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On the way to ‘zero waste’ management: Recovery potential of elements, including rare earth elements, from fine fraction of waste

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ABSTRACT

Existing schemes of solid waste handling have been improved implementing advanced systems for recovery and reuse of various materials. Nowadays, the ‘zero waste’ concept is becoming more topical through the reduction of disposed waste. Recovery of metals, nutrients and other materials that can be returned to the material cycles still remain as a challenge for future. Landfill mining (LFM) is one of the approaches that can deal with former dumpsites, and derived materials may become important for circular economy within the concept ‘beyond the zero waste’. Perspectives of material recovery can include recycling of critical industrial metals, including rare earth elements (REEs). The LFM projects performed in the Baltic Region along with a conventional source separation of iron-scrap, plastics etc. have shown that the potential of fine-grained fractions (including clay and colloidal matter) of excavated waste have considerably large amounts of potentially valuable metals and distinct REEs. In this paper analytical screening studies are discussed extending the understanding of element content in fine fraction of waste derived from excavated, separated and screened waste in a perspective of circular economy. Technological feasibility was evaluated by using modified sequential extraction technique where easy extractable amount of metals can be estimated. Results revealed that considerable concentrations of Mn (418–823 mg/kg), Ni (41–84 mg/kg), Co (10.7–19.3 mg/kg) and Cd (1.0–3.0 mg/kg) were detected in fine fraction (<10 mm) of waste sampled from Högbytorp landfill, while Cr (49–518 mg/kg) and Pb (30–264 mg/kg) were found in fine fraction (<10 mm) of waste from Torma landfill revealing wide heterogeneity of tested samples. Waste should become a utilizable resource closing the loop of anthropogenic material cycle as the hidden potential of valuable materials in dumps is considerable.

1. Introduction

1.1. Concept of ‘zero waste’

Shortage of the Earth's resources and exponentially increasing amount of produced waste demands reduction of waste, thus, recovery and reuse of materials from waste ultimately becomes an important issue of the economic interest. Furthermore, instead of waste disposal, modern waste management is on the way to implement the concept of ‘zero waste’ (Arndt et al., 2017; Burlakovs et al., 2017; Sverdrup and Ragnarsdóttir, 2014). Commonly used definition of the ‘zero waste’ concept was proposed by the Zero Waste International Alliance in 2004 (ZWIA, 2015) as follows: *‘Zero waste’ is a goal that is ethical, economical, efficient and visionary, to guide people in changing their lifestyles and practices to emulate sustainable natural cycles, where all discarded materials are designed to become resources for others to use.*

Many industrial and urban activities including households generate considerable amount of solid waste every day all over the world even if the recycling and energy generation from solid waste is increasing (Bhatnagar et al., 2013; Burlakovs et al., 2017; Kriipsalu et al., 2008). According to the study done by Laurent et al. (2014), the annual total solid waste generation worldwide is approximately

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17 Gt with a prediction to reach about 27 Gt by 2050. Nevertheless, solid waste discarded daily by the society contains several important constituents, e.g., valuable metals such as zinc, copper, nickel, chromium and lead can be found everywhere. Not only the prices have become higher in the market, but as stated in the study done by Boesch et al. (2014) the trend of increasing prices of natural resources is expected to be even more pronounced in the near future. It brings the necessity to shift the traditional linear models followed by our society not only into closed-loop models but also consider the recovery of all valuable resource man have lost as waste products from our daily activities but also through history (Burlakovs, 2012; Burlakovs et al., 2017). There is also a crucial need to look into waste reservoirs (soils, sludge, slag and ash from incineration, bottom sediments, soils from landfills, waste from ore mining etc.) that are fully contaminated with metals and nutrients as potential secondary stocks of such valuable constituents. This lead us into the topic of the paper and introduction of the “Beyond the zero waste concept” which encourages recovery of all materials lost during the entire life cycles of different products manufactured, which are still available in different sinks (landfills, sediments of rivers, ocean, etc.) (Bhatnagar et al., 2013; Burlakovs et al., 2016; Ghosh et al., 2016; Kriipsalu et al., 2008). The long-term goal is to apply such innovative approach in an environmental and economic efficient way, making use of the accumulated knowledge, including reuse/recycling of materials bound in urban and rural structures. This will include landfill mining, glass mining, harbour and bay mining as well as seafloor mining (Burlakovs et al., 2017).

1.2. Criticality of elements

Thousands of landfills as potential sources of environmental pollution can become primary objects for both, remediation and landfill mining (LFM), in the future. Metals including rare earth elements (REEs) are fundamental to economy and growth (Mikhaylov, 2010) as well as often are essential for maintaining and improving technological processes especially concerning so-called ‘green technologies’ (EC, 2014). However, REEs of anthropogenic origin pose threats through water pollution and their spatial distribution is especially wide in metropolitan areas (Song et al., 2017).

LFM can be described as ‘a process for extracting minerals or other solid natural resources from waste materials that have been previously disposed of by burying into the ground’ (Krook et al., 2012). The process involves the excavation, screening, and separation of material from older landfills (Burlakovs et al., 2015; Hogland, 2002). The comparison of pilot results from LFM and industrial sites in Latvia, Estonia and Sweden is given, screening type of analysis give a bit of the step further to shed the light on REEs and other elemental contents in fine fraction of excavated dump masses. Analysis of excavated waste is an initial step for scientists and entrepreneurs to re-inject lost material of growing concern within the EU and across the globe back into the economic cycle (Burlakovs et al., 2015; Hogland, 2002; Krook et al., 2012).

