INVESTIGATION OF MUNICIPAL SOLID WASTE (MSW) AND INDUSTRIAL LANDFILLS AS A POTENTIAL SOURCE OF SECONDARY RAW MATERIALS

Heikki Särkkä [1,*], Tommi Kaartinen [2], Esa Hannus [1], Sami Hirvonen [3], Tuire Valjus [4], Jouni Lerssi [4], Giovanna A. Dino [5], Piergiorgio Rossetti [5], Zoe Griffiths [6], Stuart T. Wagland [6] and Frederic Coulon [6,*]

[1] South-Eastern Finland University of Applied Sciences, Patteristonkatu 3 D, 50101 Mikkeli, Finland
[2] VTT Technical Research Centre of Finland, Biologinkuja 7, 02150 Espoo, Finland
[3] Metsäsairila Oy, Arkistokatu 12, 50100 Mikkeli, Finland
[4] Geological Survey of Finland (GTK) Po.Box 96, 02151 Espoo, Finland
[5] University of Torino, Earth Science Department, via Valperga Caluso 35, 10125 Torino, Italy
[6] Cranfield University, School of Water, Energy and Environment, Cranfield, MK430AL, United Kingdom

ABSTRACT
Many of the secondary raw materials (SRM) in landfills constitute valuable and scarce natural resources. It has already been recognised that the recovery of these elements is critical for the sustainability of a number of industries and SRM recovery from anthropogenic waste deposits represents a significant opportunity. In this study, the characterisation of the different waste fractions and the amount of SRM that can potentially be recovered from two landfill sites in Finland is presented. The first site was a municipal solid waste (MSW) landfill site and it was specifically investigated for its metals, SRM, plastics, wood, paper, and cardboard content as well as its fine fraction (<20 mm). The second site was an industrial landfill site containing residual wastes from industrial processes including 1) aluminium salt slag from refining process of aluminium scrap and 2) shredding residues from automobiles, household appliances and other metals containing waste. This site was investigated for its metals and SRM recovery potential as well as its fine fraction. Results suggest that the fine fraction offers opportunities for metal (Cr, Cu, Ni, Pb, and Zn) and SRM extraction and recovery from both landfill site types while the chemical composition of the industrial waste landfill offered greater opporutinity as it was comparable to typical aluminium salt slags. Nevertheless, the concentrations of rare earth metals (REE) and other valuable elements were low even in comparison with the concentrations found in the Earth's crust. Therefore mining landfill sites only for their metals or SRM content is not expected to be financially viable. However, other opportunities, such as waste-derived fuels from excavated materials especially at MSW landfill sites, still exists and fosters the application and feasibility of landfill mining.

1. INTRODUCTION

The issue of resource security has come to the forefront of the debate as Critical Raw Materials (CRM) and Secondary Raw Materials (SRM) supply is fundamental to maintain and develop EU economy. For example, recent studies predicted that the depletion of silver, copper and platinum group metals (PGM´s) reserves will occur in the next 17-30 years and a peak in supply will be required within the next 20-35 (Sverdrup et al., 2014a,b; Sverdrup and Ragnarsdottir 2016). Sverdrup and Ragnarsdottir (2016) further stressed the importance of metals recycling, metal conservation and elimination of dissipative losses so that the society can become more sustainable with respect to metals supply. For PGM alone, they predict that extraction will reach a maximum in the period 2020–2050 and that market supply will peak in 2070–2080. For copper, the peak production estimates are much closer, from 2031 to 2042. In a longer perspective, taking into account price and recycling, the supply of copper to society is expected to run out sometime after 2400 (Sverdrup et al., 2014b). Thus, considering the increasing scarcity and raising prices of SRM, their recovery from anthropogenic deposits such as urban and mine

