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Statistical comparison of leaching behavior of incineration bottom ash using seawater and deionized water: significant findings based on several leaching methods

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Abstract

Bottom ashes generated from municipal solid waste incineration have gained increasing popularity as alternative construction materials, however, they contain elevated heavy metals posing a challenge for its free usage. Different leaching methods are developed to quantify leaching potential of incineration bottom ashes meanwhile guide its environmentally friendly application. Yet, there are diverse IBA applications while the *in situ* environment is always complicated, challenging its legislation. In this study, leaching tests were conveyed using batch and column leaching methods with seawater as opposed to deionized water, to unveil the metal leaching potential of IBA subjected to salty environment, which is commonly encountered when using IBA in land reclamation yet not well understood. Statistical analysis for different leaching methods suggested disparate performance between seawater and deionized water primarily ascribed to ionic strength. Impacts of leachant are metal-specific dependent on leaching methods and have a function of intrinsic characteristics of incineration bottom ashes. Leaching performances were further compared on additional perspectives, e.g. leaching approach and liquid to solid ratio, indicating sophisticated leaching potentials dominated by combined geochemistry. It is necessary to develop application-oriented leaching methods with corresponding leaching criteria to preclude discriminations between different applications, e.g., terrestrial applications vs. land reclamation.

Keywords: Seawater; Leaching methods; Incineration bottom ashes; Metal leaching; Utilization

1. Introduction

Incineration may achieve an overall 70-90% of volume reduction of municipal solid waste (MSW), during which a small volume of residue (~20-25% by mass) namely incineration bottom ashes (IBA) are generated [1]. Recycling of IBA is widely practiced with increasingly adopted waste to energy (WTE) facilities in the globe, while management practices for it vary in different jurisdictions, and there is still need for legislation on recycling of IBA [2].

One challenge in the development of IBA legislation is that, IBA may vary a lot in their quality ascribed to different applied technologies for incineration and the variation of feedstock for burning [3], while reserve most of the metals (>80%) at higher concentrations from the resource [4-6]. When the total content of elements is taken as a basis, IBA should be considered as hazardous waste from the perspective of eco-toxic hazard properties. Note that, however, substances should be in solution first in order to exert a potential eco-toxic effect, alternative assessments of wastes may consider the leached concentrations instead [7]. European Council Decision 2003/33/EC lists the waste acceptance criteria based on element-leached concentrations for the different categories of waste: inert wastes, nonhazardous wastes, hazardous wastes, pursuant to the Directive of 1999, as showed in Table 1. As such, IBA may not be considered as non-hazardous waste based on leaching data (EU) at pH = 7-12 [7]. National legislation has been implemented to regulate utilization of IBA in the Netherlands and in France, however, in the Netherlands, IBA is placed in a special category, because it does not always meet the regulatory requirement [4].

Table 1

Another challenge in the development of IBA legislation is that, IBA utilization has to depend on its impact (mainly ascribed to element leaching) to the environment. There are varieties of applications being currently performed, such as substitutes of non-constructive aggregates, sub-base materials for road pavements, embankment fillings, concrete production or coast erosion protection, with involvement of distinct environmental boundaries [1, 8-11]. Accordingly, many lab-based leaching methods have been developed to simulate the IBA leaching potential when it is applied in the field. In general, these leaching methods may be classified into several categories, such as the batch leaching tests under the natural pH, the pH-static leaching tests, and the column leaching tests, etc. For instance, column leaching tests are considered as simulating the flow of percolating groundwater through a porous bed of the targeting material [12]. On the contrary, batch leaching tests are static leaching method which generates chemical data at equilibrium for mechanistic applications. As compared to batch leaching tests under natural pHs, the pH-static leaching tests intend to establish the complicated leaching profile as a function of pH values, useful to quantify the IBA leachability [13, 14].

Although pH is the most relevant factor on the assessment of the differences between leaching methods due to its strong control on the pollutant release [15], there are many other factors which may significantly affect the leaching results, such as particle size distribution, liquid to solid (L/S) ratio, leaching reagents, apparatus and scale, mixing time, leaching procedures, etc. [4, 16-20]. So far, many of these factors have been specified among various available methods designed for leaching analysis. However, it yields various leached data which is hard to cross-compare among one another and quantify the exact hazard associated with IBA [21]. In this regard, regulations imposed on IBA utilization must be scenario specific to map each other

between the laboratory leaching results and the specific application. Furthermore, leaching method development for IBA classification in the laboratory is still at their preliminary stage of development, whereas factors that are considered in their leaching procedures are rather limited. For instance, in the case of IBA application in land reclamation, bottom ashes are in fact exposed to seawater environment. Leaching methods using deionized water (DI) as the basic leachant may not be feasible any longer. According to Schmukat et al. [22], ionic strength (in terms of NaCl) and salinity significantly influenced the release metal(loid)s in the water phase for copper slag. Cetin et al. [23] also mentioned that a raised ionic strength may decrease the surface negativity of the fly ash and released Ba^{2+} , Cu^{2+} and Zn^{2+} ions from the solid surface into the aqueous solution by electrostatic interaction. It is hereby of great interest for investigation of seawater-leaching scenario, as IBA for land reclamation may be considered a viable strategy to “close the waste loop” in land-scarce countries/regions, such as Singapore and Japan.