Availability of metals is limited on the Earth – it is an undeniable fact (Ragnarsdottir et al., 2012). Generic wisdom keeps the world on a boundary that individual mines one day will go empty. It is also evident that industry uses lesser and lesser ore grades and needs smaller concentrations to be economically viable when prices of metals go up and scarcity of resources pushes up. In a closer perspective such elements as REEs, which mostly are produced in China, as well as elements like In, Sb and some other marked as critical will reach a shortage already in next few decades (Ragnarsdottir et al., 2012). Thus, improvement of technologies unarguably will move forward a decline of raw material supplies, including various metals, however, the society needs to think of material recycling that is already lost from calculations – recycling of landfilled waste masses.

1.3. Circular economy and recovery of elements

Circular economy has got an important attention through the report ‘Towards the Circular Economy’ (EMAF, 2013). The key idea of linear economy is unsustainable from both the material and environmental point of view. Linear thinking regards resources in a “take, make, dump”. It is non-justifiable in a world where resources become increasingly scarce with considerable environmental impacts from extracting (Lovins et al., 2013). Circular economy closes the loop in cyclical manner: an idea firstly stated in the area of industrial ecology (Ayres and Ayres, 2002). Boesch et al. (2014) state that whereas energy recovery from waste material has received much attention in the last years and is rather advanced technologically than the potential of material recovery from by-products of waste management processes. Incineration residues has often been neglected from recovery but for scrap however, it is also source of many metals and phosphorous. As the waste contains valuables including metals, critical elements and REEs – the content of dumps can become in the future fundamental in economic terms and essential for developing industrial technologies of recovery through LFM. It happens in time when resource depletion is comparably near (Burlakovs et al., 2017).

The largest volume of old landfills consists of fine granulometric composition and even colloidal material (Arina et al., 2014; Burlakovs, 2012; Burlakovs et al., 2013; Burlakovs and Vircavs, 2011, 2012a,b). This fine fraction material (among them clay and colloidal matter) contains considerably large amounts of valuable metals including REEs (Ziyang et al., 2015). Earlier studies dealing with sequential extraction procedure revealed that most of REEs in soils (e.g., in colloidal fractions of floodplains) are bound in the residual fraction, followed by the reducible, the oxidizable and the water soluble/exchangeable/carbonate bound fraction (Mihajlovic et al., 2017). Khan et al. (2017) used chemometric approach to determine elevated levels of REEs in Malaysian former mining areas that in general might be considered as a secondary type of mono-landfills. Similarly, geochemical mobility of heavy metals in bottom sediment of the Hoedong reservoir in Korea have been studied recently under the supervision of Lee et al. (2017). The aim of the current study was to choose as much as possible homogenized mass of fine grained dump material in order to estimate the content of major, minor elements and REEs in fine fraction from excavated waste separated during the LFM projects, earlier described by Burlakovs et al. (2015, 2016).

Concentration of metals, critical and REEs was analysed and compared. Sequential extraction was performed in order to determine potential easy extractability of valuable metals. Main results show that fine fraction of waste might have certain interest of recovery if technological development evolves in future.

1.4. Technologies of element recovery

A number of physical, chemical and hybrid techniques to extract metals from solid phase is available in the scientific literature (Andreottola et al., 2010; Kirkelund et al., 2009; Vandevivere et al., 2001; Zhang et al., 2009; Yoo et al., 2013). However, even though several researchers have been studying different techniques to extract/recover metals, research has been mostly conducted with artificially polluted media, which makes it difficult to predict functionality in real conditions and upscale the process (Øygard et al., 2008). Furthermore, results obtained with the use of artificially polluted solids can be considerably different when comparing with real conditions in which contamination is usually present in a multi-element form (Vandevivere et al., 2001), mainly when dealing with complex media such as soil, slags, sewage sludge and harbour bottom sediments. Among the different techniques, Vandevivere et al. (2001) have used a strong transition metal chelating agents to recover Pb, Zn, Cu and Cd from real contaminated media whereas Andreottola et al. (2010) have studied the use of chemical oxidation, electrochemical oxidation and electrokinetics under different conditions to remediate dredged materials from Venice Lagoon. Other are studies that focused also on dredged marine bottom sediments and the feasibility of using chemical complexation washing agents to extract metals (Yoo et al., 2013). Studies about remediation of metals from bay sediments and the main parameters playing important roles on the process performance were conducted at laboratory scale (Zhang et al., 2009), where the use of chemical complexation reagents such as ethylenediamine tetra acetic acid disodium salt (EDTA-2Na), sodium dodecyl-sulphate (SDS), acetic acid (HAc), oxalic acid ($H_2C_2O_4$), ammonium acetate (NH_4Ac) and ammonium oxalate ($(NH_4)_2C_2O_4$) were studied. As it can be observed in the literature, there have been some investigations focused on harbour sediments and Kirkelund et al. (2009) studied in laboratory scale the use of electro-dialytic process as extraction strategy to tackle Cu, Zn, Pb and Cd strongly bound to anoxic sediments.

Several researchers have focused on the potential for extraction/removal of metals from ashes and slags generated in municipal solid waste incineration plants through different techniques including thermal and hydro-metallurgy (Kuboňová et al., 2013), dry discharge of bottom ashes followed by a series of magnets and Eddy currents (Meylan and Spoerri, 2014; Morf et al., 2013), fly ashes acid leaching (Meylan and Spoerri, 2014), multi-stage dust collection technique (Okada and Nishimoto, 2013) and a novel combined approach using bio-electrochemical systems followed by electrolysis reactors (Tao et al., 2014). The application of wet high intensity magnetic separation to remove As, Cu, Pb, and Zn from a sandy loam soil from a mining site was studied by Sierra et al. (2014). Extraction of Cu from artificially contaminated sediments through the use of electro kinetic process driven by a galvanic cell (Yuan et al., 2009) was also reported. As previously mentioned, there are several authors that have investigated different methods to extract metals from solid phase materials. Xu et al. (2017) reviews the development of electrokinetic and bioleaching techniques, analyses advantages and limitation of the heavy metals-contaminated sewage sludge treatment. However, there is no literature and, therefore, a lack of knowledge when considering fine-grained fraction of excavated dumps and the potential of metals recovery. Some research on metals availability and composition of both bottom sediments from Oskarshamn Harbour and fine-grained fraction from Swedish and Estonian (Fathollahzadeh et al., 2014; Kaczala et al., 2017) as well as Latvian excavated landfills (dumps) have been performed. The initial results have shown that approximately 50% and 40% of excavated waste volume is presented by size fractions under 40 mm and 10 mm respectively (Burlakovs et al., 2013, 2015, 2016). Furthermore, the preliminary studies conducted have shown that metals such as copper, zinc can be abundant suggesting further studies on how to separate the right fractions of each metal and extract them.