wastes disposal sites is essential. In fact a great amount of waste can be regained as practical and valuable SRM by enhancing the recovery processes from industrial, extractive and municipal solid waste (MSW) landfill sites especially if we consider that Europe is highly dependent on the imports of certain raw materials including rare earth elements (REE) and SRM. Europe has between 150,000 and 500,000 landfill sites, with an estimated 90% of them being "non-sanitary" landfills, pre-dating the EU Landfill Directive of 1999 (Jones et al., 2013). These older landfills tend to contain municipal solid waste and often lack any environmental protection technology. To avoid future environmental and health problems, many of these landfills will soon require expensive remediation measures. This situation does present us with an exciting opportunity for a combined resource-recovery and remediation strategy, which will drastically reduce future remediation costs, reclaim valuable land, while at the same time unlocking billions of tonnes of valuable resources contained within these landfills (Gutiérrez-Gutiérrez et al., 2015; Dino et al., 2016; Dino et al., 2017). There is however to date no inventory available of SRM and CRM present in EU landfills. There has been only very limited knowledge around best practice and how to manage the excavation and recovery of valuable materials.

There are still challenges in enhanced landfill mining (ELFM). We need to understand more about each of the stages involved: the exploration, separation, transformation and up cycling technologies, and how these can be applied in the best way in dealing with the differing urban and industrial landfill sites. For instance, to recover recyclable materials such as metals and plastics, we need to consider their chemical degradation as they may not be suitable for conventional recycling. The recyclable materials are in a moist environment and emerge with soil/clay covering and attachments. As such, we need to find the best cleaning approaches.

There are also policy challenges in establishing legal frameworks for ELFM but concerted action is underway to overcome them (EURELCO, 2017). The potential of ELFM was presented to the European Parliament last year. It has received backing from the the European Commission in May 2017 by acknowledging in their 'Closing the Loop – EU Action Plan for the Circular Economy' that an increment in reuse and recycling of key waste streams has to be undertaken and made a specific reference to ELFM. When considering ELFM, some actions should be forecasted to asses the sustainability of the mining opportunity: (i) the estimation of the amount of types of waste materials; (ii) the characterisation and localization of the different wastes present in landfill; (iii) their potential recovery and treatability for their utilization (Kaartinen et al., 2013). Burlakovs et al. (2017) further critically summarised LFM challenges from historical sites and driving paradigms of LFM from 'classical hunting for valuables' to 'perspective in ecosystem revitalization'. Krook et al. (2012) also reviewed the main trends, objectives, topics and findings of 39 research papers on landfill mining published during the period 1988–2008. Together with a follow up review by Krook and Baas (2013), they conclude that urban mining and landfill mining show high potential but the state-of-the-art

is still too theoretical and requires large pilot scale demonstration to assess and demonstrate the performance of such activities in practice.

Several materials and energy fractions can be exploited from landfills. Wagner and Raymond (2015) are among the first authors to present an economically succesful LFM case study where metals were recovered. In another study, Leme et al. (2014) showed that the integration of waste-to-energy plant to a LFM project is highly dependent on the MSW treatment fees due to high installation, operation and maintenance costs. Quaghebeur et al. (2013) investigated different valorization options for the excavated materials from the REMO site in Houthalen (Belgium). The results showed differences in the composition and the characteristics of the waste materials with regard to type of waste (MSW versus IW) and the period during which the waste was stored. For the plastics, paper/cardboard, wood and textile recovered, waste-to-energy was the most suitable valorization route. Quaghebeur et al. (2013) also concluded that metals, glass/ceramics, stones and other inert materials can be valorized as well if the materials will be separated from each other. The amount of combustibles in the excavated waste confirmed the large potential of waste-to-energy for landfill mining. Fine fractions from the industrial waste contained high concentrations of heavy metals (Cu, Cr, Ni and Zn) and thus offer opportunities for metal extraction and recovery. Also the biochemical methane potential (BMP) of the fine fractions from two Finnish landfills have been investigated and showed good opportunities for further energy from waste recovery (Sormunen et al., 2008; Mönkäre et al., 2016). The present paper presents two ELFM pilot case studies carried out in Finland. One of the site was a MSW landfill site (Metsäsairila) and the other one an industrial waste (IW) landfill site (Vierumäki). Detailed site investigation of the two sites was carried out to evaluate the potential SRM resources that can be exploited from different landfills. The described characterisation was part of a wider activity related to the Smart Ground H2020 project (Grant number 641988) which aims, together with other objectives, to foster resource recovery from both urban solid waste landfill sites and mine waste disposal sites by (i) improving the availability and the accessibility of data and information on SRM amount in EU anthropogenic deposits and (ii) integrating data from existing databases and new information collected into a single EU database.