To statistically understand the metal leaching potential caused from seawater, a series of experiments were carried out with several leaching methods in the paper. IBA was collected from 5 source locations across Singapore, subjected to both batch leaching tests (*EN 12457-1:2002* at L/S 2 and *EN 12457-2:2002* at L/S 10) and the column leaching tests (*DD CEN/TS 14405:2004*). Two specific leachants i.e. DI and seawater, were intentionally applied to all three leaching methods, simulating the IBA utilization as supplementary construction materials under two scenarios – terrestrial and ocean applications, respectively, for deterministic assessment of the leaching behaviors and the associated impacts on the leaching standards.

2. Methodology

2.1. Sampling and IBA preparation

The IBA used for this study was collected from multiple sources, including four incineration plants (IP1-4) under operation in Singapore and a metal recovery plant (MR) as well, which is processing IBA from three out the four plants in Singapore. All IPs are WTE plants using MSW as the primary fuel equipped with moving-grate stoker furnaces. The IBA sampling scheme was developed based on *BS EN14899:2005* Characterisation of waste – Sampling of waste materials – Framework for preparation of a sampling plan. Detailed sampling schedule could be found in Table 2 below.

Table 2

Sampling scheme included, (a) 22 daily samples in the first month and 45 weekly samples from IP1 afterward until a year, (b) 22 daily samples in first month and 44 weekly samples from IP2 afterward until a year, (c) 4 weekly samples from the other two incineration plants (IP3 and IP4), and (d) 4 monthly samples followed by 6 weekly samples from MR. IBA samples from all IPs were directly collected from the incineration plants at conveyors after ash extracting followed by quenching process. The other samples were processed IBAs from MR, which were subjected to overbelt magnet separation and eddy current process for respective ferrous metal and non-ferrous metal removal before collection from discharge hoppers. The IBA from all four IPs and MR contained grate siftings, boiler ash, economizer ash, and grate ash combined at the quenching tank. Approximately 50 kg of IBA was collected for each sample from all resources. The IBA was immediately dried at 40 °C for 72 hr upon collection, sieved based on five size fractions (refer to section 2.2), and then separately stored in box containers with caps. The weight of the oversized fraction, i.e. >50 mm, was not used in the study. Except for IBA samples from MR, the

ferrous metals for IP1-4 were removed using a customized magnetic separator and non-ferrous metals by hand in laboratory, respectively. Before leaching tests were performed, all IBA samples were crushed with a jaw crusher to below 4 mm (BB 100, Retsch). Combination of IBA samples (made of separate samples, which was achieved by mixing equal fraction of each sample crushed by weight) was used for this test.

2.2. IBA characterization

Moisture content (%) of IBAs was obtained by weight difference before and after oven drying at 105 °C for 24 h. Particle size distribution was determined with sieving of five size divisions, 0-2 mm, 2-4 mm, 4-20 mm, 20-50 mm and >50 mm. Total carbon (TC) and total organic carbon (TOC) were measured with a TOC analyzer (multi N/C® 2100/2100S, Analytikjena). Rotor mill (ZM 200, Retsch) was used to reduce the IBA sample size below 150 µm before total element measurement. Total element content of each sample was determined by mixing digestion using HCl/HNO₃/HF (US EPA Method 3052). The digested solution was first filtered through 0.45 µm filter nylon membrane followed by dilutions before analysis for cations using Inductively Coupled Plasma-Mass Spectrometer (ICP-MS) (Nexlon300D, Perkin Elmer).

2.3. Batch and column leaching tests

Batch and column leaching tests of IBA were scheduled as shown in Table 2. Batch leaching test was carried out with liquid to solid (L/S) ratio 2 for 6 samples MRW1-6 (*EN12457-1*), and with L/S ratio 10 for 10 samples IP1C1, IP1C2, IP2C1, IP2C2, IP3C, IP4C, and MRM1-4 (*EN 12457-2*), while using seawater and deionized (DI) water as leachants, respectively. In brief, the material was mixed with seawater or DI in a 1-L capped bottle at an L/S ratio of 2 L kg⁻¹ or 10 L

kg⁻¹ on an end-over-end tumbler for 24 h. The eluate was then vacuum-filtered with the 0.45 µm filter paper subjected to analysis using ICP-MS (Nexlon 300D, Perkin Elmer) and IC analyzer (IC882 Compact IC Plus, Metrohm) for respective trace elements and salt contents. This test was conducted triplicates. Column leaching tests (*DD CEN/TS 14405:2005*) were conducted simulating the percolation scenario for 8 samples, IP1C1, IP1C2, IP2C1, IP2C2, IP3C, IP4C, MRMC, and MRW1. This test is designed to provide the release of substances as a function of L/S under specified conditions intended to ensure local equilibrium between the substances in the solid and aqueous phases and at the natural pH imposed by the waste itself. In this test, approximately 700-800 g of IBA was packed in a column (size × I.D. = 30 cm × 5 cm) equipped with a layer of glass beads of approximately 1 cm at both ends (to avoid dead volume) in a standardized manner. The leachant (seawater) was percolated in up-flow through the column at a specified flow rate (12 mL min⁻¹) up to a fixed L/S ratio (10). The eluate was collected in several separate fractions. Seawater was used in this test. The execution time of the test was approximately 30 d for L/S = 10 l kg⁻¹.

