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The leaching potential of sewage sludge and municipal waste incineration ashes in terms of landfill safety and potential reuse

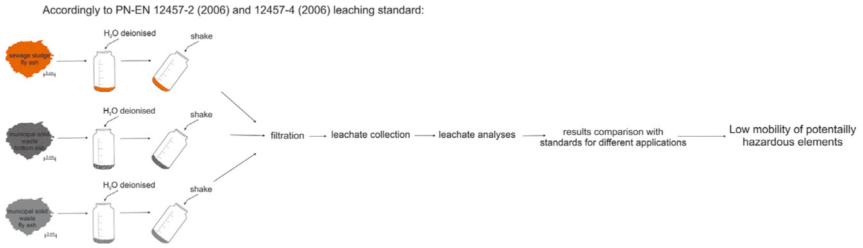
Monika Kasina [a,*], Bartłomiej Kajdas [b], Marek Michalik [a]

[a] Institute of Geological Sciences, Jagiellonian University, Gronostajowa 3a, 30-387 Kraków, Poland
[b] Centre for Environmental Education, Jagiellonian University, Gronostajowa 5, 30-387 Kraków, Poland

HIGHLIGHTS

- The European norm EN-12457 was used as an indicator of potentially hazardous elements mobility.
- Studied ashes indicate low mobility of potentially hazardous elements.
- Studied ashes fulfil requirements for safe landfilling due to low leachability.
- Mineral and chemical composition of ashes and their pH influence low mobility of elements.

GRAPHICAL ABSTRACT



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ABSTRACT

Incinerations residues from different types of materials (sewage sludge incineration ash and municipal waste incineration ashes) can either be by-products used in industry, or can pose a serious environmental problem related to their composition and the presence of potentially hazardous elements. State regulations and standards indicate whether material is inert, non-hazardous or hazardous. These standards, however, do not provide a complete overview on the leaching behavior of potentially hazardous elements in the environment. This study presents the result of batch experiment performed in accordance with the PN-EN 12457-2 (2006) and PN-EN 12457-4 (2006) standards. The results indicated that the leachability of elements is strongly dependent on the mineral composition of the waste product (the concentration and composition of soluble phase), the chemical composition (the mobility of hazardous elements and their affinity to soluble minerals), and the pH. To ensure environmental safety a thorough characterization of the waste is required followed by qualitative assignment to a particular waste type based on available guidance. Furthermore, to avoid leaching of potentially harmful elements into soils or surface water, it is also paramount to perform environmental impact assessment of wastes used as by-product in industry e.g., as building or road construction materials (aggregate) and fertilizers.

1. Introduction

The production of municipal wastes and sewage sludge increases year on year due to population growth and industrial and technological development. This leads to environmental challenges related to the treatment of wastes. The principle of the circular economy and sustainability highlights the importance of -a- recycling valuable products from the waste stream to protect natural resources, -b- reusing waste in other industrial applications, and -c- reducing the impact of waste on the environment by a reduction in landfilling and the more efficient use of water and energy, following the assumptions of Agenda 2030 (A/RES/70/1, 2015).

In 2018, 12.5 million tonnes of municipal waste was collected in Poland (a 4.3% increase compared to 2017) (GUS, 2018), and 10.5 million tonnes of sewage sludge (a 1% increase compared to 2017) (GUS, 2019), while over 50% of municipal solid waste incineration (MSWI) and 11% of sewage sludge is still landfilled (GUS, 2018; 2019). There

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[*] Corresponding author.
E-mail address: monika.kasina@uj.edu.pl (M. Kasina).

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are different waste management options e.g. reusing, recycling, composting, combustion, landfill disposal. The least favorable option is landfilling (Gehrmann et al., 2017), with its possible side effects on the environment (Simon and Müller, 2004).

Incineration of wastes and sewage sludge is an important option in waste management. It leads to the sanitization of potentially dangerous materials by the breakdown of toxic organic substances (Sabbas et al., 2003), as well as a significant reduction of the mass and volume of waste by 70% and 90%, respectively (Chimenos et al., 1999). In addition, energy can be recovered to produce electricity or heat. As a result of the thermal treatment, bottom ash (BA), fly ash (FA) and air pollution control (APC) residues are produced as incineration residues. The residues are around 30% by mass of the input materials inserted into the furnace (Kanhar et al., 2020), where BA is usually 90% of the all-solid residues produced during waste incineration, the FA and residues removed with flue gases from the furnace such as APC residues form 10% of the total (Chandler et al., 1997) in incinerated municipal solid waste, while 85% of FA and 15% of APC are produced as a result of sewage sludge incineration (Kasina et al., 2019). BA is usually considered a non-hazardous material that contains up to 10% elemental metals and can be considered as a future waste-based source (Kowalski et al., 2017) for application in many fields of industry such as construction, building materials or cement productions (e.g., Abubakar and Baharudin, 2012). FA, on the other hand, generally contains more valuable and critical elements in comparison to Earth materials, but less in comparison to the content of currently exploited ores (Kasina et al., 2019). FA is considered hazardous (Raclavská et al., 2017) due to elevated concentrations of potentially hazardous trace elements (Saqib and Bäckström, 2016). When incineration residues cannot be further processed and reused, they must be disposed of without posing a threat to the environment. To control and alleviate the possible negative effects of the disposed waste and to take appropriate countermeasures against pollution through the application of European standards PN-EN 12457-2 (2006) and PN-EN 12457-4 (2006), a one stage batch test was established. To learn about the release potential of hazardous elements and estimate whether the incineration residues can still be used in industry without posing a threat to the environment, or whether end-of-life products fulfil the requirements for landfilling following the Council Decision 2003/33/EC (2003) laboratory tests were perform accordingly to the mentioned above standards. Considerable amount of studies focus on this topic, however detailed characterization of incineration residues and their leachates is still important and challenging due to the multiple factors affecting the final composition of ashes, including:

- technology
- the characteristic of the area from which the waste originates (rural or industrial)
- the level of public awareness and the related degree of recycling of waste-based raw materials
- the level of development of a region (e.g. waste collected in EU countries will have different characteristics and composition than waste produced in India), and thus the challenges of comparing of the obtained results with those obtained in other countries.
- seasonal changes (summer/winter),

2. Materials and methods

For the batch experiments, BA and FA were collected from three waste-to-energy incineration plants:

a. A sewage sludge incineration plant located in a ca. 800,000 population city (ISSA), where the sludge dried to 36% of dry mass is incinerated in a fluidized bed boiler (supplier Pyrofluid™) operating at 850–900 °C.
b. An MSWI plant located in a ca. 2 million population city (MSWI 1), which operates at 850–1150 °C using a grate furnace (supplier Krüger W-MARK 5).
c. An MSWI plant located in a ca. 800,000 population city (MSWI 2), where a grate furnace operates at 850–1000 °C (supplier Doosan Lentjes).

2.1. Batch experiment with accordance to PN-EN 12457-2 (2006) and PN-EN 12457-4 (2006) standards

FA and BA incineration residues were examined with accordance to the PN-EN 12457-2 (2006) and PN-EN 12457-4 (2006) standards, respectively. The procedure is based on a single stage leaching at liquid (L) to solid (S) ratio of 10 to 1 for materials with particle size distribution lower than 4 mm (PN-EN 12457-2 (2006)) and lower than 10 mm (PN-EN 12457-4 (2006)). In terms of the eluent, deionized water was used in this study.

2.2. Sample characterization and preparation

For the laboratory experiments, five samples of FA from the ISSA plant, two samples of BA and three samples of FA from the MSWI 1 plant, and four samples of BA and two of FA samples from the MSWI 2 plant were studied. In Table 1, details of the sample type, affiliated to the sample waste code in accordance to EU standards (European Commision, 2000 – consolidated version), the locality and sampling periods are listed.

Prior to the batch experiment, the BA samples (1 kg of an averaged sample each) were ground in a tungsten carbide mortar and sieved through a 4 mm mesh size sieve to fulfil the requirements for a standard concerning particle size; and the FA samples (1 kg of an averaged sample each) were sieved without grinding due to the small particle size. In the next step, 100 g of each averaged sample was soaked in 105 °C to remove the excess of moisture and achieve mass stabilization.

To determine the amount of material to be used during the leaching procedure, it was necessary to determine the ratio of dry matter content (DR %) of a given sample according to the formula:

$$DR = 100 \times MD/MW$$

where MD refers to the mass of the dried sample in kg, and MW indicates the mass of the analytical sample before drying in kg.

It was also necessary to determine the humidity ratio (HR %) of a given sample, in accordance with the formula:

Table 1
Overview of the samples selected for the experiment.

