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Physiochemical characterization and heavy metals leaching potential of municipal solid waste incinerated bottom ash (MSWI-BA) when utilized in road construction

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Physicochemical Characterization and Heavy Metals Leaching Potential of Municipal Solid Waste Incinerated Bottom Ash (MSWI-BA) when Utilized in Road Construction --Manuscript Draft--

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Abstract:	In this study, the physicochemical properties, microstructure and heavy metal leaching potential of various MSWI-BA particle sizes were detected. The environmental risks that possibly result from the utilization of MSWI-BA aggregate in road construction were discussed. The air-dried MSWI-BA was sieved into four groups, including 4.75-9.5 mm, 2.36-4.75 mm, 0.075-2.36 mm and < 0.075 mm. X-ray Fluorescence (XRF), X-ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) analyses were conducted. It was found that the main elements of MSWI-BA are Ca, Si and Al; the major heavy metals are Zn, Cu, Cr and Pb; and the main mineral compositions are quartz and calcite. Overall, above characteristics were shown to be independent of MSWI-BA particle size, however, the micro-pores, attached particles and hydration products increased with the decrease of particle size. The standard leaching test and a	

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Physicochemical Characterization and Heavy Metals Leaching Potential of Municipal Solid Waste Incinerated Bottom Ash (MSWI-BA) when Utilized in Road Construction

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Abstract: In this study, the physicochemical properties, microstructure and heavy metal leaching potential of various MSWI-BA particle sizes were detected. The environmental risks that possibly result from the utilization of MSWI-BA aggregate in road construction were discussed. The air-dried MSWI-BA was sieved into four groups, including 4.75-9.5 mm, 2.36-4.75 mm, 0.075-2.36 mm and < 0.075 mm. X-ray Fluorescence (XRF), X-ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) analyses were conducted. It was found that the main elements of MSWI-BA are Ca, Si and Al; the major heavy metals are Zn, Cu, Cr and Pb; and the main mineral compositions are quartz and calcite. Overall, above characteristics were shown to be independent of MSWI-BA particle size, however, the micro-pores, attached particles and hydration products increased with the decrease of particle size. The standard leaching test and a simulated leaching experiment with four solid/liquid ratios were implemented to study the leaching behavior of Zn, Cu, Pb, and Cr. Results showed that the leaching characteristics of selected metals were affected by the species of metal, MSWI-BA particle size, solid/liquid ratio and test method. The MSWI-BA aggregate is indicated as an appropriate substitute material for natural aggregate in road construction due to the low leached metal concentrations.

Keywords: MSWI-BA (municipal solid waste incinerated-bottom ash); physicochemical properties; microstructure; leaching characteristics; heavy metals; road construction material

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1. Introduction

Over the years, municipal solid waste (MSW) management has become an increasingly important issue that is indicated as a core problem with sustainable development of most cities. Four major methods, including composting, landfills, recycling and incineration are used to deal with solid waste. Compared with the first three, incineration can effectively decrease the waste mass and volume by approximately 70% and 90%, respectively, and simultaneously convert waste into electrical energy (Zhang et al. 2010). Due to this, incineration is still recommended as an effective treatment for MSW disposal in many countries around the world (Hartmann et al. 2015). However, two main by-products including fly ash (MSWI-FA) and bottom ash (MSWI-BA) are still produced during the incinerating process. Specifically, MSWI-BA accounts for nearly 80-90% of the total mass, which means quantities of MSWI-BAs are still left over requiring further treatment even after incineration (Li et al.2012; Caprai et al.2017). At this point, it is also important to minimize the health and environmental effects by managing the ash in an environment-friendly way. Unfortunately, most of MSWI-BA around the world is commonly disposed of in landfills as it is typically the most convenient and inexpensive alternative.

In 2016, the collection and transportation of MSW in cities around China was almost 2.04 billion tons, and approximately 0.74 billion tons were processed by incineration, accounting for 36% of the total waste (China Statistical Yearbook 2017). This means that nearly 0.2 billion tons of MSWI-BA were left requiring treatment for which the space needed would be 9.21 million m³ (calculated by assuming a density of 2.17 g/cm³) and assuming all of this bottom ash would be landfilled (Shi et al. 2004). This poses a serious threat to the environment and is a public health management challenge in cities.

Extensive research has been conducted investigating the basic characteristics of MSWI-BA, and some positive results were reported. Forteza et al. (2004) and Lam et al. (2010) have confirmed that the MSWI-BA aggregate and natural aggregate have very similar properties, which make possible to utilize it as construction material. With the great demand for construction materials in recent years, various ways for utilization of MSWI-BA have been developed, such as fill material for embankment or subgrade (Lin et al. 2012) and aggregate in asphalt mixture (Huang et al. 2006; Hassan and Khalid 2010) or cement concrete (Tasneem et al. 2017; Ciarón et al. 2016). Hu et al. (2018) implemented an experimental study to investigate the property and treatment mechanism of using MSWI-BA aggregate for soil treatment. It was found that the engineering properties of treated soil enhanced with the increase of MSWI-BA proportion. Becquart et al. (2009) focused on the great potential of using MSWI-BA in road construction based on its mechanical behavior. However, it was suggested that the properties and heavy metal leaching of MSWI-BA should be carefully considered when using it as sub-base aggregate. Results reported by Forteza et al. (2004) indicated that MSWI-BA was an alternative material for road construction, but appropriate particle sizes and potential negative environmental effects should be fully considered. Thus, although MSWI-BA aggregate has been indicated as an appropriate material for road construction, the influence on engineering properties and environment are still two key considerations that need further investigation.

1 It is known that the particle size distribution is one of the key considerations for road construction
2 materials' selection. The particle size of MSWI-BA varies with production areas, time of
3 production and the MSW source. However, the MSWI-BA particle size is generally 9.5mm
4 (minus), which is a good match with most aggregate materials used in road construction. Xue et
5 al. (2009) carried out a laboratory evaluation using MSWI ash substitution for both aggregate ($>$
6 4.75mm) and filler ($\leq 0.075\text{mm}$) and reported that nearly 8-16% of MSWI ash was guaranteed to
7 meet the requirements for Stone Matrix Asphalt (SMA) mixtures. An et al. (2015) investigated
8 the effects of partially replacing fine aggregates ($< 4.75\text{mm}$) with MSWI-BA in both Hot
9 Mixture Asphalt (HMA) and Portland Cement Concrete (PCC) and found the optimum
10 proportions of using MSWI-BA in HMA and PCC were 20% and 10%, respectively. Liu et al.
11 (2014) tested the properties of asphalt mixture containing MSWI-BA aggregates of 2.36 mm
12 (minus) and 2.36-9.5 mm and recommended that the optimum substitution rate was between
13 10%-20%. According to these researchers, the appropriate particle sizes of MSWI-BA for using
14 as a material in road construction are 0-2.36, 2.36-4.75 mm, 0-4.75 mm and 0-9.5 mm.
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21 The potential environmental effects of recycling MSWI-BA in road construction is another
22 important concern, since previous studies have reported that MSWI-BA typically contains heavy
23 metals such as zinc (Zn), chromium (Cr), nickel (Ni), cadmium (Cd), lead (Pb), copper (Cu),
24 mercury (Hg) and stannum (Sn) some of which are at relatively high concentrations (Nan 2015;
25 Yang et al. 2018a). It is indicated that any variation of environmental conditions can lead to the
26 release of heavy metals from MSWI-BA into soils, and surface or ground water, then producing
27 potential effects of human toxicity and eco-toxicity (Nan 2015; Allegrini et al. 2015; Birgisdóttir
28 et al. 2006). Thus, the leaching of heavy metals from MSWI-BA should to be carefully assessed
29 before utilization.
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35 To make better utilization of MSWI-BA as a road construction material in Nanjing, China, the
36 first objective of this study was to evaluate the potential of using the ash as a substitute material
37 for natural aggregate in road construction. This was achieved by investigating the basic properties
38 of MSWI-BA for four different particle-size groups, which are the particle-size ranges most
39 commonly used in China. In this work, separate analyses were carried out for the elemental and
40 mineral compositions, and microstructure of MSWI-BA samples. The second objective of this
41 study was to assess the potential environmental negative effect of recycling MSWI-BA as road
42 construction material. This was studied in terms of the leaching behavior of selected heavy metals
43 that vary with MSWI-BA particle size, solid/liquid ratio and test method. Herein, both the
44 standard leaching test and simulated leaching experiment were conducted. The results from this
45 study also provide a foundation for further studies.
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52 **2. Materials and methodology**