3. Results and discussion

3.1. IBA characterization

Table S1 shows characterization of different IBA samples which have been used for the study. Based on laboratory separation processes with IBA samples from IP1-4, ferrous metals accounted for 7.18-13.90% by weight and non-ferrous metals accounted for 0.85-1.84% by weight, respectively. These values fall into the typical range of literatures [1], however, IBA from IP4 presented both lower ferrous and non-ferrous metal fractions as compared to the others likely because less metal sources from its feedstock for burning. The IBA size distributions were

well matched with those of natural aggregates (sands: 25.52-64%; gravels: 36-65%; and coarse gravels: 0-22.54% for all IBA samples, respectively) [24]. As compared to samples from the four IPs, IBA collected from MR possessed much less percentage (0-12%) on larger size fractions (i.e. 20-50 mm and above), likely ascribed to more accumulation of ferrous and non-ferrous metals within the big size particles and thus being more recovered. Bar sizer and dry recovery in MR may be also contributed to size breakdown for certain IBA particles. Burnout organics level in terms of loss of ignition (LOI) was determined at range of 1.07-2.32% in four IPs, agreeable with values presented by best available techniques [25], due to application of advanced combustion control systems, which would be able to regulate feeding and combustion rate to achieve a complete burn out of the refuse (NEA, 2017). Moisture content was measured at 16.23-25.52% for IBA from IP1-4, determined by operational processes [26, 27], which in overall was higher than those found in MR due to dry recovery process with the latter ones. Among the 21 trace metals under investigation (Table S1), concentrations of Ba, Cu, and Zn were measured as high as >1000 mg/kg, whereas Tl and Se as low as <1.0 mg/kg, largely dependent of the original source of burning. The heterogeneity of metal distributions among IBA likely induce complicated geochemical environment, which would affect their respective leaching potential.

Leaching performance of IBA depends on its chemical properties and the leaching method applied. Chemical association of elements in the solid matrix of IBA will vary for different samples therefore it is important to statistically investigate the impacts of leachants (DI and seawater) on leaching with multiple samples [28]. In addition, element leaching is highly dependent on the leaching methods, whereby the particle size, L/S ratio, time, pH, mixing/agitation approaches, scale and the type of leachants are the dominant factors affecting

the leaching results. For all experiments carried out in the study, IBA samples were prepared with an upper size limit of 4 mm achieved by mechanical crushing, whereas all leaching tests under the natural pH conditions. With pre-treatment of IBA samples, relatively homogenous samples in term of physical size are obtained and tested. In contrast to pH-static leaching test, the test methods used in this study were carried out without pH control. Therefore, pH of leachate is determined by the source properties of IBA and its interaction with the leachants used. For all batch leaching tests it was assumed that an equilibrium state would be attained in the end, therefore time effects are ignored. As for the scale, all samples were prepared with the volume ranged within 1 order of magnitude difference at lab scale, i.e. 90 g and 350 g for all batch leaching tests at respective L/S ratio of 10 and 2 while around 700-800 g for the column leaching tests. The scale effects were deemed negligible in the current study. In the following sections (3.2-3.5) leaching effects with respects of different leaching methods, L/S ratios and leachant types were statistically analyzed and then compared between each other, to unveil their characteristic leaching potential. We acknowledge that for certain comparison studies the IBA samples used in different leaching methods were not identical, for instance the ones in batch tests at L/S 2 vs. L/S 10 or column tests (Table 2). Under the circumstances, the resulted effects due to factors of IBA samples were considered random however complied with the same normal distribution. And indeed, bulk properties such as moisture, LOI, TC, TOC together with size distribution and total elements among different IBA samples from IP1-4 and MR in the current study were found to be not statistically significantly different between each other (p -value >0.05, Table S1).

3.2. Batch leaching tests at L/S 2

IBA metal leaching concentrations from batch tests at L/S ratio 2 using DI and seawater are shown in Fig. 1. 13 elements were investigated including Ag, As, Ba, Cd, Cr, Cu, Hg, Ni, Pb, Sb, Se, Tl, and Zn for 6 IBA samples obtained from MR. Based on *t*-test (p -value = 0.037), there existed statistically significant difference between seawater and DI leaching. Metal leached amounts using DI followed a decreasing order as, Cu (1-10 mg/kg) > Ba, Ni, Cr, Pb, Zn (0.1-1.0 mg/kg) > Sb (0.01-1.0 mg/kg) > As, Cd, Se (0.001-0.01 mg/kg) > Ag, Hg, Tl (<0.001 mg/kg). Such ranking under seawater was changed as, Cu (1-10 mg/kg) > Ba, Cr, Ni, Pb (0.1-1.0 mg/kg) > Sb (0.01-1.0 mg/kg) > As, Cd, Se (0.001-0.01 mg/kg) > Ag, Hg, Tl, Zn (<0.001 mg/kg). Zn distribution in IBA (Fig. S1) was most abundant (2665.02-3332.48 mg/kg) while its leaching was not prominent under DI, and was even completely inhibited under seawater. By contrast, Cu was the most leachable element under both leaching environments, likely due to its activity with divalent format (Cu^{2+}) [29] together with the formation of Cu-dissolved organic carbon complexes in the IBA leachate [30, 31]. Other than Zn, Pb was partly inhibited as well under seawater, both of which were attributed to pH buffering capacity of seawater [32]. By contrast, Tl was noted with significant increment in its leaching by seawater. Minor incremental leaching was also found with Ba, Ni and Sb. Clearly, seawater significantly modified the leaching potential of Zn, Pb and Tl based on batch leaching test at L/S 2. Due to the same IBA resource from MR, the difference among the leached amounts across IBA samples for each metal is not significant.