2. Materials and methods

2.1. Description of sampling sites and waste extraction

Two study sites were chosen to collect the samples: I) Högbytorp mixed type industrial/municipal landfill in Sweden; II) Torma municipal landfill in Estonia (Fig. 1).

Högbytorp landfill is located 40 km northwest of Stockholm (60°32′N, 17°37′E) in Sweden. It was established in 1964, and occupies an area of around 30 ha and contains municipal as well as industrial waste. The landfill consists of a complex mixture of organic and inorganic waste such as contaminated soil, oily sludge from car washes, organic materials, paper, wood and plastics, metals scrap, ash, medical waste, hazardous waste disposal, and wastes from municipalities, industries and households (Jani et al., 2016).

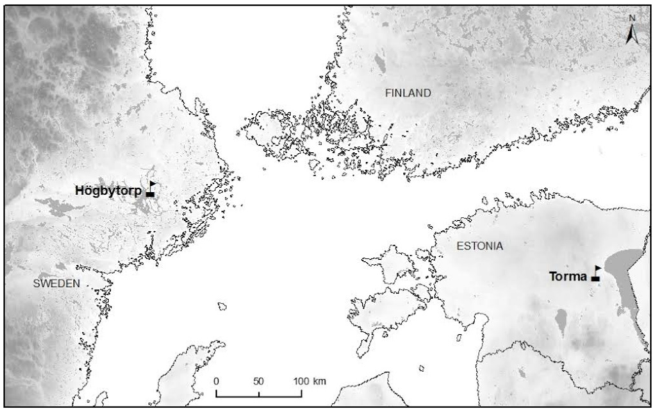

Fig. 1. Location of the study sites: Högbytorp landfill in Sweden and Torma landfill in Estonia.

Torma landfill is located in Jõgeva County (58°51′11.2″N, 26°52′37.9″E) in Tartu municipality, Estonia. It was established in 2001. The landfill has an area of 6.2 ha. The landfill contains three cells of different age with municipal solid waste as the main source of waste (Jani et al., 2017).

Excavation was done from the vertical waste walls (at Torma landfill) or heaps (at Högbytorp landfill) in a number of test-pits at the sampling sites after preliminary planning (Burlakovs et al., 2015, 2016). The equipment used for excavation from test-pits included tread excavator or tractor with a bucket size of around 1 m^3. A layer of weathered waste was removed to create so-called ‘fresh cut’ as well as a topsoil 0.3–0.5 m was removed. Derived waste was shredded, separated and homogenized to be prepared for further analytical studies. Regarding landfills, the waste of size 0–40 mm usually is assumed as a fine fraction. During this study, after mechanical sieving method following proportions were identified: 80% of waste consisted of pieces smaller than 10 mm, but 20% of waste was of size 10–40 mm. It was possible to identify some metals, mostly Fe, Al, Cu, visually during the sorting; amount of these metals in the fine fraction (0–40 mm) was calculated about 0.6%. Taking into account manual and visual assessment of waste as well as potential economic value at industrial scale, for further research only fraction of <10 mm was prepared.

2.2. Applied analytical procedure

Samples of the fine fraction (<10 mm) of waste were prepared as described in previous subsection in order to analyse content of metals and metalloids, including REEs, to be suitable for material recovery in a distant future. Samples collected at selected landfill sites, Torma and Högbytorp, were processed in a specific manner (Fig. 2) and analysed using inductively coupled mass spectrometry (ICP-MS) and atomic absorption spectrometry (AAS) as described previously in literature (Burlakovs et al., 2015, 2016; Kilbride et al., 2006; Mester and Sturgeon, 2003; Radu and Diamond, 2009).

Total content of elements was analysed after wet acid-based digestion procedure as follows: each sample (1,0000 ± 0,0050 g) was poured over with 50 ml of concentrated HNO_3 (65% w/v, Merck) and 5 ml of concentrated H_2O_2 (30% w/v, Merck). After hold

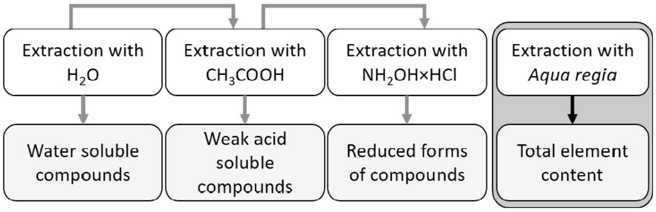

Fig. 2. Schematic overview of the waste sample sequential extraction.

for 24 h, solutions were heated on heating block (Biosan) until half of volume evaporated. Adding of HNO_3 and heating was repeated until complete sample mineralization. Solutions were filtered through membrane filters (0.45 μm, Simplepure). Each sample was prepared in triplicate, as well as blank samples were prepared for each batch of samples.

Modified sequential extraction was carried out as follows (after Chen and Ma, 2001; Favas, 2013; Malandrino et al., 2011; Masto et al., 2015; Okoro et al., 2012; Tessier et al., 1979):

Step 1: Fraction of water soluble (easily available) compounds. A sample of waste (3.0000 ± 0.0050 g) was extracted by heated (40 °C) deionised water in a glass beaker for 2 h on a mechanical shaker.