No.		Sample	Type	Waste code	Sampling
Sewage sludge incineration plant, located in ca. 800,000 inhabitants city					
1.	ISSA	SZ01	FA	19 01 14	November 2015
2.		SZ02	FA	19 01 14	March 2016
3.		SZ03	FA	19 01 14	July 2016
4.		SZ04	FA	19 01 14	December 2016
5.		SZ05	FA	19 01 14	October 2017
Municipal waste incineration plant, located in a ca. 2 million inhabitants city					
6.	MSWI 1	KZ06	BA	19 01 12	May 2016
7.		KZ09	BA	19 01 12	April 2017
8.		KPL02	FA	19 01 13*	May 2016
9.		KPL03	FA	19 01 13*	April 2017
10.		KPO1	APC	19 01 07*	December 2015
Municipal incineration plant, located in ca. 800,000 inhabitants city					
11.	MSWI 2	KRZ7	BA	19 01 12	November 2016
12.		KRZ10	BA	19 01 12	June 2017
13.		KRZ11	BA	19 01 12	June 2017
14.		KRZ12	BA	19 01 12	June 2017
15.		KRP5	FA	19 01 13*	June 2017
16.		KRP6	FA	19 01 13*	June 2017

* fly ash containing hazardous substances.

$$\mathrm{HR} = 100 \times (\mathrm{MW}{-}\mathrm{MD})/\mathrm{MD}$$

For a single-stage test, the amount of previously averaged total MW mass containing 0.090 kg ± 0.005 kg of dry matter was used.

The calculated amount of each sample was placed in a polypropylene bottle, together with the appropriate amount of deionized water as a solvent. The amount of deionized water (DI) used during the leaching was calculated using the formula:

$$\mathrm{DI} = (10\text{–}\mathrm{HR}/100) \times \mathrm{MD}, \mathrm{at}\ (\mathrm{L/S}) = 10\ \mathrm{l/kg}$$

The sample and solvent were mixed together in tightly tapped 1 l polypropylene bottles and mixed in a rotary stirrer for 24 h at room temperature. After mixing, each sample was weaned for 15 min to allow sedimentation. Afterwards, the leachate was filtrated through 0.45 μm membrane filters (Whatman) using a pressure filtration kit (Glassco) and a vacuum pump with a capacity of 2.5 atm (KNF N86KT.18).

Immediately after filtration, the leachate parameters (volume, temperature, pH, conductivity and total dissolved solid) were measured using Elmetron CP-401 multimeters (Elmetron, Poland), calibrated before each measurement with standard buffering solutions (pH 4 and 7). 100 ml of each filtrated leachate was placed in 150 ml polypropylene bottles, sealed, additionally protected with parafilm and stored in a fridge until sending for analysis.

2.3. Analytical methods

The chemical composition, BA and FA was obtained using inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma atomic emission spectroscopy (ICP-AES), performed by Bureau Veritas Minerals (formerly AcmeLabs Analytical Laboratories) in Vancouver, Canada. To determine the chemical composition of leachates, ICP-MS analyses were performed by Activation Laboratories Ltd. (Actlabs) in Ancaster, Canada.

X-ray diffraction (XRD) was used for mineralogical characterization of BA and FA samples. The analyses were performed at the Institute of Geological Sciences, Jagiellonian University in Krakow, Poland using a Philips X'Pert (APD type) diffractometer with a PW 3020 vertical goniometer equipped with a curved graphite crystal monochromator (CuKα radiation, analytical range 2–64° 2Θ, step 0.02°, counting time 2 s/step). Phase identification was obtained using Philips X'Pert software (associated with the ICDD database). The interpretation of the qualitative results was based on SEIFERT AutoQuan 2.62 (BGMN) software.

3. Results and discussion

The composition of the starting material is given in Table 2 and the concentrations of elements after the leaching procedure and the percentage of leached elements in the leachate are listed in Table 3. The leachability was calculated using the following formula.

$$\mathrm{R} = \ \mathrm{c(solvedmg/L)} \times \mathrm{V\ (L)} \times \Big(\mathrm{c(totalmg/kg)}^{-1} \times \mathrm{m(samplekg)}^{-1}$$

3.1. Mineralogy of the starting materials

The ISSA sewage sludge ash samples (SZ01, SZ02, SZ03, SZ04 and SZ05) contain quartz, feldspar, hematite, whitlockite and amorphous phase as major components, whereas anhydrite, calcite, gypsum and halite were present as minor mineral phases (Kasina et al., 2019).

Notable differences in composition between the FA from MSWI 1 and FA from sewage sludge incineration were noticed. Accordingly, two samples (KPL02 and KPL03) selected for batch experiment contained apatite, anhydrite, larnite and Fe oxides as major phases, while the minor components were quartz, calcite, gehlenite, halite and sylvite.

The BA samples (KZ06 and KZ09) selected for the experiment from MSWI 1 were composed of quartz, larnite, calcite, minerals of the melilite group, feldspar, sulfates (anhydrite, basanite, gypsum), Fe oxides, wollastonite and amorphous phase that accounted for ~55% of all BA components. Whereas the APC sample (KOP01) contained halite, sylvite, apatite, anhydrite, basanite, calcite, cordierite and amorphous phase which was almost 54% of all detected components.

The main component in the BA of MSWI 2 was amorphous phase, largely silicate glass with Al, Ca, Fe, Mg, Na and K detected as minor components. The main crystalline phases were quartz, calcite, anhydrite and pyroxene (diopside), mullite and feldspar.

The FA (KRP5 and KRP6) was mostly composed of silicate glass, quartz, calcite, feldspar, gypsum, anhydrite and portlandite.

3.2. Leaching behavior of ashes

The mechanism of leaching kinetics is very complicated and strongly dependent on various physical and chemical parameters (Chandler et al., 1997; Kalbe et al., 2008) where pH values seem to be a an important factor (Dijkstra et al., 2006). The pH of the solution after leaching vary depending on the sample from over 9 in ISSA sample to 11–12 in ashes from MSWI, details are listed in Table 3. According to Izquierdo and Querol (2012) the pH of the ash–water system is controlled by the Ca/S concentrations in ashes. Ca is significantly enriched in relation to S content indicating strongly alkaline ashes.

The composition of the leachates varied depending on the starting material. Accordingly, the ISSA leachates exhibited high concentrations of alkaline elements Na and K, and Ca and Mg (Table 3). This elemental suite could originate from dissolution of sulfates, carbonates and chlorides or the amorphous material rich in Si and P, also containing Ca, K, Na, Mg, Al, Ti, S, Cl, Mn and Ba (Kasina et al., 2019). However, the mobility of elements such as Si, Al, Fe, Mn and Ti in the ISSA samples was low. We, thus infer that the presence of Na, K, Ca and Mg in the leachates is associated with the dissolution of sulphates, carbonates and chlorides. Further material characterization will be performed in the next stage of the research. This will provide unequivocal evidence of the element donors in the leachates.

In terms of the MSWI 1 samples, alkaline and alkaline earth elements such as Ca, Na and K were also quite mobile (Table 3). This is strongly related to the dissolution of soluble sulfates and silicates, as also suggested by Lo and Liao (2007). In the FA from MSWI 2, the highest leachability was also observed for Ca, Na and K; while in the BA the highest leachability was measured for Ca. Other major elements except for Ti showed low leachability (Table 3).

The overall concentration of metals and potentially hazardous elements in the ISSA samples was quite low, except for Ba and Cr (Table 3), while the extraction was quite low, and the leachability was at the level of few to dozens of percent for selected elements. Dermatas and Moon (2006) suggested that the leachability of Cr(III) is inversely proportional to lime content. Same authors also suggested that pozzolanic reaction can lead to Cr(VI) precipitation and its immobilization (Dermatas and Moon, 2006). In the case of Cr, we assumed both its presence in the less mobile form of Cr (III) or Cr (IV) immobilization as a result of high content of pozzolanic elements.

The leaching potential of most of the hazardous elements in the MSWI 1 samples was low, except for As, Mo, Cd and Ni where we measured hig concentrations in the leachate when compared to the starting samples. (Table 3).

Interestingly, high leachability of Mo, Sb and Se and Hg were measured in the ISSA samples (Table 3). The concentrations of Se and Hg in the leachate from the MSWI 1 and MSWI 2 samples were also significantly elevated.

An interesting relation between Ca and V and Ca and Sb was noted by Kalbe and Simon (2020) indicating that the solubility of Sb and V is related to the Ca concentration due to the formation of $\mathrm{Ca(Sb(OH)_6)_2}$

Table 2
The chemical characteristic of metals, potentially hazardous elements and Rare Earth Elements (REE) in the starting materials.