Fig. 1

3.3. Batch leaching test at L/S 10

Metal leaching concentrations from batch tests at L/S = 10 are shown in Fig. 2. IBA samples used for batch tests at L/S 10 were from multiple collection points so that the leached amounts for each element were highly heterogeneous among different IBA samples. Analogous to L/S = 2, *t*-test was performed with leaching data at L/S = 10 (*p*-value = 0.0022) indicating that, statistical significant difference was also there between seawater and DI leaching. Heavy metals from L/S 10 exhibited in overall higher leaching than those counterparts from L/S 2, ranking high to low with DI as, Ba, Cu, Pb, Zn (1-10 mg/kg) > Cr, Ni (0.1-1.0 mg/kg) > Sb, Se (0.01-1.0 mg/kg) > As, Cd, Hg (0.001-0.01 mg/kg) > Ag, Tl (<0.001 mg/kg); while ranking high to low with seawater as, Ba, Cr, Cu, Sb (1-10 mg/kg) > Ni, Se, Zn (0.1-1.0 mg/kg) > As, Pb (0.01-1.0 mg/kg) > Cd, Hg (0.001-0.01 mg/kg) > Ag, Tl (<0.001 mg/kg). Higher release is ascribed to the higher L/S ratio whereby element leaching was dominated by the partitioning coefficient. There were big differences as much as 3 orders of magnitude for Cu, Hg, Pb, Sb, Tl and Zn on their leached amounts between seawater and DI, while with lower leaching for Cu, Pb and Zn and higher leaching for Hg, Sb and Tl when seawater is used instead of DI as leachant, suggested the complicated effects. Seawater with its high salt contents like Na⁺, K⁺, Ca²⁺, Mg²⁺, Cl⁻ and SO₄²⁻, presents a strong buffering capacity on pH in the solution, hereby metal leaching would be altered according to their respective pH-dependent solubility [33]. The final leaching concentrations of heavy metals, however, likely determined also by second reactions such as surface adsorption with Fe and Al (hydr)oxides [31], thus affecting the trace metal leaching [34-36]. In this regard, seawater presented with large amount of cations and anions (the conductivity of seawater leachate was 49.0 ± 0.9 ms/cm while DI leachate was 4.6 ± 0.8 ms/cm for batch leaching test at L/S 10) may significantly change the surface charge of minerals [23].

Noteworthy that, such changes are both ion specific and mineral specific, for example, a large

concentration of Cl may decrease the net charge of the hydroxide surface [31]. Base on the current leaching test results, the interpretation of the multi-leaching behaviors (whereby Cu, Pb and Zn leached more while Hg, Sb and Tl leached less) may be a function of one or all of the following parameters: ionic strength, electronegativity, system pH and the adsorption surfaces [37]. However, leaching performance of different IBA samples varied for the six elements, which could be caused by the differences in IBA chemical properties.

Fig. 2

The modification onto the metal leaching imposed by seawater appeared to be more significant at L/S 10 than L/S 2, as evidenced from the metal ranking lists and the leached magnitudes. It suggests that a higher L/S ratio tends to exaggerate the gaps between DI and seawater on metals leaching potential. This is potentially because of the higher pH buffer capacity when the amount of seawater was increased and larger differences in conductivity of leachate formed when higher L/S ratio was applied in the batch leaching test. pH of seawater leachate decrease 11.2 ± 0.2 to 10.1 ± 0.5 while pH of DI leachate remained at around 11.6 ± 0.6 to 11.8 ± 0.7 when L/S ratio increased from 2 to 10 in batch test. Conductivity of seawater leachate remained at around 57.3 ± 0.8 ms/cm to 49.0 ± 0.9 ms/cm while for DI leachate reduced 16.6 ± 0.9 ms/cm to 4.6 ± 0.8 ms/cm when L/S ratio increased. Existing leaching tests such as Toxicity characteristic leaching procedure (*TCLP*) and Leaching Environmental Assessment Framework (*LEAF*) Test Methods are designed to mimic field conditions (landfill or terrestrial applications) or to challenge the waste material for determination of its intrinsic properties. The results in this study suggested the differences in the leachant would significantly influence the leaching performance of IBA. As a

result, it is important to specify the IBA leaching criteria in association with different leaching methods to mockup application scenarios.

3.4. Column leaching tests

Figure 3 shows the cumulative leached amounts for heavy metals along with different L/S ratios at 0.1, 0.2, 0.5, 1, 2, 5, and 10 during column tests using DI and seawater, respectively. The pH values were recorded lower under seawater than DI, while their differences were getting larger with the increased L/S ratio, as a result of buffering capacity from increased seawater breakthrough [38]. As such, the metal leaching behavior changed according to their respective pH-dependent solubility under the new pH. As mentioned earlier, the difference on metal leaching behaviors may also be attributed to the interactions of leachant components with dissolved minerals from the IBA [36], leading to secondary reactions for competitive sorption and re-precipitation [31]. However, the effects from the leachant may be less significant than that from the IBA own properties. At least, the release trend (in terms of the curve shape) from seawater maintained analogous to that from DI. By contrast, the difference of cumulative leached amount for certain metals among the IBA samples was as much as 2-3 logs of magnitude, such as Ag, Cr, Hg, Pb and Zn. It is worth mentioning that the slope of the leaching curves at its later stage indicated their further potential correspondingly for enhanced or inhibited leaching beyond the test range. In this regard, special attentions perhaps have to be paid to Cr, Hg, Sb and Tl, as their leaching potential turned to be dramatically enhanced at later stage of the column tests under seawater.