2. MATERIALS AND METHODS

2.1 Landfill sites description

The first site, Metsäsairila, is a MSW landfill site located in the South-Eastern region of Finland, nearby the City of Mikkeli. MSW buried in the site is collected from approximately 55 000 inhabitants. The site has been operating since beginning of 1970´s and is divided in two distinct cell areas: a closed one and an active operational one. The active cell area is located in the northern part of the landfill. The active cell is membrane-lined with a mixture of bentonite and moraine on the bottom structure; in contrast the closed area is located on swamp. Both active and closed cells have collection system for leachate. Landfill

gas which is mainly collected from the closed cell and used for combined heat and electricity production on site. The height of the waste filling was estimated to be around 20-25 meters in the closed cell and between 6 and 10 m in the active cell. The closed cell is currently being capped with a layer of clay and silt moraine and will be completed in 2018. The surface area of the closed cells is around 8 ha while the active cells surface area is around 3 ha. The active area has received waste since 2007. The second site, Kuusakoski Oy's, is an industrial landfill site located in Vierumäki, southern Finland. The site started receiving waste in 1974 and has been closed in three stages in 1989, 1990 and 1991. The wastes disposed of in the landfill are residues from industrial processes including 1) aluminium salt slag from refining process of aluminium scrap and 2) shredding residues from automobiles, household appliances and other metals containing waste. The area of site is estimated to be approximately 2.5 hectares. Typical to a landfill of this age, there are no engineered bottom isolation layers at the landfill, and a peat layer has been used as a compacting bottom structure. The height of the waste filling was estimated to be ranging between 5 to 8 meters. After completion, the waste was covered with a layer of clay functioning as a sealing layer, moraine and a layer for vegetation. Today, the landfill site is reminiscent to a typical young forest.

2.2 Sampling, sorting and analysis of collected samples

Geophysics characterization was carried out at Metsäsairila landfill site as described previously by Lahti et al. (2005). By using geophysics it was possible to direct the sampling to the most appropriate points of the landfill site and also to get broader information of the physical properties of the landfill material. The geophysics characterization was carried out only in the closed cell area as in the active cell it was too many confounding factors to make the geophysical field measurements. Electrical resistivity tomography (ERT), Induzed polarization tomography (IPT), Magnetic and Electromagnetic (EM) methods were used together in order to get the best result in searching the metal containing areas (Lerssi et al., 2016). Gravity method was used to determine the bedrock level and also the thickness of the landfill material (Vanhala et al., 2005; Valli and Mattsson, 1998). By using gravity it was possible to determine the maximum drilling depth to avoid the damages on the landfill bottom. Five sampling points were then drilled by hydraulic piling rig in the areas with the highest conductivity and total magnetic intensity (Figure 1). Samples with codes DH1, DH2a and DH3 were from the closed part of the landfill site and samples with codes DH6 and DH7 were from the currently operational part of the landfill for waste disposal.

The amount of waste materials collected at each sampling point is summarized in Table 1. Samples were moved to sorting point where they were manually sorted by sieves to different particle size categories (>100 mm, 20-100 mm and <20mm) and waste fractions (metals, wood, paper, plastics, textile, soil and others). Waste fraction separation was done to fractions size of 20-100 mm and >100 mm.

FIGURE 1: Topographic map of Metsäsairila MSW landfill site obtained using laser scanning based DEM with 2 metres resolution overlaid with aerial photography (orange dots: location of the sampling points).

Analysis of the fine material samples (<20mm) for critical raw materials (CRMs) content was carried out by an external laboratory (ALS Finland Oy, Finland). Reference method used was based on US EPA 200.8, CSN EN ISO 17294-2 and US EPA 6020 (measurements were done by inductively coupled plasma mass spectrometry (ICP-MS).