Sample [mgkg^{-1}]	ISSA					MSWI 1					MSWI2					
	SZ01	SZ02	SZ03	SZ04	SZ05	KPL02	KPL03	KPO1	KZ05	KZ09	KRP5	KRP6	KRZ7	KRZ10	KRZ11	KRZ12
	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	APC	Bottom ash	Bottom ash	Fly ash	Fly ash	Fly ash	Bottom ash	Bottom ash	Bottom ash
Major elements																
Si	163,600.00	165,100.00	189,400.00	181,600.00	177,600.00	96,902.50	74,673.30	83,779.80	223,879.80	266,096.60	152,849.10	149,440.00	282,254.80	288,979.60	277,724.90	261,613.40
Al	41,700.00	43,600.00	39,200.00	45,300.00	46,300.00	38,564.10	41,314.90	39,569.20	50,625.30	36,183.60	43,801.20	55,333.40	30,417.50	28,460.20	47,768.70	47,610.00
Ti	5900.00	5500.00	5100.00	5900.00	5800.00	11,560.70	11,081.50	10,782.00	7128.10	3594.00	9703.80	11,081.50	2995.00	3114.80	6169.70	4013.30
Fe	110,800.00	99,700.00	87,100.00	102,600.00	97,400.00	128,616.00	86,676.00	116,733.00	970,212.00	417,303.00	164,265.00	168,459.00	243,252.00	260,727.00	167,061.00	236,262.00
Mg	20,900.00	22,100.00	22,800.00	22,600.00	20,300.00	13,326.30	14,291.10	14,170.50	9527.40	9768.60	12,361.50	12,904.20	10,733.40	10,251.00	13,567.50	11,155.50
Mn	900.00	900.00	800.00	800.00	800.00	696.60	619.20	774.00	696.60	464.40	619.20	774.00	464.40	541.80	464.40	541.80
Ca	90,300.00	96,200.00	88,500.00	70,900.00	78,400.00	251,322.50	281,996.00	252,609.50	129,558.00	113,613.50	197,983.50	196,053.00	99,313.50	94,737.50	109,824.00	108,966.00
Na	5300.00	5500.00	5300.00	4700.00	5000.00	30,496.20	33,167.40	27,454.00	38,361.40	53,572.40	24,560.20	22,631.00	54,388.60	60,102.00	56,317.80	55,130.60
K	15,000.00	15,900.00	14,300.00	15,500.00	16,000.00	32,453.00	30,627.00	30,129.00	5893.00	7221.00	23,323.00	20,335.00	7636.00	8134.00	9379.00	7636.00
Rare Earth Elements (REE)																
Sc	6.00	6.00	5.00	6.00	6.00	63.00	51.00	3.00	3.00	2.00	3.00	3.00	2.00	2.00	2.00	2.00
Y	15.20	16.30	15.80	18.80	17.30	11.30	11.80	12.30	9.20	9.00	10.50	12.00	10.50	10.10	11.50	35.80
La	19.00	21.80	20.80	22.20	22.90	14.60	17.00	14.80	16.60	11.80	17.20	16.40	12.00	14.80	15.70	13.70
Ce	39.00	43.60	41.80	44.40	42.80	25.80	30.80	30.00	51.60	35.30	30.90	30.30	35.20	47.20	58.60	37.50
Pr	4.03	4.47	4.26	4.88	4.34	2.76	2.88	2.82	3.06	3.65	2.79	2.94	2.59	2.69	2.97	2.52
Nd	15.30	16.80	15.80	18.60	17.40	10.40	10.30	9.70	9.50	13.20	10.10	10.90	9.10	9.70	9.90	9.10
Sm	2.83	3.39	3.01	3.51	3.17	1.53	1.55	2.04	1.49	1.40	1.64	1.96	1.69	1.68	1.65	1.56
Eu	0.61	0.62	0.63	0.73	0.63	0.56	0.63	0.64	0.45	0.43	0.46	0.57	0.54	0.44	0.51	0.45
Gd	3.06	3.24	3.03	3.68	3.26	1.47	1.58	1.34	1.50	1.74	1.68	1.74	1.68	1.82	2.51	1.63
Tb	0.45	0.48	0.45	0.51	0.50	0.40	0.41	0.41	0.25	0.27	0.30	0.32	0.32	0.30	0.32	0.28
Dy	2.45	2.69	2.48	2.97	2.91	1.15	1.21	1.32	1.10	1.30	1.32	1.65	1.45	1.32	1.33	1.36
Ho	0.51	0.56	0.54	0.61	0.57	0.23	0.23	0.19	0.23	0.28	0.24	0.27	0.30	0.30	0.28	0.32
Er	1.57	1.62	1.68	1.86	1.59	0.66	0.66	0.77	0.99	1.63	0.90	0.92	1.78	3.94	1.41	1.28
Tm	0.22	0.24	0.24	0.27	0.26	0.09	0.10	0.10	0.11	0.10	0.11	0.12	0.12	0.13	0.12	0.12
Yb	1.60	1.62	1.83	1.88	1.75	0.65	0.64	0.71	0.68	0.73	0.74	0.96	0.86	0.93	0.98	0.79
Lu	0.23	0.24	0.26	0.28	0.26	0.09	0.10	0.11	0.11	0.10	0.11	0.12	0.13	0.14	0.15	0.12

Minor elements																
Ag	12.50	12.50	12.00	14.40	13.50	12.10	12.90	13.60	0.80	2.10	5.30	6.80	2.10	1.90	6.10	2.40
As	16.20	14.10	11.60	14.00	15.10	13.20	11.30	19.30	3.50	2.80	9.70	8.80	2.90	2.50	1.90	2.90
Ba	1204.00	1446.00	1255.00	1144.00	1057.00	84.00	86.57	1039.00	2088.00	1046.00	932.00	1108.00	1385.00	1224.00	1925.00	1185.00
Be	2.00	2.00	1.00	2.00	2.00	1.00	1.00	1.00	2.00	2.00	3.00	1.00	1.00	1.00	2.00	1.00
Bi	10.50	7.20	8.70	8.90	9.20	21.10	15.50	16.00	0.40	0.40	7.50	7.20	0.50	0.60	0.40	1.50
Cd	7.00	4.80	5.00	5.70	6.90	35.70	36.50	43.30	0.50	2.40	30.10	29.00	8.80	0.90	1.40	1.40
Co	27.20	32.00	41.50	28.70	27.00	20.50	39.50	24.80	63.80	40.40	20.00	27.50	58.20	20.90	41.30	33.30
Cr	1046.52	779.76	827.64	526.68	547.20	478.80	389.88	410.40	314.64	396.72	417.24	471.96	307.80	294.12	417.24	437.76
Cs	4.70	6.90	5.90	4.10	4.60	2.70	2.10	2.70	1.10	1.60	2.70	2.50	2.10	2.10	1.60	1.70
Cu	665.50	666.60	570.30	654.80	627.10	258.70	300.90	299.10	641.40	1057.70	407.10	399.10	450.60	799.10	1135.60	1101.20
Ga	8.90	7.40	8.10	7.80	8.30	6.50	4.10	8.10	8.70	9.30	8.10	9.50	4.10	5.50	9.20	7.70
Hf	6.40	6.40	7.10	8.60	7.90	4.10	3.70	3.50	6.00	5.00	5.30	6.00	4.50	5.60	10.30	14.70
Hg	0.01	0.01	0.06	0.05	0.04	0.04	0.01	0.01	0.01	0 0.01	1.43	0.06	0.01	0.31	0.01	0.27
Mo	25.50	20.60	18.30	21.50	22.30	12.10	11.60	14.40	5.60	3.60	9.80	9.90	3.70	3.30	2.70	4.90
Nb	8.50	9.40	9.90	10.70	9.50	11.60	10.80	11.40	9.90	7.50	10.20	11.70	7.10	7.00	8.60	7.50
Ni	119.50	86.80	79.30	102.40	119.20	40.50	36.90	46.40	29.70	48.90	53.20	47.90	33.70	94.20	24.30	54.20
Pb	138.00	127.40	125.90	151.60	143.20	327.90	235.60	461.60	35.00	170.30	1423.50	681.10	365.60	240.30	311.10	623.90
Rb	50.10	54.20	52.00	57.80	55.20	50.40	49.10	45.30	17.90	25.20	35.60	32.00	23.30	32.00	27.00	22.30
Sb	6.90	7.80	7.10	5.90	6.60	339.60	329.00	194.00	18.10	14.10	132.10	112.30	12.70	10.70	17.80	15.60
Se	4.10	8.50	7.70	2.40	5.40	3.80	2.50	4.20	0 0.5	0 0.5	2.60	1.90	0.50	0.50	0.50	0.50
Sn	364.00	473.00	147.00	379.00	318.00	595.00	553.00	536.00	178.00	103.00	225.00	169.00	77.00	66.00	202.00	78.00
Sr	534.70	548.10	511.00	491.70	512.50	466.30	550.70	511.20	897.20	489.40	349.00	388.40	380.50	426.30	885.00	405.30
Ta	0.70	0.70	0.70	0.70	0.60	2.70	1.70	2.30	1.50	0.80	0.90	1.10	0.70	0.70	0.80	1.00
Th	5.70	6.20	6.10	6.60	5.80	4.20	4.50	4.10	3.30	3.50	4.60	4.80	3.90	4.10	4.30	3.90
Tl	0.90	0.80	0.70	0.80	1.00	0.10	0.10	0.10	0 0.1	0 0.1	0.20	0.10	0.10	0.10	0.10	0.10
U	8.30	9.40	7.70	8.40	8.90	1.40	1.60	1.60	1.30	1.30	1.90	1.80	1.70	1.50	1.60	1.80
V	62.00	71.00	76.00	89.00	68.00	28.00	21.00	30.00	23.00	19.00	43.00	40.00	26.00	22.00	25.00	30.00
W	9.10	137.20	136.20	8.40	7.50	5.20	73.00	43.50	614.80	214.70	6.80	7.00	534.10	44.60	284.80	216.80
Zn	4472.00	3938.00	3550.00	3918.00	4025.00	6940.00	5002.00	8067.00	415.00	768.00	4489.00	4227.00	1232.00	1003.00	1285.00	1269.00
Zr	250.30	246.60	278.00	344.70	309.70	161.00	158.00	140.60	270.30	210.00	201.50	249.00	180.90	238.10	420.20	636.3