53 **2.1. Materials**

54 MSWI-BA samples used in this study were collected from a waste incineration power plant in
55 Nanjing, China. To reduce the moisture content, the fresh MSWI-BAs were stored in a quenching
56 bath for 7 days before sampling and then air-dried in the lab for another 90 days before
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1 pretreatment. Both sampling and preparation were carried out in accordance with Chinese
2 standard methods (HJ/T 20-1998) 1998 and (HJ/T 298-2007) 2007.
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4 As shown in Figure 1, the air-dried MSWI-BA sample was actually a mixture of slag, ceramics,
5 glass, non-ferrous and ferrous metal and other non-combustible or unburned substances, with a
6 light gray appearance. Due to the incomplete combustion or agglomeration during the
7 incineration process, a few parts were grainy and irregular.
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10 **2.2. Methodology**

11 **2.2.1. Pre-treatment**

12 Air-dried MSWI-BA samples were pre-treated by three steps: (1) impurities including glass,
13 ceramic and magnetic particles were removed by hand; (2) a sieving test was carried out to
14 analyze the particle size distribution, by the method (T 0302-2005, JTG E42-2005) 2005; and (3)
15 all the screened samples were classified into four particle-size groups with maximum particle size
16 ranges of: 4.75-9.5 mm, 2.36-4.75 mm, 0.075-2.36 mm and <0.075 mm.
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23 **2.2.2 Chemical Composition**

24 It has been reported that the properties of MSWI-BA mostly depend on its chemical composition
25 (Le et al. 2018), so that the chemical composition analysis was firstly conducted in this study.
26 After pre-treatment, MSWI-BA samples with four different particle sizes were dried to constant
27 weight in an oven at 105°C, then reduced into powder. The powder samples were subsequently
28 passed through a sieve with a mesh size of 0.075 mm. To prevent contamination and humidity
29 from air, the powder samples were separately stored in plastic bags prior to the chemical
30 composition analysis (Kayode et al. 2018). The elemental and mineral compositions of bottom
31 ash samples with different particle sizes were respectively analyzed by using X-ray fluorescence
32 (XRF) and X-ray Diffraction (XRD) techniques (Yang et al. 2018c).
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40 **2.2.3 Microstructure Characterization**

41 To explore if there were differences in the microstructures of MSWI-BA samples with different
42 particle-size ranges, the powder samples were separately examined by scanning electron
43 microscopy (SEM) in this study. Prior to SEM, the four groups of MSWI-BA powder samples
44 were dried in an oven at 105°C for 6 hours, then gold-plated in a high vacuum environmental
45 chamber.
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50 **2.2.4. Heavy Metal Leaching Tests**

51 To assess the potential environmental risk of major heavy metals leaching toxicity from
52 MSWI-BA aggregates, the leaching tests in this study were conducted in two phases. The first
53 phase investigated the leaching characteristic of MSWI-BA samples by using the standard
54 method; and the second phase explored the effect of the moisture content in roadbed on the
55 leaching characteristic of MSWI-BA samples by using a specially designed experiment.
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1 The standard method for determining the leaching toxicity of MSWI-BA samples was Horizontal
2 Vibration Extraction Procedure (HVEP, HJ 557-2010) 2010, which actually is an extraction
3 method to assess the potential leaching over a short time (Hassan and Khalid 2010). Pre-treated
4 MSWI-BA samples were passed through a sieve with a mesh size of 3 mm. 100 g of each group
5 of screened sample was set with 1 L distilled water in a flask, then taken onto a horizontal
6 vibration machine working at a frequency of 110 + 10 times/min and an amplitude of 40 mm, for
7 8 hours. After the vibration procedure, all the mixtures were cooled at room temperature for 16
8 hours, then filtered with a 0.45 µm filter paper and stored in the refrigerator at 4°C before
9 digestion.

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14 Another environmental experiment was designed to simulate different moisture contents in a
15 roadbed onsite. Four different solid/liquid ratios of 1:10, 1:20, 1:30 and 1:40 were chosen. 100 g
16 of MSWI-BA aggregates with four particle-sizes were set with 1, 2, 3 and 4 L distilled water in
17 glass containers with lids, respectively. A contact time of 28 days was selected as there was little
18 change in most parameters between 27 and 28 days. Every 24 hours, 10 mL of leachate sample
19 was collected using a tube from each container, during the whole experiment. And 3 mL nitric
20 acid was added to keep the leachate sample stable before storing in the refrigerator at 4°C.

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25 After the heavy metal leaching tests, all the leachate solution samples were digested by
26 microwave digestion and the concentrations of chromium (Cr), copper (Cu), zinc (Zn) and lead
27 (Pb) in samples were then detected by inductively coupled plasma mass spectrometry (ICP-MS).
28 It should be noted that Cr, Cu, Zn and Pb were selected because the MSWI-BA samples showed
29 high concentrations of these four heavy metals, through the results from the previous elemental
30 composition analysis in this study.

