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Impact of pH on leaching of inorganic constituents from MSW incineration bottom ash: An Indian case study

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ABSTRACT

Assessment of contaminant leaching from municipal solid waste (MSW) incineration bottom ash (IBA) is crucial in determining its fate. Investigating the role of pH to examine the fluctuations in contaminant leaching would enhance understanding of the leaching behavior of IBA due to the changes in surrounding environment. However, leaching from IBA has chiefly been studied at its intrinsic pH. Present study examines the influence of pH on leaching of contaminants from IBA collected from two incineration plants of Delhi using CEN/TS 14429 (2015). The test results revealed the amphoteric nature of Pb and Zn, exhibiting high leaching at low and high pH, whereas low leaching at neutral to moderately high pH. The leaching concentration of Cd, Cr, Cu, and Ni increased under acidic pH conditions but was much lower at neutral to alkaline pH. The leaching pattern of Sb resembled Mo, with their concentrations decreasing under acidic pH, increasing until moderately high pH, and decreasing again under very high pH. The variation in the leaching of chlorides and sulfates with pH is insignificant in comparison to trace metal(loid)s. The test results further demonstrated that fresh IBA, having alkaline pH between 10-12, is suitable for disposal in non-hazardous waste landfills however, IBA having pH between 8-10, may have to be disposed in hazardous waste landfills due to aggravated leaching of antimony. Moreover, caution is indispensable when co-disposing IBA with MSW as degradation of MSW can cause pH to shift towards the acidic range which exacerbate the contaminant leaching from IBA.

Keywords: Incineration bottom ash, Leaching, pH, Heavy metals

1   INTRODUCTION

Municipal solid waste (MSW) incinerated bottom ash (IBA) is the primary residue obtained from the incineration of MSW. Presently in India, IBA is co-disposed with incineration fly ash in sanitary landfills, open dumpsites, or mining pits with no prior assessment of contamination potential to the sub-surface environment due to lack of scientific studies on IBA within the country (Gupta et al., 2021a). Previous studies on IBA (del Valle-Zermeño et al., 2013; Tang et al., 2015; Vaitkus et al., 2018; Van Gerven et al., 2005; Zekkos et al., 2015) have suggested that it can prospectively substitute natural raw materials in earthworks like embankments, road construction, landfill cover material, etc. If the same could be ascertained for Indian conditions, it would substantially reduce the footprint of future landfills and contribute towards sustainable development.

Leaching studies are deemed mandatory for assessing the options of disposal and reuse of IBA (Blasenbauer et al., 2020). Leaching from IBA has primarily been studied at its intrinsic pH (Arickx et al., 2006; Caviglia et al., 2019; Hyks et al., 2011; Lidelöw & Lagerkvist, 2007; Santos et al., 2013), as it simulates the release of contaminants when IBA comes in contact with water having pH close to neutral and helps adjudge its reuse and disposal options. However, investigating the fluctuations in contaminant leaching due to pH changes would enhance the perception of leaching from IBA due to changes in the surrounding environment encountered throughout its life cycle (van der Sloot et al., 1997; Dijkstra et al., 2006; Ai et al., 2019). The European Committee for Standardization (CEN) and US Environmental Protection Agency (USEPA) have established the standard protocols for pH-dependent leaching (CEN/TS 14429, 2015; US EPA, 2013), which emphasizes the importance of the same. Quina et al.

(2009) suggested that pH-dependent leaching offers a better understanding of the environmental impact of IBA compared to the total content. Total content represents available elements determined by aqua regia acid digestion, whereas in pH dependent studies, leaching of elements at varying pH of the leachant is determined.

The review of the literature (Tang et al., 2015; Vaitkus et al., 2018, Gupta et al., 2021b) demonstrates wide variability in the characteristics of IBA from diverse origins attributable to the differences in the composition of MSW, operating conditions of the incinerator, etc. However, the authors believe that the present study can be used as a benchmark and the results of this study would certainly provide a spectrum of concentrations of contaminants of concern under varying pH conditions. Gupta et al. (2021b) have studied the leaching characteristics of IBA from Indian MSW incineration plants, but the study is limited to intrinsic pH conditions. Hence, the present work investigates the pH-dependent leaching characteristics of IBA from incineration plants in Delhi to ascertain its long-term impact on the environment.