Table 3
The concentrations of elements after the leaching and the percentage of leached elements in the leachate.

Sample		ISSA					MSWI 1					MSWI 2					
		SZ01	SZ02	SZ03	SZ04	SZ05	KPL02	KPL03	KPO1	KZ05	KZ09	KRP5	KRP6	KRZ7	KRZ10	KRZ11	KRZ12
		Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	APC	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Bottom ash	Bottom ash	Bottom ash
pH in the leachate		9.47	9.30	9.20	9.41	9.36	12.27	12.56	12.08	11.17	12.09	12.42	12.62	10.56	11.29	11.38	12.15
Major elements																	
Si	mg/L after leaching	30	20	20	20	20	800	800	700	70	50	400	200	40	80	100	40
	% share	0.18	0.12	0.11	0.11	0.11	8.26	10.71	8.36	0.31	0.19	2.62	1.34	0.14	0.28	0.36	0.15
Al	mg/L after leaching	0.555	1.78	0.894	1.22	1.33	8	8	7	64	23.8	4	3.53	29.9	74.7	217	0.4
	% share	0.01	0.04	0.02	0.03	0.03	0.21	0.19	0.18	1.26	0.66	0.09	0.06	0.98	2.62	4.54	0.01
Fe	mg/L after leaching	1.00	1.00	0.90	1.00	0.80	40.00	40.00	40.00	4.00	2.00	20.00	10.00	2.00	4.00	6.00	2.00
	% share	0.01	0.01	0.01	0.01	0.01	3.11	4.61	3.43	0.04	0.05	1.22	0.59	0.08	0.15	0.36	0.08
Ti	mg/L after leaching	0.01	0.01	0.01	0.01	0.01	0.40	0.40	0.40	0.04	0.02	0.20	0.10	0.02	0.04	0.06	0.02
	% share	0.00	0.00	0.00	0.00	0.00	0.03	0.04	0.04	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.00
Mg	mg/L after leaching	109.00	66.10	54.60	108.00	82.90	8.00	8.00	7.00	0.70	0.50	4.00	2.00	0.40	0.80	1.00	0.40
	% share	5.21	3.00	2.40	4.79	4.08	0.60	0.56	0.49	0.07	0.05	0.32	0.15	0.04	0.08	0.07	0.04
Mn	mg/L after leaching	0.01	0.01	0.01	0.01	0.01	0.40	0.40	0.40	0.04	0.02	0.20	0.10	0.02	0.04	0.06	0.02
	% share	0.01	0.02	0.01	0.01	0.01	0.57	0.65	0.52	0.06	0.04	0.32	0.13	0.04	0.07	0.13	0.04
Ca	mg/L after leaching	263.00	221.00	216.00	151.00	193.00	3000.00	3000.00	8870.00	300.00	161.00	3440.00	1380.00	217.00	300.00	400.00	265.00
	% share	2.91	2.30	2.44	2.13	2.46	11.94	10.64	35.11	2.32	1.42	17.38	7.04	2.19	3.17	3.64	2.43
Na	mg/L after leaching	55.30	38.90	27.40	27.20	31.70	2170.00	2320.00	3190.00	102.00	70.80	3060.00	1280.00	123.00	125.00	140.00	130.00
	% share	10.50	7.08	5.20	5.82	6.38	71.16	69.95	116.19	2.66	1.32	124.59	56.56	2.26	2.08	2.49	2.36
K	mg/L after leaching	68.60	49.40	39.30	57.60	53.20	2550.00	2250.00	3740.00	27.80	18.50	3340.00	1240.00	34.70	41.40	48.30	39.60
	% share	4.57	3.10	2.75	3.71	3.32	78.58	73.46	124.13	4.72	2.56	143.21	60.98	4.54	5.09	5.15	5.19

Sample		ISSA					MSWI 1					MSWI 2					
		SZ01	SZ02	SZ03	SZ04	SZ05	KPL02	KPL03	KPO1	KZ05	KZ09	KRP5	KRP6	KRZ7	KRZ10	KRZ11	KRZ12
		Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	APC	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Bottom ash	Bottom ash	Bottom ash
pH in the leachate		9.47	9.30	9.20	9.41	9.36	12.27	12.56	12.08	11.17	12.09	12.42	12.62	10.56	11.29	11.38	12.15
Rare Earth Elements (REE)																	
Sc	mg/L after leaching	0.10	0.10	0.09	0.10	0.08	4.00	4.00	4.00	0.40	0.20	2.00	1.00	0.20	0.40	0.60	0.20
	% share	16.67	16.67	18.00	16.67	13.33	63.49	78.43	1333.33	133.33	100.00	666.67	333.33	100.00	200.00	300.00	100.00
Y	mg/L after leaching	0.0004	0.0004	0.0003	0.0003	0.0002	0.0100	0.0100	0.0100	0.0010	0.0007	0.0050	0.0040	0.0006	0.0010	0.0020	0.0006
	% share	0.03	0.02	0.02	0.02	0.01	0.88	0.85	0.81	0.11	0.08	0.48	0.33	0.06	0.10	0.17	0.02
La	mg/L after leaching	0.0001	0.0003	0.0001	0.0004	0.0001	0.0052	0.0040	0.0040	0.0047	0.0002	0.0020	0.0407	0.0002	0.0004	0.0006	0.0002
	% share	0.01	0.02	0.00	0.02	0.00	0.36	0.24	0.27	0.29	0.02	0.12	2.48	0.02	0.03	0.04	0.01
Ce	mg/L after leaching	0.000	0.002	0.000	0.002	0.000	0.025	0.005	0.006	0.007	0.001	0.004	0.056	0.001	0.001	0.001	0.001
	% share	0.01	0.04	0.00	0.05	0.00	0.98	0.15	0.21	0.14	0.02	0.14	1.85	0.02	0.02	0.02	0.02
Pr	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.02	0.02	0.02	0.02	0.02	1.45	1.39	1.42	0.13	0.05	0.72	0.34	0.08	0.15	0.20	0.08
Nd	mg/L after leaching	0.0003	0.0039	0.0002	0.0016	0.0002	0.0333	0.0040	0.0040	0.0057	0.0015	0.0042	0.0115	0.0002	0.0009	0.0006	0.0014
	% share	0.02	0.23	0.02	0.09	0.01	3.20	0.39	0.41	0.60	0.11	0.42	1.06	0.02	0.09	0.06	0.16
Sm	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000	0.0000	0.0000	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.04	0.03	0.03	0.03	0.03	0.00	0.00	0.00	0.27	0.14	1.22	0.51	0.12	0.24	0.36	0.13
Eu	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.16	0.16	0.14	0.14	0.13	7.14	6.35	6.25	0.89	0.47	4.35	1.75	0.37	0.91	1.18	0.44
Gd	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.03	0.03	0.03	0.03	0.02	2.72	2.53	2.99	0.27	0.11	1.19	0.57	0.12	0.22	0.24	0.12
Tb	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.22	0.21	0.20	0.20	0.16	10.00	9.76	9.76	1.60	0.74	6.67	3.13	0.63	1.33	1.88	0.71
Dy	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.04	0.04	0.04	0.03	0.03	3.48	3.31	3.03	0.36	0.15	1.52	0.61	0.14	0.30	0.45	0.15

