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Chemical association and mobility of trace elements in 13 different fuel incineration bottom ashes

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HIGHLIGHTS

- Bottom ashes from 13 Swedish waste incineration facilities were collected.
- Trace elements contents followed the sequence as Cu > Zn > Pb > Cr > Ni > Sb > As.
- Standard leaching indicates Cr had strongest mobility followed by Pb, Sb and Cu.
- Large amount of Cu is found in organic bound phase in 31% of samples.
- Amounts of unstable trace elements in biofuel bottom ashes are comparatively lower.

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ABSTRACT

The release of trace elements from waste incineration bottom ash is problematic during utilization/landfilling. Thirteen bottom ashes (from various waste fuels and wooden/mixed wooden fuel) were investigated with respect to the total content, leaching behaviour by standard leaching procedure (EN 12457-3), and chemical association of trace elements by sequential extraction. Results showed that the content of trace elements in household/or industrial waste bottom ashes were of high level in comparison to in wooden/mixed wooden fuel/mixed wooden waste ashes. Type of fuel being treated greatly impacts the total inventory of trace elements. On average, trace element content in 13 ashes followed the decreasing order; Cu > Zn > Pb > Cr > Ni > Sb > As. In this study the average total content of Zn, Pb, Cu and Cr was higher in grate bottom ash treating household/industrial waste in comparison to fluidized boilers ash using same waste, however, there were too few data points and variation in data was large. By Standard leaching procedure, an excessive amount (more than disposal limit) of leached Cr, Pb, Sb and Cu (mostly in household/industrial waste ash) was observed in 6, 5, 5 and 4 of the 13 samples, respectively. Correlation coefficients (r) found between total and water leachable contents for Cu, Sb and As were 0.8, 0.7 and 0.6 respectively. Sequential extraction indicated that residual was the major fraction mostly, however, considerable amounts of trace elements had the potential to leach out. A large fraction of arsenic (57% based on average values) in 5 samples (mostly in waste/virgin wood and mixed wooden waste/fuel) and Zn (49% based on average values) in 4 of 13 samples (mostly household/or industrial) were found in the fractions that are easily available (acid soluble and exchangeable). Further, a considerable amount of Cu in 4 samples were found associated with the organic-bound phase. Dissolved organic matter might play an important role in leaching of Cu during utilization/landfilling. Moreover, principal component analysis (PCA) showed that fuel type affects the association of trace elements in bottom ash. Amounts of labile trace elements in wooden/mixed wooden fuel/waste bottom ashes were comparatively lower than other fuel bottom ashes. None of the samples exceeded the limit of disposal with respect to DOC leaching while chlorine in two and sulphate in three samples (household/industrial) exceeded limit. LOI (550 °C) values were higher for bottom ash from grate facilities probably due to no-pre-treatment of the waste fuel. While comparatively low values of LOI (1000 °C) in few samples implies that the oxidation might have outweighed the loss of carbonates.

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1. Introduction

Incineration for waste to energy is a widely developed method for solid waste management in Sweden [1]. In addition to its unique benefits of mass and volume reduction of waste, it can also provide electricity and/or heat. In Sweden 48% of household waste, 40% of industrial waste (excluding mine waste) and 75% of recovered waste wood (construction and demolition) are treated through incineration [2,3].

As a result of thermal treatment of waste, in addition to fly ash and air pollution control residues, a significant amount of bottom ash is generated [4]. The formation of bottom ash generally ranges from 250 to 420 kg/ton of feed waste, without including the grate siftings (5 kg/ton) and boiler ash (2–12 kg/ton), which usually are included [5]. In 2013, a total of 902 760 tons of bottom ash (including slag) was produced in Sweden [1]. Bottom ash can be used in construction (e.g. road base layers and parking lots) or as an alternative to natural gravel, due to its suitable geotechnical properties [5]. In Sweden 60% of the total bottom ash production is utilized as construction material at landfills, 8% in road construction and surfacing applications, while other uses include capping of mine waste and backfilling of underground oil storage facilities [6]. However, from an environmental perspective, leaching potential of trace elements, inorganics such as chlorine and sulphate and dissolved organic carbon compounds in bottom ash are essential to discuss. Trace elements in bottom ash are far less mobile than those in fly ash [5], but since bottom ash forms about 85–90% of the incineration residues [7], the total amounts of potentially toxic trace elements contained in bottom ash are considerable, such as that of Cu and Zn. Large release of trace elements and chlorine has been reported to occur in the 1st decade of bottom ash application or landfilling [8,9] in comparison to the natural gravel application. Such release may pose a risk to surface and ground waters. Therefore, to avoid any adverse environmental impact and to improve the quality of bottom ash, it is crucial to determine the content, mobility and chemical association of trace elements, and also the chemical characteristics of various waste fuel incineration ashes, since it will help to understand the migration behaviour of trace elements.