Step 2: Fraction of weak acid soluble compounds. Sediments of waste left from the 1st step of sequential extraction were carefully collected into a glass beaker and extracted with 40 mL of 0.11 M CH_3COOH for 16 h on a mechanical shaker (Tachometer RPM PSU-20, Biosan).

Step 3: Fraction of reduced compounds. Sediments of waste left from the 2nd step of sequential extraction were collected into a glass beaker and extracted with 2 ml of 0.5 M $NH_2OH \times HCl$ for 16 h on a mechanical shaker.

After each step, double filtration of extracts using a paper filter and a membrane filter (0.45 μm, Simplepure) was done; the extracts were filled into a polypropylene tubes and diluted with deionised water up to 50 ml and acidified with 0.7 M HNO_3. Each analytical solution was made in triplicate and, at each step, blank samples were prepared in the same manner. All obtained extracts until analyses were kept in a cool place (+4 °C).

Residual fraction was calculated taking into account total content of an element and data derived by sequential extraction.

Concentration of major and minor elements (Ca, Al, Fe, Ba, Cu, Cr, Fe, K, Na, Mg, Mn, Pb, Zn, Co, Ni, Cd; core and refractory) in analytical solutions of waste was analysed using AAS (Perkin Elmer AAnalyst200). ICP-MS device (Perkin Elmer ELAN DRC-e) was employed to detect concentration of Ba, Sr, Rb, As, Cs, Th and REEs (Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Yb, Lu) (Burlakovs et al., 2016; Dean, 2005; Kabata-Pendias and Pendias, 2010; Kealey and Haines, 2005). Quality of the analytical measurements was ensured by the analysis of certified reference material IAEA-336 Lichen containing reference values on wide spectra of major and minor elements, including some REEs. In this study, besides the range of REEs, the elements not exceeding concentration of 1000 mg/kg in waste were listed as minor elements, but those at higher concentrations were assessed as major elements.

2.3. Approach of data analysis

Data analysis was performed by using extended MS Excel data analysis tool QI Macros to generate standard statistical analysis. Box-whisker plotting was chosen as one of the most appropriate visually descriptive statistical approach for the comparative assessment of element measurement data sets. For more objective assessment of data, outlier values were excluded, thus, box-whisker plots indicate minimum, maximum and median concentration of elements as well as the range between 25th and 75th percentiles.

3. Results and discussion

Results confirmed that wide range of chemical elements are accumulated in fine fraction of waste; nevertheless, differences were distinguished between both study sites as Högbytorp in Sweden is an industrial and mixed waste landfill remaining from the end of 20th century, but Torma in Estonia is still in operation and it is purely municipal landfill that works according to the current EU waste legislation (e.g., Landfill Directive, 1999; Waste Framework Directive, 2008).

3.1. Content of major elements in fine fraction of waste

Total content of major elements in fine fraction of waste from landfills revealed that significantly higher content of Ca (respectively, 42–94 g/kg), Al (9–22 g/kg), Cu (2.1–5.4 g/kg), Na (1.9–4.6 g/kg), K (1.1–3.0 g/kg) and Zn (1.2–2.8 g/kg) can be found in waste samples from Högbytorp, while Fe (8–72 g/kg) was found in higher concentration in samples from Torma, but detected amount of Mg (5.1–17.5 g/kg for Högbytorp and 2.7–12.3 g/kg for Torma) was similar for samples from both study sites (Fig. 3).

Fine fraction of waste from Högbytorp landfill, thus, have 2–4 times larger concentration for some major than in Torma. These results are reasonable due to metal separation at landfills prior dumping. Ca prevalence in samples from Högbytorp can be explained by large proportion of construction waste in the whole waste mass. Fine fraction of waste is mainly composed of clayey and organic matter that bounds metallic elements easily, thus, considerable amount of Fe, Al, Mg, Cu and Zn can be present. Furthermore, Ca, Fe, Al, Mg, Na and K are basic constituents of soil geochemical

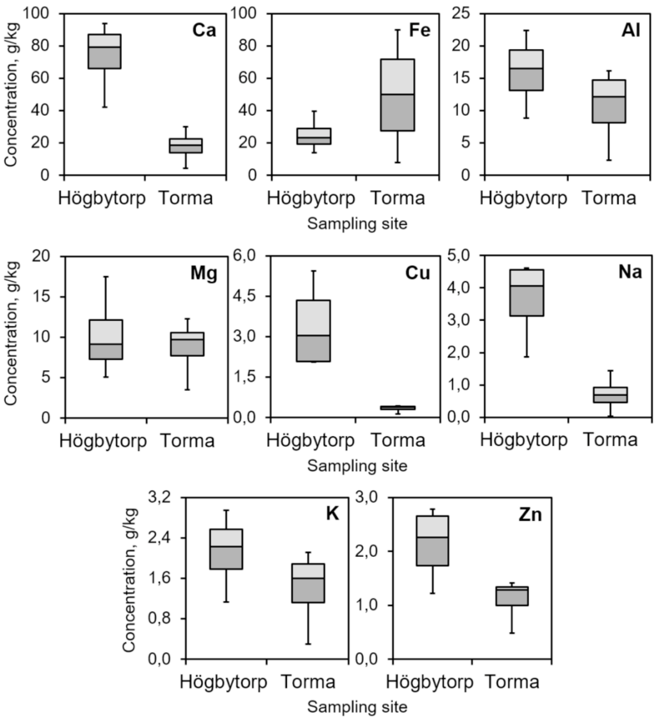

Fig. 3. Content of major elements in fine fraction of waste from Högbytorp and Torma landfills (excluding outliers, indicating minimum, maximum, median and range between 25th and 75th percentiles).