2 MATERIAL COLLECTION AND METHODOLOGY

2.1 Material Collection

MSW incinerated bottom ash samples was collected from two MSW incineration (MSWI) plants in Delhi, referred to as Plant A and Plant B. The water-quenched IBA was kept for air-drying under a shed for about a week and later screened through a 4.75 mm sieve. The screening through a 4.75 mm sieve was targeted to eliminate gravel-like fraction (i.e., coarser than 4.75 mm), and to obtain soil-sized fraction of IBA (i.e., sand, silt, and clay) based on Indian Standards for soil testing (IS: 2720-Part 4, 1985). The present study is limited to the soil-sized fraction, which constitutes 65-70% of the total IBA and can be used in geotechnical applications. Figures 1(a) to 1(c) illustrate the on-site processing of IBA and around 3 tonnes of soil-size fraction were obtained. Representative samples were obtained by coning and quartering for experimental studies.
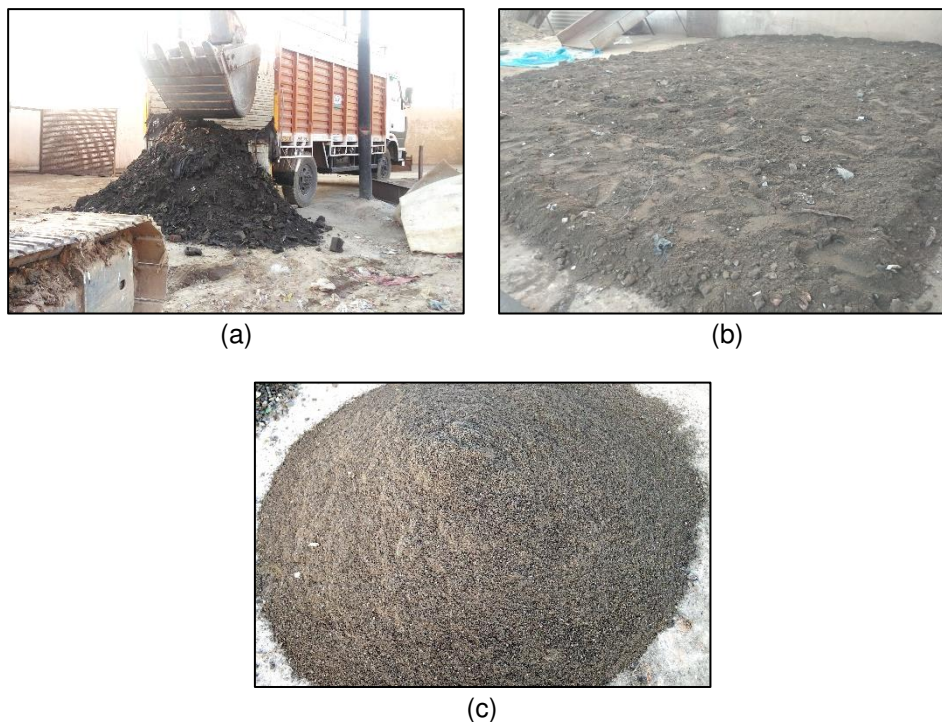

(a)

(b)

(c)

Figure 1. (a) *Unloading of IBA from trucks,* **(b)** *IBA spread in thin layers for air-drying,* **(c)** *representative IBA sample after screening from 4.75 mm sieve*

