Environmental perspectives of recycling various combustion ashes in cement production – a review

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1 **Environmental perspectives of recycling various combustion ashes in cement production –**
2 **a review**

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Abstract

Recycling different types of ashes for cement production has gained increasing attentions worldwide in a bid to close the waste loop. It minimizes waste landfilling and meanwhile produces useful secondary materials with reduced costs. Ascribed to the presence of elevated metal concentrations, however, it also receives negative inclination for their reuse. Herein, recycling various combustion ashes, such as municipal solid waste incineration fly ashes (MSWI FA), municipal solid waste incineration bottom ashes (MSWI BA), coal fly ashes (CFA), coal bottom ashes (CBA), blast furnace slags (BFS), biomass ashes (BIOA), sewage sludge ashes (SSA) and different co-combustion ashes (CCA), were comprehensively reviewed, from environmental perspectives combined with statistical data analysis (e.g. bulk components, trace metals, leaching potential, and etc.), to quantitatively explore their feasibility during cement production. It was unveiled that pozzolanic contents were predominant which highly fluctuated in their composition based on the ash type, limiting the replacement at maximum of 5-10 wt.%. Considering total metal criteria, heavy metal contents posed challenges as secondary raw materials for blended cements. However, in consideration of metal leaching criteria, exothermic pozzolanic reactions in the second phase of blended cement would sufficiently alleviate their leaching potential, ensuring the environmental feasibility. Apart from the above, treatment costs have to be evaluated in nexus of multiple factors, whereas government policies play significant roles in valorization of recycling ashes. From sustainability perspective, life cycle assessment promises the overall strategy on ash utilization in cement industry.

Keywords: combustion ashes, cement, pozzolanic properties, heavy metals, environment perspectives

1. Introduction

Past decades have seen increasing application of waste material for application in cement, comprised of a variety of industrial byproducts such as fly ashes and bottom ashes, silica fume and blast furnace slags (BFS), gypsum, waste glasses and mine tailings etc. (Garcia-Lodeiro et al., 2016; Mehta, 1998; Schneider et al., 2011; Sobolev et al., 2007). Back in 1937 coal fly ashes (CFA) were first recognized in the US as a valuable source of raw material and/or pozzolan for concrete (Phair, 2006). To date 30% of all fly ash generated is recycled for use as filler or pozzolan in concrete (Scheetz et al., 1999). According to the European Cement Association, up to 5% of primary raw material in clinker can be replaced by mineral ashes contained in the waste derived fuel, saving primary raw materials and avoiding 1.4 mega tons of mineral ashes that otherwise would have been landfilled (de Beer et al., 2016). Among the vast number of studies on cement/concrete with waste material, significant portions of them are relevant to fly ash and slag (Fig. 1). Moving forward with global urbanization and growing industrialization, the infrastructure of solid waste management experiences evolution from open disposal to integral management hierarchy (Tam and Tam, 2006). More waste materials herein investigate on their potential as resources in the cement industry, yet the investigation is rather limited (Fig. 1). These materials are generally pozzolanic due to richness of silicon, iron, aluminum, calcium and/or sulfur in their compounds, which would chemically react with calcium hydroxide in the presence of moisture to form complex possessing cementitious properties (Cheerarot and Jaturapitakkul, 2004; Schneider et al., 2011). Incentives are primarily derived by increasing demands on cement products in countries like China and India, whereby a sum of 1.5 – 2.0 G ton cement consumption per annum is estimated within the next a few decades (Schneider et al., 2011). Also, addition of certain of these materials e.g. magnesium oxide may yield more durable

composite cement (Phair, 2006). All in all, manufacture of cement by replacement of recycled materials helps to close the loop on many waste products with an industrial symbiosis at minimized cost and energy input (Chertow, 2000; Phair, 2006).

Fig. 1

2. Fundamentals of Portland cement

Portland cement is the most common type of cement in general usage because it is the basic ingredient of concrete, mortar, stucco and most non-specialty grout (Baghchesaraei and Baghchesaraei, 2012). It is composed of >90% ground fine power (around 10 micron) from Portland cement clinker and $\leq 5\%$ gypsum as set retarder together with minor constituents in less than 5% for different application control dependent on various standards (Huntzinger and Eatmon, 2009). Its name is originated from Portland stone, a type of building stone quarried on the Isle of Portland in Dorset, England in the early 19th century.

2.1. Cement types

Two major standards are used for classification of cement, called U.S. standard the ASTM C150 and EU standard EN-197. Other than the two, most countries set their own standards for cement production which is generally based on the U.S. and/or EU standard, except specifications on certain items (Juenger et al., 2011), for instance in UK, the level of Cr (VI), which is considered toxic and major skin irritant, may not exceed 2 mg/L (Baghchesaraei and Baghchesaraei, 2012), while in Swiss which permits the secondary materials for cement production under the Swiss guidelines, Cr value may not exceed 150 mg/L in clinker (van der Sloot et al., 2011). ASTM

C150 specification covers eight types of Portland cement, Type I (common usage), Type II (moderate sulfate resistance/moderate heat of hydration), Type III (high early strength), Type IV (low heat of hydration), Type V (high sulfate resistance), together with three air-entraining cements Type IA, Type IIA and Type IIIA for the same uses as Type I, II and III respectively (ASTM C150/C150M-16e1, 2016). EN197 cement types encompass CEM I, II, III, IV and V representing Portland cement, Portland-composite cement, Blast furnace cement, Pozzolan cement and Composite cement respectively covering up to 27 products (CEN, 2000). It is noteworthy that chemistry-based criteria are adopted among the standards which necessitate binder-specific composition in the cements rather than those performance-based (Juenger et al., 2011). Apart from the cements aforementioned, there are several other types which were lately emerged since 20th century, echoing environmental sustainability, such as calcium sulfoaluminate cements, alkali-activated cements and hybrid alkaline cements (Shi et al., 2011).

2.2. Chemistry and performance of Portland cement

Major components in Portland cement are listed in Table 1 (Baghchesaraei and Baghchesaraei, 2012; Chen, 2006). Portland cement contains four cementitious complexes, including tricalcium silicate (C_3S), dicalcium silicate (C_2S), tricalcium aluminate (C_3A) and tetracalcium aluminoferrite (C_4AF) (Wang et al., 2016). The affluence of components in Portland cement ranks as $CaO > SiO_2 > Al_2O_3, Fe_2O_3 > MgO, TiO_2, SO_3, Na_2O, K_2O$, and etc.

Table 1

Mechanisms relevant to pozzolanic properties in Portland cement are extremely complicated due to the presence of many other minerals in the clinker, including some alkali sulfates, free lime,

periclase, and etc. Albeit their small quantity, they may have some significant influences on the hydration process of clinker and the overall properties of concrete made from Portland cement (Chen, 2006). Their detailed influences will be further addressed in the paper where second materials were reviewed for cement production (Chapter 3). Pertaining to fundamentals for Portland cement, interaction during hydration can be simplified with individual chemical reaction with respect to the major component as shown in the following (Wang et al., 2016), where the $(CaO)_x \cdot (SiO_2)_y \cdot (H_2O)_z$ (C-S-H) complex is the key hydration product that contribute to the strength of cementitious products.

Figure 2 indicates a comprehensive kinetics associated with formation of different components alongside the pozzolanic reactions under cement hydration, while the rates of heat and porosity evolutions are also illustrated. Cementitious complexes start to accumulate rapidly after a few hours of cement hydration, and about 70% of C-S-H structures are formed in a week.

Fig. 2

3. Combustion ashes for potential use in cement

3.1. Potential waste materials – types and current status

Potential waste materials used in cement require a well matching quality standard associated with ingredients and composition (ASTM C150/C150M-16e1, 2016; CEN, 2000). The presence

of physicochemical properties similar to conventional raw materials is essential to favor the pozzolanic reaction. For example, the qualified waste material should contain an adequate content of amorphous SiO_2 , Al_2O_3 and Fe_2O_3 to assure its chemical reactivity meanwhile enough small particle sizes to guarantee the sufficiently large reactive surface area for the pozzolanic reaction (Madani Hosseini et al., 2011; Wang et al., 2016). In this regard combustion ashes carry an advantage than most of the other wastes in which their components may already present a limitation.