Fig. 3

3.5. Metal leaching potential under different methods

Certain IBA samples, i.e., IP1C1, IP1C2, IP2C1, IP2C2, IP3C, and IP4C, since they performed with both batch ($L/S = 10$) and column leaching tests, are further compared between each other as shown in Fig. 4. Two special situations were noted associated with two groups of metal species, (a) metals dependent of leaching methods (batch or column) while independent of the leachants (seawater or DI), (b) metals dependent of both. Ba (except for IP1C2 with seawater leaching), Cu, Pb and Zn exhibited higher leaching in batch tests (1.0-10 mg/kg) from all IBA samples, disregard of DI or seawater. These metals were meanwhile ranked amongst the most affluent (Table S1). Contrastively, Ag, Ni, Se and Tl (except for IP4C with DI) demonstrated higher leaching in column tests, independent of leachants as well. Other than aforementioned, there existed certain metal (i.e. Sb) showing dependent on both the leachant and the leaching method during the column tests using seawater, whereby a higher leaching was always noted. As for the rest metals, such as As, Cd, Cr and Hg, their concentrations presented mixed trends out of the two situations, likely dominated by their own IBA properties. Indeed, the role of ionic strength on metal release (particularly for those with relative high concentrations in the IBAs) may be significant yet less than other factors e.g. pH (Fig. 3), L/S ratio (Fig. 1 vs. Fig. 2), and etc., based on their magnitude of changes [31]. Note that, the leached amounts for Ag, As, Cd, Hg and Tl were extremely low (<0.001 - 0.01 mg/kg) and deemed stable to surrounding geochemistry. As such, they carried less significance of comparison as opposed to those with high release and sensitivity by different leaching conditions.

Fig. 4

3.6. Metal leaching potential with different leachants

Statistical comparison of leaching data among different leaching methods provides deeper insights into effects from factors of ionic strength coupled with L/S ratio and leaching approaches, as shown in Table 3 and Fig. 5-7 (Ag is not shown). Plots of metal concentrations between DI and seawater were firstly made with respects of batch tests at L/S = 2 (Fig. 5), L/S = 10 (Fig. 6) and the column tests (Fig. 7).

Fig. 5-7

3.6.1. L/S ratio effects

Comparison of batch tests between L/S 2 and L/S 10 enlightens the seawater effects associated with L/S ratios while avoiding the influence by different leaching methods (Fig. 5 & 6).

Seawater effects were metal-specific on their leaching potential while also a function of L/S ratios (Fig. 5). A higher L/S ratio in batch leaching tests tended to exaggerate the difference on metal leaching between seawater and DI. Certain metals shared the same trends (promotion or inhibition) between the two L/S ratios, i.e., Cr, Cu, Ni, Pb, Sb, Tl and Zn, which simultaneously carried with a higher leaching potential under DI (0.1-10 mg/kg). It suggested that ionic strength from seawater tend to pose higher influence on those high-leachable metal species.

3.6.2. Leaching method effects at L/S = 2 and 10

Under a fixed L/S ratio, comparison of batch and column tests may further enlightens understanding of effects due to the operation approaches. Figure 6 hereby shows the plots for 13 metals on their leached amounts between DI and seawater from both batch and column tests at the same L/S = 2. Departed data points above the 45° slope (dash line) were noted for a few

metals, such as Ba, Ni (Batch tests), Sb and Se (column tests), suggesting increased leaching facilitated by seawater. Statistically comparison of the metal leaching results can be found in Table 3, whereas column tests using seawater tended to leach out more metals or if not, at least comparable amounts (except for As). These findings however were contradicted to those from batch tests where several metals were inhibited instead, like Cd, Cu, Pb and Zn.

Batch and column tests was compared with a fixed L/S ratio at 10 too (Fig. 7). With a raised L/S ratio from 2 to 10, leaching of Cd, Ni and Sb was further enhanced in column tests by seawater. On the other end, as compared to L/S = 2, seawater under L/S = 10 in batch tests has reversed the leaching style of certain metals, such as Ba, Cd and Hg (Fig. 6 vs. Fig. 7). Due to the different leaching approaches, the leaching geochemistry would be different (e.g. pH and leachate components) [39, 40]. Also, substances are continuously removed from the column, but not from the batch tests. To this end, it led to a constant disequilibrium in the column however not the case presumed in batches.

3.6.3. Statistical comparison

Table 3 was made to summarize the statistical analysis results based on linear regression modelling. The linear regression coefficients regarding the plots with Figs. 5-7, herein, provided a quantitative assessment of the variation: if the coefficient = 0.9-1.1, comparable release (=) between the two leachants was assigned; if the coefficient >1.1, significantly higher metal release (+) would be assigned; and if the coefficient <0.9, lower metal release (-) would be assigned, as shown in Table 3.

Table 3

Under batch tests at L/S 2, 6 metals (As, Ba, Cr, Ni, Sb, Tl) were noted with significantly higher release, 3 (Ag, Se, Zn) with comparable release while the other 4 (Cd, Cu, Hg, Pb) with significantly lower release in the presence of seawater. The inventory under batch tests at L/S 10 was noted with significant higher release for 7 metals (Cd, Cr, Hg, Ni, Sb, Se, Tl), comparable release for 2 (Ag, As) and significant lower release for 4 (Ba, Cu, Pb, Zn) in the presence of seawater. Slightly more metals may be favored for their leaching in the presence of seawater, given the metals inventory of 7 “+” vs. 4 “-” and 6 “+” vs. 4 “-” with L/S 10 and L/S 2, respectively. Taking account of the metal distributions, it was interestingly found that seawater likely had a preferred inhibition on those metals concurrently with a higher distribution, such as Cu, Pb and Zn.