Many studies have been devoted to the evaluation of chemical and leaching characteristics of bottom ash. Leaching characteristics of Korean and Japanese municipal solid waste incineration (MSWI) bottom ashes were compared by Shim et al. [10]. The leaching of Pb exceeded the regulatory level, and leached amounts of trace elements followed the decreasing order Pb > Cr > Cd for both countries. Zhang et al. [7] reported that leaching of trace elements such as Pb, Cu and Zn from MSWI bottom ash was pH dependent and that dissolution/precipitation reactions were the mobility controlling mechanisms. Yao et al. [11] stated that leaching of Zn from MSWI bottom ash was low in alkaline and neutral conditions, while significant release was observed during acidic conditions. Further, sequential extraction procedures of bottom ash indicated that fractionation of trace elements in different phases also depend upon the input waste fuel [10,11]. Su et al. [12] showed that in MSWI bottom ash, carbonate bound, organic-matter bound and residual fractions were the predominant phases for Cu. Lundholm et al. [13] reported that about 5% Cu, 15% Cr, and 60% As were volatilized during pure CCA-wood incineration. Further, Cu was retained in bottom ash as copper arsenate, copper oxide and double oxide, while Cr forms oxides and As predominantly formed refractory arsenates with calcium, copper, and potassium. In another investigation [14] scanning electron microscopy and energy dispersive X-ray spectroscopy (SEM/EDX) techniques were employed to determine the association of Cr, Cu and As in CCA wood incineration mixed bottom/fly ash. It was observed that Cr existed inside the structure of the matrix particles (some Cr also found in micro wood pieces in ash), while Cu in addition to these places, was also was found condensed on surface of coarser ash particles and As was found associated with Ca in the ash. Similarly, Lassesson and Steenari [15] investigated Cu association in MSWI bottom ash using X-ray absorption spectrometry and X-ray diffraction (XRD/XAS) and reported that it was a mixture of Cu(0), Cu(I), and Cu(II) in the relative concentrations of 10–20% Cu(0), 25–35% Cu(I), and 55–65% Cu(II) and were most likely oxides (Cu_2O, CuO, $CuCr_2O_4$).

MSWI bottom ash has often been the focus of chemical association studies and limited work is reported about association of trace elements in wooden/mixed wooden fuel incineration bottom ash such as virgin wood, recovered waste wood (RWW) or mixed wooden fuels including peat, bark and wood chips. Since these fractions are part of waste fuel being used for heat production in Sweden, determination of chemical association or grouping of trace elements in different fractions of these bottom ashes is also important.

Generally release of trace elements has been reported to be affected by geochemical characteristics such as mineral composition, acid neutralization ability, weathering conditions and intrinsic properties of elements in bottom ash. Several studies have stated that leaching from bottom ash is usually solubility controlled and factors such as, leachate pH, liquid solid ratio (L/S), extractant type and concentration, contact time and solid matrix might affect the leachability [7,16,17].

Bottom ash, in particular, contains a significant amount of unburned organic matter. Total organic carbon (TOC) and dissolved organic carbon (DOC) contents must be kept low for possible reuse purposes. Belevi et al. [18] reported a DOC level of 200–800 mg/l in leachate from MSW bottom ash; while in another study it was reported to be in excess of 2000 mg/l [19]. Dabo et al. [9] observed that leached DOC concentration from bottom ash decreased with time and reported a value of 500 mg/l in the first month and after rapid decrease over time it reached a final mean value of 10 mg/l after 10 years. According to European regulations [20], TOC and DOC must be <5% and <0.5%, respectively, for waste to be acceptable in landfills. DOC may also enhance pollutants mobility from incineration residues when landfilled or used in construction [21]. It has been reported that DOC facilitates the leaching of trace elements through formation of metal-DOC complexes especially for Cu, which makes it very important to evaluate DOC release from residues [7,12,21]. Utilization of bottom ash is also limited by excessive chlorine and sulphate contents. Average chloride content in MSWI bottom ash is more than 10 g/kg dw. Limit values of chlorine and sulphate for bottom ash to be inert are 0.8 g/kg dw and 1 g/kg dw, respectively [22,23]. Further, excessive chlorine causes metal and equipment corrosion so its removal is necessary.

The aim of this study is to discuss the chemical characteristics, leaching behaviour and chemical association of trace elements in 13 Swedish solid waste incineration bottom ashes through the following work.

(a) Determination of total content of trace elements (Zn, Pb, Cu, Cr, As, Sb, Ni), composition and loss on ignition of 13 bottom ashes, resulting from the incineration of various waste materials such as virgin wood, recovered waste wood (RWW), mixed wooden waste (peat, bark, wood chips), household, industrial, and mixed waste.
(b) Standard leaching of bottom ashes at L/S 10 with water according to method pr EN12457-3.
(c) A modified four step sequential extraction method for determination of chemical association of trace elements in bottom ash.