



Heavy metal mobility and valuable contents of processed municipal solid waste incineration residues from Southwestern Germany



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ABSTRACT

As conventional end-of-life disposal, municipal solid waste (MSW) incineration residues can be problematic due to potential release of toxic compounds into the environment. Using municipal solid waste incineration residues as urban-mine of valuable metals (e.g. precious metals) could provide a trash-to-treasure possibility. The objectives of the study are to (i) determine the contents of different contaminant metallic elements (Zn, Cu, Ba, Pb, Cr and Ni) in four size fractions of MSW incineration residues and discuss their mobility potential by using the modified BCR sequential extraction method; (ii) investigate the level of valuable critical contents (precious metals, rare earth elements, etc.) in these wastes. We also characterized mineralogy and elemental composition of four different grain size fractions (0–0.5, 0.5–2.0, 2.0–4.0 and 4.0–16.0 mm) of processed municipal solid waste incineration residue (PIR) from the Southwestern region of Germany, using X-ray fluorescence, X-ray powder diffraction and different spectroscopic techniques. Among all studied size fractions, grains smaller than 2 mm contained higher amounts of total extractable heavy metals in most cases. The most important finding of the study is that the total contents of Cu, Au and Pt in the incineration residues reached economically profitable levels (5.1 g/kg, 21.69 mg/kg and 17.45 mg/kg, respectively).

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1. Introduction

In many industrialized countries, thermal waste treatment is considered one of the mainstays of waste management strategy. Municipal solid waste (MSW) apart from recyclable paper, plastic, metal and organic waste, is combusted to generate electricity, heat and/or process steam. Incineration of MSW reduces the volume of waste by over 90%, and the mass by up to 85% (Chandler et al., 1997; RenoSam and Rambøll, 2006; Ginés et al., 2009). Continuous developments of technology over the recent decades have advanced the incineration process to assure that the byproduct flue

gas meets air quality standards (Belevi and Moench, 2000). As a result, for nations short on landfill space, incineration instead of MSW landfilling is a more reasonable solution. In Europe, for instance, waste incineration has grown steadily. Since 1995, the amount of municipal waste incinerated in the EU-27 Member States has risen by 32 million tons or 100% and accounted for 64 million tons in 2014 (Eurostat, 2016). In Japan, due to the limited habitable land and pressure of waste volume reduction, up to 75% of MSW is incinerated, producing approximately 6 million tons of residue annually (Sakai and Hiraoka, 2000).

However, the incineration process is not the final stage of waste treatment of MSW (Hjelmar, 1996). Although most organic-based materials are destroyed by complete oxidation to carbon dioxide and water vapor during incineration, inorganic compounds remain present at substantial amounts (Chandler, 1997). The major

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environmental concern has shifted from air emissions during incineration to the solid residues, since those residues are commonly disposed in landfills, potentially leading to soil and water pollution (Van der Sloot et al., 2001). Even though leaching of residues could be controlled, long-term environmental impacts remain unclear. There will be more stringent regulations and measures that restrain the MSW incineration residues disposal as landfills in the near future. Also, securing landfill sites is more and more difficult, particularly in populated areas (Sakai and Hiraoka, 2000). Overall, there is a discernible trend to reutilize MSW incineration residues for different applications with the emerging recycling philosophy.

The composition of MSW incineration residues is influenced by the MSW source and incineration process as well as other factors. MSW incineration residues include bottom ash, grate siftings, boiler and economizer ash, fly ash and air pollution control (APC) residues (Chandler et al., 1997). Each residue is produced from a different process step during incineration. Strategies to reuse MSW incineration residues have been previously reviewed and discussed (Ferreira et al., 2003; Lam et al., 2010) and these residues have significant potential for the recovery of scrap metals (Shen and Forssberg, 2003; Allegrini et al., 2015). Among all applications, utilization of MSW incineration residues as construction materials is widely studied (Ferreira et al., 2003; Lin et al., 2003; Forteza et al., 2004; Ginés et al., 2009; Jinyoung Kim, 2014). However, disposal and utilization of the MSW incineration residues are both limited by their composition. A number of studies reported the presence of hazardous metals in MSW incineration residues, including Zn, Pb, Cd, and Cu (Bosshard et al., 1996; El-Fadel et al., 1997; Meima and Comans, 1999; Crannell et al., 2000; Dijkstra et al., 2006). Moreover, in regions where natural sources such as clay, sand and gypsum or other secondary sources (ash from coal power stations) are abundant, utilization of MSW incineration residues for construction material might lack economic benefit. Therefore, two aspects are important for the management of MSW incineration residues: (i) knowledge of the total and leachable metals (hazardous components) in MSW incineration residues and (ii) evaluating other potential uses and values of MSW incineration residues.

Over the last few decades, some researchers have reported the occurrence of precious metals and rare earth elements in MSW incineration residues (Greenberg et al., 1978; Zhang et al., 2001). Concentration levels of these elements in waste solids were usually considered too low compared to those present in natural ores, thus, there was a limited economic benefit to recover these metals from MSW incineration residues. However, different metal-containing waste streams now need to be taken into account as resource of metals regardless of their low grade and quality, because many economies in countries and areas including the European Union, United States, Japan and Korea are confronted with an increasing supply risk of critical raw materials (Tercero, 2013). Economic utilization of waste streams such as MSW incineration residues, which can be considered as new and unexplored resources, may play a crucial role in alleviating the resource scarcity stress and securing sustainable uses and supplies of these critical metals in the future.