Unmanned Aerial Vehicle (UAV) photogrammetry survey of the Vierumäki industrial landfill site was conducted for visualisation of topography before the physical exploration of site was carried out. Topographic and morphologic 3D characterization of the site will obtain a detailed reconstruction of the topographic surface. This will give better overview about structure and composition of the investigated pilot site. Photogrammetry is a viable alternative for calculating landfill volume which is useful for the SRM´s volume evaluation on site. Figure 2 shows an orthophoto of the Vierumäki industrial landfill site with cell size of 5 x 5 cm.

Sampling at Vierumäki industrial landfill site was done with an excavator from five sampling points to cover the landfill area as well as possible with limited amount of time and resources (Figure 3).

During the excavation, it was noticed, that the landfill had well defined layers which were attributed to the aluminum salt slag and the shredding residues (Figure 4). The

TABLE 1: Amounts of aggregate waste materials collected at the Metsäsairila MSW landfill site.

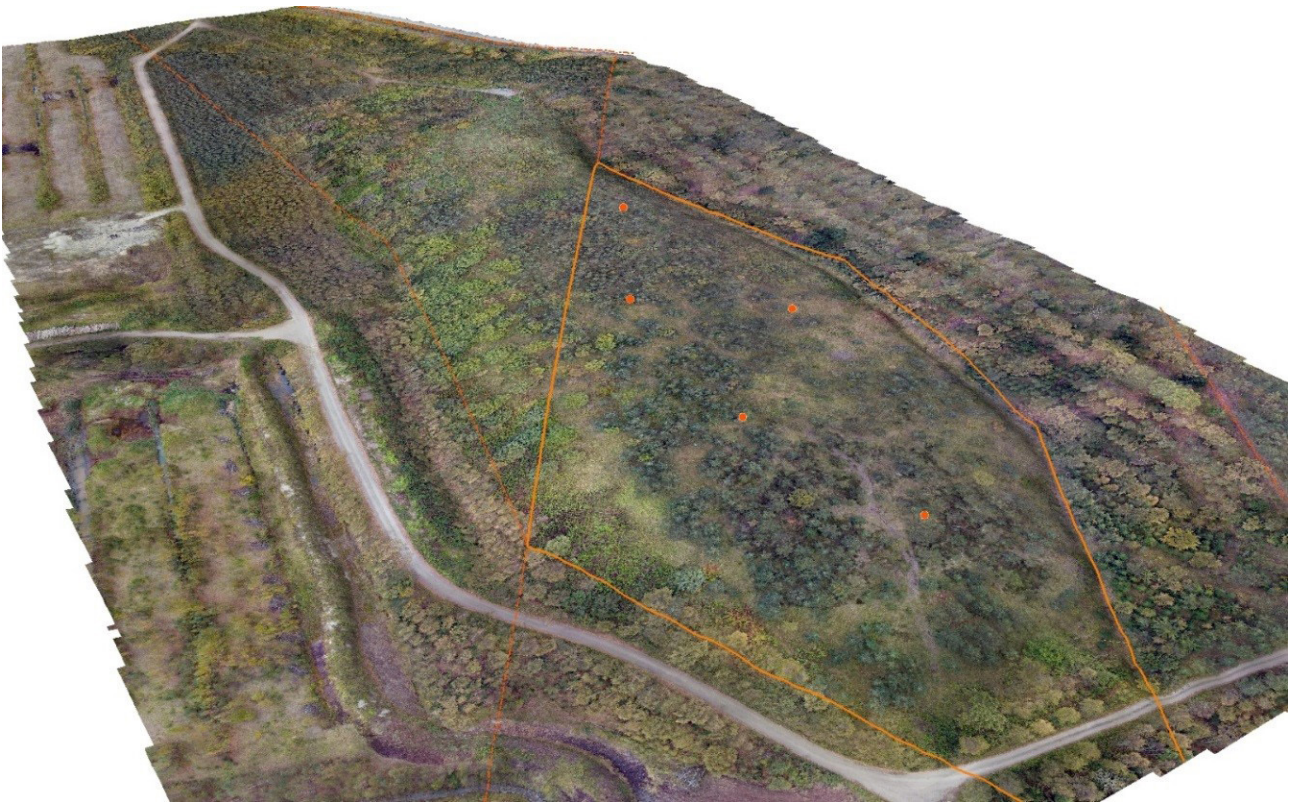| Sample ID	Sample depth (m)	Amount of aggregate waste materials (kg)
DH1	3.5-17	406.0
DH2a	3-12	192.3
DH3	2.5-10	277.4
DH6	0.2-5	282.2
DH7	0.2-5	284.4

