



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



Sergey Abramov^{a,b,1}, Jing He^{b,c,*,1}, Dominik Wimmer^b, Marie-Louise Lemloh^b, E. Marie Muehe^{b,d}, Benjamin Gann^e, Ellen Roehm^b, Rainer Kirchhof^f, Michael G. Babechuk^{g,h}, Ronny Schoenberg^g, Harald Thorwarth^f, Thomas Helle^e, Andreas Kappler^b

^a Microbiology Department, Faculty of Biology, Lomonosov Moscow State University, 1-12 Leninskie Gory, 119991 Moscow, Russia

^b Geomicrobiology, Center for Applied Geosciences, University of Tuebingen, Sigwartstr. 10, 72076 Tuebingen, Germany

^c School of Environmental Studies, China University of Geosciences, Lumo Road 388, Wuhan, China

^d Earth System Science, Stanford University, 397 Panama Mall Mitchell Building 101 CA 94305-2210 Stanford, USA

^e Novis GmbH, Vor dem Kreuzberg 17, 72070 Tuebingen, Germany

^f University of Applied Forest Sciences Rottenburg, Schadenweilerhof 1, 72108 Rottenburg am Neckar, Germany

^g Isotope Geochemistry, Department of Geosciences, University of Tuebingen, Wilhelmstr. 56, 72074 Tuebingen, Germany

^h Department of Earth Sciences, Memorial University of Newfoundland, Alexander Murray Building, St. John's A1B 3X5, Canada

ARTICLE INFO

Article history:

Received 8 February 2018

Revised 12 July 2018

Accepted 6 August 2018

Keywords:

Municipal solid waste

Incineration residue

Sequential extraction

Heavy metals

Precious metals

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

* Corresponding author at: School of Environmental Studies, China University of Geosciences, Lumo Road 388, Wuhan, China.

E-mail address: jhe11@foxmail.com (J. He).

¹ Co-first authors.

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 reuse 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

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