3.1.1. Fly ashes

In incinerators, the inorganic components of the solid fuel are carried away by the flue gas stream and retained in emission control systems to form materials that are commonly known as fly ashes (Li et al., 2017). Dependent of the feedstock for burning, the types of fly ashes can be further classified into two major streams, CFA and municipal solid waste incineration fly ashes (MSWI FA). CFA accounts for 5-20% of feed coal and is typically accounted for 70-85% up to 90-95 wt.% of the total ash generated (Mikulčić et al., 2016; Yao et al., 2015). Despite of the presence of traceable amount of As, Cr, Cu, Ni and Zn in CFA, CFA leachates exhibited negligible ecotoxicity (Skodras et al., 2006). As such, it remains exempt from regulation in USA as a hazardous waste according to Resource Conservation and Recovery Act (RCRA), 42 USC 6901-6991, and US Environmental Protection Agency (EPA) (Vassilev and Vassileva, 2007). About 20% of the fly ash generated is being used in concrete production, while other uses include road base construction, soil amendment, zeolite synthesis, and use as a filler in polymers (Cho et al., 2005). Despite the existence of these opportunities, a large fraction of the fly ash

produced worldwide is still unused and disposed of as waste or stored in landfill (Mikulčić et al., 2016).

MSWI FA is generally accounted for about 1-5% of the input waste (Andreola et al., 2008; Dou et al., 2017). Distinct from CFA, MSWI FA is normally characterized by a high content of chlorides, sulfates and significant amounts of dangerous substances, such as heavy metals and toxic organic compounds like dioxins and furans that are carcinogenetic to humans (Margallo et al., 2015a), which make it a hazardous waste (Huang et al., 2018). In particular, the amount of major heavy metals i.e. Pb and Zn from MSWI FA were reported as much as 14,000 and 21,000 mg/kg, respectively (Margallo et al., 2015b). In this regard, the stabilization or removal of heavy metals from MSWI FA has been considered as an essential process before its utilization. Furthermore, fly ash is highly reactive and vulnerable for mineralogical alteration when in contact with the environment (Bontempi et al., 2010).

3.1.2. Bottom ashes and Slags

The inorganic components of the solid fuel remain in the combustion chamber are commonly known as bottom ashes (Li et al., 2017). Typical bottom ashes include coal bottom ashes (CBA) and municipal solid waste incineration bottom ashes (MSWI BA). CBA forms when 15-20% of the coal ash produced in the boiler falls to the bottom of the furnace where it is removed. In many countries CBA is reported as nonhazardous, solid or inert wastes and used widely in construction applications albeit the absence of legal certainty. Global production of coal combustion products was approximately 780 M tons in 2011, with effective utilization 415 M tons or 53% of total production (10.6-96.4% effective utilization rate within countries reported)

(Heidrich et al., 2013). For municipal solid wastes, 80-90% of the residues (approximately 30% by mass) produced from the combustion process is bottom ash. In this regard, 440 M tons of MSWI BA was generated worldwide in 2016 (Lynn et al., 2017). The bottom ash from MSWI was a stony material that contained stone, brick, ceramic, glass and unburned organic matter (wood, plastic and fibre), etc. The component of bottom ash was similar to the cement material, the utilization of bottom ash for alternative cement material was a potential method (Li et al., 2016). At present, the application of MSWI BA is especially extensive in EU countries, like UK (up to 45% for blended cement), France (up to 50% for aggregates in concrete) and Portugal (up to 50% for cement clinker) (Lam et al., 2010).

In general, slag is a waste product from the pyrometallurgical processing of various ores in the blast furnace (Piatak et al., 2015). It is estimated that annual world iron slag output in 2014 was on the order of 310 to 370 million tons, and steel slag about 170 to 250 million tons, based on typical ratios of slag to crude iron and steel output (USGS, 2015). The current utilization rate of steel slag is heterogeneous among different countries, close to 100% for countries like US, Japan, Germany and France as opposed to 22% in China. In these developed countries, a half of slag has been applied for road project while the other half for sintering and recycling of iron (Gao et al., 2011). However, there are many other slags from different thermal processes, which carry promising usage in cement industry, road construction, civil engineering work, fertilizer production, and landfill daily cover etc., such as the MSWI FA slag which is further melted from gasification process or vitrification process (Shih et al., 2013), and coal slag which is a by-product from entrained-flow coal gasification (Wei et al., 2014; Wu et al., 2015).

In the paper, twelve types of combustion ashes were reviewed as potential secondary materials for cement production, including MSWI FA, MSWI BA, CFA, CBA, BFS, biomass ashes (BIOA), sewage sludge ashes (SSA) and different co-combustion ashes (CCA) (five types).

3.1.3. BIOA, SSA and CCA

BIOA is the inorganic remains after carbonization of the biomass such as rice husks (Ajiwe et al., 2000; Vassilev et al., 2010), sugar cane bagasse (Frías et al., 2011), empty palm fruit bunch (Tay and Show, 1995), beech wood chips, corncobs, marine microalgae (Vassilev et al., 2010) and such. As a general trend, most trace elements are less enriched in BIOA than in coal ash and the reason for that is mostly associated with lower amount of inorganic matter. (Vassilev et al., 2014). Comparable economical cements could be made from BIOA replacing up to 20% by weight of the Portland cement.

One of the best methods of sewage sludge disposal is incineration since it achieves large reduction of volume, odor minimization and thermal destruction of toxic organic compounds (Fytili and Zabaniotou, 2008; Khiari et al., 2004), leaving approximately 30% of the solid remain as SSA. SSA has been used to manufacture bricks, to incorporate into concrete mixtures and as a fine aggregate in mortar (Fytili and Zabaniotou, 2008). Monzo et al. (2003) reported that SSA behaves as an active material due to the pozzalonic properties, producing and increase of compressive strength compared to control mortar. High sulfur content of SSA ($\%SO_3 > 10$) seems to have little influence on compressive strength of mortars containing SSA (Fytili and Zabaniotou, 2008).

The co-combustion can be of different constituents that will have a direct relationship with the type and quality of CCA produced and its trace element composition. The agricultural and wood fuels co-combustion did not increase the amount of harmful metals in the bottom ash and all the analyzed bottom ashes fulfilled the requirements for the relatively tight Finnish regulations for forest and field fertilizers (Kortelainen et al., 2015).

3.2. Bulk contents of combustion ashes

Bulk contents of various combustion ashes, with reference to cement contents, have been elaborately quantified in many literatures which are summarized in Table 2. The value range of each component was adopted based on 95% confidence interval (95% CI) wherever applicable. Briefly, the bulk contents mainly include CaO, SiO₂, Al₂O₃, Fe₂O₃, SO₃, MgO, Na₂O and K₂O, whereas CaO and SiO₂ are usually the two dominant components, together occupying 50 % by weight or more. There were a few exceptions that Al₂O₃ in coal and volcano ashes shared a higher fraction than CaO, with a range of 18.5-32.6% and 14.3-17.2%, respectively. Organic fraction was described with loss of ignition (LOI), frequently with higher variation than inorganics. Case by case, there exist some other inorganics (Cl and P) and metal oxides in the combustion ashes, which could own a significant portion, shown in Table 2. Ash composition presented statistically high heterogeneities among different types. Also, a wide span of value ranges was noticed for the major components from each type. Ratio of the high-end value over the low-end value (H/L) of the major components was herein calculated indicating their composition stability. It was found that most components in ashes had an H/L ratio less than 3.0-5.0. Such ratios for MSWI FAs with respect to SiO₂ (29.0) and for coal ashes with respect to CaO (5.0 for CFA and 18.7 for CBA), however, were much higher, therefore posing a great

