C and Site D, respectively. Metals concentrations and VS content were compared to assess the existence of a relationship between each other. The correlation coefficients ($R \leqslant 0.20$) observed indicates that the concentration of metals is unaffected by the content of VS content. An increase in the VS content does not increase or decrease the metal content in the landfilled material and there is not a specific percentage range of VS that presents higher concentrations the critical metals.

Even though the results could not demonstrate that higher VS content results in an increase in the concentration of critical metals within the landfilled material, this does not mean that the organic matter does not affect the retention of these metals in the landfill. VS content analysis takes into account plastics, paper and other materials that are within the sample and so it cannot be assumed that all VS account for the organic compounds that can retain metals. Organic matter and microbial ecology are very important in the absorption and mobilisation of metals (Bradl et al., 2005; Bozkurt et al., 1999) and so further work on microbial activity is required.

3.3. Opportunities for metal recovery

The total amount of metals that could be recovered from the four landfills was calculated using the quantity of wastes buried (or disposed of) in each LFS. Reviewing the market price of the obtained metals an estimated value by assuming a high degree of purity (>99%) was then calculated (Table 3). The most valuable metals are the commonly known precious metals including Pt, Au, Pd and Ru. Despite only 5 tonnes of PGMs estimated from the four landfills sites there is a good opportunity of significant revenue of $148,246,000. For the REEs, Nd would present the highest value with $9,183,000 followed by Dy, Eu, Pr and Y, while for the other critical metals; Li would generate more revenue with $13,722,000 and Co having a value of $3,370,000.

Nevertheless caution is needed in extrapolating these potential revenues as at the end of the extraction process the metals are concentrated in acid solution which must be treated in order to separate the metals of interest. Therefore, the viability of reusing these metals is restrained to the costs of extraction and treatment and the degree of purity of the metal (DEFRA, 2012). Achieving a level of purity above 99% becomes a major challenge; therefore the cost of recovering these critical metals individually may exceed the theoretical gross value estimated in the landfill. Table 4 summarises and compares the activities of extraction of materials from landfill with the activities of conventional mining. Although some of the activities are similar for both cases, LFM will involve a greater risk of environmental impact due to gas release. Therefore LFM will result in higher extraction costs. Considering obtaining only critical metals as the marketable product from both scenarios it can be

assumed that the revenues from LFM the four studied landfills are not enough to be economically viable.

Other metals such as Cu, Al, Ag, Au, Ni and Zn could be used to increase and supplement the economic value of LFM counteracting the losses of extracting single critical metals.

With regards to the key challenges in metal recovery the metal content depends mostly on waste composition which varies between landfills and could make difficult to ensure a typical concentration of critical metals for landfills. Nonetheless, Krook et al. (2012) found some patterns in the composition of landfilled waste; 50–60% weight of soil-type material, 20–30% weight combustibles, 10% weight inorganic materials and less weight percent for metals (Krook et al., 2012), which help to identify patterns in metal content. Also, as shown in Table 2, major differences in metal concentrations between landfills were found for the metals not considered as critical which are more frequently used and at different quantities.

Another concern is in proving that the recovery of these metals from a landfill site is economically viable. As stated previously, due to the low concentrations of critical metals the only way to make LFM economically viable is to target more materials to recover, specifically refuse-derived fuels (RDF) for energy recovery and materials as part of an ELFM approach. It has been previously suggested that the only materials from a landfill which can provide enough profits are the ones that can be used as supplementary waste fuel due to the clear market for energy (Krook et al., 2012; Van Passel et al., 2013; Chapman et al., 2010, 2011). The use of RDF sourced from landfilled materials in advanced thermal conversion processes has been investigated (Bosmans et al., 2014), highlighting the attractive potential for upcycling excavated materials as an alternative to conventional energy recovery. Therefore, the recovery of this material for energy recovery and/or upcycling together with critical metals would increase the economic benefits and viability of the project.

