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A. Keulen (**Eindhoven University of Technology and Van Gansewinkel Minerals**)

A. van Zomeren (**ECN**)

P. Harpe (**ASCEM, Rhenen, The Netherlands**)

W. Aarnink (**Van Gansewinkel Minerals, Eindhoven, The Netherlands**)

H.A.E. Simons (**LBPSIGHT, Nieuwegein, The Netherlands**)

H.J.H. Brouwers (**Eindhoven University of Technology, Department of the Built Environment, The Netherlands**)

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A. Keulen<sup>a,b,\*</sup>, A. van Zomeren<sup>c</sup>, P. Harpe<sup>d</sup>, W. Aarnink<sup>b</sup>, H.A.E. Simons<sup>e</sup>, H.J.H. Brouwers<sup>a</sup>

<sup>a</sup> Eindhoven University of Technology, Department of the Built Environment, Eindhoven, The Netherlands

<sup>b</sup> Van Gansewinkel Minerals, Eindhoven, The Netherlands

<sup>c</sup> ECN, Petten, The Netherlands

<sup>d</sup> ASCEM, Rheden, The Netherlands

<sup>e</sup> LBPSIGHT, Nieuwegein, The Netherlands

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## ABSTRACT

Municipal solid waste incineration bottom ash was treated with specially designed dry and wet treatment processes, obtaining high quality bottom ash granulate fractions (BGF) suitable for up to 100% replacement of natural gravel in concrete. The wet treatment (using only water for separating and washing) significantly lowers the leaching of e.g. chloride and sulfate, heavy metals (antimony, molybdenum and copper) and dissolved organic carbon (DOC). Two potential bottom ash granulate fractions, both in compliance with the standard EN 12620 (aggregates for concrete), were added into earth-moist concrete mixtures. The fresh and hardened concrete physical performances (e.g. workability, strength and freeze-thaw) of high strength concrete mixtures were maintained or improved compared with the reference mixtures, even after replacing up to 100% of the initial natural gravel. Final element leaching of monolithic and crushed granular state BGF containing concretes, showed no differences with the gravel references. Leaching of all mixtures did not exceed the limit values set by the Dutch Soil Quality Degree. In addition, multiple-life-phase emission (pH static test) for the critical elements of input bottom ash, bottom ash granulate (BGF) and crushed BGF containing concrete were assessed. Simulation pH lowering or potential carbonation processes indicated that metal (antimony, barium, chrome and copper) and sulfate element leaching behavior are mainly pH dominated and controlled, although differ in mechanism and related mineral abundance.

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## 1. Introduction

One of the sustainability strategies of the European Union (EU) is developing a circular economy. Potential waste and secondary materials are promoted to be re-used or recycled and subsequently applied within comparable or new processes and or applications. Hence, initial waste materials are regarded as potential new resources. This approach will ultimately lower the amount of necessary primary materials and potentially reduce the amount of materials that go into landfills. In relation to this strategy, the EU Construction Products Regulation (CPR 305/2011/EU) has come into force. This EU regulation attempts to obtain more knowledge and junction, creating a generic and level playing field between EU

member states in regard to the re-use application of waste and or secondary materials (related to environmental quality) within processes.

The Netherlands is already facilitating the re-use of many types of secondary materials within construction works by a clear and workable regulation regarding the application of building materials (“(SQD) Soil Quality Degree,” 2015). In addition, specific actions are initiated by the government to stimulate re-use in a sustainable way. One example is the re-use of Municipal Solid Waste Incinerator (MSWI) bottom ash, where the Dutch industry has signed a ‘green-deal’ with the central government to improve (towards the year 2020) the physical and environmental quality of the treated ashes. Initiating a more environmental, economically efficient and sustainable bottom ash use in constructions e.g. open granular applications for road base layers and/or secondary aggregate in asphalt and cement concrete applications.

A general, conventionally dry treated MSWI bottom ash is mainly composed non-combustible materials e.g. slag, stone, glass,

\* Corresponding author at: Eindhoven University of Technology, Department of the Built Environment, Eindhoven, The Netherlands and ECN, Petten, The Netherlands.