Ho	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.20	0.18	0.17	0.16	0.14	17.39	17.39	21.05	1.74	0.71	8.33	3.70	0.67	1.33	2.14	0.63
Er	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.06	0.06	0.05	0.05	0.05	6.06	6.06	5.19	0.40	0.12	2.22	1.09	0.11	0.10	0.43	0.16
Tm	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.45	0.42	0.38	0.37	0.31	44.44	40.00	40.00	3.64	2.00	18.18	8.33	1.67	3.08	5.00	1.67
Yb	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.06	0.06	0.05	0.05	0.05	6.15	6.25	5.63	0.59	0.27	2.70	1.04	0.23	0.43	0.61	0.25
Lu	mg/L after leaching	0.0001	0.0001	0.0001	0.0003	0.0001	0.0040	0.0040	0.0040	0.0004	0.0004	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.43	0.42	0.35	1.24	0.31	44.44	40.00	36.36	3.64	3.96	18.18	8.33	1.54	2.86	4.00	1.67

Sample		ISSA					MSWI 1					MSWI 2					
		SZ01	SZ02	SZ03	SZ04	SZ05	KPL02	KPL03	KPO1	KZ05	KZ09	KRP5	KRP6	KRZ7	KRZ10	KRZ11	KRZ12
		Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	APC	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Bottom ash	Bottom ash	Bottom ash
pH in the leachate		9.47	9.30	9.20	9.41	9.36	12.27	12.56	12.08	11.17	12.09	12.42	12.62	10.56	11.29	11.38	12.15
Minor elements																	
Ag	mg/L after leaching	0.03	0.02	0.02	0.02	0.02	0.80	0.80	0.70	0.07	0.05	0.40	0.20	0.04	0.08	0.10	0.04
	% share	2.40	1.60	1.67	1.39	1.48	66.12	62.02	51.47	87.50	23.81	75.47	29.41	19.05	42.11	16.39	16.67
As	mg/L after leaching	0.01	0.01	0.01	0.02	0.01	0.10	0.10	0.10	0.01	0.01	0.05	0.04	0.01	0.01	0.02	0.01
	% share	0.34	0.43	0.44	1.33	0.68	7.58	8.85	5.18	2.86	2.50	5.15	4.55	2.07	4.00	10.53	2.07
Ba	mg/L after leaching	0.10	0.16	0.17	0.07	0.11	0.46	0.40	6.50	0.16	1.70	0.84	0.28	0.05	0.13	2.77	0.35
	% share	0.09	0.11	0.14	0.06	0.11	5.48	4.62	6.26	0.08	1.63	0.90	0.25	0.03	0.11	1.44	0.29
Be	mg/L after leaching	0.01	0.01	0.01	0.01	0.01	0.40	0.40	0.40	0.04	0.02	0.20	0.10	0.02	0.04	0.06	0.02
	% share	5.00	5.00	9.00	5.00	4.00	400.00	400.00	400.00	20.00	10.00	66.67	100.00	20.00	40.00	30.00	20.00
Bi	mg/L after leaching	0.04	0.04	0.03	0.03	0.02	1.00	1.00	1.00	0.10	0.07	0.50	0.40	0.06	0.10	0.20	0.06
	% share	3.81	5.56	3.45	3.37	2.17	47.39	64.52	62.50	250.00	175.00	66.67	55.56	120.00	166.67	500.00	40.00
Cd	mg/L after leaching	0.001	0.001	0.001	0.001	0.002	0.040	0.040	0.040	0.004	0.002	0.020	0.010	0.002	0.004	0.006	0.002
	% share	0.14	0.30	0.27	0.22	0.23	1.12	1.10	0.92	8.00	0.83	0.66	0.34	0.23	4.44	4.29	1.43
Co	mg/L after leaching	0.001	0.001	0.001	0.001	0.000	0.020	0.020	0.020	0.002	0.001	0.009	0.006	0.001	0.002	0.003	0.001
	% share	0.03	0.02	0.01	0.02	0.01	0.98	0.51	0.81	0.03	0.02	0.45	0.22	0.02	0.10	0.07	0.03
Cr	mg/L after leaching	0.07	0.06	0.05	0.05	0.04	5.02	4.40	2.00	0.20	0.10	1.00	0.94	0.10	0.20	0.30	0.10
	% share	0.07	0.08	0.06	0.09	0.07	10.48	11.29	4.87	0.64	0.25	2.40	2.00	0.32	0.68	0.72	0.23
Cs	mg/L after leaching	0.006	0.005	0.005	0.003	0.005	0.206	0.170	0.631	0.001	0.001	0.360	0.107	0.002	0.001	0.004	0.001
	% share	1.30	0.72	0.76	0.74	1.03	76.30	80.95	233.70	1.19	0.81	133.33	42.80	0.78	0.54	2.61	0.58
Cu	mg/L after leaching	0.03	0.02	0.02	0.02	0.02	0.80	0.80	0.70	0.07	0.05	0.40	0.20	0.12	0.16	0.10	0.05
	% share	0.05	0.03	0.04	0.03	0.03	3.09	2.66	2.34	0.11	0.05	0.98	0.50	0.27	0.20	0.09	0.04
Ga	mg/L after leaching	0.003	0.005	0.004	0.005	0.005	0.040	0.040	0.040	0.019	0.011	0.020	0.010	0.012	0.017	0.051	0.002
	% share	0.36	0.67	0.46	0.60	0.59	6.15	9.76	4.94	2.16	1.20	2.47	1.05	2.95	3.11	5.54	0.26
Hf	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.02	0.02	0.01	0.01	0.01	0.98	1.08	1.14	0.07	0.04	0.38	0.17	0.04	0.07	0.06	0.01
Hg[a]	mg/L after leaching	0.03	0.02	0.02	0.02	0.02	0.80	0.80	0.70	0.07	0.05	0.40	0.20	0.04	0.08	0.10	0.04
	% share	3000.00	2000.00	333.33	400.00	500.00	20,000.00	80,000.00	70,000.00	7000.00	5000.00	279.72	3333.33	4000.00	258.06	10,000.00	148.15
Mo	mg/L after leaching	0.90	0.98	0.69	0.95	0.97	0.48	0.40	0.40	0.04	0.02	0.35	0.16	0.03	0.04	0.06	0.04
	% share	35.10	47.38	37.65	44.14	43.45	39.42	34.48	27.78	7.14	5.56	35.41	15.76	8.38	12.12	22.22	7.14
Nb	mg/L after leaching	0.0007	0.0006	0.0005	0.0005	0.0004	0.0200	0.0200	0.0200	0.0020	0.0010	0.0090	0.0060	0.0010	0.0020	0.0030	0.0010
	% share	0.08	0.06	0.05	0.05	0.04	1.72	1.85	1.75	0.20	0.13	0.88	0.51	0.14	0.29	0.35	0.13
Ni	mg/L after leaching	0.04	0.04	0.03	0.03	0.02	1.00	1.00	1.00	0.10	0.07	0.50	0.40	0.06	0.10	0.20	0.06
	% share	0.33	0.46	0.38	0.29	0.17	24.69	27.10	21.55	3.37	1.43	9.40	8.35	1.78	1.06	8.23	1.11
Pb	mg/L after leaching	0.06	0.12	0.04	0.02	0.00	1.29	1.44	61.50	0.04	0.17	2.51	4.27	0.02	0.04	0.24	1.11
	% share	0.41	0.90	0.30	0.15	0.03	3.93	6.11	133.23	1.21	1.02	1.76	6.27	0.06	0.18	0.78	1.78
Rb	mg/L after leaching	0.12	0.07	0.07	0.09	0.09	4.20	3.65	7.18	0.04	0.03	5.13	1.80	0.05	0.06	0.06	0.05
	% share	2.36	1.36	1.39	1.56	1.64	83.33	74.34	158.50	2.01	1.06	144.10	56.25	2.11	1.73	2.20	2.23
Sb	mg/L after leaching	0.01	0.03	0.03	0.02	0.03	0.04	0.04	0.04	0.01	0.01	0.02	0.01	0.03	0.02	0.02	0.00
	% share	1.54	3.35	3.75	3.20	4.20	0.12	0.12	0.21	0.72	0.71	0.15	0.09	2.17	1.89	1.12	0.29
Se[a]	mg/L after leaching	0.09	0.26	0.29	0.13	0.20	0.80	0.80	0.70	0.07	0.05	0.40	0.20	0.04	0.08	0.10	0.04
	% share	21.88	30.00	38.18	54.58	36.30	210.53	320.00	166.67	140.00	100.00	153.85	105.26	80.00	160.00	200.00	80.00