2.2   Experimental Study

Static pH-dependent leaching tests were conducted on IBA samples using CEN/TS 14429 (2015) and about 50 gm of IBA samples was mixed with the 500 ml leachant for a total contact time of 48 hrs. The tests were performed at pH ranging from 4 to 12 (4, 5, 6, 7, 8, 9, 10, and 11.5). The leachant consisted of a combination of nitric acid (0 to 5 ml) and deionized water. The amount of nitric acid added to maintain the required pH was determined from pre-titration experiments. The suspension obtained was filtered through a 0.45-micron filter, and the eluate was analyzed for pH, sulfates, chlorides, and heavy metal(loid)s. Silver nitrate titration and turbidimetric methods were used to determine chlorides and sulfates, respectively, as per APHA et al. (2012). Heavy metal(loid)s were determined using inductively coupled plasma mass spectroscopy (ICP-MS). The leaching test was performed twice on each sample. Each determination of chlorides, sulfates, and heavy metal(loid)s was made thrice to ensure the accuracy of the results, and the mean values were reported. The variation in pH value by ± 0.2 units was rounded off to the nearest integer when reporting the results. For Ex. pH of 5.2 and 4.8 is considered equivalent to 5. Gupta et al. (2021b) identified ten contaminants of concerns (CoCs), namely, Cd, Cr, Cu, Mo, Ni, Pb, Sb, Zn, Cl^- and SO_4^{-2}, which should mandatorily be investigated for IBA. Hence, the present work investigates the role of pH on these contaminants only.

Furthermore, the total content of the trace metal(loid)s was determined on IBA samples using aqua-regia digestion. Herein, the IBA samples were pulverized to pass 75 μm and microwave digested using aqua regia (i.e., 3 parts of HCl and 1 part of HNO_3). The filtered samples were analyzed for concentration of trace metal(loid)s using ICP-MS. The total content of chlorides and sulfates was determined using BS: 1377-3 (2018). The leachability (or L') of each contaminant was calculated as the ratio of the maximum leaching concentration of the contaminant obtained from pH-dependent test to its total content and expressed as percentage.

The leaching test results were further compared with the European Union limits (EU Council, 2003) for inert waste landfills (EU_IW), non-hazardous landfills (EU_NHW) to assess disposal of IBA.

3   RESULTS AND DISCUSSION

Figure 2 presents the results of pH-dependent leaching study and its comparison with the corresponding total content. The dashed lines represent the curve obtained by fitting experimental data into polynomial curves. These curves are similar to solubility curves for the respective elements (Quina et al., 2009; Dou et al., 2017). The obtained trend of the curves is interpreted using literature studies (Dijkstra et al., 2006; Meima & Comans, 1999; Zhang et al., 2008) with reference to precipitation/dissolution and surface complexation models which have been found helpful in predicting the factors controlling leaching of contaminants from IBA. The maximum (max) and minimum (min) leaching concentration of the contaminant, the ratio of maximum to minimum leaching concentration (max/min), the total content of the contaminants (TC), and the leachability of contaminant (L') are presented in Table 1. A higher value of maximum to minimum ratio demonstrates a significant influence of pH on the leaching concentration of the particular contaminant.

The leaching of Cadmium (Cd) decreases with an increase in pH. The results suggest that Cd is mainly released under acidic conditions. At moderate to highly alkaline conditions, the leaching of Cd decreases significantly by 2 to 3 orders; hence, Cd leaching is not a matter of concern for intrinsic pH conditions of IBA. Previous studies (Ai et al., 2019; Meima & Comans, 1999) reveal that insoluble otavite ($CdCO_3$) defines the leaching of Cd for alkaline pH conditions. Dijkstra et al. (2006) and Zhang et al. (2008) noted that the surface complexation model accurately predicts Cd leaching compared to the precipitation/dissolution model, which overestimates the same. The studies also noted that Cd is adsorbed by hydrous ferric oxide in the pH range of 6-12. The leachability of Cd is approximately 70%, indicating that the majority of Cd is available for leaching at pH<4.