In this study, we characterized mineralogy, elemental composition of five different grain size fractions (<0.5, 0.5–2.0, 2.0–4.0, 4.0–16.0 and >16.0 mm) of an end-of-life material (waste), which is the tailing of MSW incineration residues (bottom ash, fly ash and APC residues) after the recyclable metal scrap had been removed by a German metal recovery plant. We performed modified BCR procedure to extract heavy metals from each fraction to investigate the mobility potential of those contaminants under different extraction conditions. Moreover, this study provides insights for management of processed MSW incineration residues with regard to

opportunities of utilizing solid hazardous waste as valuable metals mines to approach sustainable use of critical materials (e.g. precious metals, rare earth elements, etc.). To simplify the name of the material, we call it PIR, representing Processed (Municipal Solid Waste) Incineration Residue.

2. Material and methods

2.1. Origin of PIR

PIR was kindly provided by a MSWI residue processing plant, which processes regional MSWI residues from southwestern areas of Germany (Hessen and Baden-Württemberg) by ageing, sieving, crushing (>2 mm fractions), magnetic recycling and eddy current separation to recover ferrous and non-ferrous metals. After recycling, the rest of all fractions are piled together prior to disposal in landfills or further use as construction material (Holm and Simon, 2017). The PIR material for the present work had been stored for 6 months (from late spring until late autumn) at an open-air site (3 months before and 3 months after the recycling process). Approximately 100 kg of bulk samples were taken from different depths of the pile (1–2 m) from the storage site and kept in the laboratory in five sealed plastic buckets at room temperature. Before characterization, PIR in each of five buckets was mixed and 2 kg of PIR from each of five plastic buckets were transported to one empty container and mixed. Mixed PIR were air-dried in the fume hood for three days.

2.2. Determination of pH

The PIR pH was directly measured using a benchtop pH-meter (inoLab pH 7110, WTW GmbH) equipped with a Mettler Toledo electrode (InLab Easy DIN) in a settling suspension of 10 g of PIR (air dried) in 25 mL of MQ-H₂O (solid to water ratio of 1:2.5) after 1 h, 24 h and 7 days in triplicates.

2.3. Characterization of different grain size fractions

2.3.1. Pretreatment of PIR – size fraction separation

The mixed PIR was air-dried in the fume hood for 3 days and sieved into fractions of <0.5 mm, 0.5–2.0 mm, 2.0–4.0 mm, 4.0–16.0 mm and >16.0 mm (fractions I, II, III, IV and V, respectively) with an analytical vibration screening instrument (Vibratory Sieve Shaker Analysette 3, Fritsch GmbH). Before analysis, all fractions were dried at 105 °C for more than 24 h and milled by planetary mill (Fritsch Pulverisette, Fritsch GmbH) to fine powder (<2 mm). The powdered samples were stored in 50 mL, polypropylene centrifuge tubes (Orange Scientific or SLG Süd-Laborbedarf Gauting) at room temperature in desiccator dryer with silica gel prior to other analysis.

2.3.2. Mineralogy

The mineral composition of milled PIR fractions was determined by XRD, which was carried out with a Bruker D8 Discover GADDS XRD²-microdiffractometer. The device is equipped with a Co-K α radiation source, a primary graphite monochromator, and a 2-dimensional HI-STAR-detector (Berthold et al., 2009). Crystalline mineral phases were identified with the internal database of the EVA software (version 10.0.1.0).

2.3.3. Elemental analysis

The major elemental composition of milled PIR fractions was determined by X-ray fluorescence (Bruker AXS S4 Pioneer XRF device, equipped with a 4 kW Rh-tube). For analysis, 6.0 g of sample powder were mixed and homogenized with 1.2 g of wax and

pressed to a pellet with 300 kN/m². The data was analyzed using an internal database from Bruker AXS (MultiRes-Vac34) with specific reference materials.

To determine trace elements, milled fractions were digested by a microwave-assisted technique with aqua regia. The digests were analyzed by ICP-QMS (Thermo iCap-Q). Detail of microwave digestion procedures and pretreatment of samples are described in SI.

Supplementary data associated with this article can be found, in the online version, at <https://doi.org/10.1016/j.wasman.2018.08.010>.

Aqua regia digests were analyzed by ICP-OES (SPECTROBLUE TI, Ametek) to quantify Au and Pt. Measurement of elements of typical environmental concern (Table 1) was done via MP-AES (4100, Agilent).

2.4. Sequential extraction of metals

For sequential extraction of metals from PIR we used a modified BCR protocol described by Rauret et al. (1999) and Oyeyiola et al. (2011) similar to Smeda and Zyrnicki (2002) who applied the modified scheme to fly ash. Milled dry samples of the PIR fractions (1.000 ± 0.001 g) were extracted in duplicates under oxic conditions. For each extraction step, two blank samples without PIR were performed. The detailed protocol for the sequential extraction is presented in Table S1.

3. Results and discussion

3.1. Particle size distribution

In order to assess the particle size distribution of PIR, we used sieve analysis to separate PIR into five different fractions I to V (<0.5, 0.5–2.0, 2.0–4.0, 4.0–16.0 and >16.0 mm) and found a distribution of 16.0% (fraction I), 30.2% (fraction II), 21.2% (fraction III) and 30.1% (fraction IV; Fig. 1). Fraction V was not further investigated since it contained mainly big ceramics and glass pieces and it accounted for only a very small part (2.5%) of PIR.