Under column tests at L/S 2, 7 metals (Ba, Cr, Hg, Pb, Sb, Se, Zn) were assigned for significantly higher release similar to those in the batch tests, while the number assigned for significantly lower release dramatically reduced to 1 (As), suggesting a more favorable leaching environment by seawater. This trend was even reinforced at L/S 10, where 10 metals (Ag, Ba, Cd, Cr, Hg, Ni, Pb, Sb, Se, Zn) out of 13 presented higher release with seawater. It should be noted that, however, most metals with a higher distribution from IBA samples used to be inhibited in batch tests with seawater, were now among those with significant higher release in the column tests. It is possibly a result from the constant unbalance state within the columns in favor of metal mobility particularly under the high ionic strength. The descending pHs in column test with time under seawater, may also be responsible for the observed enhancement in overall metal leaching.

3.7. Metal leaching potential with an increased L/S ratio from 0.1 to 10 in the column tests

Column tests with seawater generally indicated a favorable condition for enhanced metal leaching potential as compared to batch tests, which was further strengthened as L/S ratio raised from 2 to 10 (Table 3). To depict the full scenario, we plotted the metal leaching profiles along with increased L/S ratios from 0.1 till 10 ($L/S = 0.1, 0.2, 0.5, 1, 2, 5, \text{ and } 10$) showing in Fig. 8.

Fig. 8

Transitional trends were noted on the metal release with the increase of L/S ratios, but metal specific. Firstly, certain data aggregated on the 45° dashline, such as Cd and Cu, suggesting a stable performance independent of the leachant. Second, for a few metals, their data spread over a wide area, like Cr and Pb. As for the distribution, substantial data points of Ba, Hg, Tl and Zn slightly fell above the 45° dashline, while most data points of Sb fell below it. Another pattern of distribution was noted with As and Se, whereas an increased dispersivity happened with the increase of L/S ratios. Mixed patterns were also found. For instance, Ba started to skewed up until certain L/S ratio, while Sb presented a more complicated concave shape. Different metal patterns indicated various responses to seawater as compared to DI, as a function of their respective distribution and speciation in solubility controlling mineral phases of the IBA [28, 41-44].

Table 4 summarizes the element leaching potential with column tests under different L/S ratios (0.1-10) between DI and seawater, with a linear regression analysis on these data. Analogous to Table 3, quantitative assessment of the variation on linear regression coefficients was provided in Table 4: if the coefficient = 0.9-1.1, comparable release (=) between the two leachants was

assigned; if the coefficient >1.1 , significantly higher metal release (+) would be assigned; and if the coefficient <0.9 , lower metal release (-) would be assigned.

Table 4

The inventory provides a platform to track the transition trend for each metal. Indeed, there existed an interesting trend for most metals. With ranking L/S ratios low to high, all metals, except for Ag, Cd and Se, followed an apparent one-way variation of leaching potential statistically different from their primary ones (L/S at 0.1). The transitions may be further classified into two-phase and three-phase variations. For instance, As was initially inhibited (L/S 0.1-2.0) by seawater until reached to comparably leaching to those by DI (L/S 5 & 10), undergoing a two-phase variation. Cr and Ni were the only two metals which experienced the three-phase variations in this regard, from inhibition through comparable to enhanced release by seawater.

It is possible to check Table 4 via another angle i.e. the initial trend of metal release. Ascribed to a small volume of seawater passing through (e.g. at L/S 0.1) meanwhile a limit contacting time at the beginning, significant changes would be hardly possible on the metal leaching behavior. Herein, As, Cr, Cu, Hg, Ni and Pb likely carries a higher sensitivity to seawater or seawater-modified geochemical conditions, e.g. pH variation and the presence of competitive species, as their release was instantly affected under seawater [45-47]. Other than that, there were no significant differences between seawater and DI for the leaching of Ag, Ba, Cd, Pb, Sb, Se, Tl and Zn at the beginning, thought to be less sensitive. The switching rate of leaching status between two phases, rapid or slow, in fact indicating the different mechanisms relevant to metal

leaching capacity and kinetics. For instance, it took only about 1 and 2 L/S for respective Cr and Ni to yield three-phase transition, while 5 L/S for Ag to yield two-phase transition, suggesting a retarded effect from the latter. As a matter, metal leaching kinetics was controlled from a combination of several factors, including metal distribution and speciation, pH sensitivity, partitioning coefficient, competitive sorption capacity, and etc. [48-52]. Among these factors, pH was often the dominant one affecting the metal leaching performance. As seen in Fig. 3, the growing pH difference caused by seawater (toward lower) affected metals with higher pH sensitivity (e.g. Cr) with more rapid kinetics.

4. Conclusions

- (1) Metal leaching behaviors from IBA were different among different leaching methods, while statistically correlated to the leachant types, i.e., seawater or DI.
- (2) Based on the same leaching method, however, the effects by seawater was minor as compared to those exerted from IBA intrinsic properties.
- (3) For batch tests with $L/S = 2$, seawater effects were noted while not as obvious as those with $L/S = 10$. For most metals with higher leached amounts (0.1-10 mg/kg), there was a similar trend between batch leaching tests of $L/S = 2$ and 10. For instance, seawater promoted the leaching of Cr, Ni, and Sb while inhibiting leaching of Cu, Pb and Zn under both L/S ratios.
- (4) For column tests, however, it was apparent that seawater tended to enhance most metal leaching as compared to DI. While such effects become more obvious along with increased L/S ratios.

- (5) Given above, the leaching standard using DI as the default leachant may not be appropriate for IBA application scenario in land reclamation, as it lead to a bias to the metal leaching potential in IBA associated with seawater environment.
- (6) Scenario-based leaching methods and the associated leaching criteria shall be established for IBA application.
- (7) Multiple safety factors may be considered for leaching criteria established based on laboratory compliance test using seawater for different trace metals in reference to DI.

