ASSESSING THE OPPORTUNITIES OF LANDFILL MINING AS A SOURCE OF CRITICAL RAW MATERIALS IN EUROPE

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SUMMARY: Many of the metals in landfill 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. Arsenic (which is an essential part of the production of transistors and LEDs) is predicted to run out sometime in the next five to 50 years if consumption continues at the present rate. Nickel used for anything involving stainless steel and platinum group metals (PGMs) used in catalytic converters, fertilisers and others are also identified as critical materials (CM) to the EU economy at risk of depletion However, despite the increasing demand, none of this supply is supported by recycling. This is due to the high cost of recovery from low concentrations when compared to conventional mining. As demonstrated by the two pilot case studies of this study, mining landfill sites only for their metals content is not expected to be financially viable. However, other opportunities such as Waste-derived fuels from excavated materials exist which if combined , form the concept of 'enhanced landfill mining'. have the potential to be highly energetic. The energy potential is comparable to the levels of energy of Refuse-Derived Fuels (RDF) produced from non-landfilled wastes.

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. Considering their increasing scarcity and raising prices, their recycling and 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, mining and municipal landfill sites especially if we consider that Europe is highly dependent on the imports of certain raw materials including

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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 be contain with 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). 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.

Mining solely for metals is not expected to be financially viable (Kaartinen et al., 2013; Gutiérrez-Gutiérrez et al. 2015). Other opportunities exist that together form the concept of ‘enhanced landfill mining’: waste-derived fuels from excavated materials can be highly ‘energetic’; the land can be reclaimed and the soil remediated, making it available for development (Jones et al., 2013). There are however challenges in enhanced landfill mining. We need to understand more about each of the stages involved: the exploration, separation, transformation and up cycling technologies, and how these can be best applied 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 were 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 enhanced landfill mining (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 mining of either municipal solid waste (MSW) or industrial waste (IW) disposal sites, 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). 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 IW landfill site (Vierumäki). Detailed site investigation of the two sites was carried out to evaluate the potential SRM resource that can be exploited. The described characterisation framework is 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 (2) 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 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 characterisation 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. 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.

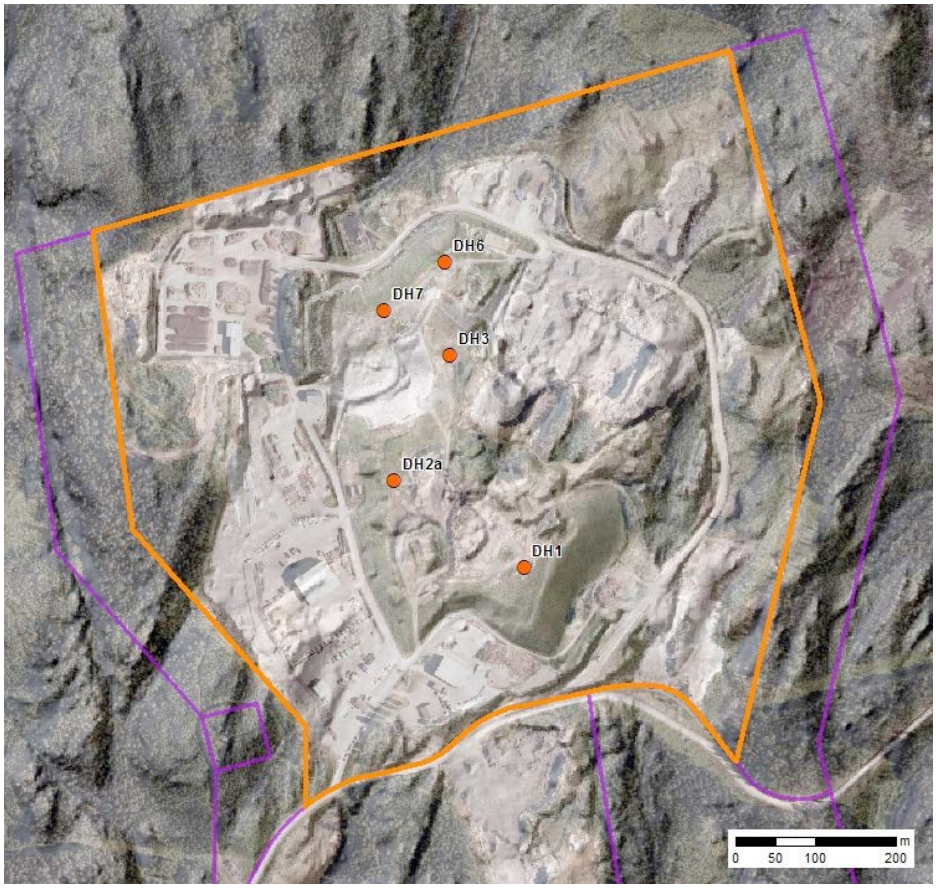

Figure 1: Topographic map of Metsäsairila MSW landfill site obtained using laser scanning based DEM with 2 metres resolution overlaid with aerial photography (orange dots: location of the sampling points).