3.2. Mineralogy

Based on XRD analysis, seven major mineral phases could be identified in each grain size fraction: quartz (SiO₂), hematite (Fe₂O₃), calcite (CaCO₃), magnetite (Fe₃O₄), anhydrite (CaSO₄), akermanite (Ca₂Mg(Si₂O₇)), and gehlenite (Ca₂Al₂SiO₇). Additionally, the diffraction patterns obtained by XRD analysis of fractions I–IV (Fig. S1) showed a high number of small signals, which originate from crystalline compounds that are below detection limit.

In most cases, formation of (hydr)oxides during incineration contributes to the alkalinity of the solid residue (Chandler et al., 1997). The use of hydrated lime as flue gas cleaner can also lead to an extremely high pH value of APC residues (Belevi et al., 1992). The high pH value of the PIR initially prevents the mobilization of some metals. Formation of calcite starts immediately in the APC and can be prolonged over the time span of temporary storage or landfilling of the material (Johnson et al., 1995).

3.3. pH of PIR

During the carbonation process, alkaline minerals capture atmospheric CO₂ (e.g. calcium hydroxide into calcite (Rendek et al., 2006)) leading to a decrease of the pH value (Meima and Comans, 1998) and partial dissolution of sorbed heavy metals (e.g. Cd, Pb, Zn, Cu and Mo (Meima and Comans, 1999)). This explains the pH value of incineration residues decreased from 11.7 (without ageing, sample from MSWI plant) to 10–11

(10.69 ± 0.01, 10.79 ± 0.01 and 10.53 ± 0.03, suspension measured after 1 h, 24 h and 7 days, respectively) (aged PIR, this study).

The oxidation of Fe during ageing of MSW incineration residues promotes formation of Fe oxides (e.g. hematite and magnetite). This process is accompanied with sorption of heavy metals on Fe minerals, including Pb, Cd, Zn, Ni, Mo, Cr and Cu (Sabbas et al., 2003). Similar sorptive properties are also displayed by other mineral phases, including Fe and Al (oxy)(hydr)oxides, amorphous aluminosilicates and calcium aluminosilicate mellilite group, including akermanite and gehlenite (Sabbas et al., 2003; Zhou and Haynes, 2010).

3.4. Elemental analysis

3.4.1. Overview

The elemental composition of PIR as determined by XRF, MP-AES, ICP-OES and ICP-QMS is presented in Table 1. The major elemental constituents (>10,000 mg/kg) in the incineration residues are Si, Ca, Fe, Al, Na, Mg, K, P and Ti, except the non-determined light elements such H, C, N and O. Minor constituents (1000–10,000 mg/kg) include Zn, Cu, Mn, Ba and Pb. Trace constituents (<1000 mg/kg) include Cr, Ni, Sn, V, Mo, Ce and many other elements. The observed concentrations of Na, Mg and K are fairly typical for MSW bottom/fly ash or APC system residues. The concentrations of Si and Al are above typical ranges that were commonly observed in such wastes (Chandler et al., 1997). Ca concentrations are greatly above typical ranges for bottom/fly ashes (Crannell et al., 2000) but are typical for APC system residues (Quina et al., 2008). This may result from the mixture of bottom ash, fly ash and APC residues prior to waste disposal in landfills. APC residues, which contain high levels of calcium, may associate with bottom and fly ash during the disposal and ageing, thus leading to an apparent higher level of Ca in PIR. The concentrations of minor elements (e.g. Zn, Cu, Ba, Pb) were within the range of reported values for bottom/fly ash and APC residues (Allegrini et al., 2014). The concentrations of valuable trace elements such as precious metals and rare earth elements will be discussed in Section 3.5.

3.4.2. Element distribution in different size fractions

As shown in Table 1, Si concentration in these four size fractions positively correlates with particle size of the incineration residues. Glass, ceramics and other Si-containing components contribute to the high Si content in the municipal solid waste. General properties such as high melting temperature, high hardness, high moduli of elasticity and chemical resistance prevent these materials being easily ground into smaller particles (Black and Kohser, 2011). Thus there is a typical trend to have higher Si content in larger size fractions of bottom ash and slag of waste incinerations (Syc et al., 2015).

In contrast, the concentrations of Ca, Al, Zn, Nd, Ti, Cd, Tl and some other trace metals in these four size fractions decrease with increasing grain size, showing an opposite trend as the distribution of Si. Arsenic concentration follows a similar trend, except that fractions III and IV appear to contain an equivalent quantity of this element. As for Ba, V and Mo, size fraction II (0.5–2.0 mm) contains the highest levels of these hazardous elements among all fractions. The concentration of Pb, Cr and Ni vary significantly in the fraction II. At the same time the higher content of these metals seems to be more typical for fine fractions (I and II).

3.5. Sequential extraction of hazardous metals

Heavy metals are an integral part of MSW incineration residues (Lam et al., 2010). Untreated wastes ash can be harmful to environment by releasing these heavy metals at landfills. It is essential to

Table 1
Metal content of typical environmental concern, REEs, precious metals and other critical metals in each of the four size fractions of PIR. Values reported in mg/kg with standard deviation. Total content of metals in original PIR (I–IV fractions, >97% of weight of material; right column) calculated on the base of mass ratios of the size fractions (Chart 1) and content of metals in each of the four size fractions.