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Tables and Figures

Table 1. Metal leaching limits based on Council Decision 2003/33/EC (EUR-Lex, 2003; Dou et al., 2017).

Country/ Region	Remarks	L/S = L/kg	Cl ⁻	F ⁻	SO ₄ ²⁻	As	Ba	Cd	Cr	Cu	Hg	Mo	Ni	Pb	Sb	Se	Zn
			mg/kg														
Council Decision 2003/33/EC	Inert waste	2	550	4	560	0.1	7	0.03	-	0.9	0.003	0.3	0.2	0.2	0.02	0.06	2
		10	800	10	1000	0.5	20	0.04	0.5	2	0.01	0.5	0.4	0.5	0.06	0.1	4
		Percolation*	450	2.5	1500	0.06	4	0.02	-	0.6	0.002	0.2	0.12	0.15	0.1	0.04	1.2
	Non-hazardous waste	2	10000	60	10000	0.4	30	0.6	4	25	0.05	5	5	5	0.2	0.3	25
		10	15000	50	20000	2	100	1	10	50	0.2	10	10	10	0.7	0.5	50
		Percolation*	8500	40	7000	0.3	20	0.3	2.5	30	0.03	3.5	3	3	0.15	0.2	15
	Hazardous waste	2	17000	200	25000	6	100	3	25	50	0.5	20	20	25	2	4	90
		10	25000	500	50000	25	300	5	70	100	2	30	40	50	5	7	200
		Percolation*	15000	120	17000	3	60	1.7	15	60	0.3	10	12	15	1	3	60
*: unit with mg/L;																	

Table 2. Sample collection, sample preparation with respects of different leaching tests.

Sample ID	Collection points	Daily samples	Weekly Samples	Monthly samples	Total samples	Sample preparation	Batch leaching		Column leaching
							L/S 2	L/S 10	
IP1C1	IP1	22 (1 st month)	16 (2 nd -5 th month)	0	38	5-month combination: 1 st month + 2 nd -5 th month	-	+	+
IP1C2	IP1	0	29 (6 th -12 th month)	0	29	7-month combination: 6 th -12 th month	-	+	+
IP2C1	IP2	22 (1 st month)	16 (2 nd -5 th month)	0	38	5-month combination: 1 st month + 2 nd -5 th month	-	+	+
IP2C2	IP2	0	28 (6 th -12 th month)	0	28	7-month combination: 6 th -12 th month	-	+	+
IP3C	IP3	0	4 (1 month)		4	1 month combination with 4 samples	-	+	-
IP4C	IP4	0	4 (1 month)		4	1 month combination with 4 samples	-	+	-
MRM1	MR	0	0	1	1	1 st month sample	-	+	+
MRM2	MR	0	0	1	1	2 nd month sample	-	+	+
MRM3	MR	0	0	1	1	3 rd month sample	-	+	-
MRM4	MR	0	0	1	1	4 th month sample	-	+	-
MRMC	MR	0	0	4	4	4-month combination: 1 st -4 th samples	-	-	+
MRW1	MR	0	1	0	1	1 st week sample after 4 month samples	+	-	+
MRW2	MR	0	1	0	1	2 nd week sample after 4 month samples	+	-	-
MRW3	MR	0	1	0	1	3 rd week sample after 4 month samples	+	-	-
MRW4	MR	0	1	0	1	4 th week sample after 4 month samples	+	-	-
MRW5	MR	0	1	0	1	5 th week sample after 4 month samples	+	-	-
MRW6	MR	0	1	0	1	6 th week sample after 4 month samples	+	-	-
IP1-4: incineration plant 1 to 4; “+”: indicates the IBA sample which was subjected to the specific leaching test; “-”: no leaching tests carried out with the IBA samples.									

Table 3. Summary of element leaching potential with different leaching standards between DI and seawater.

Types of leaching tests	Leached metals under both DI and seawater													Inventory			pH ^{&}
	Ag	As	Ba	Cd	Cr	Cu	Hg	Ni	Pb	Sb	Se	Tl	Zn	+	=	-	
Batch, L/S 2																	
Trend	=	+	+	-	+	-	-	+	-	+	=	+	=	6	4	4	-
Coefficient	NA	1.2122	1.5549	0.8098	1.8708	0.7827	0.31	1.535	0.2534	1.6845	1.0356	2.7624	NA				0.9675
Correlation (<i>R</i> ²)	NA	0.5707	0.7688	-0.152	0.655	-0.101	0.3029	0.6451	0.1804	-2.051	0.5439	0.9357	NA				-0.723
Batch, L/S 10																	
Trend	=	=	-	+	+	-	+	+	-	+	+	+	-	7	2	5	-
Coefficient	1	0.999	0.5873	1.5665	3.9669	0.2589	1.712	1.1307	0.0049	6.39	7.4565	4.1194	0.0016				0.8621
Correlation (<i>R</i> ²)	1	0.6643	-1.742	-0.405	-0.158	0.3964	-0.221	0.9481	-0.923	-2.576	-0.143	0.7528	0.1047				0.0113
Column, L/S 10																	
Trend	+	=	+	+	+	=	+	+	+	+	+	=	+	9	4	0	-
Coefficient	1.2423	0.6444*	1.7492	1.2761	4.869	1.0053	2.0924	1.1863	1.2667	8.5984	1.8694	1.0058	1.641				0.9795
Correlation (<i>R</i> ²)	0.9810	-4.067	0.5572	0.6884	0.8472	0.9438	0.7887	0.6714	0.2886	0.6999	-0.413	0.9810	0.8770				0.8685
Column, L/S 2																	
Trend	=	-	+	=	+	=	+	=	+	+	+	=	+	7	5	1	-
Coefficient	1.0726	0.7788	1.4699	1.0395	1.7675	0.9252	1.1937	1.0523	1.1368	1.2421	1.1427	1.0346	1.4606				0.9653
Correlation (<i>R</i> ²)	0.8327	-0.095	0.4711	0.8718	0.8376	0.9565	0.9626	0.6851	0.5312	0.7259	0.6487	0.9384	0.9042				0.7700
“+”: Seawater leaching caused higher release whereas the Trendline coefficient was >1.1																	
“-”: Seawater leaching caused lower release whereas the Trendline coefficient was <0.9																	
“=”: Seawater and Deionized water leaching showed comparable release whereas the Trendline coefficient was at 0.9-1.1																	
*: The coefficient was double checked with data dispersivity to assign the comparison result due to correlation (<i>R</i> ²) <0.5000																	
&: pH value is log-based so that its trend was indicated via comparison between coefficient and 1 echoing its high sensitivity. If the coefficient <1, “-” is assigned; otherwise “+” is assigned when >1 while “=” if the coefficient equal to 1.																	
NA: Not Available																	