E-mail addresses: [arno.keulen@vangansewinkel.com](mailto:arno.keulen@vangansewinkel.com) (A. Keulen), [vanzomeren@ecn.nl](mailto:vanzomeren@ecn.nl) (A. van Zomeren).

ceramic, sand and metallic metals. Where especially metal recovery is of high economical value regarding the extraction of valuable scarce resources e.g. copper, lead, molybdenum, zinc, aluminum and iron (Allegri et al., 2014). The final mineral ash composition primarily consists of silicon, calcium, iron and aluminum containing structures e.g. quartz, calcite, hematite and ettringite (Chang and Wey, 2006; Chimenos et al., 1999; Funari et al., 2014; Kuo et al., 2013; Tang et al., 2015). The mineral compositions can vary time-to-time between the incinerators in relation to solid waste input and incineration conditions. Additionally, these conditions also effect the abundance and moderate concentrations of heavy metals and salts that are within the ashes (Funari et al., 2014; Margallo et al., 2015).

Treated mineral bottom ashes are to some extent comparable to the most widely used raw concrete aggregates and are therefore possibly useful as mineral additions in various construction material applications (Abbà et al., 2014; Bertolini et al., 2004; Cioffi et al., 2011; Florea, 2014; Forteza et al., 2004; Hassan and Khalid, 2010; Kuo et al., 2013; Pera et al., 1997; Syahrul et al., 2010; Toraldo et al., 2013). As such, a well-designed bottom ash granulate could potentially play an important role in the sustainable progress within Portland cement mixtures, as an aggregate alternative for primary sand and gravel. Additionally, this relative new ash application could stimulate reuse of the relatively fast growing (millions of tons) bottom ash quantities produced world-wide and that are currently and mainly stored in landfill sites (Oehmig et al., 2015).

To the authors knowledge, only a few available recent studies deal with mechanically treated MSWI bottom ash fractions in concrete, where currently no significant and satisfying results have been obtained. Research does show that, overall improved ash-containing concrete performance is obtained with wet treated ashes in comparison with only dry treated (Rubner et al., 2008; Sorlini et al., 2011; Zhang and Zhao, 2014). Where Kuo et al. (2015) and Yang et al. (2012) attribute the difference to the finding that the liquid phase extracts and reduces a large part of the available and potentially disturbing salts, heavy metals and fine particles, present within the ashes. To summarize literature on bottom ash containing concretes compared with the Portland reference systems the following main drawbacks have been observed:

- Strong and substantial decline of fresh concrete workability (Yu et al., 2014; Zhang and Zhao, 2014).
- Severe matrix expansion and cracking by hydrogen gas production (Müller and Rübner, 2006; Yu et al., 2014).
- Cement hydration retardation by abundant disturbing substances (van Eijk, 2001).
- Severe increase of matrix porosity and permeability due to gas bubble formation (Müller and Rübner, 2006; Rubner et al., 2008; Yu et al., 2014)
- Moderate to high loss of mechanical strength (Rubner et al., 2008; Sorlini et al., 2011; Yu et al., 2014; Zhang and Zhao, 2014).

Despite these drawbacks, the mentioned studies also report multiple ash related upsides e.g., relatively good particle distribution for concrete application, equal to slightly lower material density compared to concrete aggregate (Sorlini et al., 2011), moderate to high ash particle abrasion properties, pozzolanic reaction of bottom ash particles (Zhang and Zhao, 2014) and very low economical material costs when applied as granulate.

Knowing both disadvantages and advantages, the authors elaborated a new and promising approach; applying a specially developed dry and wet treatment on the ash before being utilized in earth-moist concrete mixtures. This treatment approach and related concrete design (which will be performed on large scale and real life production pilots) has never been considered.

Until now, all published work focuses on laboratory-scale production by applying relatively wet (high consistency) concrete mixture designs (liquid/binder ratios of  $\geq 0.4$ – $0.6$ ) instead of dry (low to zero consistency) earth-moist designs. Interestingly, research on the earth-moist concrete production and related products are rarely published (Hüsken and Brouwers, 2008), due to difficulties to produce and simulate this production process at the laboratory scale. In practice, the production requires an extremely high pressure compaction in combination with an ideal particle packing and mixture consistency.

When applying the treated ash, the related earth-moist concrete material properties could trigger an optimized result and consequently ash usage can then be favored. Identifying the potential synergy of this approach: firstly, earth-moist concrete has no slump or measureable workability and consistency is mainly determined by the compaction rate in combination with visual inspection. Hence, workability loss initiated by the ash at its higher water demand is not of influence on the fresh concrete performance. Secondly, the relatively high porosity of earth-moist concrete is able to capture potential hydrogen gas production from the bottom ash, preventing matrix expansion and related crack formation. Thirdly, the dry and wet ash treatment reduces the amount of disturbing substances within the bottom ash, therefore accordingly the potential negative interferences on the cement hydration can be minimized or neglected. Therefore, the following most important parameters (substances) in bottom ash treatment which influence the concrete performances that need to be controlled to obtain valuable bottom ash granulate fractions (BGF) are:

- Recovery of as much of the non-ferrous (aluminum and zinc) and ferrous metallic metals as possible that cause potential concrete expansion, cracking and pop-out problems due to hydrogen formation (Florea, 2014; Rubner et al., 2008).
- Extraction of the majority of fine and coarse unburned organic particles and their released organic structures that potentially disturb cement hydration processes (van Eijk, 2001).
- Reduce the total percentage of very-fine, mainly organic micrometer particles. These particles could interfere with the needed particle packing approach, increase water demand in the mixture, lower fresh concrete workability and retard cement hydration.
- Optimize bottom ash particle-size distribution to obtain an appropriate mineral granulate replacement for natural gravel.
- Reduce the amount of potential leachable salts, heavy metal and organic structures that influence the cement hydration or can leach into soil and groundwater systems (Cornelis et al., 2008; Dijkstra et al., 2006b; Meima and Comans, 1999, 1998; Shim et al., 2003).

Given these characteristics, this research focusses on treatment optimization of raw bottom ash where (1) all of its initial disturbing substances are selectively removed and (2) the (BGF) material is tuned to an ideal particle-size distribution. Furthermore, bottom ash granulates are first produced in a pilot experiment which combines specially designed dry and wet treatment processes. In a second pilot production, pre-fabricated earth-moist concrete elements are produced by replacing various mass percentages (0–100%) of the natural gravel by the BGF.

The overall aims of the present work are:

- Investigate the suitability of the designed bottom ash treatment processes.
- Characterize the initial BGF material properties and their performance within open granular and concrete application.

- Place emphasis on the leaching emissions and mechanisms in the first, second and multiple-use phases of the concrete products containing BGF as gravel replacement.

The novelty of this work is primarily the comprehensive approach to develop earth-moist concrete containing treated bottom ash. Different physical and chemical parameters important for concrete development are combined to obtain a novel bottom ash treatment process for substituting sand/gravel in concrete.

## 2. Materials and methods

### 2.1. Treatment process for MSWI bottom ash

Physical and chemical characterization of fresh MSWI bottom ash is of great importance, in producing qualitative useful concrete granulates (Florea, 2014; Rubner et al., 2008; Tang et al., 2015). Therefore, raw, untreated ash requires upgrading treatments due to its relatively heterogenic and chemically instable properties. One method, for example, is extracting all the non-mineral disturbances e.g. unburned and metallic materials. In this study, a sequence of treatment steps is performed to obtain useful granulates. Within this pilot production, up to one thousand tons of granulate was produced from MSWI bottom ash from a Dutch municipal solid waste incinerator located in Duiven. The ash was treated stepwise with dry and wet separation processes (explained below in the steps 1 to 4), producing optimal bottom ash granulate fractions (BGF) which were further characterized and tested as granulate in earth moist concrete. The performed BGF production process is a combination of current common practice (Biganzoli et al., 2013; Hu et al., 2008; Rem et al., 2004) and new developed dry and wet mechanical separation techniques composed of four separate treatment steps (described below). An overview of the processes and the different bottom ash fractions is shown in Fig. 1. Note that the total material input of 43% (M,-) within the wet treatment process increased to a 49% output. This effect is caused by water washing and saturation of the relatively porous BGF and the generated sludge fraction, which has a 50% water content.

### Step 1. Weathering of fresh MSWI bottom ash

Freshly produced and quenched MSWI bottom ash was stored outside and naturally weathered for  $\geq 3$  months. During this period, the ash slowly dries and neutralizes towards a more physically and chemically stable material (Chimenos et al., 2003; Meima and Comans, 1999; Saffarzadeh et al., 2011). This step further enhances the decrease of leaching potential of inorganic elements such as heavy metals (Aberg et al., 2006; Comans et al., 2000; Dijkstra et al., 2006b; Piantone et al., 2004). Additionally, during this time period and related high pH  $\geq 11$ , significant oxidation of metallic ferrous and non-ferrous metals occurs.

### Step 2. Dry separation MSWI bottom ash and metal recovery

Weathered bottom ash was first mechanically crushed and particles (<400 mm) of mainly 'unburned' and metallic materials were extracted using a screener and powerful overhead magnet. The generated mineral fraction (BMF) was further dry separated, in the same screener, into two fractions, BMF 0–12 mm and 12–31.5 mm. Fraction 0–12 mm was additionally separated into two fractions, BMF 0–2/3 mm (which was not further treated) and BMF 2–12 mm. Both fractions 2–12 mm and 12–31.5 mm were further processed using magnets, eddy current systems and fluff extraction apparatus to extract the maximum amount of unburned and metallic materials e.g. ferrous, non-ferrous and stainless steel (Allegrini et al., 2014). Finally, both fractions were mixed together resulting in a BMF 2/3–31.5 mm bottom ash fraction.