FIGURE 2: 3D site topography based on DTM with 50 cm resolution and draped orthophoto.
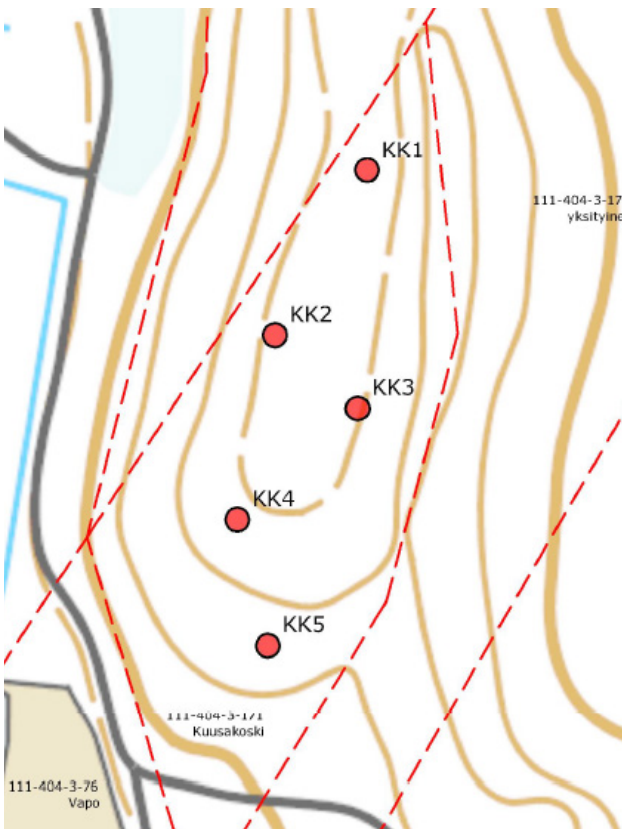

FIGURE 3: Locations of sampling points at the industrial landfill. Red dots show the sampling points and the dashed line surrounding the dots show the rough borders of the industrial landfill.

FIGURE 4: Landfill layers at the Vierumäki industrial landfill.

estimated height of each layer on each sampling spot was recorded to enable calculations of material amounts in the landfill.

After excavating the cover layers (moraine and clay) the two waste layers were mixed together to obtain a representative composite sample from the excavated waste materials as summarised in Table 2.

The composite samples were manually sieved to different particle size categories >100 mm, 20-100 mm and <20mm. The two largest particle size categories, >100 and 20-100 mm, were sorted to different waste fractions (metals, combustibles, soil and others). The fine fractions <20 mm and the combustible fractions (20-100 mm and >100 mm combined from each sampling point) were analysed for Al, Mg, Cu, Sb, Co and Cr by XRF. A composite sample of all fine fraction samples and one composite sample from all combustible samples were also analysed for Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Yb, Pt, Pd, Ru, In, Ag and Au by Aqua Regia dissolution and subsequent analysis by induced coupled plasma mass spectrometry (ICP-MS) as described in Kaartinen et al. (2013). In addition, the calorific values of the combustible samples were determined with a bomb calorimetry by ALS Finland Oy (European Standards, 2011). The calorific value is an important quality attribute as it indicates the amount of recoverable energy from waste.