Van Passel et al. (2013) states that a substantial economic potential exists for landfill mining projects and describes several economic methodologies for exploring the economic potential of landfill mining (Van Passel et al., 2013), including private and social costs and benefits, where the inclusion of recovered critical metals definitely increases the profits. Furthermore, the technology performance (efficiency) is an uncertainty that keeps the companies out of these projects (Krook et al., 2012). Recovered landfilled waste must pass through a screening process to obtain the soil-type fraction where the critical metals are extracted. Therefore, technologies for excavation and materials processing are required to extract and segregate the waste from the landfill site efficiently in order to obtain a relatively clean sub-product (soil-type fraction) and marketable materials (recyclables). Technologies including wind shifters and magnet separation have not been proven to be very efficient for separating the waste from the soil-type in good quality sub-products. This indicates that more advanced stationary processing plants would be required instead or tailored separation techniques must be developed to achieve a high level of quality in the excavated materials (Krook et al., 2012; Jones et al., 2013).

Once the soil-type material has been separated, the critical metals will be extracted. Selection of the most efficient solution will be crucial for recovering the metals of interest. According to their chemical properties the metals have more solubility in certain leaching solutions and it is important that the selected solution used for extracting the metals will recover as much amount as possible (Spooren et al., 2013). Additionally, high purity single critical metals compounds must then be separated from the leaching solution. Individual REEs are very difficult to obtain since they possess similar properties (both chemical and physical). Techniques such as ion-exchange and solvent extraction have been developed to

Table 3
Total average concentration of analysed metals in a landfill site, ores typical metal content, market prices and estimated value of their concentration found in the four landfills.

Metal	LFS concentration (mg/kg)	Ore concentration (mg/kg)	Price (US$ kg^{-1})[a]	Reference	Estimated tonnage	Potential value (US$)
Li	19.55	–	63.37		216.54	13,721,938
In	0.07 ± 0.05	1–100	705	Metal-Pages Ldt. (2014)	0.54	377,625
Sb	9.04 ± 6.15	27,000	9.20	Metal-Pages Ldt. (2014)	94.01	864,858
Co	12.11 ± 6.06	500–3000	33.00	The London Metal Exchange (2014)	102.11	3,369,724
Sc	2.92 ± 1.14	20–130,000	–	–	–	–
Y	5.64 ± 1.86	2600–180,000	60.00	Metal-Pages Ldt. (2014)	54.03	3,241,736
La	8.98 ± 2.61	18,000–300,000	10.00	Metal-Pages Ldt. (2014)	93.12	931,230
Ce	20.43 ± 6.54	1600–33,000	11.00	Metal-Pages Ldt. (2014)	213.75	2,351,297
Pr	2.25 ± 0.61	6000–98,000	150	Metal-Pages Ldt. (2014)	22.50	3,374,352
Nd	1.93 ± 0.58	690–16,000	92.50	Metal-Pages Ldt. (2014)	99.27	9,182,590
Sm	0.49 ± 0.17	220–1100	26.50	Metal-Pages Ldt. (2014)	18.99	503,200
Eu	1.88 ± 0.62	650–21,000	1000	Metal-Pages Ldt. (2014)	4.36	4,364,719
Gd	2.92 ± 1.14	20–130,000	133	Metal-Pages Ldt. (2014)	18.24	2,416,433
Tb	0.22 ± 0.07	43–5300	820	Metal-Pages Ldt. (2014)	2.11	1,733,833
Dy	1.26 ± 0.43	430–47,000	475	Metal-Pages Ldt. (2014)	11.97	5,685,171
Ho	0.19 ± 0.07	16–11,000	–	–	–	–
Er	0.59 ± 0.19	13–29,000	46.80	MetalPrices.com (2014)	5.74	268,601
Tm	0.07 ± 0.02	3–4900	–	–	–	–
Yb	0.47 ± 0.16	3–34,000	–	–	–	–
Lu	0.06 ± 0.02	<2200	–	–	–	–
Pt	0.02 ± 1.78	2–4	46,945	The London Metal Exchange (2014)	0.15	6,875,315
Pd	0.59 ± 0.39	2–7	28,296	MetalPrices.com (2014)	4.99	141,333,181
Ru	0.002 ± 0.004	0.3–0.7	2196	MetalPrices.com (2014)	0.02	37,536
Ag	3.05	–	752	MetalPrices.com (2014)	33.55	25,232,782
Au	0.15 ± 0.25	5–30	44,530	MetalPrices.com (2014)	1.41	62,938,638
Al	14,768 ± 5021	20–130,000	2.032	The London Metal Exchange (2014)	132,308	268,850,383
Cu	1468 ± 2539	5000–20,000	7.01	The London Metal Exchange (2014)	19,170	134,397,036

[a] US$ kg^{-1} of metal with approximately 99% purity.