### Step 3. Wet separation and washing treatment

The BMF 2/3–31.5 mm was the input for the wet separation process. For this treatment, a specially designed mobile water separating-cleaning plant was built. It is a water consuming process with a liquid to solid ratio (L/S) of approximately 1:2 (contact-time  $\pm 10$  min). By washing and scrubbing potential initial material disturbances e.g. very fine particles and mobile organic and inorganic leachable contaminants (organic acids, heavy metals and salts) were removed. This method was applied earlier by several other authors (Aguar et al., 2009; Lin et al., 2011; Sorlini et al., 2011; Yang et al., 2012). The pollutants were mainly concentrated in the sludge fraction, resulting in a coarser residue with an overall

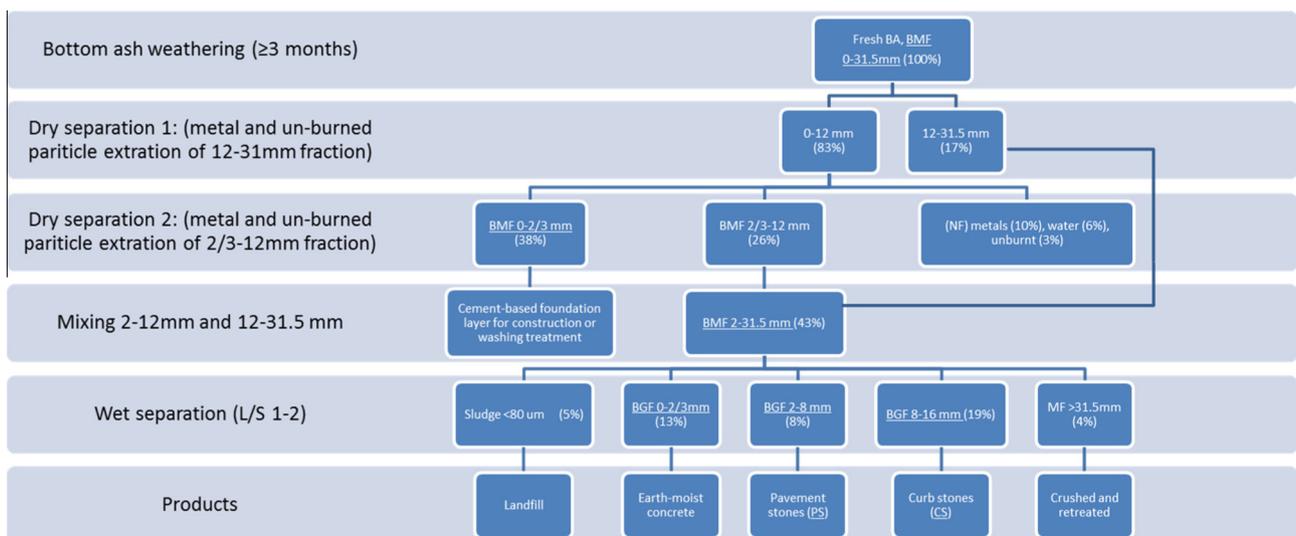


Fig. 1. Mass balance of bottom ash granulate production and treatment process.

better environmental quality. Fig. 1 shows that five different fractions were produced: sludge <80  $\mu\text{m}$ , three fractions of BGF 0–4 mm, 2–8 mm, and 8–16 mm, and an oversize BMF >31.5 mm.

#### Step 4. Final granulate treatment for concrete granulate

Finally, both coarse fractions (BGF 2–8 mm and 8–16 mm) were additionally treated before being used in Portland cement (alkali based) concretes. Due to washing, the bottom ash was further crushed or broken to release and additional fraction of metallic non-ferrous and ferrous particles that were initially covered by ash agglomerates. These metals needed to be extracted from the material to obtain a higher metal recovery and reduce their potentially damaging effect on the concrete. Metallic metals (e.g. Al and Zn) potentially initiate hydrogen gas production that can subsequently lead to expansion problems with fresh and hardened concrete and interfere with the cement hydration reaction (Müller and Rübner, 2006). Therefore, both BGF fractions were treated with magnetic separator systems to further reduce the metallic metal contents.