3. Results and Discussions

3.1. Elemental Composition

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39 The elemental compositions of all MSWI-BA samples are listed in Table 1. As can be seen there
40 is a slight variability in the elemental compositions among the four groups of MSWI-BA samples.
41 Specifically, the major elements in MSWI-BA samples were calcium (Ca), silicon (Si) and
42 aluminium (Al), which accounted for over 70.48% of the total mass; followed by chlorine (Cl),
43 iron (Fe), sulphur (S), magnesium (Mg), phosphorus (P), titanium (Ti), potassium (K), sodium
44 (Na), and zinc (Zn), accounting for approximately 26.75-28.53%; copper (Cu), chromium (Cr),
45 lead (Pb), strontium (Sr) and barium (Ba) accounting for 0.33-1.30%. Also to place this in the
46 broader context of other available ashes, bottom ash produced from burning hard coal was
47 compared to MSWI-BA. The data reported by Azarsa and Gupta (2018) was used. Similar to
48 MSWI-BA, Ca, Si, and Al combined accounted for more than 70% of the total mass (>84% to be
49 precise). However, the major difference was that the coal-based bottom ash had about 60% Si and
50 about 10% CaO as opposed to approximately 19% Si and about 43% CaO (for the 4.75-9.5 mm
51 particle size range). Moreover, the MSWI-BA samples were obviously found containing heavy
52 metals such as Zn, Cu, Cr and Pb which accounted for 0.71-2.20% of the total mass. Four
53 samples of it is indicated that the basic elements in MSWI-BA are Ca, Si and Al. It is also
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1 implied that the surrounding soils, surface and underground water would be contaminated when
2 MSWI-BA aggregates are recycled in road construction without any pretreatment, because of
3 containing certain heavy metals. Aging treatment in outdoor conditions is recommended as an
4 effective pretreatment to reduce the carbonation, hydration and organic biodegradation reactions
5 in MSWI-BA, which results in decreases in the mobility of certain heavy metals, thus improving
6 its environmental performance (Lynn et al. 2017).
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10 Also, it should be noted that the 17 elements listed in Table 1 were the main elements in
11 MSWI-BA sample, accounting for over 99.7% of the total mass; but a total of 26 elements were
12 actually detected by the XRF analysis.
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15 Furthermore, a statistical analysis was conducted to investigate the relationship between major
16 elements (Ca, Si and Al) and the particle size ranges of MSWI-BA. As shown in Figure 2,
17 significant linear relations between elements (Ca, Si and Al) and different sizes of MSWI-BA
18 particle with high R^2 values in the range of 0.91-0.95. Specifically, Ca concentration in
19 MSWI-BA increased with the decrease in particle size, while Si and Al decreased. Moreover, the
20 reduction rate of Si was slightly greater than that of Al. Therefore, it is indicated that Ca, Si and
21 Al concentrations in MSWI-BA is dependent with MSWI-BA particle size ($p < 0.05$).
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26 3.2. Mineral composition

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28 XRD analysis of MSWI-BA samples with four different particle sizes supported the previous
29 elemental composition results, revealing the most similar mineral composition in these
30 four-group samples. Thus, one XRD pattern of MSWI-BA at 4.75-9.5 mm is given as a
31 representation (Figure 3). Herein, the major minerals that made up the MSWI-BA sample are
32 calcite (CaCO_3) and quartz (SiO_2). The same results were reported by Zhu et al. (2018), Yang et
33 al. (2018b) and Wongsa et al. (2017). Zhu et al. (2018) found that the main mineral compositions
34 of MSWI-BA are calcite (CaCO_3) and quartz (SiO_2), but a small amount of $\text{CaAl}_2\text{Si}_2\text{O}_8$, $3\text{Al}_2\text{O}_3 \cdot$
35 2SiO_2 and CaSO_4 which weren't seen in this research. Yang et al. (2018b) found that the main
36 mineral compositions in MSWI-BA were SiO_2 , CaCO_3 and $\text{Ca}(\text{AlO}_2)_2$, which were similar
37 comparing with those in MSWI-FA. Wongsa et al. (2017) found that MSWI-BA consisted of
38 crystalline phases of calcite (C, CaCO_3) and quartz (Q, SiO_2). These results prove that the main
39 minerals of MSWI-BA are calcite and quartz which do not change with producing areas and dates,
40 as well as the particle size. Meanwhile, it is indicated that MSWI-BA has a great potential of
41 being recycled as aggregate for use in road construction, because calcite and quartz are exactly
42 the two major mineral compositions of natural aggregate (Forteza et al. 2004; Xie et al. 2017).
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51 In addition, a statistical analysis was also conducted to investigate the relationship between the
52 two major mineral compositions and the particle size of MSWI-BA, this however showed the
53 major mineral compositions were independent with the particle size.
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56 3.3. Microstructure Characteristics

57 The microstructure characteristics of MSWI-BA with four particle sizes were conducted by SEM
58 analysis, helping develop a deeper understanding of its leaching behavior (Izquierdo et al. 2010).
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1 The SEM photographs of MSWI-BA samples, corresponding to the particle-size ranges of
2 4.75-9.5 mm, 2.36-4.75 mm, 0.075-2.36 mm and <0.075 mm, are presented in Figure 4(a), 4(b),
3 4(c) and 4(d), respectively. It should be note that all the images were obtained at the same
4 micro-size of 20 μm and the same magnification of 3000X.
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7 SEM images showed that MSWI-BA particles have irregularly shaped particles with rough
8 surface texture and a porous microstructure. Generally, a material with a presence of micro-pores
9 and flaky particles on the surface has lower strength, but the irregular surface texture should be a
10 benefit to improve the adhesion between the MSWI-BA aggregate and the bitumen/cement under
11 load, resulting in high friction angles and shear strength (Izquierdo et al. 2011). Moreover, these
12 micro-pores in MSWI-BA provide a larger surface area, which is consequently available for both
13 heavy metals leaching and adsorption process (Izquierdo et al. 2010). Thus, the skid resistance
14 properties of road pavement containing a surface layer with MSWI-BA aggregates should be
15 enhanced. In addition, granular shaped crystals that related to dynamic processes, including
16 ettringite, hydrocalumite and C-S-H phase (Bayuseno and Schmahl 2010), were observed on the
17 surface, as shown in Figure 4. Based on the SEM analysis, the numbers of micro-pores, attached
18 particles and hydration products increased with decreasing MSWI-BA particle size. Therefore, it
19 is indicated that MSWI-BA of smaller particle size contains more micro-pores, irregularly shaped
20 particles and hydration products than the larger particle size, resulting in a more stable
21 microstructure, which should benefit engineering properties.
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30 **3.4. Leaching Characteristics**

31 Leaching results of selected heavy metals including Cr, Cu, Zn and Pb from MSWI-BA samples
32 with four particle-sizes, through the HVEP test and the simulated environment experiment, are
33 presented in Figures 5-9.
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36 **3.4.1. HVEP results**

37 The results of the HVEP test were shown in Figure 5. It can be seen that the leaching
38 concentrations of selected heavy metals changed with MSWI-BA particle size. Overall, with the
39 exception of Cr, the highest concentrations of Cu, Zn and Pb were found from the group of the
40 smallest particle size (<0.075 mm). On the other hand, only the leaching concentrations of Cr and
41 Pb showed a relation with MSWI-BA particle sizes. The Cr concentration reduced with
42 MSWI-BA particle size; conversely, the Pb concentration was increased. It is indicated that the
43 MSWI-BA particle size indeed affects the leaching behavior of the selected heavy metals in the
44 HVEP test, but the degree of influence is different with various particle sizes.
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51 To evaluate the potential environmental risks of using MSWI-BA as aggregate in road
52 construction, the leaching data were firstly compared to the Identification Standards for
53 Hazardous Wastes-Part 4: Identification for Extraction Toxicity (GB 5085) in China. As clearly
54 shown in Figure 5, all the Cr, Cu, Zn and Pb concentrations remained far below the leaching
55 value limits (shown by horizontal lines) for hazardous wastes. Thus, the MSWI-BA with all the
56 four particle sizes can be classified as a non-hazardous waste. Moreover, according to Chinese
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Standard (GB/T 25032-2010), all the leaching concentrations met the requirements for recycling MSWI-BA as aggregate in the construction industry. These results show that after simple treatment, MSWI-BA can directly be used as aggregate in road construction. This is based on the short-term leaching characteristics of selected heavy metals.