background, therefore, elevated concentrations (measured in g per kg) of these elements in waste samples were foreseen. Major elements such as Fe, Cr, Mn, Al, Cu and Zn are among the highest production rates of all the metals in the world and there is rising interest to extract and to amass them by new environmentally sustainable ways after creating new technologies in future. Thus, waste recycling can be assessed as one of potentially promising options especially taking into account targets of national policies worldwide that more and more often are changing their orientation towards recycling of materials, including waste and polluted soil, rather than to dump them (Arndt et al., 2017; Sverdrup and Ragnarsdóttir, 2014). Taking into account derived results from Högbytorp and Torma landfills, the prospective interest could arise especially regarding extraction of Fe and Al, if the LFM procedures are taken as the complementary process of full revitalization and resource recovery (Burlakovs et al., 2017; Sverdrup and Ragnarsdóttir, 2014). The lack of interest is dominating at economic consideration as well as wide range of arguments declare that organic soil usually has similar amount of major elements. However, the soil formation is a slow process (ca 10 mm/100 yr) that is worryingly slower than soil erosion (ca 100-1000 mm/100 yr) in agricultural areas (Brantley et al., 2007). The argument to extract major elements from soil or mining areas (which requires removing of fertile topsoil) then lose the confidence versus the fact that soil is an unrenewable resource at the scale of one human generation and in some regions even more (Arndt et al., 2017).

3.2. Content of minor elements in fine fraction of waste

Results indicated that elements not exceeding 1000 mg/kg (excluding REEs) in waste samples were Cd, Co, Cr, Mn, Ni and Pb. Significantly higher concentration of Mn (respectively, 418–823 mg/kg), Ni (41–84 mg/kg), Co (10.7–19.3 mg/kg) and Cd (1.0–3.03 mg/kg) was detected in waste samples from Högbytorp landfill, while Cr (respectively, 49–518 mg/kg) and Pb (30–264 mg/kg) were found in higher amount in samples from Torma landfills (Fig. 4).

Fine fraction of waste from Torma landfill has larger amount of some minor elements that may be linked to several reasons. Authors draw out hypothesis that Torma landfill consist of more organic waste that is not biodegraded, therefore, colloidal fine fraction of waste (soil) cap may contain a large portion of Cr, Pb (and major element Fe) (Pansu and Gautheyrou, 2016), but in case of Högbytorp landfill these elements were washed away due to leaching through years passed by.

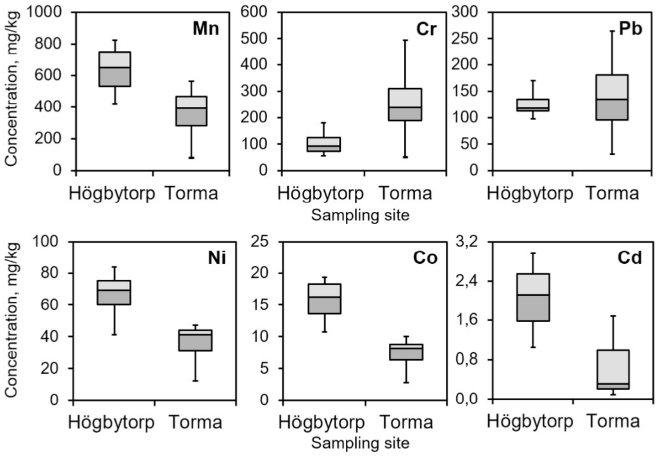

Fig. 4. Content of minor elements in fine fraction of waste from Högbytorp and Torma landfills (excluding outliers, indicating minimum, maximum, median and range between 25th and 75th percentiles).

Various resources including soil, sand, gravels, boulders and ores are used to build the infrastructures and technological items important for society, including real estate, transport roads, railways and bridges. More than 15 Bt of such material are moved every year, among them also materials containing relatively high content of valuable chemical elements. Industrial activities have become a geologic force that promotes the move to order more material than all of the geological forces combined (Hooke, 2000; Wilkinson, 2005). Even non-rare major and minor elements are of high interest when we think about the future developments and population growth (Arndt et al., 2017; Wood, 2014). Research on elemental content and market values of materials, including metals, in waste or landfill material is rarely comparable directly. Previous studies have estimated approximate values; for example, it was stated that about 300 million tons of copper is bound in controlled and other waste repositories globally (in tailings and slag heaps) corresponding to more than 30% of the remaining reserves in known copper ores (Frändegård et al., 2013; Kapur and Graedel, 2006; Sverdrup and Ragnarsdóttir, 2014). Contents of metals might pose different levels of risk to environmental health and safety of human and animal health due to various, usually complicated and even hidden or unknown history of pollution sources at contaminated brownfields, landfills and dump sites, as well as it should be estimated taking into account climatic, geographical and geological specifics of each certain site. Thus, direct comparison among the data from different sites is very complicated (Carr et al., 2008; Kaczala et al., 2017).

3.3. Content of REEs in fine fraction of waste

It was estimated that summary content of all quantified REEs (Ce, Cs, Dy, Er, Eu, Gd, Ho, La, Lu, Nd, Pr, Sc, Sm, Tb, Th, Y, Yb) was about 40 mg/kg in fine fraction of waste from Torma landfill and twice as much (>80 mg/kg) in samples from Högbytorp landfill allowing to assume that older waste contains larger concentration of REEs that is associated also with type of waste and sorting or absence of sorting of waste. Average concentration of REEs was detected from 0.09 mg/kg and 0.25 mg/kg (Tb), respectively, for Högbytorp and Torma, up to 27.2 mg/kg and 17.25 mg/kg (Ce). Proportionally, such REEs as Ce, La, Nd, Pr, Sc, Gd and Sm from cerium group that are listed as light REEs were prevailing, but heavy REEs from yttrium group were detected in negligible amounts, except Y itself (Fig. 5). Different reasons can influence such element distribution; firstly, light REEs are more common in industrial use and, therefore, they are more abundant in fine fraction of waste which is the final stage of the life cycle of materials (goods). Secondly, heavy REEs naturally are less dominant in the Earth's core (Bowen, 1979).