Element	Size fractions of PIR				Total content in original PIR (in mg/kg)
	I	II	III	IV	
<i>Elements of typical environmental concern (quantified by MP-AES), mg/kg</i>					
Zn	7189.91 ± 118.12	5548.11 ± 325.75	4663.18 ± 151.22	3451.32 ± 715.11	4969.33
Cu	4785.37 ± 291.08	7665.10 ± 2047.18	4021.10 ± 2015.01	3169.08 ± 317.98	5124.94
Ba	1773.25 ± 987.91	2193.01 ± 131.14	1922.07 ± 181.28	1788.00 ± 103.05	1939.26
Pb	1105.28 ± 163.84	1445.90 ± 965.05	1122.48 ± 342.38	755.3 ± 42.11	1103.73
Cr	538.10 ± 28.2	1099.21 ± 1014.57	511.62 ± 138.85	325.30 ± 35.64	639.40
Ni	422.63 ± 34.86	694.76 ± 414.49	325.46 ± 82.59	214.14 ± 11.55	420.84
Sn	165.11 ± 11.87	195.3 ± 38.39	201.59 ± 6.96	90.89 ± 7.63	158.56
V	36.88 ± 0.81	56.21 ± 19.73	36.53 ± 0.66	31.06 ± 0.77	40.97
Mo	29.99 ± 0.77	43.65 ± 11.91	30.47 ± 1.71	22.59 ± 1.10	31.99
As	14.7 ± 0.42	12.81 ± 0.18	11.24 ± 0.2	11.03 ± 0.59	12.23
Cd	6.09 ± 0.28	2.71 ± 0.1	1.85 ± 0.28	0.83 ± 0.01	2.49
Tl	0.07 ± 0.002	0.06 ± 0.01	0.03 ± 0.01	0.03 ± 0.001	0.05
<i>Major elements (quantified by XRF), mg/kg</i>					
Si	197 600.00	324100.00	363000.00	513400.00	371389.97
Ca	378800.00	253900.00	239400.00	165400.00	243369.25
Fe	84500.00	174700.00	162200.00	109800.00	136703.22
Al	134800.00	96400.00	91100.00	89500.00	99422.79
Na	18500.00	17300.00	22100.00	43400.00	26767.40
Mg	25800.00	16500.00	17200.00	18100.00	18681.61
K	17600.00	16800.00	17100.00	14100.00	16138.37
P	39200.00	14300.00	13600.00	7100.00	15966.05
Ti	18900.00	16000.00	15200.00	10000.00	14408.85
Mn	1800.00	3500.00	4100.00	1500.00	2711.66
<i>Rare Earth Elements (REEs; quantified by ICP-QMS), mg/kg</i>					
Ce	27.02 ± 2.04	28.4 ± 1.04	27.32 ± 0.78	24.59 ± 0.81	26.74
Nd	22.64 ± 3.80	21.47 ± 6.00	12.11 ± 0.09	12.48 ± 0.21	16.86
La	18.76 ± 0.15	17.16 ± 0.27	17.46 ± 1.67	14.51 ± 0.67	16.65
Y	7.11 ± 0.05	7.90 ± 0.20	8.12 ± 0.21	6.29 ± 0.32	7.31
Pr	5.07 ± 0.64	5.19 ± 1.22	3.05 ± 0.03	3.38 ± 0.02	4.15
Sm	4.25 ± 3.38	1.87 ± 0.09	1.76 ± 0.07	1.82 ± 0.07	2.22
Gd	3.07 ± 0.28	1.91 ± 0.07	2.15 ± 0.46	2.55 ± 0.04	2.35
Sc	2.27 ± 0.02	2.42 ± 0.06	2.28 ± 0.04	2.07 ± 0.07	2.26
Dy	2.01 ± 0.46	1.95 ± 0.53	1.29 ± 0.06	1.11 ± 0.05	1.56
Er	0.66 ± 0.01	0.77 ± 0.02	0.79 ± 0.02	0.82 ± 0.03	0.77
Yb	0.63 ± 0.02	0.68 ± 0.03	0.62 ± 0.01	0.52 ± 0.02	0.61
Eu	0.36 ± 0.003	0.37 ± 0.01	0.37 ± 0.01	0.35 ± 0.03	0.36
Tb	0.24 ± 0.01	0.28 ± 0.02	0.29 ± 0.04	0.24 ± 0.01	0.26
Ho	0.22 ± 0.002	0.26 ± 0.03	0.34 ± 0.10	0.20 ± 0.01	0.25
Tm	0.08 ± 0.0002	0.09 ± 0.001	0.09 ± 0.001	0.08 ± 0.001	0.08
Lu	0.07 ± 0.0007	0.09 ± 0.005	0.08 ± 0.002	0.07 ± 0.002	0.08
<i>Precious metals (quantified by ICP-OES), mg/kg</i>					
Au	18.67 ± 2.19	28.82 ± 1.62	29.83 ± 2.55	10.93 ± 0.42	21.69
Ag	9.57 ± 0.79	8.84 ± 1.35	6.32 ± 1.2	8.38 ± 2.21	8.29
Pt	20.57 ± 3.71	21.42 ± 2.19	18.64 ± 2.3	11.16 ± 0.64	17.45

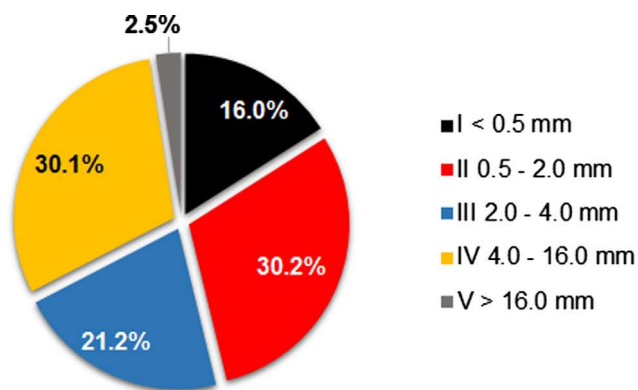


Fig. 1. Grain size distributions of PIR: mass ratios of fraction I, II, III, IV and V.

quantify the leachable hazardous metals present in the PIR and to study their mobility as a function of the physio-chemical property of solid wastes under different leaching conditions.