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. 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).

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

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.

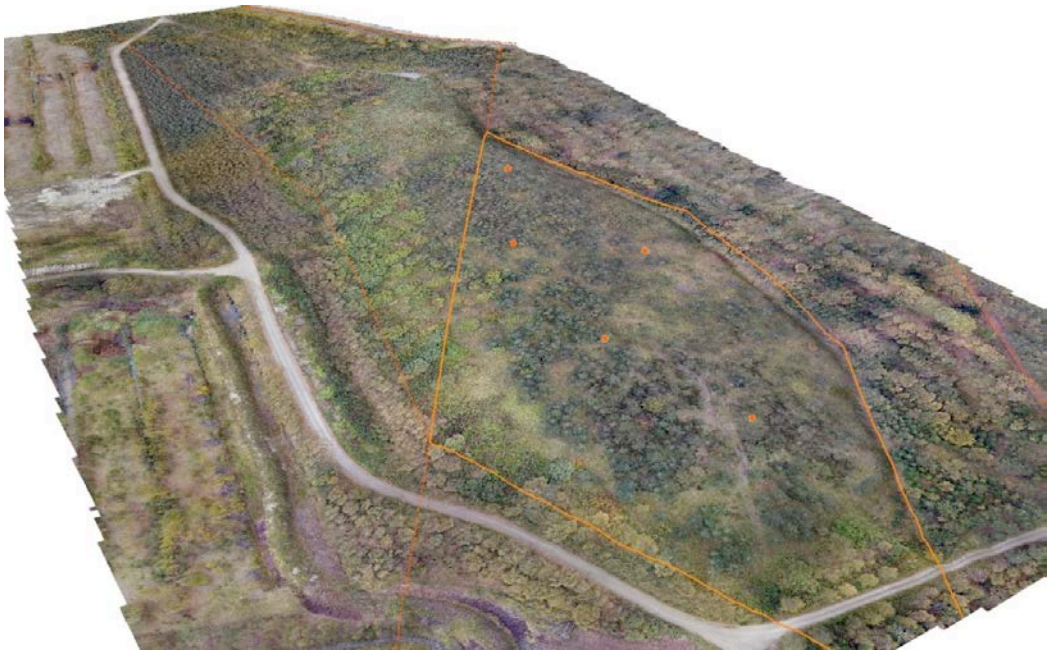

Figure 2: 3D site topography based on DTM with 50 cm resolution and draped orthophoto

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).

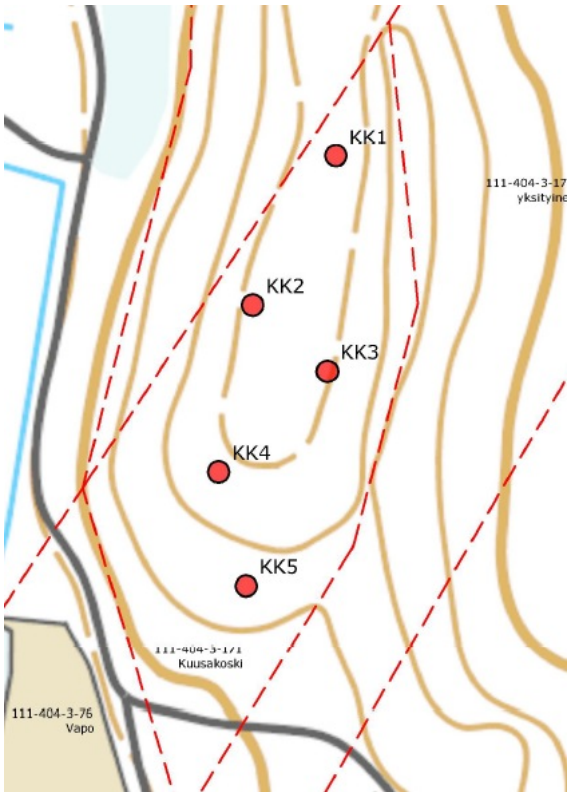

Figure 3**:** Locations of sampling points at the industrial landfill. Red dots show the sampling points and the dashed line surrounding the dots show the rough borders of the industrial landfill.