In general, REEs were detected in lower concentration in waste from Högbytorp and Torma landfills if compared with the average concentration of REEs in the Earth's crust (Lide, 2009), but it should be taken into account that averages for the Earth's crust are calculated including high concentration zones of REEs such as carbonatites, volcanic rocks and hydrothermal ores that contain huge amount of elements in comparison to common soils or sediments (Sverdrup and Ragnarsdóttir, 2014).

3.4. Bioavailability of elements in fine fraction of waste

Results acquired during the studies of waste from Högbytorp and Torma landfills on total content of elements and element distribution by fractions can be defined as a screening or preliminary assessment of manner how elements can act in environment that is

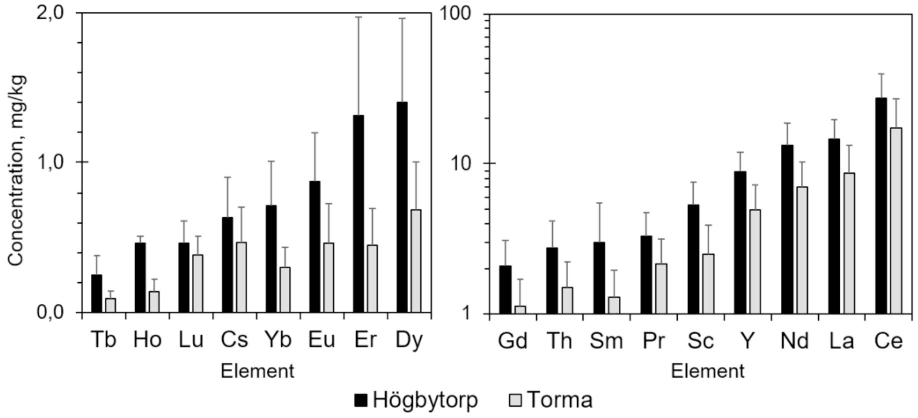

Fig. 5. Total content of REEs in fine fraction of waste from Högbytorp and Torma landfills.

important for describing mobility potential of elements and at the same time to distinguish recovery potential of resources, e.g., water soluble, acid soluble and reducible parts are easy to be extracted while other are characterized by lower extractability (Kabata-Pendias and Pendias, 2010; Pansu and Gautheyrou, 2016). Analysis of fine fraction of waste revealed that many of major and minor elements (Al, Co, Cr, Cu, Fe, Pb, Zn) mostly were bound in residual fraction which means their limited mobility and low bioavailability (Fig. 6).

Derived data indicated that abundance of water soluble compounds can be attributed to such elements as Al, Cu, Fe, K, Mg, Na, Ni and Pb, while weak acid soluble compounds were extracted almost for all major and minor elements, except for Fe, however, distinctions between sampling sites were observable and might be associated with geochemical specifics. For waste samples from Högbytorp reduced forms of elements were dominating for Al, Ca, Cd, Cr, Fe, Mn, Pb and Zn, while for samples from Torma – for Ca, Cd, Fe, Mn and Zn. Assumption can be made that Al and Fe will be leached the last from the media, while Pb, Cu and Ni are less active than, e.g., Zn. Results of waste samples of both landfills indicated that Na and K were among easily bioavailable elements following by Mg, Mn and Co. Differences in fractionation between fine fraction of waste of old and relatively new landfill is likely to be influenced by soil processes, geotechnical characteristics, aeration, pH and redox potential.

Results revealed that water and weak acid extraction provide negligible amount of critical elements and REEs, with very few exceptions. First of all, elements of concern usually are toxic heavy metals which leaching may negatively influence environment *in situ*. Furthermore, if after the LFM industrial extraction is planned or site remediation, or combined operations, toxic elements such as Cd, Cr, Pb should be avoided due to their possible leaching in water streams or could be emitted as aerosols during mechanical excavation and separation. In overall, toxic and potential trace contaminants such as Pb, Cd, Cr, Zn were found in relatively low concentration and it means that safety issues during the use of fine

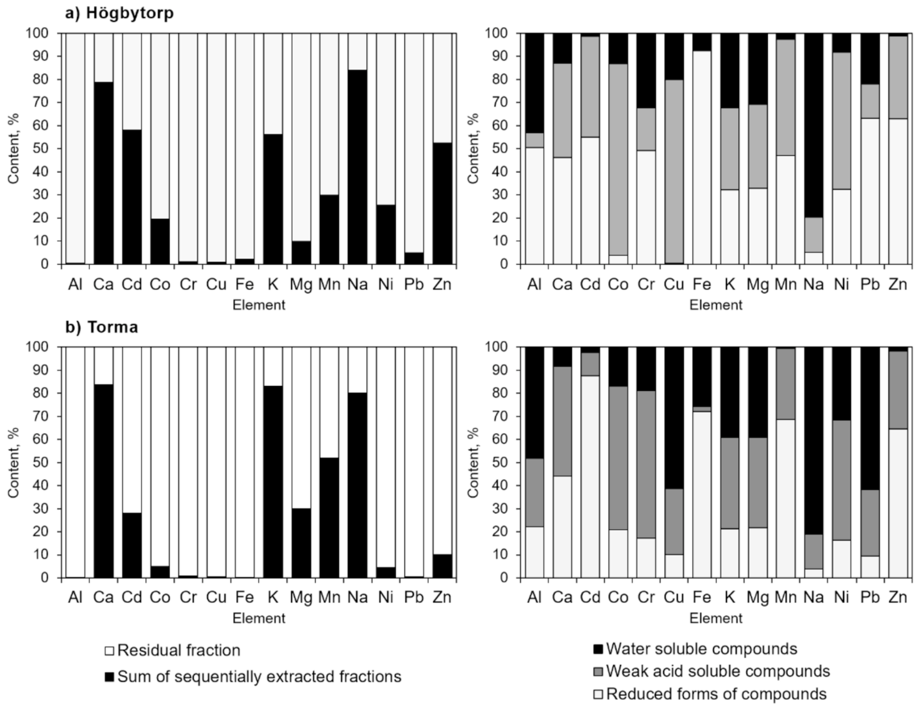

Fig. 6. Distribution of metals and metalloids by fractions of various solubility detected in fine fraction of waste from Högbytorp (a) and Torma (b) landfills.