We found that the hazardous metals Zn, Cu, Ba and Pb were present as major heavy metal contaminants in PIR (>1000 mg/kg); Cr and Ni were present at lower concentrations (100–1000 mg/kg). We therefore quantified the leachable Zn, Cu, Ba, Pb, Cr and Ni from different size fractions by using a modified BCR protocol (Table S1) consisting of four successive steps (by using water, acetic acid, the reducing agent hydroxylammonium hydrochloride and the oxidant hydrogen peroxide, respectively). Fig. 2 shows the metal extraction levels (mg per 1 kg of each separated fraction) of PIR in each extraction step. The pie charts indicate the extraction efficiency of mixed PIR (sum of amounts of leached metal from four fractions (I–IV) to the total metal content) under different extraction conditions.

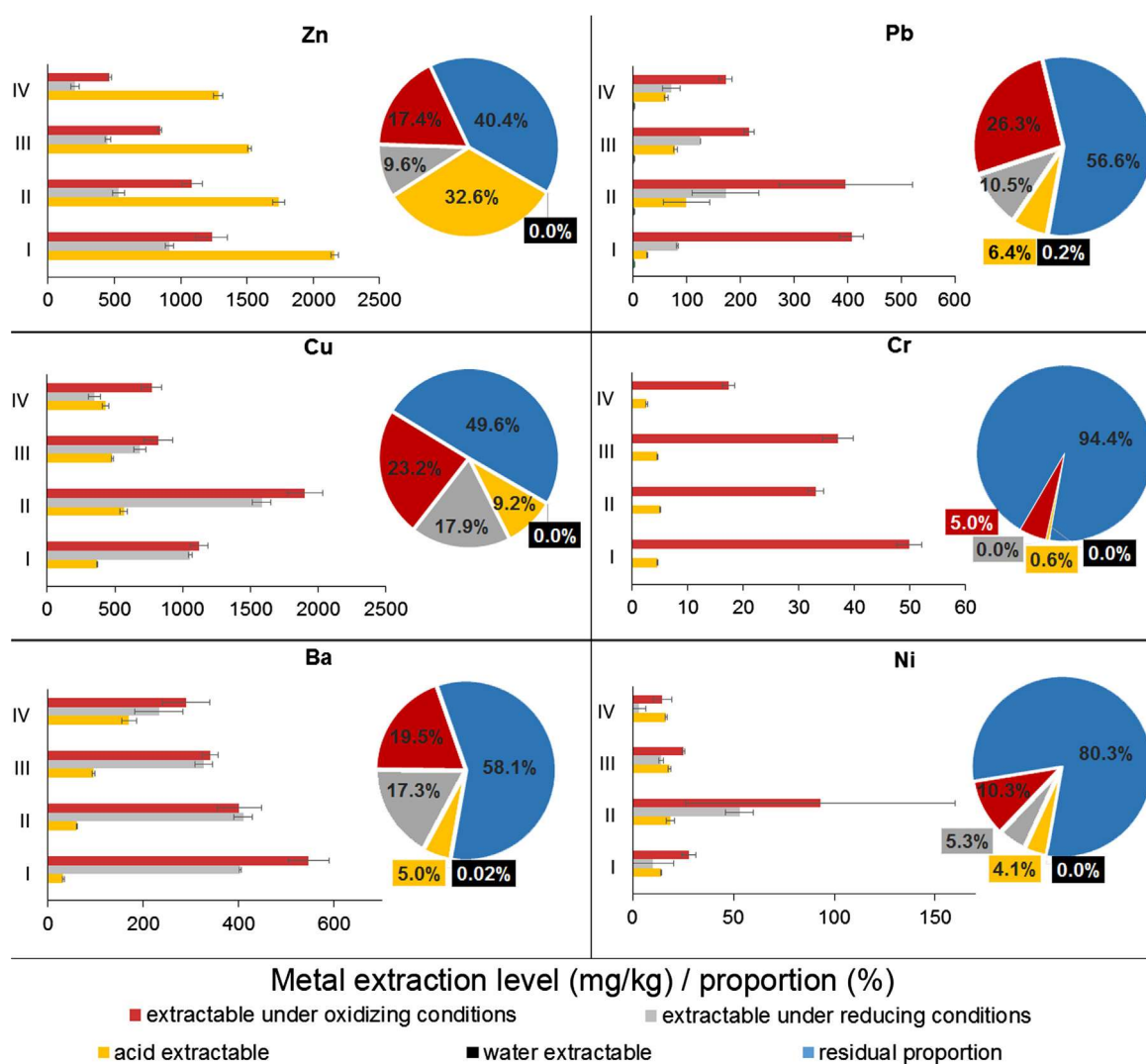


Fig. 2. Sequential extraction of Zn, Cu, Ba, Pb, Cr and Ni from four individual size fractions of PIR. Pie charts indicate the total proportion of metals (the sum of the four size fractions) under each extraction condition relative to the total metal content.