challenge in combination with other raw materials for cement production. To better control the use of different ashes in cement, the maximum allowable mixing ratio (%mixing) of the waste ashes was calculated (Table 2), based on a concept that the caused variation on the distribution of major components shall fall into their typical margins in Portland cement. Components distribution of cement raw materials (exclusion of the complementary ashes) were referenced to their corresponding peak values in Portland cement so as to achieve the %mixing. For most ashes (9 out of 12), CaO was unveiled to be the limiting component of mixing in cement, with the %mixing ranged from 8.8-13.0% for coal slags up to 18.1-23.0% for BFS. Contrastively, salt contents in MSWI BA and BIOA appeared to be the limiting component, permitting 7.0% and 5.5%, respectively. High chloride content leads to cycling, which can rapidly clog the battery of cyclonic heat exchangers and cause a plant shut-down (Ferreira et al., 2003). To make better use of the ashes with high salt content, a complementary step is recommended such as washing process using leachant as the pretreatment. Significant amounts of chloride, sulfate and excess salts can be removed from fly ash by the water washing treatment (Lam et al., 2010). It promotes the formation of a hydrate phase, and convert heavy metals into less reactive forms (Lam et al., 2010). A combined washing-sintering process for treating fly ash was investigated by Mangialardi (2001), satisfying the Italian requirements for normal weight aggregates for use in concretes. Based on current practices in cement production, a fraction range of <5% up to 10-30% of ashes are generally introduced (Hossain, 2003; Shih et al., 2003). High fraction of ashes in the mixture significantly changes the distribution of components in the cement, which may cause crucial variation of the cement performance. A maximum mixing ratio of 5-10%, hereby, seems a proper starting point.

Table 2

3.3. Trace elements of combustion ashes

Another challenge of ash usage in cement is due to the elevated heavy metals, which will raise concerns from not only the increased background trace metals, but also the increased potential for trace metal leaching. Trace metal concentrations of different ashes were hereby listed in Table 3 based on numerous literatures. Overall, 22 elements were monitored, including Ag, As, B, Ba, Be, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sb, Se, Sn, Sr, Ti, Tl, V, and Zn. Again, data range with 95% CI was adopted to indicate the metal concentrations, while the mean value was used to predict the impact of different ashes for cement production. Metal concentrations in cement were used as the benchmarks, whereby the corresponding metal concentrations from ashes at a mixing ratio of 5% were normalized, to yield a mass ratio. Normalized concentrations, if equal to or less than 1, suggests a negligible impact on metal increment. Otherwise, it suggests a significant impact. Amongst the 9 ashes investigated, elements with a significant impact (>1) were noted from 7, except for coal slags and BIOA. However, the number of significant-impact elements, and their respective increased magnitudes as well, were distinct among one another. With a decreasing order, the ranking of the number of significant-impact elements for different ashes is shown as, BFS (13 elements) $>$ MSWI FA (8 elements) $>$ other metal slags (7 elements) $>$ SSA (5 elements) $>$ MSWI BA, and CFA (3 elements) $>$ CCA (1 element). As compared to BFS, MSWI FA and SSA, ashes from coal combustion as a whole imposed limited impacts on the metal accumulation in cement production. By contrast, ashes from MSWI presented various trends dependent of ash collection points, whereby MSWI FA is of higher impacts on metal accumulation as opposed to MSWI BA. Taking account of frequency of appearance in ashes, the significant-impact metals ranked as, Cu (ASTM C150/C150M-16e1) $>$ Cd, Hg and Pb (4) $>$ Se, Sn and Zn (3) $>$ Ba, Co and Sb (2) $>$ As, Be, Cr, Mn, Mo, Ni, Sr

and Ti (ASTM C150/C150M-16e1). Furthermore, several metals from different ashes, could increase the cement background concentrations as high as more than 1 log magnitude, which include Hg (51.25), Sb (12.96) and Sn (15.79) in MSWI FA, Cr (19.35) and Se (39.73) in BFS, Sn (29.05) in SSA, As (17.31), Cd (68.94), Cu (21.75) and Pb (54.73) in other metal slags. Special attentions shall be paid on Cu, Cd, Hg and Pb, not only because of their higher frequency of appearance but also their high accumulation potential and the associated higher toxicity. For existing practices at commercial scales, CFA is predominantly used in cement production (Vargas and Halog, 2015; Wang et al., 2016; Zhuang et al., 2016), whereby metal species of Hg, Se and Sn are of concerns on their accumulation. Yet, the impacts are rather limited given the normalized concentration range at 1.16-2.36 (Table 3). Alternatively, cement production has taken account of steel and other metal slags as the secondary raw material (Piatak et al., 2015; Tsakiridis et al., 2008; Yi et al., 2012), while metal accumulation potential needs special attention during application.

Table 3

3.4. Dioxin of combustion ashes

Apart from trace metals, there exists substantial amounts of persistent organic pollutants in certain ashes like MSWI FA and CCA, such as polycyclic aromatic hydrocarbons (PAHs), methyl sulfates, polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and coplanar polychlorinated biphenyls (PCBs) (Reijnders, 2005). Among these compounds, dioxin is the most concerned one due to its higher toxicity as compared to the others (Shibamoto et al., 2007). One of the major factors influencing dioxin formation from an incinerator is the presence of chlorides in combustion materials (Shibamoto et al., 2007). Dioxin

may be formed during combustion when flammable materials mix with chloride-containing materials like polyvinyl chloride or inorganic chlorides (e.g. NaCl) (Katami et al., 2002; Katami et al., 2004). The scale of dioxin production in industrial combustion processes is linked to type of furnace or kiln, operating conditions and the type and efficiency of the air pollution control devices. The total dioxin (PCDDs/PCDFs) found from MSWI FA may be up to a few dozens of ng/g, with its toxicity equivalents (TEQ) at $>10^2$ ng/kg (Hu et al., 2012). As compared to detected PCDDs/PCDFs in cement (Karstensen, 2008), those of dioxin compounds may be a couple orders of magnitude higher. Yet, dioxins leaching from cement is usually neglected, ascribed to its low mobility in the high alkalinity and rigid environment (Reijnders, 2005; Yin et al., 2018). As for the guidelines and regulations for dioxin associated with cement industry, they are so far concerned with its air emission only (Karstensen, 2008; Pulles et al., 2006).

4. Characteristics of cement with recycled ashes

There are three major concerns associated with the cement produced from recycling combustion ashes, suitability for processing, the geotechnical behavior and the environmental hazard especially based on long-term behavior under various geochemical conditions (Ferreira et al., 2003; Juenger et al., 2011; Li et al., 2016). Recycling combustion ashes would alter the compositions in cement dependent on ash types and mixing ratio, resulting in variation of geotechnical properties during hydration and element leaching potential as well. To elucidate such variation, mechanisms associated with the pozzolanic reactions between ashes and other raw materials in cement should be well understood. Geochemical models based on Portland cement were hereby developed to predict the behavior of hybrid cement.

4.1. The geochemical perspective

Geochemical properties of ordinary cement have been comprehensively studied (Jolicoeur and Simard, 1998; Richardson, 1999; Yajun and Cahyadi, 2004). The major pozzolanic reactions for cement with recycled ashes could be similar to ordinary cement as shown in eq. 1-4. C_3S is usually hydrated much faster than C_2S (Fig. 2), contributing for early and late strength during curing, respectively (Wang et al., 2016). Using secondary materials like recycled ashes for cement production, as a matter, introduces a second phase of pozzolans (SiO_2 , Al_2O_3 and Fe_2O_3 etc.) into the original one being the homogeneous cement, which may confer additional pozzolanic reactions at later stage of cement hydration. Other than that, certain minor contents such as SO_4^{2-} , Cl^- , Na_2O , K_2O and organic compounds, may also participate in the major cement reactions, and interact with the reactions via various processes. The presence of chloride tends to reduce the compressive strength of concrete likely ascribed to the accelerating action of chloride salts on the hydration of C_3S , which caused a high concentration of hydration products in the vicinity of the hydrating cement grains, which retards the subsequent hydration and adversely affects the development of compressive strength (Al Kadhimi et al., 1988). The sulfate ions react with the hydration products of cement, namely C_3A and $Ca(OH)_2$, to produce expansive and /or softening types of deterioration, e.g. expansive ettringite, gypsum, and brucite and sometimes associated with calcite formation (Hossain, 2006). In this regard, recycling ashes for cement production likely alter the microstructure e.g. pore density and the free water regime, and thus affect the cement performance from the geotechnical perspective, such as the curing time and mechanical strength.