Table 4. Summary of element leaching potential with column tests under different L/S ratios (0.1-10) between DI and seawater.

Column leaching L/S ratios	Leached metals under both DI and seawater													Inventory			pH*
	Ag	As	Ba	Cd	Cr	Cu	Hg	Ni	Pb	Sb	Se	Tl	Zn	+	=	-	
L/S 0.1																	
Trend	=	-	=	=	-	-	+	-	+	=	=	=	=	2	7	4	-
Correlation (R^2)	0.7812	0.7503	0.3845	0.6933	0.485	0.8982	0.4409	0.2137	0.833	0.4889	0.6067	0.5462	0.8457	NA	NA	NA	0.7281
L/S 0.2																	
Trend	+	-	=	=	-	-	+	=	+	=	=	=	=	3	7	3	-
Correlation (R^2)	0.9665	0.7544	0.5836	0.7924	0.4676	0.9449	0.5493	0.3712	0.8497	0.6067	0.6976	0.5202	0.889	NA	NA	NA	0.8548
L/S 0.5																	
Trend	+	-	+	+	=	-	+	=	+	+	+	=	+	8	3	2	-
Correlation (R^2)	0.9889	0.591	0.6492	0.8299	0.7193	0.9448	0.7668	0.4201	0.8147	0.6465	0.6875	0.7782	0.9362	NA	NA	NA	0.9911
L/S 1																	
Trend	+	-	+	=	+	=	+	=	+	+	=	=	+	7	5	1	-
Correlation (R^2)	0.9834	0.4259	0.3165	0.879	0.7948	0.9652	0.898	0.5241	0.7479	0.7054	0.6663	0.8954	0.9192	NA	NA	NA	0.9088
L/S 2																	
Trend	=	-	+	=	+	=	+	=	+	+	+	=	+	7	5	1	-
Correlation (R^2)	0.8327	-0.095	0.4711	0.8718	0.8376	0.9565	0.9626	0.6851	0.5312	0.7259	0.6487	0.9384	0.9042	NA	NA	NA	0.8685
L/S 5																	
Trend	+	=	+	=	+	=	+	+	=	+	+	=	+	8	5	0	-
Correlation (R^2)	0.9594	-2.156	0.314	0.8735	0.8841	0.9525	0.7478	0.7002	0.3774	0.8342	-0.068	0.9692	0.8518	NA	NA	NA	0.9048
L/S 10																	
Trend	+	=	+	+	+	=	+	+	=	+	+	=	+	9	4	0	-
Correlation (R^2)	0.981	-4.067	0.5277	0.6884	0.8472	0.9438	0.7887	0.6714	0.2886	0.6999	-0.413	0.981	0.877	NA	NA	NA	0.7700
“+”: Seawater leaching caused higher release whereas the Trendline coefficient was >1.1																	
“-”: Seawater leaching caused lower release whereas the Trendline coefficient was <0.9																	
“=”: Seawater and Deionized water leaching showed comparable release whereas the Trendline coefficient was at 0.9-1.1																	
*: pH value is log-based so that its trend was indicated via comparison between coefficient and 1 echoing its high sensitivity. If the coefficient <1, “-” is assigned; otherwise “+” is assigned when >1 while “=” if the coefficient equal to 1.																	
NA: Not Available

Figure 1. IBA metal leaching concentrations from batch tests (L/S 2) with DI and seawater, respectively.

Figure 2. IBA metal leaching concentrations from batch tests (L/S 10) with DI and seawater, respectively.

Figure 3. Cumulative leached amount (mg/kg) for heavy metals from IBA column tests using DI and seawater, respectively.

Figure 4. Comparison of leaching behaviors from different IBA samples between batch tests (L/S 10) and column tests (at accumulative L/S 10).

Figure 5. Comparison of batch tests of L/S 2 and L/S 10 between DI and seawater.

Figure 6. Comparison of batch and column tests at L/S 2 between DI and seawater.

Figure 7. Comparison of batch and column tests at L/S 10 between DI and seawater.

Figure 8. Comparison of column tests at L/S 0.1, 0.2, 0.5, 1, 2, 5 and 10 between DI and seawater.

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