3.5.1. Metal extraction depending on particle size

Particle size is one of the crucial factors that may influence the leaching behavior of PIR. Firstly, the MSW incineration residues include bottom ash, fly ash, APC residues, etc., which exhibit different properties including grain size and composition. The distribution of toxic metal species in different residue streams depends on their chemical behavior as individual elements or compounds under certain temperature during combustion. Volatile species may be enriched in flue gas and they have longer time to interact with fly ashes, thus fine-grained residues contain higher concentration of these species (Chandler et al., 1997). Secondly, at the MSW incineration recycling plant, solids with different grain sizes had received different treatments: only the solids larger than 2 mm were crushed and magnetically sorted for metal recycling while the fraction <2 mm was not used for metal recovery and is therefore expected to contain a higher metal content. Moreover, weathering and ageing processes can occur during storage for three months after recycling process resulting in further change of composition of the waste solids (Belevi and Moench, 2000).

Our results suggest that the amount of leached Zn increased with decreasing PIR particle size (under all leaching conditions except in step 1). The Zn contained in the PIR was found not to be extractable by water (which had a pH > 10 after addition to

the PIR) suggesting that Zn occurs as insoluble species at slightly alkaline pH. Size fraction II contained the highest amount of extractable Pb and Cu under weak acidic or reducing conditions. During these two leaching steps, the levels of extractable Pb and Cu increased with decreasing particle size, among fractions II to IV. Under oxidizing conditions, the fine particles (size fractions I and II) of PIR released higher levels of Pb and Cu than the more coarse fractions (size fractions III and IV). The coarsest fraction, IV, contained the lowest amount of extractable Cr, where Cr could only be successfully extracted by acid or the oxidizing reagent hydrogen peroxide. The amount of leached Ba increased with decreasing PIR particle size under both oxidizing and reducing conditions. Conversely, acid extractable Ba increased with increasing PIR grain size. Fraction II contained the highest amount of extractable Ni (except in step 1, there was no water extractable Ni). Generally, fraction contains high total content of HM (humic matter) releases more HM under certain conditions (Table 1). For example, the highest amount of Zn was released from fraction I of PIR which contained the most Zn. The maximal amount of Cu was extracted from fraction II of PIR, which contained the highest Cu level. The same phenomenon was found for Ba, Pb and Ni.

Our results suggest that along with the fact that the fine fractions (fractions I and II) contain more metals, they also leach

more heavy metals. Therefore the fine fractions of PIR are more hazardous and they should be stored and treated separately from coarse fractions from incineration plant to final disposal site.

3.5.2. Metal extraction depending on extraction condition

3.5.2.1. Water extractable metals. Treatment of initial alkaline PIR (pH > 10) with water did not greatly change the pH of the material and the pH of extracts I–IV remained in a range of 10.5–10.8 (Fig. S2). Leachability of highly soluble species (e.g. alkali salts) is almost pH-independent (Sabbas et al., 2003). Among all the studied harmful metals, only Pb and Ba were extracted by water under such alkaline conditions. The extracted Pb may result from dissolvable species (e.g. PbO, PbCl₂, Pb₂O(OH)₂ and PbSiO₃) under alkaline conditions, which are usually found in MSW incineration ash (Ménard et al., 2006; Funatsuki et al., 2012). Although there are little publications studying the species of Ba in MSW incineration residue, we assume that the extracted Ba species could be soluble barium hydroxides or chlorides, based on the pH of extracts and our XRF results. The absence of leachable Ba in the finest fraction could be due to a relative higher concentration of sulfates in the APC residues, which came from SO₂-rich flue gas of the incineration plant. Excessive sulfates form stable and poorly soluble BaSO₄ in the finest fraction. Although the leaching efficiencies of Ba (0.02%) and Pb (0.2%) present in the water-soluble leachates was low, it has to be taken into account that the PIR had been exposed to the ambient atmosphere (air) for approximately 6 months (from late spring till late autumn) before sampling. During this time, a significant fraction of the Ba and Pb could have been leached already by water (e.g. by rain) at the storage site. Moreover, considering the great amount of MSWI being produced and disposed, the absolute amount of leached Ba and Pb can still cause environmental issues.

3.5.2.2. Extraction of metals under acidic conditions. During storage of the incineration waste, industrial sources (wastewaters, uncontrolled chemical pollution, etc.), acid rain or contact with soil solutions can be the cause for the decrease in pH value (Bruder-Hubscher et al., 2002). These anthropogenic and natural events can influence the mobility of heavy metals in PIR at storage sites or after being used as construction material.

In order to understand how pH influences the mobility of metals we determined pH values of the leachates after treatment with acetic acid (Fig. S2). The pH values of leachates obtained under acidic conditions were 5.0, 4.5 and 4.3, respectively for fraction I to IV. At this pH range the solubility of some metals increases compared to the initially highly alkaline material (Meima and Comans, 1999). The pH value of the incineration waste decreases as the material comes into contact with the acidic leaching solution and alkalinity is removed from the system (Sabbas et al., 2003).

Zinc, Cu, Pb, Ba, Cr and Ni were released under acidic conditions (after addition of 40 mL 0.11 M of acetic acid), supposedly due to neutralization of alkalinity and dissolution of carbonates and oxides that were earlier formed during the incineration and ageing process. The formation of calcite and other carbonate minerals is expected to be accompanied by co-precipitation and sorption of tested metals. Therefore its dissolution leads to the release of associated metals (Wunsch et al., 2014). Among all the studied metals, only Zn was extracted with a higher efficiency (32.6%) by acid extraction compared to the extraction efficiency under oxidizing conditions after addition of H₂O₂ as oxidizing reagent. This suggests that ZnCO₃ is one of the dominating Zn-containing components in PIR potentially posing a great risk to the environment since natural rain has a pH < 5.6 due to the CO₂, SO₂ and NO_x in the atmosphere (Charlson and

Rodhe, 1982). Although acid extractable proportions were below 10% for Cu, Pb, Ba, Cr and Ni, the large volume of MSWI waste can still leach enormous amount of hazardous metals into the landfill sites.