3.4.2. The Simulated Experiment Results

The leaching results of selected heavy metals (Cr, Cu, Zn and Pb) from MSWI-BA samples, through the simulated environment experiment, are shown in Figures 6-9. The effects of MSWI-BA particle size, solid/liquid ratio and test method on the leaching of heavy metals are analyzed and discussed.

3.4.2.1. Influence of MSWI-BA Particle Size on the Leaching of Heavy Metals

For the four particle-sizes of MSWI-BA samples, the leaching curves for Cr, Cu, Zn and Pb were similar. Overall, the leaching concentrations of Cr and Cu increased with the contact time; but Zn and Pb concentrations decreased with the contact time. As can be seen, the leaching concentrations of all four heavy metals were the highest when the MSWI-BA particle size was smaller than 0.075 mm; but the lowest leaching concentrations of Cr and Cu, and Zn and Pb were respectively found when the MSWI-BA sizes were in the range of 2.36-4.75 mm and 4.75-9.5 mm. This result implies that the leaching of selected heavy metals is not only affected by the MSWI-BA particle size, but also the basic characteristic of the heavy metal itself.

Moreover, two types of leaching processes that got affected by MSWI-BA particle sizes can be observed. For the particle sizes of 4.75-9.5mm, 0.075-2.36mm and <0.075mm, the concentrations of Cr and Cu in leachates increased rapidly with the contact time during the initial 2-3 days, and remained relatively stable in the following days till the end of the experiment; by contrast, Zn and Pb concentrations were fluctuating at the beginning and later decreased with the contact time. Similarly, for the particle size of 2.36-4.75 mm (as shown in Figure 7), Cr and Cu concentrations gradually increased with the contact time, but the leaching rates during the initial 9-10 days were higher than those observed in Figures 6, 8 and 9. With prolonging the contact time, the Zn concentrations were high in the initial 2-3 days but kept at relatively low values in the following days; however, the Pb concentrations were fluctuating in the whole process, which was different from those observed in Figures 6, 8 and 9.

Since both the leaching concentrations and leaching processes of these heavy metals were importantly affected by the MSWI-BA particle size, especially for smaller particle size, more attention should be paid to the utilization of MSWI-BA with particle size smaller than 0.075 mm, in road construction.

3.4.2.2. Influence of Solid/Liquid Ratio on the Leaching of Heavy Metals

For the simulated environment experiment, MSWI-BA samples of the same mass (100 g) were separately soaked with four different volumes of distilled water from 1 L to 4 L, which represented four solid/liquid ratios. The leaching characteristics of MSWI-BA with four particle sizes are presented in Figures 6-9. As can be seen, except for some individual sampling points,

1 overall, the leaching processes of heavy metals did not change with the solid/liquid ratio; but the
2 leaching concentrations changed with it. Specifically, the leaching concentrations of Cr and Cu
3 decreased with the increasing solid/liquid ratio, except for some individual sampling events. The
4 Cr and Cu concentrations reached to the highest and lowest levels, when the solid/liquid ratios
5 were 1:10 and 1:40, respectively, regardless of the MSWI-BA particle size. By contrast, the
6 leaching process of Zn was not affected by varying the solid/liquid ratio. Although the
7 solid/liquid ratio increased from 1:10 to 1:40, the Zn concentration still fluctuated during the
8 whole experiment. The Pb leaching from MSWI-BA of 2.36-4.75 mm and 0.075-2.36 mm were
9 obviously affected with varying solid/liquid ratio. Unlike Cr and Cu, the highest and lowest
10 concentrations of 2.36-4.75 mm were found when the solid/liquid ratio was 1:20 and 1:30 except
11 for several special point, respectively. While the highest concentration of 0.075-2.36 mm was
12 found when the solid/liquid ratio was 1:20 except for several special point, and the lowest
13 concentration when the solid/liquid ratio was 1:30 and 1:40.

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20 Based on the results, it is concluded that the basic leaching tendency of heavy metals (Cr, Cu, Zn
21 and Pb) during the simulated environment experiment cannot be easily affected by varying the
22 solid/liquid ratio; however, the leaching concentration at each sampling event is indeed affected,
23 and the extent also depends on the metal species. On the other hand, these results recommend that
24 effective treatment should be taken before the MSWI-BA aggregate is recycled in road
25 construction, and the moisture content in roadbed onsite will not affect the leaching process but
26 the leaching concentration, so more attention should be focused on achieving a lower leaching
27 concentration of Cr, Cu, Zn and Pb from MSWI-BA.

32 33 ***3.4.2.3. Influence of Test Method on the Heavy Metals Leaching***

34 As mentioned above, two leaching tests, including the HVEP test and a designed experiment,
35 were applied to investigate the leaching behaviors of selected four heavy metals from MSWI-BA
36 with four particle sizes.

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40 Overall, the leaching concentrations of Cr, Cu, Zn and Pb were apparently different in the HVEP
41 test and the designed experiment, due to different leaching periods (or contact time) and
42 experimental conditions (e.g. solid/liquid ratios, experiment procedures).

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45 With the same contact time of 24 hours, the Cr and Cu concentrations in the leachate samples
46 collected from the HVEP test were higher than those obtained from the simulated experiment,
47 except two individual sampling events for Cr; and most of Zn and Pb leaching concentrations
48 were the exact opposite. This means that the 8 hours of horizontal vibration during the HVEP test
49 can effectively increase the leaching of Cr and Cu; however, the leaching of Zn and Pb can be
50 promoted under the static leaching conditions. This also implies that more attention should be
51 paid to the leaching of Zn and Pb when the MSWI-BA aggregates are used in road construction
52 as the typical service life of a road is in between 10-30 years in China.

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55 Under the same solid/liquid ratio of 1:10, the leaching concentrations of Cr and Cu seemed to
56 relate to both the MSWI-BA particle size and contact time. Specifically, for the particle size of