4.1.1. Two phase pozzolanic reactions

During Portland cement hydration process (eq. 1-4), the presence of super-saturated Ca^{2+} concentration in the cement slurry leads to crystallization of $\text{Ca}(\text{OH})_2$ which inhibit the directional growth of C-S-H crystal from C-S-H gel formed (Wang et al., 2016). Rapid consumption of $\text{Ca}(\text{OH})_2$ before crystallization appears to be a solution to unleash such inhibition. With introduction of certain recycled ashes in cement, additional pozzolans would possibly transform $\text{Ca}(\text{OH})_2$ into other complexes via mechanisms similar to alkali activation (Fernández-Jiménez and Palomo, 2005), to alleviate $\text{Ca}(\text{OH})_2$ crystallization meanwhile to enhance concrete strength with the formed additional C-S-H complexes (Fraay et al., 1989). Firstly, dissolved OH^- will attach the ash surface to form a thin film around ash particles, followed by mineral dissolutions like SiO_2 , Al_2O_3 and Fe_2O_3 whereas reactions take place as below (Shi and Day, 2000),

It is worth noting that the above mentioned reactions generally occurred at a later stage, only after the chemical reactions (eq. 1-4) for the primary phase of cement hydration. Two reasons are responsible for the delay of second phase (i.e. recycled ashes) reaction. Firstly, congenital differences on bulk contents, surface charge, morphology, and cementitious properties like phase angle (Husain et al., 2017) between the two phases, prioritize reactions beginning with the primary phase of cement, ascribed to their higher purity, homogeneity and reactivity. Second, it takes time (only after a certain period of reactions in the primary phase) for accumulation of Ca^{2+} and OH^- concentrations, to facilitate the surficial hydration and mineral dissolution of pozzolans

in the second phase and then initiate the pozzolanic reactions. Figure 3 indicates the step-wise reactions associated with the primary and second phase in the hybrid cement with combustion ashes. It is also worth noting that, although the pozzolanic reactions associated with the second phase assist the rapid formation of C-S-H backbone structure and enhance the cement strength by additional C-S-H complexes, they decrease the ionic diffusion owing to the diminution of capillary porosity and thus retard the hydration (Wang et al., 2016).

4.1.2. Side effects from minor contents

Aside from the main pozzolans conferred by ashes, there are many other impurities dependent on different ash types, accounting for a minor fraction though while making the hydration processes more complicated. ZnO and PbO may form some hydrated compounds, which would retard the hydration reaction (Saikia et al., 2008). Naamane et al. (2016) has reported that P_2O_5 and SO_3 in SSA increase water demand and setting time compared to the control mortar, however, the compressive strengths and the degree of hydration increase with time and become superior to the control mortar at 90 days, for a replacement rate of 15%. But, a high chloride content in cement often downgrades cement quality, makes the concrete porous and decrease its strength, expedite the leaching of heavy metals from reinforced concrete to the surroundings, cause problems during the kiln operation, and accelerate the corrosion of embedded steel supports (Saikia et al., 2007; Wang et al., 2010; Wei and Cheng, 2016). First of all, chloride may cause expansion of concrete at early ages (up to 4 weeks) due to reaction with hydrated C_3A forming chloroaluminate hydrate ($Ca_4Al_2Cl_2(OH)_{12} \cdot 4H_2O$), while cause higher drying shrinkage due either to a higher surface tension of water and more probably the evaporation of water from concrete by capillary action, or to set-acceleration of the main cement compounds which form a

fine gel with a high surface tension; secondly, in the presence of chloride salts, the rate of decrease of compressive strength will be higher due to the reactions which took place between the C_3A and chloride salts, forming complex compounds of calcium chloroaluminate (Al Kadhimi et al., 1988; Loser et al., 2010). The alkali metal oxides, which have high concentrations in the ashes compared to the cement, may stimulate the formation of expansive products due to alkali-silica reactions (Saikia et al., 2008).

Fig. 3

4.2. The environmental perspective

Cement hydration processes are complicated due to the simultaneous formation of solid, liquid and porous matrix (Husain et al., 2017), whereas the formation of C-S-H gel, C-A-H gel, and C-A-F-H gel are subjected to competitive crystallization determined by their respective isotherms and kinetics. Besides, byproducts based on cementitious reactions and newly evolved different hydrates may further complicate the crystallization format, such alumina and magnesium related crystalline phases (Husain et al., 2017; Kupwade-Patil et al., 2016). An immediate impact induced by crystallization is the hardness/strength development in the cement mortar, during which trace elements will be either directly involved in various chemical processes to be chemically bounded, such as precipitations of calcium metallate (As, Mo, V) and metal hydroxides (Cd, Ni), as well as incorporation in hydration products (As, Cr, Sb, V) (Saikia et al., 2008), or physically immobilized along with the growing rigidity via solidification (Billen et al., 2015; Cerbo et al., 2016). Since recycling combustion ashes tends to decrease the capillary porosity in cement which becomes denser (Hossain and Lachemi, 2007; Wang et al., 2016), leaching of the elements from the mortar could be further decreased.

4.2.1. Trace elements leaching potential

Leaching results from raw combustion ashes and those from stabilized combustion ashes using cement are reviewed and listed in Table 4, so as to understand the stabilization effects of cement on trace metals with respect to various ashes. Note that leaching methods adopted among the investigated results are different, including TCLP method by Cerbo et al. (2016), Yang et al. (2009) and Shih et al. (2013), leaching test at $L/S = 10$ under natural pHs by Kosson et al. (2014) and Saikia et al. (2008), and the ANC method by Mofarrah & Husain (2013). We herein only carried out comparison of data before and after ash stabilization to distinguish the effects by cement stabilization, rather than cross-comparison between each other. Comparison of data was further made in Fig. 4 between stabilized ashes and raw ashes to unveil their trends of metal leaching.

Leached concentrations before cement stabilization were highly different cross metals and ashes, as a function of their primary concentration and distribution, their stability constants associated with formed chemical complexes and the subjected geochemical conditions as well. For instance, the leached amounts were found at 468.2 mg/kg for Pb in MSWI FAs as opposed to 0.002 mg/kg in CFAs. For the same type of ashes, the leached amount among metals could vary significantly up to several log magnitudes, like those in heavy oil fly ashes (HOFA). After cement stabilization, the leaching potential of most metals were substantially reduced as observed from all the investigated combustion ashes (Fig. 4). A few metals such as Cr, Cu, Pb and Zn from MSWI FA/CFA/Pb-slugs, however, showed a higher release from stabilized ashes than ashes alone based on certain literatures, likely attributed to the dissolution of hydroxides of these amphoteric metals at high pH within the mortar (Saikia et al., 2008). Yet, it is also possible that,

there would be other ingredients being introduced such as chloride, sulfate and organic fractions, involved in pozzolanic reactions together with Cr, Cu, Pb and Zn during cement hydration, which would be responsible for their enhanced mobility.

Table 4

Fig. 4

Concentrations of the leached metals, such as Ba, Cd, Cr, Pb, Sb, Se, V and Zn (Ag and Hg were both undetectable), from stabilized ashes were further compared to those from cement controls as shown in Fig. 5. Metals such as Co, Mn and Ni were noted with an interestingly higher release in cement controls suggesting the existence of the metals in more soluble phases other than hydroxide (Kogbara et al., 2014) and further adding a margin of uncertainties from cement raw materials. On the other hand, B, Cu and Mo presented an overall higher release from stabilized ashes, likely relevant to their high affluence from their primary sources, and formation of low stability complexes due to introduction of chloride and sulfate from the ashes (Al-Amoudi et al., 1995). Certain leached elements only appeared in stabilized ashes, like B and Tl, owing to their significant elevation introduced by ashes.