3.5.2.3. Extraction of metals under reducing and oxidizing conditions. Redox reactions can also play an important role for the release of heavy metals from MSW incineration residues. These reactions can involve H₂ and Fe(II) as reducing agents or O₂ and H₂O₂ as oxidizing agents (Song et al., 2016). We demonstrated that Zn, Cu, Pb, Ba and Ni were released under reducing conditions during extraction with 40 mL 0.5 M hydroxylammonium hydrochloride (Table S1). These metals could be present as metal oxides or be associated with Fe–Mn oxides. Dissolution of these minerals under reducing conditions is expected to lead to the release of associated metals (Marin et al., 1997). Surprisingly we did not identify Cr in the leachates obtained under reducing conditions although Cr oxide is often used as a protective layer preventing further oxidation of stainless steel (Chen et al., 2005). Releasing of metals associated with iron oxides (e.g. hematite and magnetite) can be accelerated in nature due to the formation of locally reducing conditions caused by industrial sources (e.g. contact with reduced wastewaters) or by anaerobic biodegradation of organic matter caught in PIR during ageing (Wan et al., 2006).

The highest metal extraction efficiencies, with the exception of Zn, occurred under oxidizing conditions. The oxidation and dissolution of metal sulfides or metals associated with sulfides can occur under oxidizing conditions (Marin et al., 1997). However, due to the fact that the mineralogical analysis did not reveal significant concentrations of sulfides, we believe that sulfides play a minor role in the incineration waste investigated in our study. Instead, the release of heavy metals under oxidizing conditions can also be explained by oxidation of elemental metals including Al, Fe and Cu (Sabbas et al., 2003).

3.6. Strategic management of PIR

3.6.1. Use of PIR as construction material or disposal in landfills

Revealed by the BCR sequential extraction analysis, metal leaching behavior can be greatly altered when changing the geochemical conditions from basic to weakly acidic. Additionally, potential leaching of hazardous components can further occur in the case of changes in redox conditions. This dynamic leaching behavior constrains the reuse of PIR as material in road construction and landscaping, by which these toxic metals may be introduced into soil and aquifers by runoff. Among all fractions of the investigated PIR, finer fractions (I and II) release more HMs under different extraction conditions. Although the PIR could be heterogeneous, there is still a trend that fraction II contains higher HMs (Cu, Pb, Cr and Ni) than the other fractions. Also the fraction II accounts for a large proportion (30.2%) of the PIR, thus, it can potentially pose great threat to the environment as disposal by releasing large absolute amount of HMs (e.g. 1 ton of PIR can release 539 g of Zn contributed by fraction II under weak acidic condition). Eliminating the finer fractions before reuse of PIR as construction materials could be beneficial for industries in consideration of meeting regulation and standard on quality of raw materials issued by authorities. Therefore, different grain size particles require specific treatments in order to prevent insufficient detoxification or excessive detoxification leading to a waste of financial resources. The reuse and the disposal strategies for these residues therefore must consider the composition of the different size fractions, the specific requirements of industry, and finally all regulations and measures of the government.

3.6.2. Waste incineration residues as urban “mine” of valuable metals and REEs

We found that Au, Ag and Pt contents in the grain size fractions I–IV of the PIR were 21.69 mg/kg, 8.29 mg/kg and 17.45 mg/kg, respectively (Table 1). This suggests that the values of Au and Pt reached a level interesting and potentially suitable for economic metal recycling. Therefore, the PIR along with ores and other secondary materials can be considered as a source of valuable metals (Shen and Forssberg, 2003; Cui and Zhang, 2008; Boduen et al., 2014). Iron oxide copper gold ore deposits in Australia, Africa South and North America contain from 0.1 to 3 mg/kg of Au (Shaofeng and Shuixing, 2016). Slag-dust dumps of pyrometallurgical conversions in Siberian region (Russia) contain platinum group metals and Au varying from 1.0 to 2.2 mg/kg, 0.4–1.2 g/kg of Ni, and 2.0–3.7 g/kg Cu (Boduen et al., 2014). Most of the Pt is typically obtained from ores with a Pt content of 10–20 mg/kg (Hermann Renner, 2000) suggesting that the extraction of Pt from PIR is potentially economically beneficial. The relatively low Ag content in our incineration waste (8.29 mg/kg) suggests that PIR may lack economic interests for Ag recovery since Ag extraction usually reaches profitable levels when extracted from raw materials with >100 mg Ag/kg (Purcell and Peters, 1998). At the same time the concentrations of Au and Pt that we found in PIR (21.69 and 17.45 mg/kg, respectively) were significantly higher than that from comparable material from a Swiss MSW incineration plant (0.4 ± 0.2 mg/kg of Au and 0.059 ± 0.022 mg/kg of Pt (Morf et al., 2013)) as reported. The calculation of the final quantities of these metals was made with using of three separately taken and pre-treated samples. However, we believe that additional analyses of different batches of PIR taken at different times of the year are required in order to get reliable results since the quantified values of Au and Pt are important for seriously considering the application of PIR recovery techniques.