3. RESULTS AND DISCUSSION

3.1 Metsäsairila MSW landfill site

The geophysical characterisation carried out at Metsäsairila provided significant new information of the landfill waste layers composition in both horizontal and vertical directions, especially for identifying the best locations for the presence of metals and determining the dimensions waste materials that should be excavated. Figure 5 shows the 3D ERT results together with the magnetic data and the bedrock topography interpreted from gravity data. The sampling places were selected within the areas with high magnetic intensity and electrical conductivity as it was indicating high metal content within the buried waste materials.

The mass distribution of the different waste fractions from the five core samples is summarised in Table 3. The mass distribution was relatively similar between the samples from the different sampling points despite their location and the aged of the buried waste (i.e. DH3 was located in the old closed area while DH6 was in the active area). Sorted fractions >100 mm and 20-100 mm which were combined from all wells together consisted mainly from
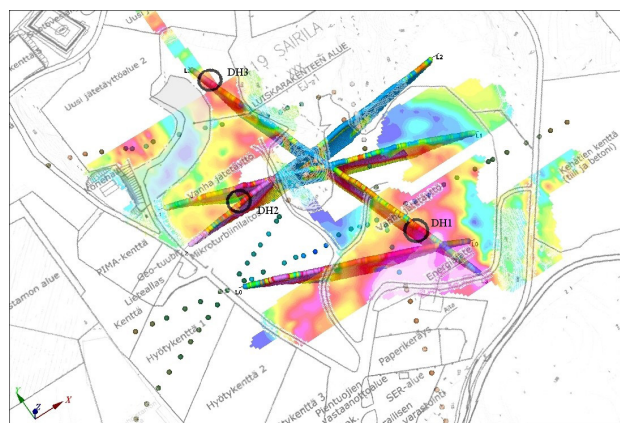
FIGURE 5: Electrical conductivity cross sections from 3D ERT profiles and total magnetic anomaly 2D map data with realized drill holes D1-D3 (sampling points). Interpreted bedrock topography of gravity profiles is visualized in the picture as coloured circles. In all the data the red colours show the high and blue colours show the low values.

energy fraction (plastic, paper, wood, cardboard, 76%), metals (5%), soil (17%) and others 2%. Results are following similar trend than research implemented by Kaartinen et al. (2013) in MSW landfill in Kuopio, Finland. They observed that amount of fine material (<20 mm) was found to be ca. 50% (w/w) which also supports previous reports of the amount of the fines (Quaghebeur et al., 2013). Fine material consisted mainly of landfilled wastes but also of the landfill cover materials (usually soil). In our study case fine fraction varied from 37% to 47% depending on the sampling point. Mainly the fine fraction included soil material but also small particles of plastic, paper and wood were present. The two main fractions were fine material (<20 mm) and the energy fraction comprised of wood, paper and cardboard, plastic and textiles. Sorted size fractions >100 mm and 20-100 mm from every sampling point had a similar waste distribution and main interesting fractions were the one considered for energy recovery and the fine material fraction (<20 mm).

The closed area of the Metsäsairila MSW site represents about 960 000 t of MSW of which metals account for 3.7% and the combustible energy fraction (wood, paper and cardboard, plastic and textiles) 42% (Table 4).

According to the analysis of metals and the fine fractions of the sorted samples, the excavated waste samples contained primarily Ba, Cr, Cu, Zn and Pb. Amounts of Ag, Au and In were rather low. The concentrations of heavy metals were lower than in the studies of Quaghebeur et al.

TABLE 2: Vertical distribution of the waste layers at Vierumäki industrial landfill.

Sampling point	KK1	KK3	KK4	KK5
Cover layers (m) (organic growth layer+moraine+clay)	0 – 1.8	0 – 1.0	0 – 1.0	0 - 1.0
Waste layer depth (m) from - to (in meters from ground)	1.8 – 5.5	1.0 - 3.5	1.0 – 5.0	0.8 - 4.5
Shredding residues layer (m) from - to	1.8 - 3.0	1.0 - 3.0	1.0 - 1.7	0.8 - 2.8
Aluminum salt slag layer (m) from - to	3.0 - 5.5	3.0 - 3.5	1.7 - 5.0	2.8 - 4.5
Mass of composite sample to manual sorting (kg)	531	252	288	242

TABLE 3: Weight distribution of different waste fractions in collected aggregate samples.