Table 4
Comparison of the activities involved in landfill mining and normal mining.

Landfill mining	Mining
Identification of suitable sites for mining	Identification of the site (exploration, analysis, etc.)
Capital costs	*Capital costs*
Site preparation	Site preparation
Rental or purchase of reclamation equipment (diggers, lorries, conveyer belts, etc.)	Rental or purchase of reclamation equipment (excavators, dozer drills, wheel loader, cutting machine, mills, etc.)
Rental or purchase of personnel safety equipment	Rental or purchase of personnel safety equipment
Rental or purchase of environmental protection equipment (mitigation equipment)	Rental or purchase of environmental protection equipment (mitigation equipment)
Construction or expansion of materials handling facilities (sorting, shredding, washing, etc.)	Construction of processing plants
Rental or purchase of hauling equipment	Haul roads/site work
Permitting fees	Electrical system
Final restoration costs	Rental or purchase of hauling equipment
	Permitting fees
	Restoration costs
	Contingency
Operational costs	*Operational costs*
Labour (e.g. monitoring, testing, equipment operation and materials handling)	Labour (e.g. monitoring, testing, equipment operation)
Equipment fuel and maintenance	Supplies and materials (e.g. explosives)
Energy	Administration
Administrative and regulatory compliance expenses	Equipment fuel and maintenance
Worker training in safety procedures	Energy
Hauling costs	Worker training in safety procedures
Landfilling residual waste which cannot be reclaimed	Hauling costs
Insurance	Waste management

separate these metals, where solvent extraction is the most accepted since is easier to use in large volumes. After solvent extraction the remaining will be a solution containing the individual metals of interest but may still have the presence of impurities that must also be removed by other chemical or physical processes (Xie et al., 2014).

Finally, risks imposed by excavating and extracting material from a landfill concerns the government and local population. Risks such as the leaching of hazardous substances, slope stability issues, air ingress and explosive conditions as well as fugitive emission of poisonous gases such as hydrogen sulphide have attracted mainstream attention. This has created, due to a lack of regulatory measures of the subject, a demand for policies that can secure the safety of the population. Also, possible long-term effects that landfill mining may impose have not been fully studied restraining these projects since companies and governments cannot ensure safety of the population and the environment (Krook et al., 2012).

4. Conclusions

From the four landfill sites analysed, concentrations of REEs in each landfill do not differ significantly between each other and overall Ce was the most abundant rare metal. PGMs concentrations ranged between 0.36 and 0.70 mg kg^{-1}, Pd was the most abundant PGM. Significant quantities of Al and Cu were observed.

Analysis of the mobility of these critical metals through the landfill suggested that these metals are not being vertically transported completely since concentrations remained similar through the whole range of depth in the four LFSs. No clear pattern was identified between metal concentrations and the depth of the waste inferring that leachate does not significantly solubilises these metals and moves them through the landfill. Metal mobilisation in landfills is due to complex interactions between chemistry and microbiology, and in the case of the metals assessed in this study further work is required to fully understand these effects.

It is suggested that the recovery of only critical metals may not be economically viable. However the recovery of these materials along with other valuable metals (Al, Cu, Ag and Au) and additional materials of value, such as those suitable for use in RDF, may result in a viable business model. It is worth noting that the studied landfills contain mixed MSW and C&I waste which makes them less rich in critical metal, while mono-landfills of industrial waste could present higher concentrations of these metals which extraction may not require the recovery of other valuable materials.

Acknowledgments

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