Copper is also another metal of interest to recover from the PIR. The total Cu concentration in fractions I–IV was 5.1 g/kg. This value is similar to the Cu content in the porphyry deposits (5.0–7.5 g/kg of Cu (Cooke et al., 2005)), which are widely used for Cu mining. All four PIR size fractions contained comparatively equal quantities of valuable metals, including Cu, and therefore all tested fractions can be used for metal recovery. Thus, PIR can be considered as a prospective feedstock for recovering of precious metals and Cu by conventional techniques.

The total content of REEs in PIR fractions I–IV was approximately 82.51 mg/kg. The most abundant elements were Ce, Nd and La with concentrations of 26.74 mg/kg, 16.86 mg/kg and 16.65 mg/kg, respectively. The results are consistent with a previous study that was made for treated MSW incineration residues from a Danish MSW incineration plant which found that the total concentration of REEs in the treated material was approximately 110 mg/kg (Allegrini et al., 2014). The concentration of individual REEs in the PIR was lower than the content of REE that is typically found in ores. For example, the concentration of the REE oxides CeO₂, Nd₂O₃ and La₂O₃, in the apatite ores in Russia (Kola Peninsula) reached 45 g/kg, 12.5 g/kg and 7 g/kg respectively (McGill, 2000). Therefore, economically feasible recovery of REEs from PIR requires development of advanced effective techniques.

In order to determine the potential financial value of PIR, we evaluated the current stock prices of different metals and critical REEs (Table 2). According to this estimate, the value of precious metals, light and heavy REEs, and some other critical metals (including Cu) in PIR can reach 1915.92 \$ per ton of material. Of particular interest is the recovery of Au and Pt which were found at high concentrations in PIR. The disuse of other critical metal resources like REEs also has to be avoided following current guidelines and policies within the European Union [9]. For this reason novel hydrometallurgical processes (leaching, bioleaching,

Table 2

Value of precious metals, light and heavy REEs, and some other critical metals (including Cu) that could be potentially recovered from PIR (I–IV grain size fractions) according to stock prices (year 2016).

Metal	Metal price, \$ USD/g	Potential value in PIR, \$ USD/ton
<i>Precious Metals^{a,b}</i>		
Au	36.861	799.60
Ag	0.504	4.18
Pt	55.460	967.79
<i>Non Ferrous Metals^c</i>		
Ni	0.009	3.64
Cu	0.005	23.78
Al	0.002	46.24
Zn	0.002	9.30
Pb	0.002	1.27
<i>Other^c</i>		
Co	0.024	0.98
Mg	0.002	19.89
Mn	0.002	2.70
Mo	0.015	0.49
<i>Light REE^d</i>		
La	0.007	0.12
Ce	0.007	0.19
Pr	0.085	0.35
Nd	0.060	1.01
Sm	0.007	0.02
<i>Heavy REE^d</i>		
Dy	0.350	0.54
Sc	15.000	33.83
Total		1915.92

Source:

^a CME Group.

^b Johnson Matthey PMM.

^c London Metal Exchange.

^d HEFA Rare Earth (prices for the period May 17th–19th, 2016).

precipitation, cementation, liquid-liquid extraction, electrochemistry, and biosorption) and their combinations can be considered as a possible way of adaption and modernization of conventional recycling processes for MSW incineration residues. Application of these processes for recovery of metals from PIR can reduce the economic dependence from imported metals. This is an urgent task for countries not having their own mineral reserves. Moreover, recovery of potentially toxic metals from PIR can contribute to using PIR as a feedstock for construction and decrease anthropogenic contamination of the environment during landfilling.

4. Conclusions

In this study we predicted metal mobility from different size fractions of PIR by using sequential extraction analysis. Our results indicate that leaching efficiencies of hazardous metals in the fine size fractions I and II were higher than that from the coarse size fractions III and IV of PIR. In parallel, total contents of heavy metals in fractions I and II were also higher than those in fractions III and IV. Therefore, the fine fractions I and II, which when combined account for 46% of total PIR mass, pose higher risks than the more coarse fractions III and IV with regard to leaching of Zn, Pb, Cu, Ba and Ni. The largest amount of Zn was present as acid extractable species in all fractions while a predominance of leachable Pb, Cu, Cr, Ba and Ni had been found under oxidizing conditions from most of samples, with the exception of Ni for which similar amounts are present as acid extractable and oxidizable species in the finest size fraction. Thus, before disposal and reuse of PIR as construction material, decontamination processes should be applied to control the environmental risk that PIR may cause, especially for the fine particles and specifically for Zn, that is already extractable by weak

acid, and for other metals, which can easily leached by rainwater. We also demonstrated that the original PIR contains economically profitable levels of Cu, Au and Pt (5.1 g/kg, 21.69 mg/kg and 17.45 mg/kg, respectively), especially in the size fractions of <2 mm for Pt and Cu, and in the fractions of 0.5–4.0 mm for Au. These values are competitive with the valuable metal contents in other secondary resources making the process of their extraction from PIR economically suitable (Shen and Forssberg, 2003; Cui and Zhang, 2008; Boduen et al., 2014). Heavy non-ferrous metals can be concentrated by physical separation. Following magnetic density separation can separate Au from the other metals (Cu, Zn and etc.). Then concentrate can be directly smelted in Cu and Au smelters (Muchova et al., 2009). Hydrometallurgical methods can also be considered. Gold, for example, could be leached in an alkaline environment by cyanide (Gökelma et al., 2016) and Cu by ammonia (Xua et al., 2016). Thus, the present study provides important insights into potential environmental problems but also new opportunities associated with municipal solid waste incineration residues towards a secure, sustainable and economical waste management.

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Conflict of interest

None.

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