Chromium (Cr) leaching is observed to be elevated in the acidic pH range. The leaching concentration of Cr decreases with an increase in pH, with the lowest values observed close to neutral pH and insignificant differences observed in Cr concentration for the alkaline pH range. The leaching of Cr decreases at higher pH because of insoluble minerals like ettringite, which absorbs it (Quina et al., 2009). Zhang et al. (2008) suggested that both precipitation/dissolution and surface complexation models are essential to predict the leaching behavior of Cr. The study illustrates that Cr is absorbed by

hydrous ferric oxide (FehOCrOH$^+$) in the pH range of 3-7 by surface complexation phenomenon, while chromium oxide (Cr$_2$O$_3$) governs the solubility of Cr in the alkaline pH range. The leachability of Cr is low (4-8%), suggesting that most of the chromium remains in the solid matrix and is unavailable for leaching.

Copper (Cu) leaching is observed to be highest in acidic conditions, and the concentration decreases with an increase in pH. Dijkstra et al. (2006) and Zhang et al. (2008) suggested that the solubility of Cu is strongly affected by the presence of dissolved organic carbon (DOC). Furthermore, Dijkstra et al. (2006) observed that surface complexation, i.e., absorption by ferrous and aluminum hydroxides controls the leaching of Cu. The study also suggested that malachite (Cu$_2$OH$_2$CO$_3$) controls the leaching of Cu for pH above 8. On the contrary, Zhang et al. (2008) noticed that the precipitation/dissolution model predicts the leaching of Cu more accurately than the surface complexation model, and the leaching is controlled by malachite in the pH range of 5-7 and copper oxide for pH above 7. Leachability of Cu is in the range of 15-20%.

Molybdenum (Mo) leaching is highest around pH 7 to 8, whereas it decreases in acidic and alkaline conditions. A similar pattern was observed by Ai et al. (2019) and Van Gerven et al. (2005). The leaching of Mo at pH < 6 is controlled by iron molybdate (Fe$_2$(MoO$_4$)$_3$) or wulfenite (PbMoO$_4$) (Dijkstra et al., 2006; Meima & Comans, 1999). At higher pH, calcium molybdate (CaMoO$_4$) and oxyanion substitution of sulfate in ettringite define the leaching characteristics of Mo in IBA (Van Gerven et al., 2005). Akin to Cd, the leachability of Mo is high (65-70%), suggesting that it is readily available for leaching at pH 7-8.
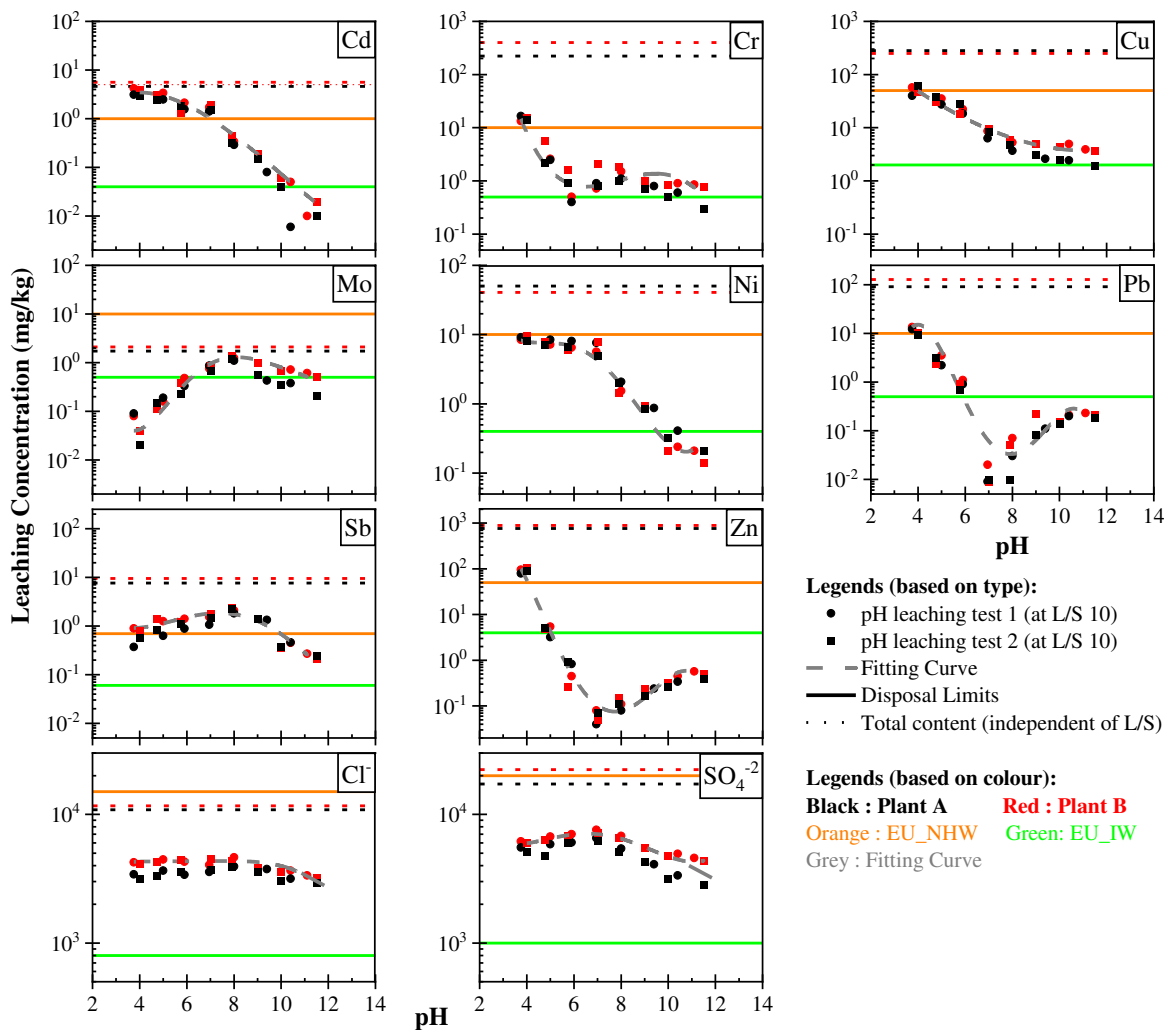