Waste fractions	DH1	DH2a	DH3	DH6	DH7	Average
>100 mm	**111.51**	**68.23**	**50.03**	**69.57**	**81**	**76.1**
metal	6.54	9.3	3.75	2.45	1.7	4.7
wood	8.9	11	3.4	5.06	13.6	8.4
paper and cardboard	8.15	11.92	4.27	5.52	8.8	7.7
plastic	44.2	30.4	30.96	41.2	27.8	34.9
textiles	13.92	4.38	5.73	8.99	28	12.2
soil	29.8	1.23	1.92	6.35	1.1	8.1
others	0	0	0	0	0	0
20-100 mm	**124.71**	**52.7**	**101.76**	**78.84**	**75.18**	**86.6**
metals	2.82	3.19	6.76	2.22	1.54	2.8
wood	25.49	8.02	12.6	20.9	14.52	13.6
paper and cardboard	12.37	6.77	12.5	8.7	10.5	8.5
plastic	30.2	19.8	36.4	20.5	14.44	20.2
textiles	18.11	4.07	4.41	2.8	5.2	5.8
soil	34	10.12	25	19.9	26.18	23
others	1.72	0.73	4.09	3.82	2.8	2.6
<20 mm	**169.8**	**71.4**	**125.6**	**133.8**	**128.2**	**125.8**
Total mass (kg)	**406.02**	**192.33**	**277.39**	**282.21**	**284.38**	**288.5**

TABLE 4: Estimated amounts of the different waste materials in the closed area of the Metsäsairila MSW landfill site.Uc iam te

	Average (%)	Estimated total amount (t)
Metals	3.70	35 474
Wood	7.93	76 088
Paper and cardboard	6.39	61 366
Plastic	21.92	210 430
Textiles	5.78	55 490
Soil	11.66	111 891
Others	0.75	7 169
Fine fraction	41.40	397 440
Total	100	955 348

(2013) and Gutiérrez-Gutiérrez et al. (2015). Specifically, Gutiérrez-Gutiérrez et al. (2015) assessed the content of four UK MSW landfill sites and reported concentrations for rare earth elements (REE) (including Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu) of 220 ±11 mg/kg, PGM (Pt, Pd, Ru) 2,1 ± 0,2 mg/kg, and other critical metals such as Li, Ln, Sb, Co of 156 ±7 mg/kg. In this study at Metsäsairila MSW landfill site, the REE concentration was twice lower (87±13 mg/kg). Concentrations of Pt, Pd and Ru were also lower, less than 1.5 mg/kg. However Ce was found to be the most abundant rare metal in our study as in Gutiérrez-Gutiérrez et al. (2015). This finding underlines the differences in the composition and the characteristics of the waste materials in different MSW landfills with regard to type, location and the period connected to landfilling activity. Based on these results, it is obvious that the quantity of REE and PGM that can be recovered from the waste materials will be much lower than what would be extracted by mining natural ores. In addition, at the end of the extraction process the metals are concentrated in acid solution which must be treated to separate the metals of interest. Achieving a level of purity above 99% becomes a major inconvenient to make the recovered metals highly valuable. Also concentrations are in same range than for ordinary soil so it is predicted that extracting them from fine fraction would not give extra benefit for MSW landfill mining.

3.2 Vierumäki industrial waste landfill site

In contrast to the Metsäsairila MSW landfill site, the fine fraction <20 mm had by far the greatest mass share of all the samples in the Vierumäki industrial waste landfill site (on average 74 ± 7% (n=4)). The 20-100 mm fraction represented 20 ± 7% and the >100 mm fraction 6 ± 3% of the waste samples. From visual observation, the fine fraction consisted mainly of the aluminium salt slag. Based on the field observations of the landfill layers (Table 2), a simplified cross section of the landfill site was estimated as follows: 1 m of cover layers, 1 m of shredding waste and 3 meters of aluminum salt slag. Together with the estimated landfill area of 2.5 hectares and the results from manual sorting, the masses of different material types at the landfill were estimated as shown in Table 5. Here the fine fraction <20 mm is regarded as aluminium salt slag.