fraction as recovered material has to be evaluated but does not pose direct hazards for environment as elements mostly are bound in environmentally stable compounds.

Due to low concentration (below the limit of detection), it was not possible to apply sequential extraction for all range of REEs, except for Ce, La, Nd, Sc, Sm and Y; additional data were derived for such minor elements as As, Rb, Ba and Sr (Fig. 7).

It was detected that fraction of reduced compounds was probably affected by the age of landfill, i.e., it was significantly greater for waste samples from Högbytorp (old landfill) than for samples from Torma (relatively new landfill). Proportion of easy extractable REEs such as La, Ce, Nd and Y is relatively higher for waste from Torma landfill. It can be also associated with element leaching during the time, Theoretically, it could be possible that initial amount of REEs in Högbytorp landfill was much higher and declined due to leaching.

3.5. Environmental concerns of reuse of fine fraction of waste

The expected environmental concern from eventual process of LFM regarding aspect of possible leaching of toxic heavy metals can be assessed as negligible. Therefore, one of perspective ways of valorisation of fine fraction of waste nowadays can be expected as use for landfill covering material, but extraction of valuables still is under the great discussion due to high expenses (Burlakovs et al., 2015, 2016). It is explained by deficiencies in methodology to extract metals or other valuables from municipal waste with admixture of industrial waste that has relatively low levels of valuable materials and, e.g., REEs or coloured metals are rather mixed with Ca, Fe and Al in fine fraction of soil and waste. However, REEs industrially are used in very wide spectrum; their prices are volatile, but recycling rates are extremely low. Results on content and proportion of major, minor elements and REEs in landfills tested in screening studies confirm principles of geochemical association regarding general distribution of elements in the Earth's matters. Geological sources of REEs mainly are specific alkaline igneous and secondary deposited rocks (Bowen, 1979); however, the current study deals with purely anthropogenic loads as the fine fraction of waste is coming from the alloys, electronic wastes, nanomaterials and many other sources (Arndt et al., 2017; Burlakovs et al., 2017; Carr et al., 2008; Ridings et al., 2000; Sverdrup and Ragnarsdóttir, 2014). Proportionally higher amount of Cs, La, Nd and Y in fine fraction of waste can also mirror the proportional intensities of the use of REEs in the industry – the beginning of the circular technological loop. Preliminary evaluation of potential use of landfilled waste, dredged sediments from harbours, fly ashes as secondary source of valuable metals and/or construction material is provident due to increasing shortage of natural resources all over the world. The focus on understanding of the main aspects in terms of technologies and economy as well as potential barriers might null the hypothesis of fine-grained fractions of waste being quite promising source of metals for future (Arndt et al., 2017; Burlakovs et al., 2017; Burlakovs and Vircavs, 2011, 2012a,b). As soon as an understanding on how valuable metals are distributed in fine-grained fractions of waste or sediments and their chemical bindings better will be known, separation methods will provide economically feasible process and make it possible to tackle a much smaller volume with high concentrations of metals. According to the studies done by Bolan et al. (2014) and Øygard et al. (2008), the ability to separate fractions with low contamination from those having high contaminant concentrations varies for different soils due to different origin of contamination (e.g., mining related contaminants that occur in mineral phases versus soil contaminated with chemical spillage where contaminants occur as soluble salts and particle coatings). Soil washing is one of the widely used techniques consisting of a physical separation process that utilizes water to concentrate contaminants into a smaller soil volume by means of particle size separation, specific-gravity separation, attrition scrubbing, froth flotation or magnetic. However, according to the study done under supervision of Jensen et al. (2007) soil washing has limited success when dealing with fine fractions due to troublesome handling of such smaller fractions. Furthermore, the authors state the need for an efficient unit that makes the decontamination (extraction) of fine-grained fractions through soil washing towards the beneficial use of the washed material (in this specific case, metals recovery) economically and environmentally feasible.

Another concern might arise from significant amount of toxic substances of organic/inorganic character and for instant pharmaceuticals which also need to be removed and treated in an adequate way which might be expensive. Fine graded particles cause major problems in operational aspects in relation to soil/sediment remediation (Andreottola et al., 2010), and considering the presence of considerable amounts of fine soil fraction in landfills and the presence of metals, it is of great importance to understand how these constraints can be minimized and a method for extraction can be developed in conjunction with the business sector in order to increase economic benefits through efficiency and effectiveness. According to the study done by Zhang et al. (2009), even though various sediment technologies have been tested since the 1990's