1 4.75-9.5 mm, all the Cr and Cu concentrations were lower from the simulated experiment than
2 those from the HVEP test. For the particle sizes of 2.36-4.75 mm and 0.075-2.36 mm, the
3 leaching concentrations of Cr and Cu were lower than those from the HVEP test during the first
4 half of the simulated experiment, and higher than those from the HVEP test in the second half of
5 the experiment. For the particle size of <0.075 mm, the Cr and Cu concentrations were higher
6 than those from the HVEP test. However, the results were different for Zn and Pb. The Zn
7 concentrations in leachates collected from the simulated experiment were mostly higher than
8 those from the HVEP test. And the Pb concentrations were different with MSWI-BA particle size.
9 For the group of 4.75-9.5 mm and 2.36-4.75 mm, the Pb concentrations in the simulated
10 experiment were higher than those in HVEP test; but for 0.075-2.36 mm and smaller than 0.075
11 mm, the result was exactly the opposite.
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17 The leaching results reveal that the leaching characteristics of Cr, Cu, Zn and Pb from MSWI-BA
18 depend on many factors, not only related to the species of heavy metal, but also the MSWI-BA
19 particle size, solid/liquid ratio and test method (or leaching conditions). It is indicated that the
20 leaching concentration of heavy metal will be higher if the MSWI-BA particle size is smaller, the
21 moisture content in roadbed is higher and the contact time is longer. And for the leaching test
22 method, the HVEP test is beneficial to identify whether MSWI-BA is a non-hazardous waste and
23 whether it can be used as an aggregate in road construction, while the simulated environment
24 experiment helps to understand the leaching process in the long-term.
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28 The leaching data from the simulated experiment were also compared to the relevant limit values
29 in Identification Standards for Hazardous Wastes-Part 4: Identification for Extraction Toxicity
30 (GB 5085) and the Municipal Solid Waste Incineration Bottom Ash Aggregate (GB/T
31 25032-2010) for evaluating the potential of utilizing MSWI-BA aggregate in road construction.
32 All the metals (Cr, Cu, Zn and Pb) in the leachate samples were far below the leaching limits for
33 hazardous wastes; meanwhile, all the heavy metal concentrations met the limits for MSWI-BA
34 aggregate. Thus, the MSWI-BA used in this study can be classified as a non-hazardous waste and
35 it is feasible to directly use them as aggregate in road construction, from the long-term leaching
36 characteristics of selected heavy metals.
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43 It can be concluded that the MSWI-BA aggregate collected from Nanjing, is an appropriate
44 substitute material for natural aggregate in road construction due to the following reasons, from
45 the physicochemical characteristics and environmental perspective: (1) its chemical composition
46 is much similar to the natural aggregate commonly used in road construction; (2) the MSWI-BA
47 particle containing hydration products and rough surface should improve the performance of the
48 pavement; and (3) the leaching of the major heavy metals is below the level limits for both the
49 hazardous waste and MSWI-BA aggregate after a simple pre-treatment, in both short- and
50 long-term.
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55 **3.4.2.4. Potential Influence on Surface Water and Groundwater**

56 The leaching data from both the HVEP test and simulated experiment were further compared to
57 the Standard for Groundwater Quality (GB/T 14848-2017) and Standard for Surface Water
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1 Quality (GB 3838-2002) in China (limit values are shown in Table 2), to investigate the potential
2 environmental risks that might be caused by the utilization of MSWI-BA as an aggregate in road
3 construction.
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5 Data shows that Zn and Cu concentrations in the leachates after the HVEP test respectively
6 matched the level limit for I and II Groundwater, which are suitable for various situations like
7 potable water, irrigation, process water, etc. However, the other two metals – Cr and Pb
8 concentrations matched the requirements for different groundwater categories, depending on the
9 particle size of MSWI-BA. For the leaching data from the simulated experiment, both the
10 concentrations of Zn and Pb matched the limits for II Groundwater; and the Cu and Cr
11 respectively matched the limit values for III and V Groundwater. These results indicated that
12 although MSWI-BA is an appropriate substitute material for natural aggregate in road
13 construction, more attention should be paid because the leaching of heavy metals from
14 MSWI-BA is likely to affect the groundwater quality in both the short- and long-term, especially
15 when the road crosses the environmentally sensitive areas such as source of drinking water or
16 irrigation water, a wildlife preserve or a natural protection zone.
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23 Cu, Zn and Pb concentrations matched the limits for II Surface Water; Cr concentrations from
24 MSWI-BA of 2.36-4.75 mm and <0.075 mm, and 0.075-2.36 mm and 4.75-9.5 mm respectively
25 matched the limits for II and V Surface Water. The leaching data from the simulated
26 experiment showed that, overall, Cu and Zn concentrations matched the requirements for II
27 Surface Water, Pb concentrations matched the limits for III Surface Water, and Cr
28 concentrations matched the limits for V Surface Water. Apparently, similar results were seen
29 here for both the HVEP test and the simulated experiment, which means that a similar degree of
30 negative impact on the surrounding surface water can result by the utilization of MSWI-BA as
31 aggregate in road construction in both the short- and long-term. Therefore, effective treatments or
32 measures should be taken when the MSWI-BA aggregate is used in road construction. Some
33 cases have proved that the environmental impact assessment is an appropriate preventive measure
34 for road construction with the utilization of MSWI-BA aggregates.
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42 Results also suggest that appropriate pretreatments should be taken to reduce the leaching of
43 heavy metals while using the MSWI-BA with a particle size of smaller than 0.075 in road
44 construction, because the leaching concentrations from MSWI-BA of that particle size are always
45 higher than the other particle sizes. And at the moment, the most effective and cheapest
46 pretreatment is the natural aging.
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51 **4. Conclusion**

52 (1) The chemical (elemental and mineral) composition analysis shows that the chemical
53 composition of MSWI-BA with 4.75-9.5 mm, 2.36-4.75 mm, 0.075-2.36 mm and < 0.075 mm is
54 similar. The main elements are Ca, Si and Al, the main heavy metals are Zn, Cu, Cr and Pb, and
55 the main minerals are quartz (SiO₂) and calcite (CaCO₃).
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1 (2) The MSWI-BA samples with four particle sizes have a similar microstructure containing
2 quantities of micro-pores and irregularly shaped particles on the rough surface. Both the
3 micro-pores and attached particles increased with the decrease of MSWI-BA particle size.
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5 (3) The leaching behaviors of selected heavy metals (Cr, Cu, Zn and Pb) from MSWI-BA are not
6 only influenced by the species of heavy metal, but also the MSWI-BA particle size, solid/liquid
7 ratio and test method.
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10 (4) Based on the data from the HVEP test and the simulated environment experiment, it can be
11 concluded that MSWI-BA meets both the requirements for non-hazardous waste (GB 5085) and
12 MSWI-BA aggregate (GB/T 25032-2010). Therefore, it is feasible to use MSWI-BA aggregate as
13 substitute material for natural aggregate in road construction.
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17 (5) Potential negative impact on the surrounding groundwater and surface water may be caused
18 by the use of MSWI-BA aggregate in road construction in both the short- and long-term, because
19 of the major heavy metals leaching in MSWI-BA, so that effective pretreatment and preventive
20 measures should be taken.
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24 **Conflict of Interest:** The authors declare that they have no conflict of interest.
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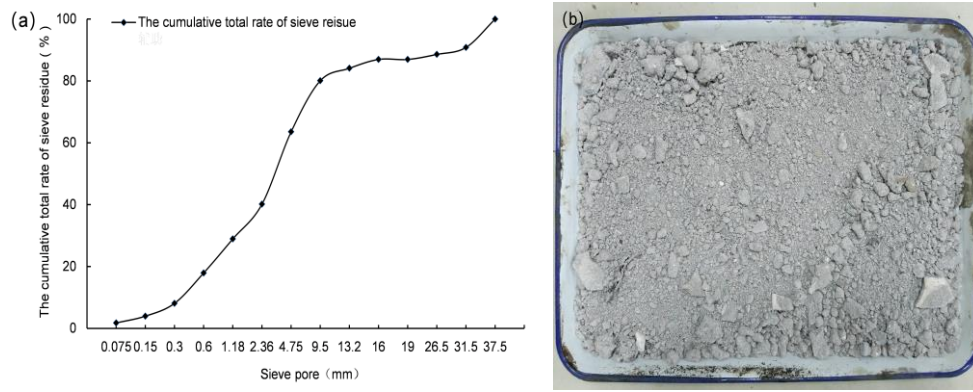


Fig 1 MSWI-BA samples after 90 days of indoor air-drying: (a) Particle size distribution of MSWI-BA samples used in this study, and (b) appearance of MSWI-BA samples

Note: Fig 1(a) was performed with Microsoft Office Excel 2010; Fig 1(b) was saved as BMP with Microsoft Office Word 2010.