Figure 2. pH-dependent leaching of contaminants from IBA (CEN/TS 14429, 2015)

Nickel (Ni) shows high leaching in acidic conditions, and the concentration decreases by an order of 2 at pH 10-11. In the alkaline condition, precipitation/dissolution and surface complexation models accurately predict the leaching behavior of Ni (Dijkstra et al., 2006; Zhang et al., 2008). Nickel hydroxide (NiOH$_2$) is the controlling factor at a pH above 7. Hydrous ferric oxide (HFO as FehONi$^+$) can adsorb Ni ions or form complexes, which can also influence the leaching of Ni in the pH range of 6-9 (Zhang et al., 2008). The leachability of Ni is 18-20%.

Leaching of lead (Pb) is amphoteric in nature, showing higher solubility under acidic and alkaline conditions and reduced leaching around neutral pH conditions. pH significantly affects the leaching of Pb, as indicated by a high maximum to minimum leaching ratio of 1300-1400. Pb leaching above pH 8 is observed to be lower than that in an acidic environment and is governed by lead hydroxide (Pb(OH)$_2$) precipitates (Zhang et al., 2008). The study also suggested that the precipitation/dissolution model predicts the leaching of Pb accurately. On the contrary, Dijkstra et al. (2006) stated that the surface complexation model may better predict the leaching of Pb and observed cerussite (PbCO$_3$) to control its leaching. The formation of complex compounds with DOC (Dijkstra et al., 2006) and the adsorption of Pb on the iron oxide surface (Oehmig et al., 2015) suppresses the leaching of Pb. The leachability of Pb is only 10-12%, indicating that the trace metal is mostly unavailable for leaching.