The average concentrations of the critical metals, REE and PGM in the fine fraction and the combustible fraction of the samples are summarized in Table 6. The fine fraction <20 mm had characteristics comparable to typical aluminium salt slags. The concentrations of REE and other valuable elements were in contrast very low even in comparison with the concentrations found in the Earth's crust (USEPA, 2012).

The average calorific value of the combustible fractions

TABLE 5: Estimation of the material amounts at Vierumäki industrial landfill site.

Material	Mass (t)
Cover layers	25 000
Shredding waste total of which	20 000
Fine fraction (Al salt slag)	15 600
Combustibles	4400
Metals	72
Soil	578
Other (mainly large pieces of Al salt slag)	379
Al salt slag class (from separate layer)	75 000

was 22±4 MJ/kg which is good compared to the heating value of other materials such as lignocellulosic materials normally ranging between 12.2 and 20.6 MJ/kg, biochar between 27.4 and 32.6 MJ/kg, plastics and synthetic rubber between 37.8 and 38.00 MJ/kg and cardboard 13.81 MJ/kg (Boumanchar et al., 2017).

4. CONCLUSIONS

Based on the characterisation of the exacavated waste samples from both landfill site types the amounts of critical metals, REE and PGM were not high enough to justify landfill mining and recovery alone. The main fractions from the MSW landfill site were by order of contribution, the fine fraction (<20 mm), followed by the combustible fraction and the metallic products (ferrous and non-ferrous products). Thus the economic viability of landfill mining could be increased by recovering additional material fractions such as plastics, paper, cardboard and wood for energy production. In contrast the aluminium content of the fine fraction of the IW landfill site is offering good opportunity for recovery. Overall, other opportunities exist that together form the concept of ELFM. Waste-derived fuels from excavated materials have the potential to be highly energetic. From both landfill sites investigated, the energy potential is comparable to the levels of energy of Refuse-Derived Fuels (RDF) produced from non-landfilled wastes. Ultimately, the mining and recovery approach leads to a further commercial opportunity in the land itself, reclaimed and the soil remediated, making it available again for housing, industrial estate development or other forms of development. Abandoned landfill sites present environmental and human health risks that can involve large taxpayer investments to clear up. Nonetheless, there are still several challenges in ELFM, which means that further research and development is needed before the full potential will be realised.

ACKNOWLEDGEMENTS

TABLE 6: Average content (%) of REEs, PGM, critical metals and others found in the Vierumäki industrial landfill site.

Element (%)	Fine fractions <20 mm average, n=4 (standard deviation)	Combustible fractions average, n=4 (standard deviation)	Typical values for aluminum salt slag (Huang et al., 2014)
Al	13 (5.4)	1.7 (0.21)	14.2
Mg	1.6 (0.17)	0.73 (0.15)	2.0
Cu	0.26 (0.07)	0.14 (0.10)	0.088
Sb	<0.01 (-)	<0.01 (-)	-
Co	<0.01 (-)	<0.01 (-)	-
Cr	0.03 (0.02)	0.01 (0.004)	0.033
Element (mg/kg)	Fine fractions <20 mm composite sample	Combustible fractions composite sample	Crustal abundance (US EPA, 2012)
Er	<0.50	<0.50	2.1
Eu	<0.50	<0.50	1.3
Au	<0.50	<0.50	0.003
Pd	<0.50	<0.50	-
La	5.6	4.5	30
Y	9.7	2.2	24
Pt	<0.50	<0.50	-
Ce	11	8.4	60
Nd	5.0	3.8	27
Ru	<0.50	<0.50	-
Pr	1.2	1.0	-
Sm	0.81	0.75	5.3
Gd	0.72	0.68	4.0
Tb	<0.50	<0.50	0.7

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