Fig. 5

Indeed, most metals present in stabilized ashes are less leachable, except Cr, Cu, Pb and Zn exhibiting enhanced release (>1 mg/kg). Yet, the metal leaching potential is highly variable associated with different ash types, where fly ashes appear to carry a higher trend of leaching as compared to the corresponding bottom ashes or slags. In addition, findings as aforementioned were primarily based on lab scale investigations, in this regard the long-term monitoring with in

situ testing/measurements are critical to achieve the first hand information to accurately assess the real leaching status quo of the end products after ash applications.

4.3. Life cycle assessments

Long-term environmental monitoring on large scale applications of recycling different ashes in cement production is rather limited. LCA is a holistic methodology allowing for the analysis of systems and products, and providing an alternative insightful view of environmental impacts and critical aspects into the benefits associated with the ash utilization for cement production under hypothetical boundary conditions (Allegrini et al., 2015). Via comprehensive integration of different conditions, the impacts of ash utilization with cement production could be studied for long term.

A direct anticipated benefit out of ash recycling, based on LCA study, is the reduction of raw materials when replacing Portland cement with ashes (Babbitt and Lindner, 2008). Benefits also include better technical and environmental performance such as reductions of emissions to water (particularly total dissolved solid), and emissions of metals to land (Babbitt and Lindner, 2008; Huntzinger and Eatmon, 2009; Mendes Moraes et al., 2010). However, the major benefit lies in the greater credit related to the avoided impacts, such as the prevention or reduction of greenhouse gas and landfilling (Turk et al., 2015). Portland cement was found to be the primary source of CO₂ emissions generated by typical commercially produced concrete mixes, being responsible for 74% to 81% of total CO₂ emissions (Flower and Sanjayan, 2007). Blended cement with ash recycling has been widely assessed with life cycle greenhouse gas emissions (García-Segura et al., 2014; Mikulčić et al., 2016). For instance, the use of alkali-activated binder

instead of ground granulated blast-furnace slag cement in concrete or in ordinary Portland cement based concrete could achieve up to 55-75% fewer emissions (Kajaste and Hurme, 2016), without accounting of life factor. Study by García-Segura et al. compared respective material emission and overall material reductions of CFA and BFS when replacing Portland cement. CFA replacement led to a lower material emission factor (4 kgCO₂/t from CFA vs. 52 kgCO₂/t from BFS) due to less processing requirement , but less reductions were achieved overall due to less amount of cement replaced as well as their durability differences (7% from CFA vs. 20% from BFS) (García-Segura et al., 2014). A more recent study using LCA by Hossain et al. (Hossain et al., 2017) indicated that about 12% of the total greenhouse gases emission can be reduced from the cement industry in Hong Kong by using waste materials to replace virgin materials (clinker). Despite many good indications from LCA studies on incorporation of ashes for cement production or replacement, assessment of the fate and transport of metals in the blended cement was still little addressed or sometimes excluded in their scenarios (Huntzinger and Eatmon, 2009). There is a need, hereby, for them to be complemented by experimental studies targeting at strength and durability check after substitution (Teixeira et al., 2016).

4.4. Regulations

Nowadays, blended cement has been widely used in EU, US, China and many other countries/regions, since regulations associated with cement production and the use of cement allow the incorporation of different types of additions directly from the factory (González-Fonteboa et al., 2016; Mukherjee et al., 2008), such as fly ash, iron and steel slag and recycled road materials. These regulations covers different legal areas with different legal regulations/criteria (Achternbosch et al., 2003), which are roughly classified into three categories

based on original incorporated sources, industrial processes and production, and downstream applications. A detailed list of these regulations encompasses building legislation, regulation on emission control in cement plants, guideline assessment of the effects of construction products on soil and groundwater, and relevant parts of the hazardous wastes legislation and waste management legislation etc., most of which concern environmental perspective much more than the geotechnical aspects (Hinojosa et al., 2014). As the key limiting chemical factors, heavy metal limit values for alternative secondary raw materials in Portland cement clinker manufacturing plants and cements from cement manufacturing plants using alternative secondary raw materials, therefore, have been thoroughly regulated (M. Achternbosch et al., 2003), in favor of their benign application. It is noteworthy however, guidelines on metal availability rather than metal capacity are increasingly adopted in context of solid wastes management meanwhile are extended to secondary material applications (Liu et al., 2015). In this regard, there is a margin for blended cement legislation moving forward to more feasible practices i.e. via the establishment of its leaching criteria. Furthermore, for some countries/regions like Australia, no regulations so far cater for the promotion of the alternative materials (van Beers et al., 2009), which present the regional imbalance in the field.

4.5. Future trends for blended cement production

Recycling combustion ashes for cement production is one of the important strategies that encompass the whole waste management hierarchy, echoing the urgent global desire to reduce, reuse and recycle waste materials. There are mixed factors to push forward the utilization of combustion ashes on one end, while on the other end dragged by slow adoption in cement industry. Generally, the drivers and barriers for ash reuses appear to fall in nine broad categories:

regulation, economics, community, technology, transportation, confidential and commercial issues, risk and liabilities, industry focus and priorities, and region specific issues (van Beers et al., 2009). From regulation perspective, more specific regulations on secondary materials have been released from different sectors in favor of the ash reuse. Meanwhile, environmental criteria become more stringent bringing new issues related to risk and liabilities. Due to unbalanced development in economies and regional specific issues, the ash utilization carries with other derivative burdens from community, technology, transportation, commercial feasibility and etc. It is quite clear that the future for recycling ashes for cement production is promising while with a lot of challenges. A good management of the ashes needs synergy of a group of elements aforementioned, whereas the general pathway for ash utilization could be perceived (Fig. 6). Two major impedances for ash application lie in environmental and technical concerns, both of which are dependent on the respective technology development. Yet, it is not necessarily the key limit for ash recycling, if the government and industry play a more positive role.

Fig. 6

5. Conclusive remarks

There is a critical point (5-10% by weight) of recycling combustion ashes for blended cement production, more than which it may cause significant changes on the cement composition. However, further considering the trace element accumulation in the blended cement, 5% mixing ratio of certain ashes only, e.g. MSWI FAs and steel slags, would significantly increase the overall trace metals from their original concentrations inside cement. LCA studies unveil that recycling ashes in general provides environmental sustainability due to diversion of ash from landfilling and reduced greenhouse gas release. Yet, environmental concerns are still high due to

563 the leaching potential of Ni, Cu, Pb or Zn from certain stabilized fly ashes using cement. To
564 promote different ash utilization in cement industry, further researches shall be concentrated in
565 technology development to enhance performance and environmental control. At the same time, it
566 is always true to look at recycling ash as business at the nexus of costs and benefits, in order to
567 achieve the final success in closing their respective loops.

568 **Tables and Figures**

569 Table 1. Major components in Portland cement clinker (Baghchesaraei and Baghchesaraei, 2012;
570 Chen, 2006).

571 Table 2. Different combustion ashes by ingredient for potential used in/with cement (Chandara et
572 al., 2009; Schneider et al., 2011).

573 Table 3. Trace elements content in different combustion ashes.

574 Table 4. Comparison of leaching on trace elements among common cement pastes, combustion
575 ashes and cement mortar mixed with combustion ashes.

576 Fig. 1. Annual number of publications on cement production with waste material (CWM) and
577 cement with fly ash, slag/bottom ash, glass, gypsum, and mining waste respectively. Data were
578 obtained from Scopus on 11th Mar 2018. Keywords for searching are “cement production AND
579 waste material”, “cement production AND waste material AND fly ash”, “cement production
580 AND waste material AND slag OR bottom ash”, “cement production AND waste material AND
581 glass” and “cement production AND waste material AND gypsum”, and “cement production
582 AND waste material AND mining waste” respectively.

583 Fig. 2. Combined plots of hydration degree of clinker phases, formation of hydration products
584 and rate of heat evolution in Portland cement paste as a function of hydration time, after Odler
585 (Odler, 1998), Mailvaganam and Rixom (Mailvaganam and Rixom, 2002) and Bullard et al
586 (Bullard et al., 2011).

587 Fig. 3. Schematic illustration of the predicted effects on the pozzolanic reactions in the second
588 phase during cement hydration at later stage. Fig. 4. Comparison of literature-reviewed trace
589 metals leached amount by different ashes and stabilized ashes using cement (data based on Table
590 4).