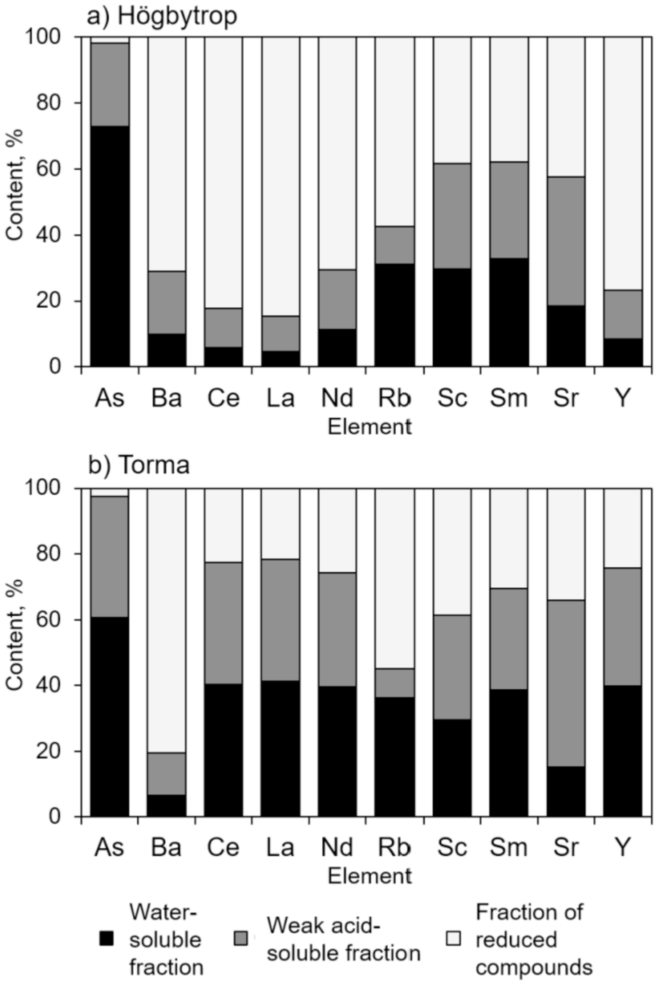

Fig. 7. Distribution of REEs and As, Ba, Rb and Sr by fractions of various solubility detected in fine fraction of Högbytorp (a) and Torma (b) landfills.

most of them have been still in experimental and validation stages emphasizing the needs to investigate and develop proper methods to combine remediation and metal recovery from fine graded waste to be applied in real conditions. Furthermore, it is stated in several studies (Ruan and Xu, 2016; Vandevivere et al., 2001) that efficiency and time required washing and extracting metals from real polluted soils and sediments were longer than the ones required when studying artificially polluted solid matrixes highlighting the importance of carrying out research dealing with real problems and real polluted media.

The fact is that the knowledge of amount of different materials and compounds in a city often is less than the knowledge of the content of in different landfill cells at the landfill site. In the zero waste and beyond the zero waste perspective the goal must be in efficient and economical way recovery all these materials mentioned become resource for others and coming generations to use (EC, 2014). The recovery and recycling must be carried out in such a way that the anthropogenic loops interfere with the sustainable natural cycles so emission to air, earth and waters will be as close to zero as possible.

Studies on speciation and potential hydrometallurgical approaches for extraction of metals, metalloids and REEs are to be continued. Only small proportion of REE deposits including loads of anthropogenic activities can be exploited using existing technologies and therefore these can be referred as ‘reserves’. For environmental purposes, it is beneficial to handle potential hazardous materials including toxic metals and REEs that exist in landfills and make sure these do not re-enter the natural environment and are recycled (Landis et al., 2003). The environmental value in itself might not be enough to cover the economic cost however other aspects such as ecosystem services restoration and real estate regain can add feasibility to the LFM projects in a perspective of circular economy (Burlakovs et al., 2017).

In general, fine fraction of waste in the outlook of future might contain economically viable amounts of REEs if the material is used as a byproduct from processing of large amount of masses suitable for extraction of such elements as Ni, Zn, Fe, Al, Co, Ti, Cr, Cu, V and possibly some other elements depending on specifics of a LFM project (Wang et al., 2017). Higher concentrations of REEs, if present in urban mines, make them to be a great secondary resource for sustainable utilization. It could become a win-win situation for both, the environment and the economy. Pyrometallurgical approach is quite expensive, while hydrometallurgical approach has an advantage of low cost of equipment investment, easy operation and good selectivity of target elements. Biometallurgical process is promising with its low cost and environmental friendliness. Moreover, some alternative new approaches as supercritical fluid, electrochemical and ionic liquid, are tested on a laboratory scale with success (Wang et al., 2017). However, complexity of urban mine waste and its heterogeneity is a limiting factor. Combining of various technologies might be a promising future solution.

The future hypothesis is that portion of ancient waste might achieve important potential that can be referred as ‘reserves or bank account’ nowadays for the future of mankind.

4. .Conclusions

Research of fine fraction of waste from Torma and Högbytorp landfills provided results on content of major and minor elements and REEs that can indicate potential value of recoverable resources. Recovery of major elements and REEs can be applied simultaneously to ongoing remediation projects of landfills and/or degraded industrial soils. Although the concentration of critical major elements and REEs is significantly lower than in mining and secondary resources mono-landfills (industrial dump) areas; the concentration of such elements as Fe, Al, Cu, Pb, Ni and some other might become of interest for extraction in more or less near future. Studies on speciation of elements have shown that some part of fine fraction of waste contains easy extractable fractions that dually can provide extraction opportunity and pose environmental concerns. Only small portion of major and minor elements combined with deposits of REEs can be exploited using existing technologies, therefore, this potential can be referred as a ‘reserve or bank account’ for the future of the world. It would be great benefit to reconsider materials that exist in landfills and return it to the economic loop thus saving resources and removing from biogeochemical anthropogenic cycle. Higher concentrations of REE, if present in urban mines, make them to become a great secondary resource applicable for sustainable utilization, useful as well as for the economy, as for the environment. The main and the most problematic limiting factor is a complexity of urban mine waste and its heterogeneity. Future solutions of recovery of REEs obviously will be a complex process consisting of combination of various technologies. The economic cost, however, still is too high, but other aspects, such as ecosystem services restoration and real estate land reclamation can add feasibility to the LFM projects in a circular economy perspective.

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