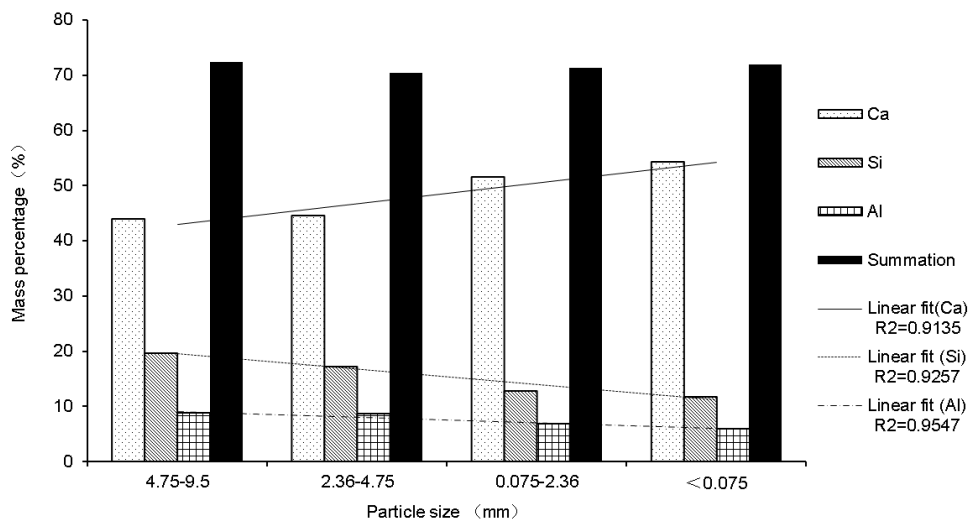


Fig 2 Relationship between the main elements and the particle sizes

Note: Fig 2 was performed with Microsoft Office Excel 2010.

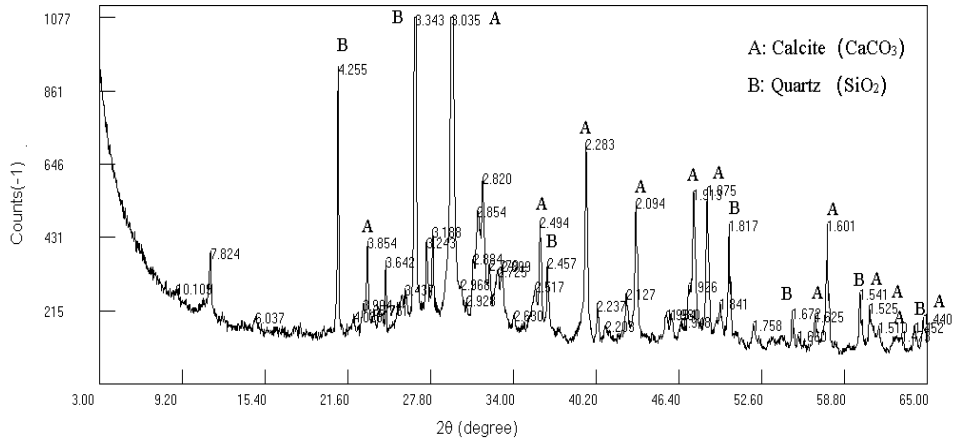


Fig 3 XRD pattern of MSWI-BA with a particle size of 4.75-9.5mm

Note: Fig 3 was saved as BMP with Microsoft Office Word 2010

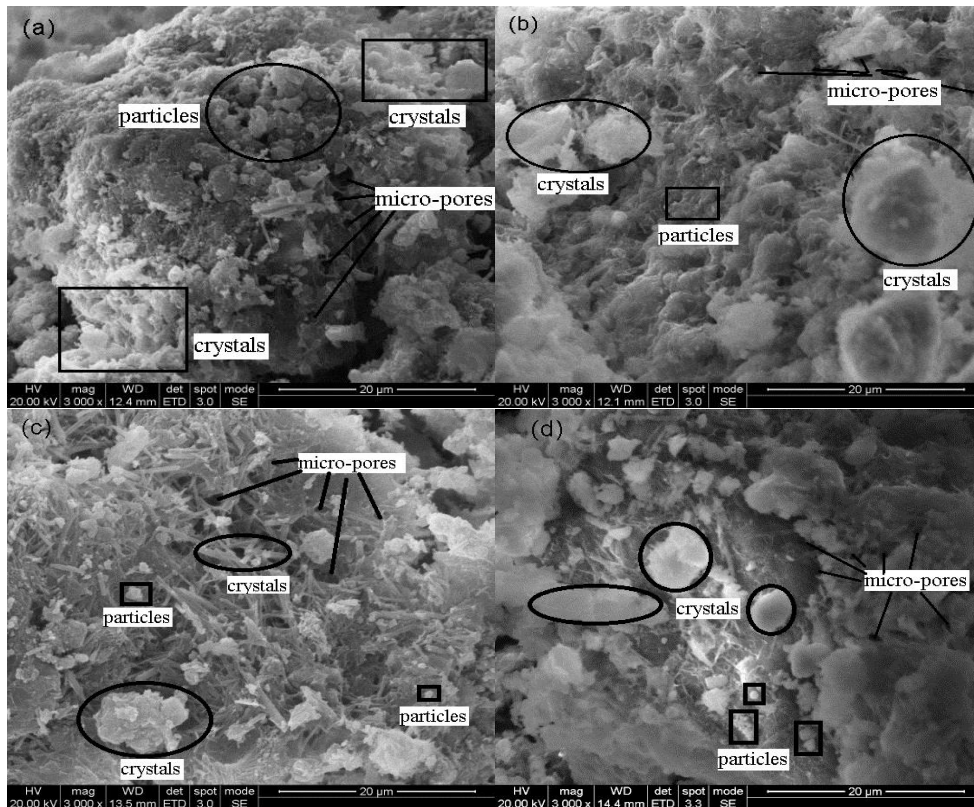


Fig 4 SEM images of MSWI-BA with four particle sizes: (a) 4.75-9.5 mm, (b) 2.36-4.75 mm, (c) 0.075-2.36 mm(c) and (d) <0.075 mm.

Note: Fig 4 was saved as BMP with Microsoft Office Word 2010.