591 Fig. 4. Comparison of literature-reviewed trace metals leached amount by different ashes and
592 stabilized ashes using cement (data based on Table 4).

593 Fig. 5. Comparison of literature-reviewed trace metals leached amount by commercial cements
594 and stabilized ashes using cement (data based on Table 4).

595 Fig. 6. A flow chart for strategy of recycling combustion ashes in cement industry (LCA: Life
596 Cycle Assessment; CBA: Cost and Benefit Analysis).

597 Table 1. Major components in Portland cement clinker (Baghchesaraei and Baghchesaraei, 2012; Chen,
598 2006).

Component	¹ Fraction (% w/w)	² Fraction (% w/w)
Tricalcium silicate (C ₃ S)	50 – 70	45 – 75
Dicalcium silicate (C ₂ S)	15 – 30	7 – 32
Tricalcium Aluminate (C ₃ A)	5 – 10	0 – 13
Tetracalcium aluminoferrite (C ₄ AF)	5 – 15	0 – 18
Magnesium oxide (MgO)	<6	
Gypsum (CaSO ₄)		2 – 10
¹ : fraction based on Chen (2006); ² : fraction based on Baghchesaraei & Baghchesaraei (2012)		

599

Table 2. Different combustion ashes by ingredient for potential used in/with cement (Chandara et al., 2009; Schneider et al., 2011).

Types		Major components (%)										Others	References
		CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	SO ₃	MgO	Na ₂ O	K ₂ O	LOI			
Portland cement		62.0-68.0	19.0-23.0	3.5-6.5	1.2-4.0	2.10-3.44	1.0-4.5	0.28-1.12	0.27-1.06	0.9 - 1.67	TiO ₂ : 0.24; MnO: 0.03-1.11; SrO: 0.07	(Aranda Usón et al., 2013; Labbaci et al., 2017; Matos and Sousa-Coutinho, 2012; Popovics, 1993; Seddik Meddah, 2015; Wild et al., 1998)	
Waste materials													
MSWI FA		14.29-42.53	1.31-37.95	2.38-8.52	0.80-4.93	3.60-12.10	1.13-3.62	2.54-12.22	2.71- 7.30		P ₂ O ₅ : 0.26-4.13; TiO ₂ : 0.56-1.36; ZnO: 0.37-1.19; PbO: 0.12-0.55; SnO ₂ : 0.44; MnO: 0.27; Cr ₂ O ₃ : 0.15; CuO: 0.14-0.39; Cl: 2.35-18.8	(Garcia-Lodeiro et al., 2016; Jin et al., 2016; Saikia et al., 2007; Saikia et al., 2015; Shi and Kan, 2009; Wan et al., 2006; Wang et al., 2010)	
	H/L ratio	3.0	29.0	3.6	6.2	3.4	3.2	4.8	2.7	-			
	% mixing	11.2-23.6%	18.4-21.1%	59.8-72.8%	75.1-87.5%	13.4-89.3%	100.0%	7.0-37.2%	11.2-32.4%				
BA		11.07-27.82	31.00-49.23	8.06-16.48	5.07-12.60	0.30-5.21	1.43-2.93	2.79- 5.76	0.72-1.54	1.48-14.52			
	H/L ratio	2.5	1.6	2.0	2.5	17.4	2.0	2.1	2.1	9.8			
	% mixing	10.5-14.9%	13.2-33.3%	23.1-65.8%	24.6-72.4%	42.7-43.1%	100%	15.3-100%	62.2-100%	5.7-100%			
FA		4.14-20.85	38.16-54.77	18.51-32.61	4.45-8.63	0.21-1.01	1.35-2.90	0.11-0.75	0.35-2.56	0-10.9			
	H/L ratio	5.0	1.4	1.8	1.9	4.8	2.1	6.8	7.3	-			
	% mixing	9.4-12.7%	11.2-20.9%	10.3-20.0%	37.7-86.2%	30.1-41.5%	100%	83.2-100%	34.5-100%	7.7-46.1%			
BA/Slag		1.16-21.71	37.37-57.56	17.74-28.01	4.66-15.58	0-7.55	0.60-3.59	0.04-0.75	0.75-1.91	1.03-4.14			
	H/L ratio	18.7	1.5	1.6	3.3	-	6.0	18.8	2.5	4.0			
	% mixing	8.8-13.0%	10.4-21.8%	12.2-21.1%	19.5-80.9%	24.6-39.0%	89.7-100%	77.8-100%	48.2-100%	23.8-100%			
Blast furnace slag		34.90-41.86	35.11-38.35	1.44-3.27	0.38-1.40	0.02-1.39	4.21-10.64	0.16-0.49	0.50-1.31				
	H/L ratio	1.2	1.1	2.3	3.7	69.5	2.5	3.1	2.6	-			
	% mixing	18.1-23.0%	20.7-24.8%	59.3-92.9%	77.3-100%	27.7-39.2%	36.3-100%	87.5-100%	76.0-100%				
Other ashes		11.64-22.25	37.26-53.44	2.83-5.94	1.67-4.14	0.82-1.92	2.86-4.14	0.52-1.62	7.38-14.73	1.98-12.94			
	H/L ratio	1.9	1.4	2.1	2.5	2.3	1.4	3.1	2.0	6.5			
	% mixing	10.6-13.1%	11.6-21.9%	81.7-100%	95.2-100%	51.1-88.2%	100%	62.7-100%	5.5-11.1%	6.4-71.3%			
Sewage sludge		18.68-37.40	12.42-26.32	2.43-14.19	1.87-6.61	3.16-6.36	2.81-4.22	0.73-2.81	0.04-1.0	1.95-36.04			
	H/L ratio	2.0	2.1	5.8	3.5	2.0	1.5	3.8	-	18.5			
	% mixing	12.2-19.6%	37.8-54.6%	28.1-73.7%	51.8-100%	31.5-100%	100%	33.2-100%	20.6-74.5%	2.2-73.3%			
Volcano		3.71- 8.71	45.26-59.02	14.27-17.22	7.74-11.41	0.14-0.71	2.69-5.25	2.39-4.18	0.92-3.32	1.10-4.95			
	H/L ratio	2.3	1.3	1.2	1.5	5.1	2.0	1.7	3.6	4.5			
	% mixing	9.3-10.1%	10.0-15.2%	21.9-27.9%	27.4-42.8%	39.0-49.1%	82.4-100%	21.5-39.8%	25.9-100%	19.0-100%			
SS + rice husk		3.94-4.52	66.23-66.80	11.58-13.30	5.57-6.37	0.88-1.02	2.00-2.31	0.71-0.78	3.99-4.26				
	H/L ratio	1.1	1.0	1.1	1.1	1.2	1.2	1.1	1.2	-			
	% mixing	9.4-9.5%	8.4-8.5%	30.6-37.1%	54.2-64.1%	52.3-55.4%	100.0%	100.0%	19.8-21.2%				
Coal + RDF		4.86	48.48	28.48	3.42	0.56	1.69	1.18	0.86	6.5			
	H/L ratio	-	-	-	-	-	-	-	-	-			
	% mixing	9.5%	13.6%	12.0%	100.0%	46.5%	100.0%	93.3%	100.0%	13.8%			
Coal + wood		13.6-14.5	43.6-45.2	21.0-21.5	3.9-4.0	0.26-0.51	2.50-2.91	7.30-7.46	0.70-1.17	0.4-0.9			
	H/L ratio	1.1	1.0	1.0	1	2.0	1.2	1.0	1.7	2.2			
	% mixing	11.0-11.2%	15.3-16.3%	16.7-17.1%	100.0%	42.1-45.7%	100.0%	11.7-12.0%	87.8-100%	60.6-100%			
Coal +boimass		4.9-20.0	25.5-48.0	7.4-25.2	1.6-8.2	0.8-1.4	0.7-3.2	2.7-3.4	2.6-3.0	4.3-8.3			
	H/L ratio	4.1	1.9	3.4	5.1	1.8	4.6	1.3	1.2	1.9			
	% mixing	9.5-12.5%	13.8-61.5%	13.8-76.9%	40.0-100%	50.8-65.7%	92.1-100%	26.9-34.7%	28.9-33.9%	10.4-22.6%			
*: the value ranges are based on 95% confidence interval (95% CI) otherwise minimum to maximum and/or individuals from literatures.													
*: Biomass includes rice husk, palm oil fuel, sugar cane bagasse, wood, corn cob, switchgrass, wheat straw, olive residue, sawdust, RDF, sunflower shell, cotton stalks, poplar, pine, cotton gin trash, olive kernel and etc.													
*: loss of ignition.													