(continued on next page)

Table 3 (*continued*)

Sample		ISSA					MSWI 1					MSWI 2					
		SZ01	SZ02	SZ03	SZ04	SZ05	KPL02	KPL03	KPO1	KZ05	KZ09	KRP5	KRP6	KRZ7	KRZ10	KRZ11	KRZ12
		Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	APC	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Bottom ash	Bottom ash	Bottom ash
Sn	mg/L after leaching	0.01	0.01	0.01	0.01	0.01	0.40	0.40	0.40	0.04	0.02	0.20	0.10	0.02	0.04	0.06	0.02
	% share	0.03	0.02	0.06	0.03	0.03	0.67	0.72	0.75	0.22	0.19	0.89	0.59	0.26	0.61	0.30	0.26
Sr	mg/L after leaching	0.84	0.63	0.55	0.49	0.57	4.30	6.57	6.46	0.46	1.24	11.40	4.98	0.68	0.70	2.00	2.39
	% share	1.57	1.15	1.07	1.00	1.11	9.22	11.93	12.64	0.51	2.53	32.66	12.82	1.79	1.65	2.26	5.90
Ta	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.14	0.14	0.13	0.14	0.13	1.48	2.35	1.74	0.27	0.25	2.22	0.91	0.29	0.57	0.75	0.20
Th	mg/L after leaching	0.0001	0.0001	0.0001	0.0001	0.0001	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.02	0.02	0.01	0.02	0.01	0.95	0.89	0.98	0.12	0.06	0.43	0.21	0.05	0.10	0.14	0.05
Tl	mg/L after leaching	0.0016	0.0005	0.0005	0.0009	0.0008	0.0040	0.0040	0.0397	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	1.76	0.67	0.65	1.18	0.82	40.00	40.00	397.00	4.00	2.00	10.00	10.00	2.00	4.00	6.00	2.00
U	mg/L after leaching	0.0002	0.0002	0.0001	0.0002	0.0002	0.0040	0.0040	0.0040	0.0004	0.0002	0.0020	0.0010	0.0002	0.0004	0.0006	0.0002
	% share	0.03	0.02	0.01	0.02	0.02	2.86	2.50	2.50	0.31	0.15	1.05	0.56	0.12	0.27	0.38	0.11
V	mg/L after leaching	0.18	0.32	0.25	0.46	0.36	0.40	0.40	0.40	0.04	0.02	0.20	0.10	0.02	0.04	0.06	0.02
	% share	2.97	4.54	3.24	5.21	5.31	14.29	19.05	13.33	1.74	1.05	4.65	2.50	0.77	1.82	2.40	0.67
W	mg/L after leaching	0.11	0.14	0.10	0.09	0.09	0.08	0.08	0.07	0.01	0.01	0.04	0.02	0.01	0.01	0.01	0.01
	% share	11.98	0.99	0.73	10.82	12.52	15.38	1.10	1.61	0.01	0.03	5.88	2.86	0.02	0.19	0.05	0.04
Zn	mg/L after leaching	0.07	0.13	0.06	0.21	0.04	2.73	2.75	11.40	0.45	0.21	2.38	5.53	0.10	0.83	0.50	0.14
	% share	0.02	0.03	0.02	0.05	0.01	0.39	0.55	1.41	1.07	0.28	0.53	1.31	0.08	0.83	0.39	0.11
Zr	mg/L after leaching	0.001	0.003	0.001	0.003	0.001	0.040	0.040	0.040	0.004	0.002	0.020	0.010	0.002	0.004	0.006	0.002
	% share	0.00	0.01	0.00	0.01	0.00	0.25	0.25	0.28	0.01	0.01	0.10	0.04	0.01	0.02	0.01	0.00

% share - leaching efficiency compared to the concentration in unreacted sample.

[a] Due to low Hg and Se concentrations and poor detection limits and sensitivity of the method obtained result indicate unrealistic % share of Hg and Se.

and CaV_2O_6 respectively. An increase in Sb and V concentrations was linked by these authors with a decreasing Ca concentration.

Similar trend was observed in studied samples in case of Sb (Fig. 1a), but very weak correlation was noted for the V (Fig. 1b), thus we may assume that different reactions influenced the V solubility.

Johnson (2003) classified Hg as typically anionic, thus relatively mobile. Their high solubility in deionized water could be dependent on the dissolution of mineral phases that can contain hazardous elements in the structure or adsorbed on components surfaces.

Although Hg concentrations in the leachates of all samples indicate mobilization due to water leaching, we have to keep in mind that reliable Hg measurement are very complicated using ICP methods and quite often marked with a significant error due to high volatility, poor detection limits and poor sensitivity of the method to mercury measurement. On the other hand the results, even if not completely credible, cannot be omitted due to the fact that Hg is of high toxicity and its elevated concentration can influence soil, water and plants, as well as human health (Bordean et al., 2003). Another very important element in environmental samples is Se, which in studied samples indicated mobilization due to water leaching. Se such as Hg is also a difficult element to measure using standard multi-element ICP analyses due to multiple oxidation states, possible volatilization, matrix interaction and also high ionization potential (Maher et al., 2016). Therefore the results of these elements have to be treated with caution.

The leaching of some elements was limited, more likely, due to the high pH of the leachate (~11), provided by the dissolution of alkaline components in deionized water. Leaching behavior of major alkali and alkaline earth elements such as Na, K, Ca, Sr is however not pH dependent (Jiao et al., 2016) and can be easily removed from the system by dissolution of soluble salts in water (Dijkstra et al., 2006; Kalbe and Simon, 2020). As suggested by many authors (e.g. Cheeseman et al., 2003; Johnson, 2003; Li et al., 2017; Saikia et al., 2006; Lynn et al., 2018), higher leachability occurs in solutions characterized by lower pH. It is related to the fact that some elements exhibit cationic properties and thus a cationic leaching pattern (Luo et al., 2019), where the elements' leachability decreases with increasing pH value of the leaching solution. However, most of metals which are of environmental concerns e.g. Pb, Cu, Ni, due to amphoteric character, reveal parabolic concentration curves in leachates versus pH value (Kalbe et al., 2008). The same trend was observed in the studied samples, however only one arm of parabola is present due to only high pH values (Fig. 2a,b,c). It is important to mention that visible parabola arm was obtained for leachates of various types of samples. It means that these elements may indicate higher solubility at low pH, while they are present in ionic form, or at higher pH, where metals are present as hydroxy-complexes (for detailed description see Dijkstra et al., 2006; Kalbe et al., 2008, Jiao et al., 2016). Additionally we have to keep in mind that some results on leaching efficiency might not be credible (e.g. when leaching efficiency exceeds 100% as for the Pb in APC sample) due to high heterogeneity of the sample or insufficient sample averaging.

Since the leaching tendency can vary from acidic to alkaline it is assumed that it is strongly dependent on the properties of metallic elements and their affinity to form cationic, amphoteric and/or oxyanionic bindings and thus is a very complex process (Jiao et al., 2016).

Immobilization of some elements, on the other hand, can be related to naturally occurring processes such as hydration or carbonization what leads to the precipitation of less soluble phases. In the case of the ISSA ashes, we may also consider the solubility of phosphate minerals and their affinity to bond with metals, and thus immobilize them in the insoluble form.

In the ISSA samples, the REE were present only in a small amount (Table 2), and their leachability was at a very low level (Table 3). However, the leachability of REE in the MSWI 1 samples was notable (Table 3), but the overall concentrations were very low (Table 2), and therefore there is unlikely that the REE will have a negative impact on the environment upon leaching nor will they be considered as potential secondary source of critical elements critical elements. In the MSWI 2 samples, the highest REE leachability was noted in the FA samples (Table 3); however, the overall concentrations were low (Table 2). Obviously, the REE in the BA were bound in less mobile fraction.

The results of the ISSA batch experiment were interpreted based on the current guidelines for landfilling. As shown in Fig. 3, the FA cannot be classified as hazardous as it does not exceed the limit values of elements (As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se and Zn) for this type of waste, and also due to the much lower concentration of the leachate elements selected for the evaluation in comparison to the limit values for non-hazardous wastes. However, it cannot be classified as inert waste due to Hg, Mo and Se concentrations exceeding the acceptable limit values for these elements (Fig. 3).