Antimony (Sb) leaching resembles Mo, with the highest values observed in the pH range of 7-8. Leaching of Sb is observed to decrease in alkaline and acidic conditions. The maximum to minimum leaching ratio is lowest for Sb amongst the studied trace metal (loid)s, indicating the most negligible influence of pH on Sb in comparison to other trace metal(loid)s. The leaching of Sb is governed by the sorption of ferric and aluminum oxides along with ettringite solubility (Ai et al., 2019, Gines et al., 2009). Dijkstra et al. (2006) and Cornelis et al. (2006) suggested that at higher pH, calcium antimonate (Ca(SbO$_3$)$_2$) is the controlling factor for the solubility of Sb. When the pH of IBA falls below 8, ettringite is transformed into gypsum, which possibly increases the leaching of Sb. The leachability of Sb is around 25-30%.

Similar to Pb, Zinc (Zn) leaching is also amphoteric, and a change in pH significantly influences its solubility as indicated by the maximum to minimum leaching ratio over 2000. Previous studies (Dijkstra et al., 2006; Meima & Comans, 1999; Zhang et al., 2008) interpreted that the precipitation/dissolution model accurately predicts the leaching of Zn, and willemite (Zn$_2$SiO$_4$) affects the leaching of Zn. The presence of Zn$_2$SiO$_4$ increases Zn solubility in acidic conditions, whereas it reduces solubility in alkaline conditions. Zhang et al. (2008) noted that the leaching of Zn reduces because of the formation of precipitates of zinc carbonate (ZnCO$_3$) for the pH range of 5-8. Most of the Zn is unavailable for leaching, as indicated by low leachability of 10-12%.

Table 1. Maximum (max) and minimum (min) leaching concentration of the contaminant, ratio of maximum to minimum leaching concentration (max/min), total content of the contaminants (TC) and leachability of contaminant (L')

	Plant A					Plant B				
	Leaching concentration, mg/kg		max/ min	TC mg/kg	L', %	Leaching concentration, mg/kg		max/ min	TC, mg/kg	L', %
	min.	Max.				min.	max.			
Cd	0.006	3.1	516.7	4.6	67.8	0.01	4.2	421.0	5.6	75.0
Cr	0.3	16.6	55.3	223.0	7.4	0.5	15.3	30.6	399.1	3.8
Cu	1.9	60.4	31.8	283.2	21.3	3.6	56.9	15.8	340.2	16.7
Mo	0.02	1.2	60.0	1.7	70.6	0.04	1.4	34.0	2.1	64.8
Ni	0.21	9.1	43.4	50.3	18.1	0.14	9.7	69.3	40.6	23.9
Sb	0.24	2.2	9.2	7.6	28.9	0.21	2.4	11.4	9.5	25.3
Pb	0.009	12.1	1343.3	90.6	13.3	0.009	13.4	1492.2	128.2	10.5
Zn	0.04	91.3	2282.5	756.9	12.1	0.05	104.1	2082.0	883.8	11.8
Cl$^-$	2923.1	3937.1	1.3	9867	39.9	3197	4638.5	1.5	10685.7	43.4
SO$_4^{-2}$	2848	6550.0	2.3	15222	43.0	4358	7575.1	1.7	18340.0	41.3

The leaching concentration of chlorides (Cl⁻) and sulfates (SO_4^{-2}) does not vary much with a change in pH, as evident from the maximum to minimum leaching ratio. The leachability of both anions varies between 40-43%. The leaching of Cl⁻ is availability controlled, and almost similar leaching concentration is observed at all the pH values (Meima & Comans, 1999; Quina et al., 2009). The leaching of SO_4^{-2} increases with the decrease in pH from 11 to 7 and then remains almost constant. A similar response was noted in other studies for the leaching of sulfates (Dijkstra et al., 2006; Meima & Comans, 1999; Quina et al., 2009). Sulfate leaching is governed by ettringite for pH above 9, whereas by gypsum for pH below 9 (Dijkstra et al., 2006; Meima & Comans, 1999).