* : the value ranges are based on 95% confidence interval (95% Cl) otherwise minimum to maximum and/or individuals from literatures.

&. Biomass includes rice husk, palm oil fuel, sugar cane bagasse, wood, corn cob, switchgrass, wheat straw, olive residue, sawdust, RDF, sunflower shell, cotton stalks, popular, pine, cotton gin trash, olive kernel and etc.

: loss of ignition.

Table 3. Trace elements content in different combustion ashes.

Trace metal	MSWI FA (Colangelo et al., 2015; Lee et al., 2011; Lindberg et al., 2015; Parés Viader et al., 2017; Ribbing, 2007; Saikia et al., 2008; Saikia et al., 2007; Shih et al., 2013; Wang et al., 2010; Wu et al., 2012)					MSWI BA (Biganzoli et al., 2012; Haha et al., 2011; Lindberg et al., 2015; Ribbing, 2007; Saikia et al., 2008)					CFA (Kermer et al.; Lederer et al.; Medina et al., 2010; Müllauer et al., 2015; Pandey et al., 2011; Świetlik et al., 2016)					CBA (Font et al., 2010; Kara et al., 2014; Matzenbacher et al., 2017; Pandey et al., 2011; Turner and Lowry, 1983; Vamvuka and Kakaras, 2011; Wei et al., 2014; Wu et al., 2015)				
	N	Mean	StD	95% CI	5% mix	N	Mean	StDev	95% CI	5% mix	N	Mean	StDev	95% CI	5% mix	N	Mean	StDev	95% CI	5% mix
Ag	2	13.31	9.32	0-97.04	-	3	70.7	105.5	0-332.7	0.27	2	18.6	25.9	0-251.2	0.07	-	-	-	-	-
As	25	32.36	46.08	13.34-51.37	-	8	219	313	0-481	0.34	12	23.8	56.7	0-59.9	0.04	15	198	424	0-432	0.31
B	-	-	-	-	-	1	120	-	-	-	6	42.52	6.16	36.05-48.99	-	7	236.1	227	26.2-446.1	-
Ba	7	416	472	0-853	-	4	377.5	6822	0-14630	0.45	2	1700	1838	0-18218	0.20	9	4850	11719	0-13858	0.58
Be	2	1.68	1.93	0-19.03	-	-	-	-	-	-	-	-	-	-	-	5	14	12.27	0-29.23	0.42
Cd	5	1.80	2.47	0-4.86	-	15	257	456	5-510	7.14	13	8.10	19.25	0-19.73	0.23	21	3.11	3.32	1.60-4.62	0.09
Co	23	17.97	17.76	10.30-25.65	-	3	102.1	171.4	0-527.9	0.28	8	69.1	113.9	0-164.3	0.19	12	45.2	40	19.8-70.6	0.13
Cr	29	146.8	138.0	94.3-199.3	-	15	272.5	316.1	97.4-447.6	0.09	13	420	883	0-954	0.14	12	172.2	102.3	107.2-237.2	0.06
Cu	23	71.2	49.3	49.9-92.6	-	13	159.5	1649	599-2592	1.12	12	1997	3259	0-4068	1.40	12	190.5	217.2	52.6-328.5	0.13
Hg	2	0.02	0.027	0-0.26	-	4	20.5	24.6	0-59.7	51.25	-	-	-	-	-	8	0.50	0.59	0.007-0.996	1.25
Mn	22	778	993	338-1218	-	7	766	617	196-1337	0.05	10	476	731	0-999	0.03	-	-	-	-	-
Mo	5	3.78	3.51	0-8.14	-	6	36.70	16.10	19.8-53.6	0.49	3	102.7	150.7	0-477.1	1.36	9	19.78	24.13	1.23-38.32	0.26
Ni	25	60.12	37.49	44.65-75.59	-	9	165.4	220.8	0-335.1	0.14	13	454	1170	0-1160	0.38	12	160.0	162.8	56.6-263.4	0.13
Pb	29	52.2	107.1	11.5-93.0	-	13	4784	7268	392-9175	4.58	12	1683	4037	0-4248	1.61	21	260.6	320.0	115.0-406.3	0.25
Sb	9	4.66	5.63	0.32-8.99	-	1	1208	-	-	12.96	-	-	-	-	-	9	65.8	109.4	0-149.8	0.71
Se	3	1.12	0.97	0-3.52	-	4	47.0	56.9	0-137.6	2.10	4	2.57	4.96	0-10.45	0.11	9	25.9	51.7	0-65.7	1.16
Sn	2	7.50	9.19	0-90.09	-	3	2369	3067	0-9988	15.79	8	50.2	133.3	0-161.6	0.33	8	354	685	0-926	2.36
Sr	1	218.0	-	-	-	2	290	297	0-2958	0.07	2	543	647	0-6356	0.12	8	1288	1398	118-2457	0.30
Ti	20	2211	2025	1263-3159	-	2	6350	7990	0-78140	0.14	8	8534	2414	6515-10552	0.19	1	4.8	-	-	0.00
Tl	4	1.70	2.03	0-4.94	-	-	-	-	-	-	-	-	-	-	-	9	6.84	5.11	2.92-10.77	0.20
V	27	103.8	69.1	76.5-131.1	-	5	97.5	102.3	0-224.5	0.05	9	127.0	86.1	105.8-238.2	0.06	10	364	363	104-624	0.18
Zn	23	291	517	68-515	-	12	16906	22631	2527-31285	2.90	12	1711	2683	6-3416	0.29	12	360.9	343.8	142.5-579.3	0.06
Trace metal	BFS (Bodor et al., 2016; Lederer et al.; Müllauer et al., 2015; Piatak et al., 2015; Piatak and Seal II, 2012)	MSWI FA (Morais, 2009; Morais et al., 2006; Takaoka et al., 2008; Wolski et al., 2004)	SSA (Herzel et al., 2016; Krüger et al., 2014; Morais, 2009; Morais et al., 2006; Takaoka et al., 2008; Wolski et al., 2004)	BIOA (Ellied et al., 2007; Kortelainen et al., 2015; Vassilev et al., 2015)	OMS (Piatak et al., 2015)	CCA (Grammelis et al., 2006; Klika, 2012; Nurmestien, 2005; Pettersson et al., 2008; Wang et al., 2017b; Zhou et al., 2017)														
Ag	-	-	-	-	0.25	10	2.20	1.28	1.29-3.12	0.01	-	-	-	-	-	-	-	-	-	-
As	15	24.8	63.3	0-59.8	0.77	8	31.5	38.1	0-63.4	0.05	13	4.30	3.24	2.34-6.26	0.01	8	11200	26390	0-33263	17.31
B	-	-	-	-	-	10	6407	3706	3756-9058	-	-	-	-	-	-	-	-	-	-	-
Ba	13	976	672	570-1382	2.35	4	4237	6313	0-14282	0.51	14	269	485	0-549	0.03	8	28028	66190	0-83365	3.37
Be	6	6.32	2.49	3.70-8.93	3.76	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Cd	13	10.22	35.4	0-31.61	5.68	2	41.2	55.3	0-538	-	10	18.59	11.98	10.02-27.16	0.55	-	-	-	-	-
Co	15	21.8	53.3	0-51.4	1.21	4	36.5	32.8	0-88.7	0.10	20	8.64	10.35	3.80-13.49	0.02	12	2324	6877	0-6694	6.47
Cr	16	2841	8324	0-7276	19.359	2	1675	2688	0-3741	0.57	20	315.8	369	143.0-488.5	0.11	10	2255	4545	0-5506	0.77
Cu	15	102.3	159.5	14.0-190.7	1.44	11	2564	3013	540-4588	1.80	23	314.7	315.6	178.2-451.2	0.22	12	30968	101630	0-95540	21.75
Hg	2	0	0	-	0.00	3	1.6	1.80	0-6.08	4.00	-	-	-	-	-	-	-	-	-	-
Mn	7	7120	9379	0-15794	9.15	4	2076	2971	0-6803	0.13	23	3070	3919	1375-4764	0.20	-	-	-	-	-
Mo	7	1.51	3.05	0-4.33	0.40	2	59.8	73.9	0-723.6	0.79	10	6.63	5.44	2.75-10.52	0.09	-	-	-	-	-
Ni	16	258	804	0-687	4.29	6	344	326	2-686	0.29	20	179.1	206.4	82.5-275.7	0.15	12	1157	2091	0-2486	0.96
Pb	15	98.3	270.2	0-248.0	1.88	5	387	424	0-913	0.37	13	16.36	19.58	4.53-28.19	0.02	10	57143	108994	0-135112	54.73
Sb	7	4.33	11.19	0-14.68	0.93	2	228	320	0-3103	2.45	10	1.16	0.64	0.70-1.62	0.01	-	-	-	-	-
Se	1	44.50	-	-	39.735	-	4.98	2.43	1.97-7.99	0.22	10	17.36	13.03	8.04-26.68	0.78	-	-	-	-	-
Sn	6	0.167	0.27	0-0.45	0.02	2	4357	6151	0-59620	29.05	10	3.90	1.76	2.64-5.16	0.03	-	-	-	-	-
Sr	7	316.1	114.1	210.6-421.7	1.45	2	324.5	3.54	292.7-356.3	0.07	10	412	387	135-688	0.09	-	-	-	-	-
Ti	7	5406	1597	3929-6883	2.45	6	7150	6447	384-1391.6	0.16	14	371.3	352.4	167.8-574.8	0.01	-	-	-	-	-
Tl	6	0.033	0.082	0-0.12	0.02	1	0.63	-	-	0.02	2	0.50	0.70	0-6.824	0.01	-	-	-	-	-
V	1	69.0	-	-	0.66	4	451	351	0-1567	0.22	14	21.26	16.76	11.59-30.94	0.01	-	-	-	-	-
Zn	15	786	2828	0-2352	2.70	11	4194	1231	1451-6936	0.72	17	462	453	229-695	0.08	12	65307	129963	0-147882	11.22