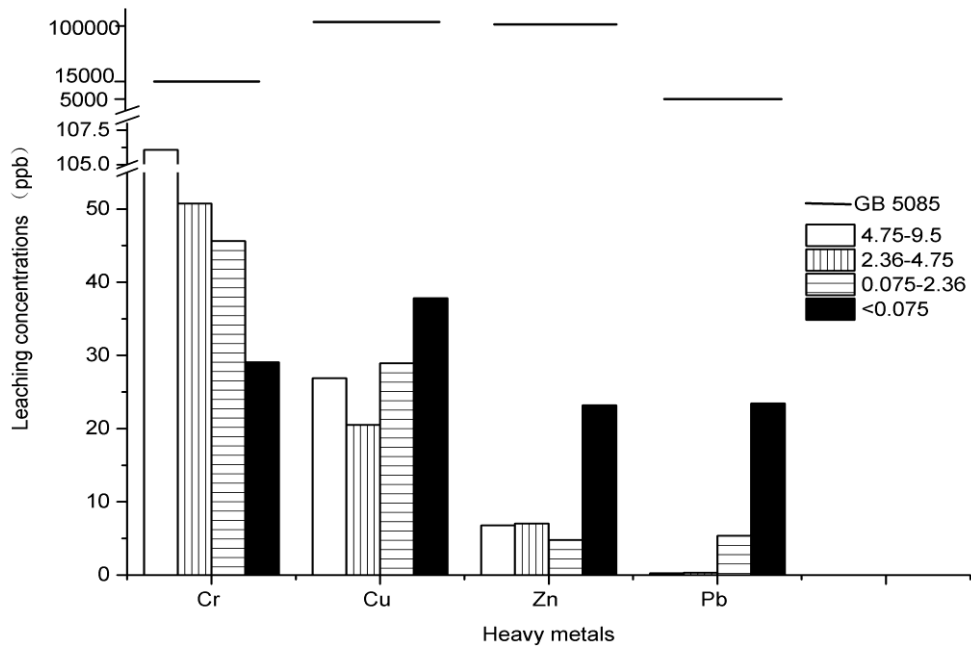


Fig 5 Results of the HVEP test and limit values in Chinese standard GB 5085

Note: Fig 5 was performed in tif format with Origin Pro 9.

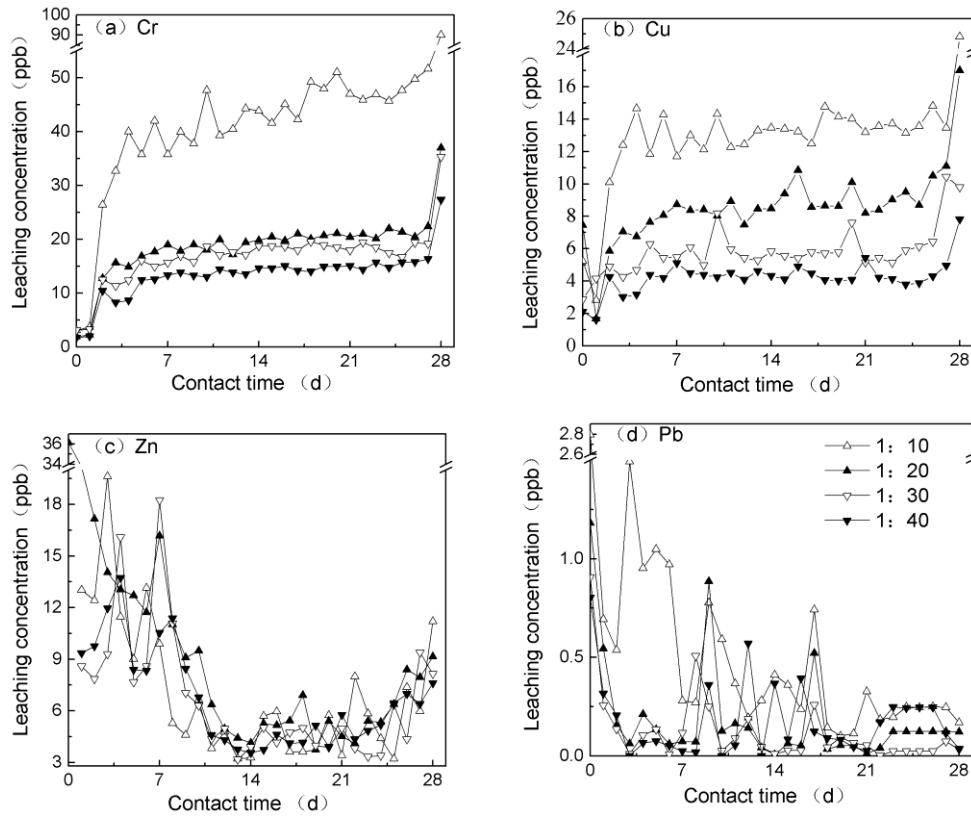


Fig 6 Leaching concentrations of heavy metals from MSWI-BA of 4.75-9.5 mm under four solid/liquid ratios, during the simulated environment experiment

Note: Fig 6 was performed in tif format with Origin Pro 9.

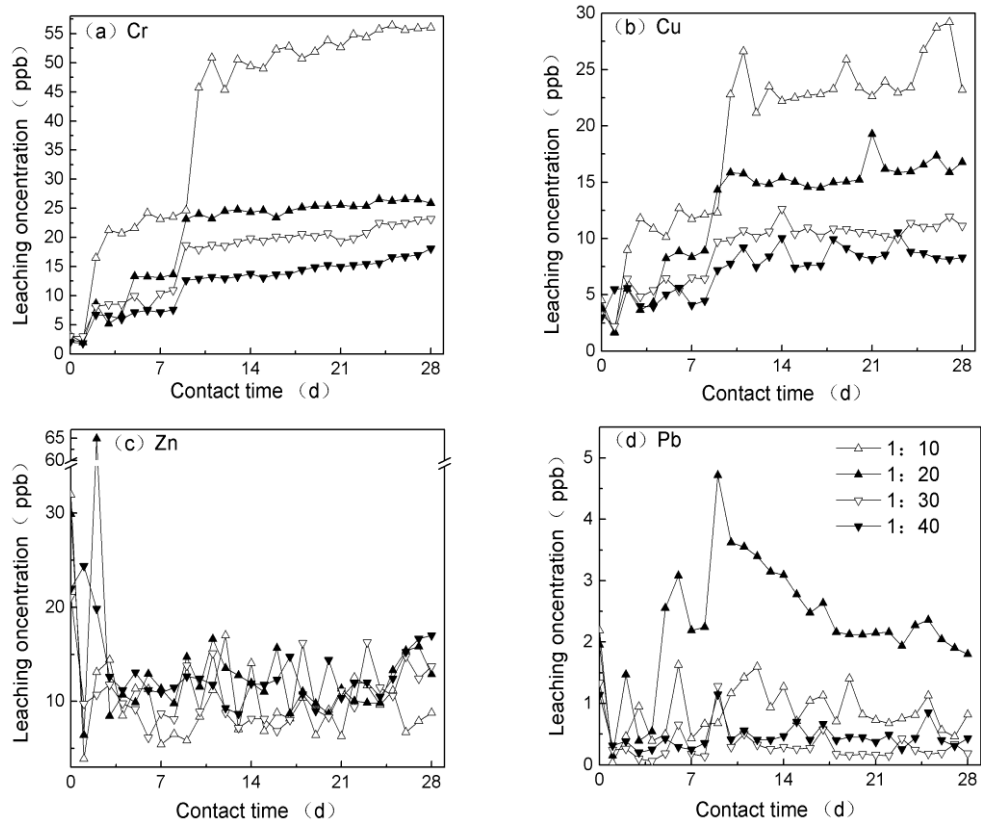


Fig 7 Leaching concentrations of heavy metals from MSWI-BA of 2.36-4.75 mm under four solid/liquid ratios, during the simulated environment experiment

Note: Fig 7 was performed in tif format with Origin Pro 9.