The comparison of leaching test results with EU_IW disposal limits illustrates that IBA is unsuitable for disposal to inert waste landfills as leaching of some of the inorganic constituents (Cr, Cu, Sb, Cl⁻ and SO_4^{-2}) consistently exceed the EU_IW limits, irrespective of the pH condition. The comparison of results with EU_NHW limits demonstrate that IBA is suitable for disposal to non-hazardous waste landfills when the pH of IBA lies above 10. However, magnification in leaching of Sb in the pH range 7 to 10, Cd and Sb in the pH range 5 to 7, while Cd, Cr, Cu, Ni, Pb, Sb, and Zn in the pH range 4-5 is concerning and significant enough to cause IBA to be categorized as hazardous. This suggests that disposal of IBA in non-hazardous waste landfills is favorable for fresh IBA with an intrinsic pH over 10. However, weathering of IBA with the passage of time can cause pH to fall in the range of 8 to 10 (Arickx et al., 2006; Loginova et al., 2019; Van Gerven et al., 2005), which can be concerning as aggravated leaching of Sb may not be favorable for its disposal to non-hazardous waste landfills.

The hazardous nature of IBA has mostly went unnoticed as most of the available studies (Caviglia et al., 2019; del Valle-Zermeño et al., 2013; Gines et al., 2009; Hyks et al., 2011; Lidelöw & Lagerkvist, 2007; Minane et al., 2017; Nikravan et al., 2020) have depicted IBA to be non-hazardous while only a few studies (Arickx et al., 2006; Sormunen & Rantsi, 2015; Tang et al., 2016) have demonstrated its hazardous nature of IBA due to exceedance in leaching of Sb. The reason for the scanty data on the hazardous nature of IBA is due to the fact that most of the studies on IBA have been conducted on fresh samples wherein Sb mostly meets the EU_NHW criteria.

The results further accentuate that it is vital to ensure that pH of IBA does not shift toward the acidic range while disposing the same. Therefore, caution is indispensable when co-disposing IBA with MSW because the generation of acetic acid, fluvic acid, and other organic acids due to the decomposition of waste within the MSW dump (Ahmed & Lan, 2012; Kulikowska & Klimiuk, 2008) may result in lowering of pH to the acidic range, which can aggravate the leaching of contaminants from IBA.

4   CONCLUSIONS

The present study investigated the effect of pH on the leaching of contaminants from incineration bottom ash. The following conclusions can be made based on the present work:

1. The leaching concentration of Cd, Cr, Cu, and Ni increases under acidic pH conditions but is much lower under neutral or alkaline pH conditions. Pb and Zn show amphoteric behavior exhibiting high leaching at low and high pH, whereas low leaching at neutral to moderately high pH. The leaching pattern of Sb resembled that of Mo, with their concentration decreasing under acidic pH, increasing until moderately high pH, and then decreasing again at higher pH conditions.
2. Amongst the metal(loid)s, leaching of Cd, Pb, and Zn is significantly influenced by pH, whereas that of Sb is least affected.
3. The variation in the leaching concentration of chlorides and sulfates with pH is insignificant in comparison to trace metal(loids).
4. The leachability of the majority of contaminants is below 40%, indicating that most trace metal(loids) and anions remain bound in the solid matrix, and hence, are unavailable for leaching throughout their life cycle. Only Cd (pH < 4) and Mo (pH of 7 to 8) show high leachability of around 70% indicating significant leaching.
5. IBA is unsuitable for disposal to inert waste landfills, irrespective of its pH. The alkaline pH (between 10-12) of fresh IBA favors its disposal in non-hazardous waste landfills. However, a reduced pH of IBA (between 8-10) due to weathering reactions can restrict its disposal to non-hazardous waste landfills due to aggravated antimony leaching.

6. It is preferable to dispose IBA in monofils as co-disposal with MSW in sanitary landfills can cause pH of IBA to shift towards the acidic range which may exacerbate the contaminant leaching from IBA.

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