The current landfilling guidelines for waste classification were also applied to MSWI 1 samples. As shown in Fig. 4, the FA samples KPL02 and KPL03 and BA samples KZ06 and KZ09 were within the limit range for elements enabling safe landfilling, despite significant increase of Hg and Se in the leachate, respectively. Only sample KPO1, an APC residue, represented material where the Pb concentrations were higher than the acceptable limit value for hazardous materials (61.50 mg/L in the leachate), and therefore should not be landfilled prior to previous fixation that disables the release of any potentially hazardous elements (Fig. 4). It is important to note the presence of high Pb concentrations can be attributed to the amphoteric behavior of this metal. It is also important to mention that metals contained in APC residues (Pb and Zn) exhibit a higher solubility under alkaline conditions as the result of the presence of amphoteric substances due to the addition of lime (Alba et al., 1997).

In Fig. 5, the leachate concentrations and the limit ranges for most elements in the MSWI 2 samples allow for safe landfilling; however, the high Hg concentrations in the FA samples classifies these materials as hazardous.

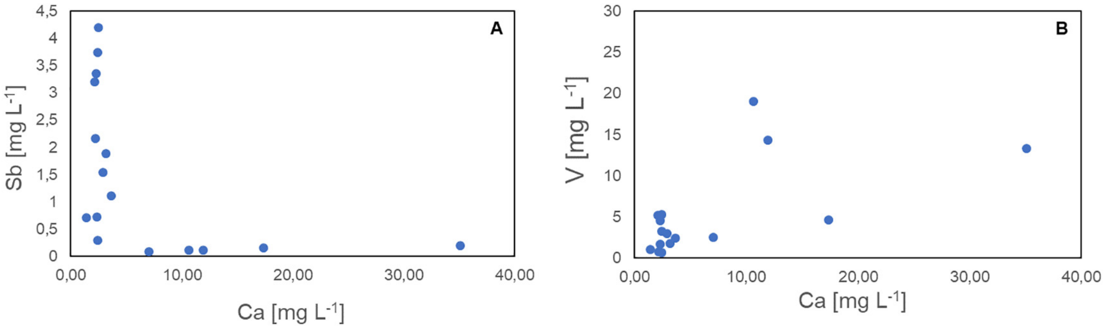

Fig. 1. A relation between Sb versus Ca (A) and V versus Ca (B).

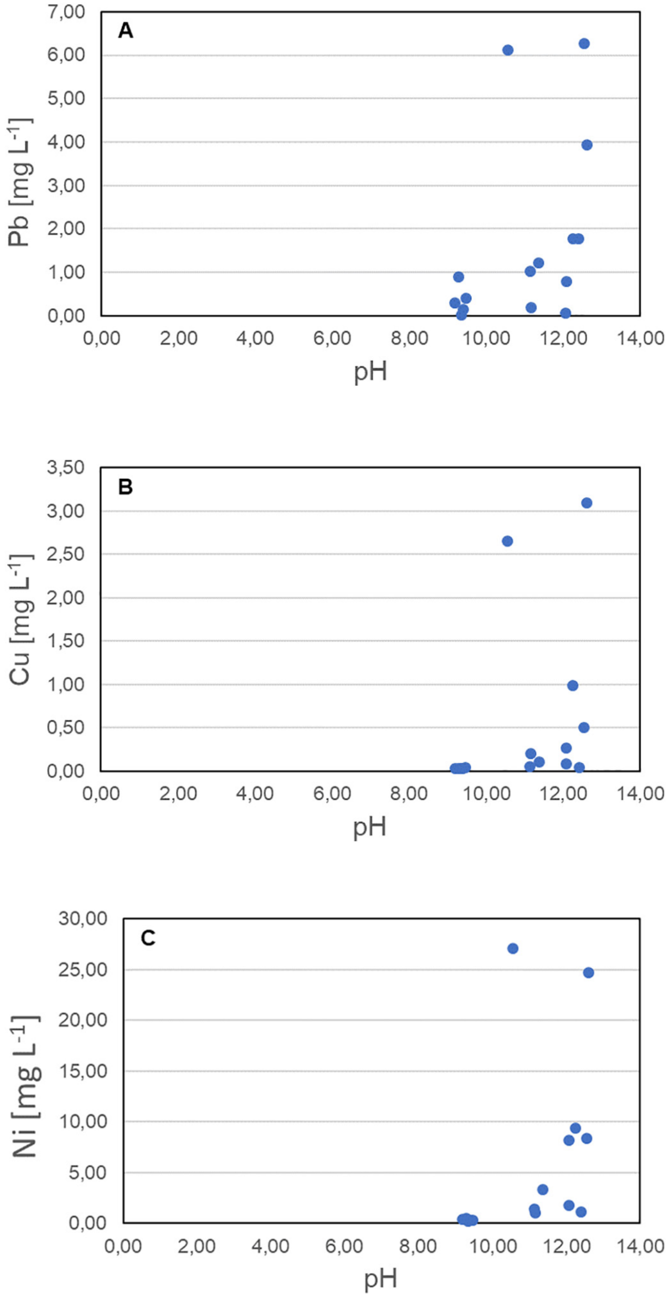

Fig. 2. Pb (A), Cu (B) and Ni (C) concentration of the leachates versus pH value in different samples.

3.3. Industrial use of ashes in accordance to the maximum acceptable limit values of potentially toxic elements in the leachate

Before their end-of-life product landfilling, BA and FA are often used in many fields of industry. MSWI ashes are widely applied in cement and concrete production (e.g. Pan et al., 2008; Keulen et al., 2016), as road construction materials (e.g. Bruder-Hubscher et al., 2001), for glass and glass ceramics (e.g. Romero et al., 2001; Silva et al., 2017) or adsorbents (Shim et al., 2003), whereas the ISSA ashes are more commonly used in agriculture (Cieślik et al., 2015).

To determine the potential use of ashes, their environmental impact must be taken into consideration. Depending on the material type and industry branch, strict requirements of element concentrations in leachate must be fulfilled before the material can serve as a by-product or additive for other industrial products.

In Figs. 3–5, the requirements for the use of ashes as road and highway substrate (accordingly to Dziennik Ustaw 796 (2015); orange dash line); for cemented ashes' use in building (Cieślik et al., 2015; purple dash line); for agriculture and reclamation works (EC Regulation 2003/2003 (2003); pink line with circles); and for ECO-ordinary Portland cement (Yang et al., 2018; grey line with triangles) are shown as examples. The results indicate that the limit values were not met for the ISSA samples as road construction material, but were met for use as an additive in cements and for agricultural purposes. The MSWI 1 samples did not meet the requirements for any usage that was taken into consideration in this study, whereas the MSWI 2 samples satisfied the requirements only for cement additives. Therefore, it can be concluded that not only the type of material (BA or FA) can show different behavior, but also the same material from different plants (e.g. BA), where the input material to the incinerator must be treated individually.

3.4. The influence of leachate on the quality of drinking water

The leachability of potentially hazardous components from waste depends on the starting material composition (mineral composition, minerals crystallinity, content of amorphous components and structure) as well as the leaching solution pH and T amongst other factors (e.g. solid to liquid ration, grain size distribution). The mobility of potentially hazardous elements can not only limit the use of waste materials as by-products, but also influence their stabilization by chemical processing, hydro-chemical solidification, carbonation, aging or using binders (Luo et al., 2019). It is also important to control the release of elements by simple interaction with water to predict, and if possible countermeasure the potential negative impact on drinking water quality. In Table 4, the main elements that affect the quality of drinking water are listed in accordance to the regulation of the Ministry for the Environment (Dziennik Ustaw Nr 204, position 1728, 2002), compared to the concentrations in the leachates from this study. Drinking water is categorized accordingly into:

A1 - Water requiring simple physical treatment, in particular filtration and disinfection.
A2 - Water requiring typical physical and chemical treatment, in particular pre-oxidation, coagulation, flocculation, decanting, filtration and disinfection (final chlorination).
A3 - Water that requires high-efficiency physical and chemical treatment, in particular pre-oxidation, coagulation, flocculation, decantation, filtration and activated carbon adsorption and disinfection (ozonation, final chlorination).

The concentration of for Fe, Cr, Se, Hg and Ba in the ISSA samples, exceeded maximum permissible limits for all water categories. Even though other elements were within the range of limit values, the leachability of this material should be controlled in order to prevent drinking water pollution. The MSWI 1 and MSWI 2 samples fulfilled the requirements only for V, and the MSWI 2 samples for Zn and As. The limit values of other elements were significantly exceeded. Therefore, even though the limit values for safe landfilling were not exceeded, each of the studied samples should undergo stabilization treatment to ensure safe landfilling of the end-of-life ash product and to fully prevent water contamination through immobilization of potentially hazardous elements.

4. Conclusions

The results obtained within the PN-EN 12457-2 (2006) and PN-EN 12457-4 (2006) standards indicate that neither the BA nor FA studied here pose threat to the environment. All samples fulfilled the requirements for safe landfilling, and they were classified as non-hazardous.