N: number of elements for data analysis; StDev: standard deviation; 95% CI: value ranges are based on 95% confidence interval; MSWI FA: municipal solid waste incineration fly ashes; MSWI BA: municipal solid waste incineration bottom ashes; OMS: other metal slags; *: for SSA, BIOA and CCA the values disregarding bottom ashes or fly ashes; †: CCA here include coal + olive kernel, coal + wood/wood wastes, coal + soybean stalk, sewage sludge/dredged sludge rice husk, sewage sludge + wood and etc.

N: number of elements for data analysis; StDev: standard deviation; 95% CI: value ranges are based on 95% confidence interval; MSWI FA: municipal solid waste incineration fly ashes; MSWI BA: municipal solid waste incineration bottom ashes; OMS: other metal slags; -: for SSA, BIOA and CCA the values disregarding bottom ashes or fly ashes; 1: CCA here include coal + olive kernel, coal + wood/wood wastes, coal + soybean stalk, sewage sludge/dredged sludge + rice husk, sewage sludge + wood and etc.

Table 4. Comparison of leaching on trace elements among common cement pastes, combustion ashes and cement mortar mixed with combustion ashes.

Leaching of trace elements from cement paste/concrete (mg/kg)		Comparison of leaching on trace elements between combustion ashes and their corresponding mortar with cements (mg/kg)																	
		MSWI FA (Cerbo et al., 2016; Saikia et al., 2008; Yang et al., 2009)						Coal FA (Kosson et al., 2014)						HOFA (Mofarrah and Husain, 2013)					
		Before	After	*2.88	Before	After	*2.88	Before	After	Before	After	Before	After	Before	After	Before	After	Before	After
pH	12.3	*2.88	12.4	12.2	*2.88	12.5	10.9	12.5	10.1	6.6	12.7	3.0	3.0	10.4	12.5	*2.88	10.6	12.6	
Ag	nd	nd			6.88	nd													
As	nd		<0.006	<0.006		na	16.4	<0.006	0.47	1.3	<0.006	0.12	0.003	na	nd		0.11	nd	
B			0.04	<0.01				0.09	20	20	0.35								
Ba	36.2		5.4	10		55.6		4.3	2.4	3.7	20				57.3			60.0	
Cd	Nd	0.16	<0.002	<0.002	18.66	0.22	0.17	<0.002	0.010	0.044	<0.002	0.012	0.003	0.07	nd	<0.2	<0.2	0.02	nd
Co		0.04										0.15	0						
Cr	0.37	1.18	0.35	0.24	24.02	1.5	1.8	0.22	0.66	0.03	0.18	0.85	0.018	18.5	0.16	1.0	<0.2	0.43	0.34
Cu	0.04	nd			7.44	0.20	0.14	0.40				1.8	0.006	16.10	0.56	4.8	<0.2	0.44	2.92
Hg												nd	nd						
Mn		0.24			58.16	0.1													
Mo	0.10		0.027	0.017			2.6	0.16	6.2	12	0.072			1.8	0.21		1.0	0.25	
Ni	0.45	0.44			30.38	0.22	0.35	0.02				233.73	0.273	0.37	0.03		0.08	0.08	
Pb	nd	1.92	0.020	0.008	12.9	1.8	0.26	0.81	<0.002	0.002	0.075	0.35	0.009	0.32	1.47	<0.04	49.1	53.3	
Sb	0.002		0.037	0.039			0.60	0.05	0.15	1.3	0.004			3.4	0.01		2.5	0.38	
Se	0.02		0.18	0.11			0.31	nd	0.75	0.055	0.11	0.093	0.003	na	nd		3.5	0.06	
Tl			<0.005	<0.005					<0.005	0.007	0.007								
V	nd		0.39	0.40			0.12	nd	1.1	1.5	0.27	911.74	0.585	0.15	nd		0.33	nd	
Zn	0.11	9.24			9.06	0.20	0.83	0.98				2.68	0.018	0.39	0.29	22.0	0.40	1.7	7.27

*: initial pH values of the leaching fluids based on TCLP standard; HOFA: heavy oil fly ashes; MSWI-CMP: vitrification slags of MSWI mixed ashes together with sludge; nd: not detectable

Fig. 1. Annual number of publications on cement production with waste material (CWM) and cement with fly ash, slag/bottom ash, glass, gypsum, and mining waste respectively. Data were obtained from Scopus on 11th Mar 2018. Keywords for searching are “cement production AND waste material”, “cement production AND waste material AND fly ash”, “cement production AND waste material AND slag OR bottom ash”, “cement production AND waste material AND glass” and “cement production AND waste material AND gypsum”, and “cement production AND waste material AND mining waste” respectively.

Fig. 2. Combined plots of hydration degree of clinker phases, formation of hydration products and rate of heat evolution in Portland cement paste as a function of hydration time, after Odler (Odler, 1998), Mailvaganam and Rixom (Mailvaganam and Rixom, 2002) and Bullard et al (Bullard et al., 2011).

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622 Fig. 3. Schematic illustration of the predicted effects on the pozzolanic reactions in the second
623 phase during cement hydration at later stage.

624

Fig. 4. Comparison of literature-reviewed trace metals leached amount by different ashes and stabilized ashes using cement (data based on Table 4).

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630

631 Fig. 5. Comparison of literature-reviewed trace metals leached amount by commercial cements
 632 and stabilized ashes using cement (data based on Table 4).

633

Fig. 6. A flow chart for strategy of recycling combustion ashes in cement industry (LCA: Life Cycle Assessment; CBA: Cost and Benefit Analysis).

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