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

Figure 4: Landfill layers at the Vierumäki industrial landfill.

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

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

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 (CSN EN 15400). 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.

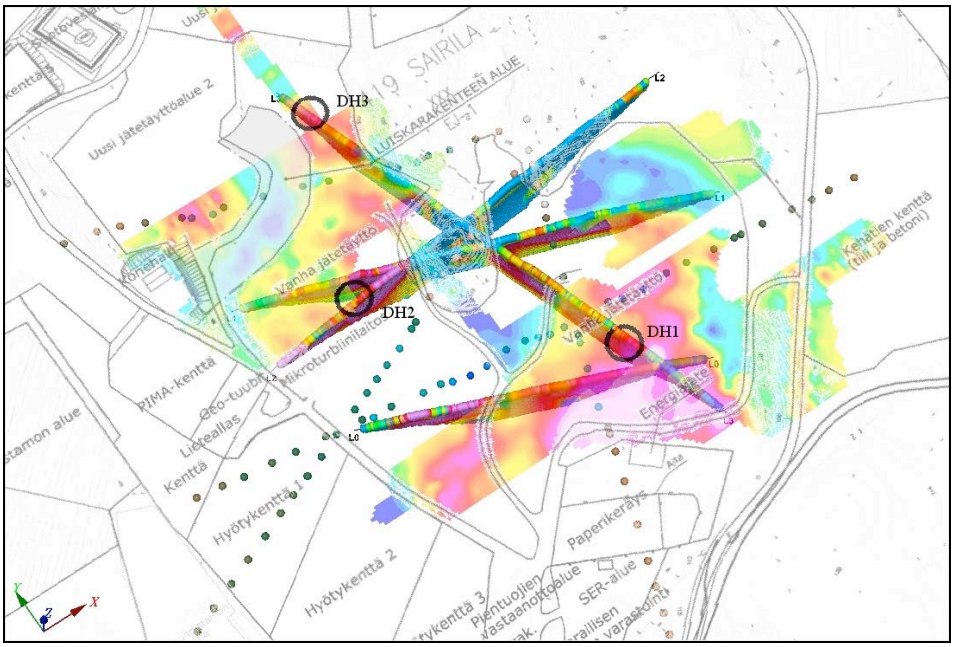

Figure 5: Electrical **c**onductivity cross sections from 3D ERT profiles and total magnetic anomaly 2D map data with realized drill holes D1-D3 (sampling points). Interpreted bedrock topography of gravity profiles is visualized in the picture as coloured circles. In all the data the red colours show the high and blue colours show the low values.

The mass distribution of the different waste fractions of the five core samples is shown in Table 3. 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).

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**

Based on the geophysical characterization of the landfill site and the average amount of waste fractions in samples DH1, DH2a and DH3, the closed part of Metsäsairila MSW landfill was identified as the most promising for landfill mining. The closed area 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).

Table 4. Estimated amounts of the different waste materials in the closed area of the Metsäsairila MSW landfill site

	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**

Further to this, the critical metals concentration comprising REE and platinum-group metals (PGM) was 87±13 mg/kg in average. Concentrations of Pt, Pd and Ru were lower than 0.5 mg/kg. 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.

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

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).

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
Dy	0.54	<0.50	3.8
Ho	<0.50	<0.50	0.8
Yb	<0.50	<0.50	2.0
Sc	1.2	0.90	16
In	<0.50	87	-
Ag	2.1	4.2	0.08

The average calorific value of the combustible fractions 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). Overall the results from the IW landfill sites showed that amount of critical raw materials (including REE and platinum group metals (PGM)) in the fine fraction (<20 mm) is not high enough to justify their recovery alone. However, the economic viability of landfill mining could be increased by recovering additional material fractions such as plastics, paper, cardboard and wood for energy production. Further to this the aluminium (Al) was the most abundant element found. However, Al concentration in the combustible fractions was low suggesting that Al was concentrated in the smallest particle sizes of the samples.

4. CONCLUSIONS

Overall for both landfill site types the amounts of critical metals, REE and PGM were not high enough to justify landfill mining and recovery alone. However, 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. In Belgium, in recent years, it took 80 million Euros to deal with the impact of five landfill sites on their immediate environment and groundwater quality. It has been estimated, based on average amounts of materials per landfill in the EU that landfills could provide up to 5% of the total needs of Europe for non-energy, non-food materials and minerals for the next 25 years. Nonetheless, there are still several challenges in ELFM, which means that further research and development is needed before the full potential will be realised.

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