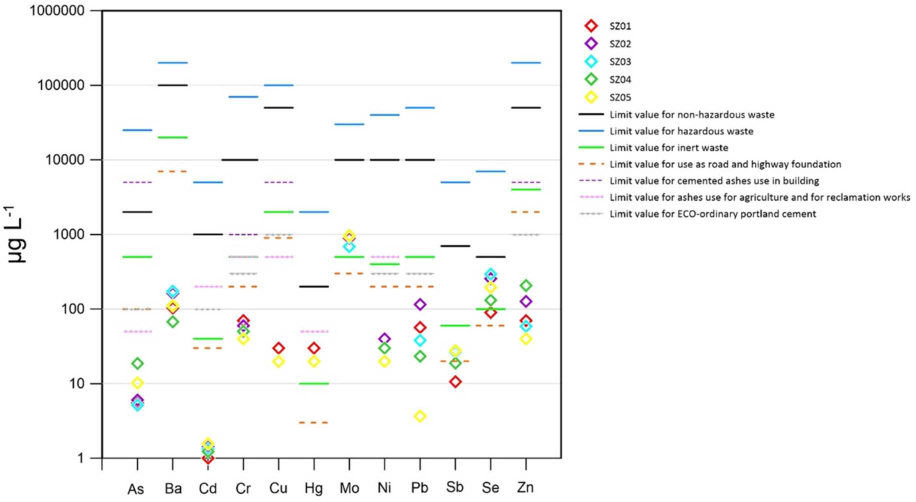

Fig. 3. The concentration of the leachate in ISSA samples (rhombs) versus the requirements for safe landfilling and usage (lines).

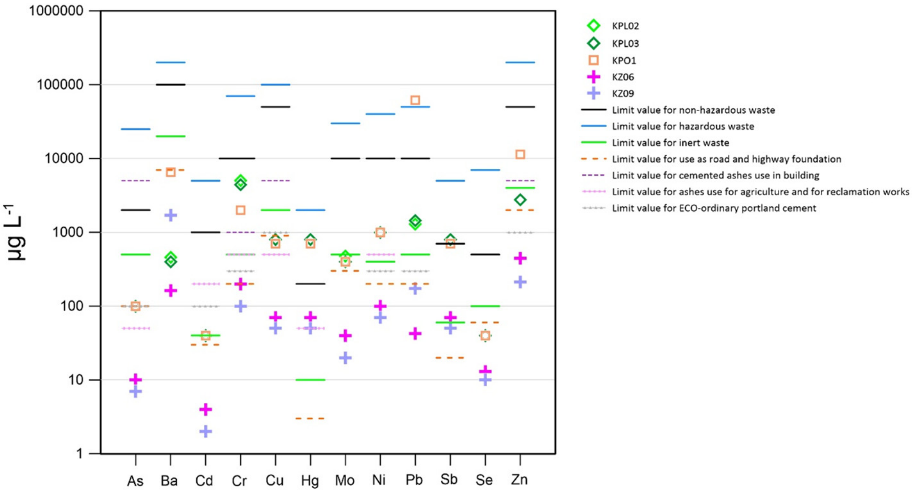

Fig. 4. The concentration of the leachate in MSWI 1 samples (crosses, squares and rhombs) versus the requirements for safe landfilling and usage (lines).

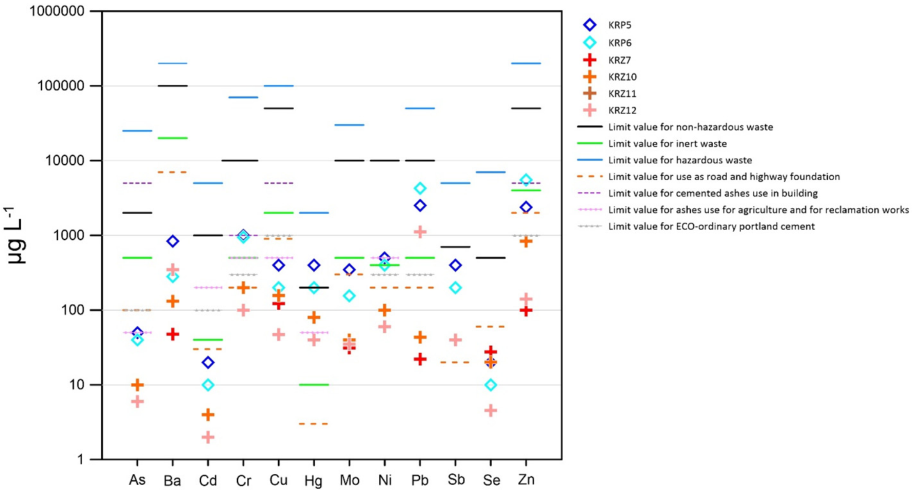

Fig. 5. The concentration of the leachate in MSWI 1 samples (rhombs and crosses) versus the requirements for safe landfilling and usage (lines).

Table 4
Elemental concentration of the leachates compared to the maximum permissible limits for drinking water according to the regulation of the Ministry for the Environment (Dziennik Ustaw Nr 204, position 1728, 2002).

Element	Water category			ISSA					MSWI 1					MSWI 2					
	A2	A3	A4	SZ01	SZ02	SZ03	SZ04	SZ05	KZ06	KZ09	KPL02	KPL03	KPO1	KRZ7	KRZ10	KRZ11	KRZ12	KRP5	KRP6
µg/L																			
As	50	50	50	5.51	6.04	5.15	18.6	10.20	<10	<7	<100	<100	<100	<6	<10	<20	<6	<50	< 40
B	1000	1000	1000	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a	n.a
Ba	10	1000	1000	103	163	173	67,2	111	162	1700	460	<400	6500	47,5	131	2770	346	837	282
Cd	5	5	5	<1	1,42	1,33	1,23	1,56	<4	<2	<40	<40	<40	<2	<4	<6	<2	<20	<10
Cr	50	50	50	<70	<60	<50	<50	<40	<200	<100	5020	4400	<2000	<100	<200	<300	<100	1000	944
Cu	50	50	500	<30	<20	<20	<20	<20	<70	<50	<800	<800	<700	122	157	<100	47,2	<400	<200
Fe	300	2000	2000	<1000	<1000	<900	<1000	<800	<4000	<2000	<40,000	<40,000	<40,000	<2000	<4000	<6000	<2000	<20,000	<10,000
Hg	1	1	1	<30	<20	<20	<20	<20	<70	<50	<800	<800	<700	<40	<80	<100	<40	<400	<200
Mn	50	100	1000	<10	12,8	<9	<10	9,2	<40	<20	<400	<400	<400	<20	<40	<60	<20	<200	<100
Ni	50	50	200	<40	< 40	<30	<30	<20	<100	<70	<1000	<1000	<1000	<60	<100	<200	<60	<500	<400
Pb	50	50	50	56,7	115	38	23,3	3,66	42,5	173	1290	1440	61,500	22,1	43,2	243	1110	2510	4270
Se	10	10	10	89,7	255	294	131	196	<70	<50	<800	<800	<700	<40	<80	<100	<40	<400	<200
V	1000	1000	1000	184	322	246	464	361	<40	<20	<400	<400	<400	<20	< 40	<60	<20	<200	<100
Zn	3000	5000	5000	<70	127	58,7	206	<40	445	213	2730	2750	11,400	<100	833	502	140	2380	5530

n.a – Not analyzed.

Batch experiment in accordance with the PN-EN 12457-2 (2006) and PN-EN 12457-4 (2006) norms indicate that the leachability of elements is strongly dependent on:

- The mineral composition of the waste product, including the content and composition of soluble phases. The mineral composition of BA and FA enables the trapping of potentially hazardous elements within the mineral structure or adsorbed on the surface, therefore decreasing their mobility.
- The chemical composition (mobility of hazardous elements and their affinity to soluble minerals).
- The pH value that influence the leaching efficiency.

Even though the leachability of most elements is low, and their concentrations in the leachates enable the classification of waste as inert, they may drastically influence the surface water use for consumption. The concentrations of almost all elements in all studied samples exceeded the government regulations, and therefore before usage or end-of-life landfilling the waste must be stabilized. Otherwise, metals may be released not only directly from the landfills, building and road construction materials, or fertilizers, leading to the pollution of soils and surface water used for consumption.

CRediT authorship contribution statement

Monika Kasina: Conceptualization, Methodology, Data curation, Formal analysis, Investigation, Validation, Visualization, Writing – original draft, Writing – review & editing. **Bartłomiej Kajdas:** Methodology, Data curation, Formal analysis, Investigation, Writing – review & editing. **Marek Michalik:** Methodology, Data curation, Formal analysis, Investigation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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