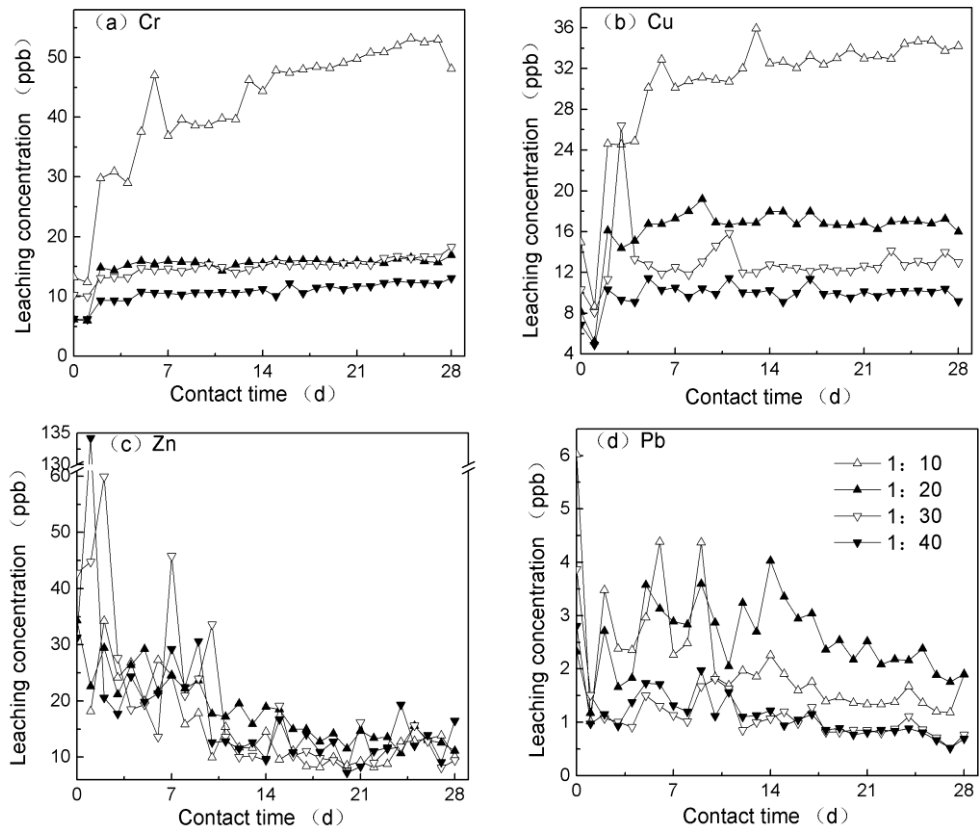


Fig 8 Leaching concentrations of heavy metals from MSWI-BA of 0.075-2.36 mm under four solid/liquid ratios, during the simulated environment experiment

Note: Fig 8 was performed in tif format with Origin Pro 9.

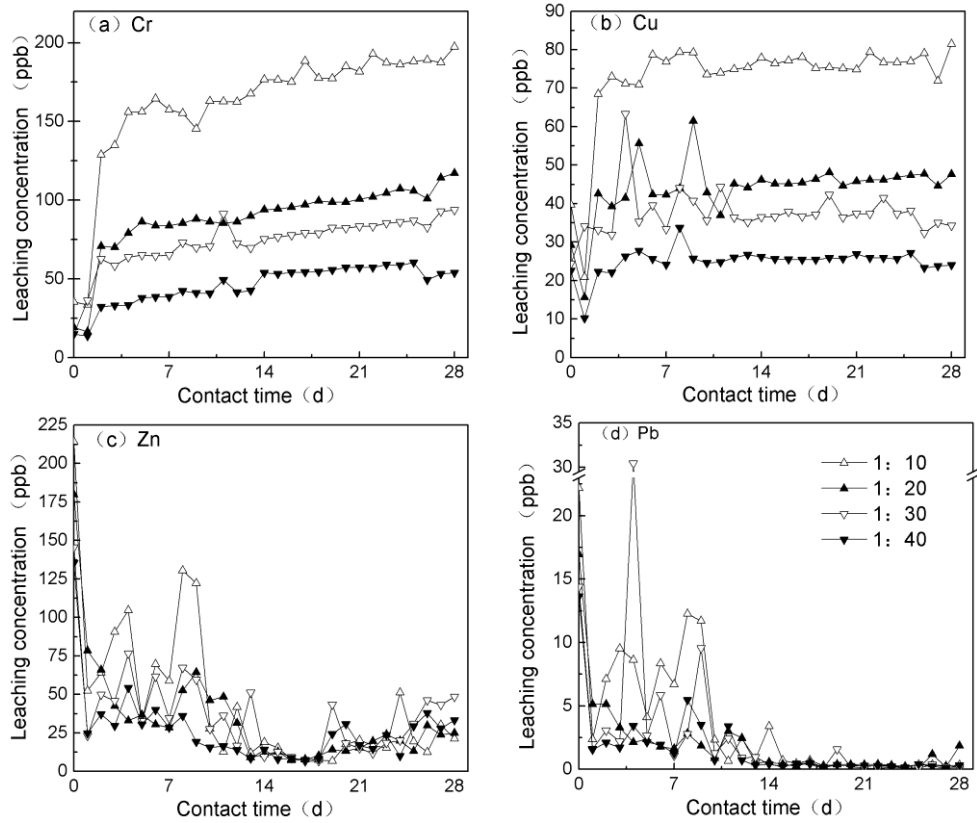


Fig 9 Leaching concentrations of heavy metals from MSWI-BA smaller than 0.075 mm under four solid/liquid ratios, during the simulated environment experiment

Note: Fig 9 was performed in tif format with Origin Pro 9.

Table 1 Elemental compositions of MSWI-BA with four particle sizes

Element	Mass Percentage (%)			
	4.75-9.5 mm	2.36-4.75 mm	2.36-4.75 mm	<0.075 mm
Ca	43.97	44.56	51.55	54.27
Si	19.60	17.27	12.85	11.75
Al	8.82	8.65	6.94	5.97
Cl	5.67	4.88	4.78	4.84
Fe	4.77	5.65	5.92	5.54
S	3.61	3.49	4.49	5.03
Mg	2.61	2.44	2.26	2.93
P	2.23	3.90	2.66	1.96
Ti	1.63	1.65	1.98	1.99
K	2.53	2.46	2.16	2.05
Na	3.22	2.14	1.59	1.79
Zn	0.48	1.92	1.32	1.28
Cu	0.10	0.12	0.22	0.14
Cr	0.10	0.13	0.17	0.13
Pb	0.03	0.03	0.19	0.06
Sr	0.04	0.08	0.12	0.07
Ba	0.44	0.41	0.60	0
Total	99.85	99.78	99.8	99.73

Table 2 Limit values in Standards for both the Groundwater and Surface in China

Heavy metal	Limit values (mg·L ⁻¹)									
	GB/T 14848-2017					GB 3838-2002				
	I ≤	II ≤	III ≤	IV ≤	V >	I ≤	II ≤	III ≤	IV ≤	V ≤
Cr	0.005	0.01	0.05	0.1	0.1	0.01	0.05	0.05	0.05	0.1
Cu	0.01	0.05	1	1.5	1.5	0.01	1	1	1	1
Zn	0.05	0.5	1	5	5	0.05	1	1	2	2
Pb	0.005	0.005	0.01	0.1	0.1	0.01	0.01	0.05	0.05	0.1