## 2.2. Materials

The pavement stones were produced with a blended Portland cement, a CEM III and the curbstones with a blended CEM II; both in combination with a very small addition of plasticizer (fatty acid type organic molecule). Furthermore, groundwater, natural river sand 0–4 mm, and rounded gravel fractions of 2–8 mm and 8–16 mm were used. Within the mixture design, natural aggregate replacement was done by addition of the two bottom ash granulate fraction (BGF), a 2–8 mm and 8–16 mm, picture of fraction is shown in Fig. 2.

## 2.3. Concrete mixture design and production

Concrete mixtures with the addition of two types of BGF 2–8 mm and 8–16 mm were tested. Replacement levels of both the original natural gravel types for BGF were 0%, 40%, 70% and 100% by mass of volume. All recipes are described in Table 1. Two element products were produced and tested; curbstones (CS) with dimensions of 1000 \* 200 \* 100 mm (length \* height \* width) and pavement stones (PS) with dimensions of 210 \* 80 \* 70 mm, shown in Fig. 3.



Fig. 2. Bottom ash granulate fraction 8–16 mm.

Both elements were constructed out of two layers of concrete, a specific high performance top layer which constitutes around 10% and a bottom layer which constitutes around 90% of the total construction. BGF was added as gravel replacement in the bottom layer concrete, within the top layer BGF is at the moment not applicable as this is not in compliance to the current Dutch standards. Within the analyses, strength tests were performed on the total layer construction and all other tests were specifically performed on the BGF containing bottom layer. The numbers behind the material abbreviations (0–100%) in Table 1 address the total amount of natural granulate replacement by BGF, within the tested concrete mixtures. Within the reference mixture 0% of BGF was used.

Factory scale production was chosen over laboratory experiments to overcome scaling problems (difficult to obtain representative products at lab-scale), and was performed at the site of 'de Hamer' in the Netherlands. Mixing was done by a wheeler batch mixer, around 1 m<sup>3</sup> per batch, with total batch times of 3 min. Fresh concrete was compacted within 10 min after production. After production, the specimens were kept in a climate chamber for approximately 24 h at 25–30 °C with a relative humidity >60%. From the climate chamber, elements were stored outside and sealed with a plastic sheet, to reduce unwanted water evaporation. In total, several hundred pavement stones and up to one hundred curbstones were produced.

## 2.4. Physical material testing

The material properties of the used natural sand and gravels and produced BGF fractions were analyzed in accordance with NEN-EN 12620 (2002+A1:2008 NL). Additionally, BGF fractions were tested in accordance with the Dutch product standard BRL 2507 ("BRL 2507 AEC granulaat als toeslagmateriaal voor beton," 2013) which was specifically developed as a supplement guideline for use of bottom ash fractions within Portland cement and or blended cement concrete mixtures. The final concrete performance was measured for multiple material parameters, in accordance with their product standards. For strength measurements, in accordance with the standard, specimens were first water soaked for 24 h. Total water saturation reduces possible internal stresses which could lead to unwanted deviations. Strengths were measured in accordance to the standard at 7 and 35 days. Flexural strength for curbstones (3 measurements per data point) was measured in accordance with the BRL 2314 (2005, class 2, mark T). Tensile strength for pavement stones (8 measurements per data point) were measured in accordance with BRL 2312 (2005). In addition, pavement stones were tested on freeze/thaw deicing salt resistance (3 measurements per data point) in accordance with NEN-EN 1338:2003 (E) annex D. As a preparation for the freeze/thaw measurements, stones were vertically cut in the middle (obtaining a smooth and representative surface) to test only and specifically the bottom BGF containing surface layer. All tested concrete specimens were randomly taken from the production.

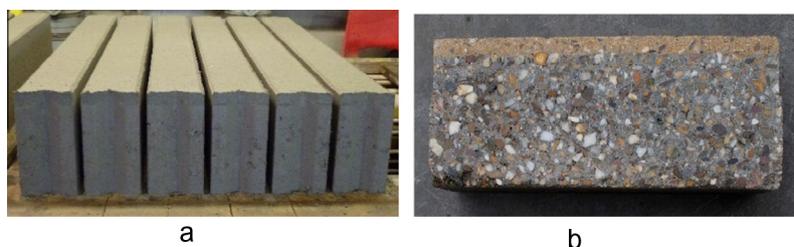
## 2.5. Chemical material testing

For the leaching analysis all granular and/or monolithic samples were taken from both the granulate and/or concrete materials. Granular samples were crushed to <4 mm before testing, in accordance with the appropriate test method. In the case of hardened concrete,  $\varnothing$ 100 mm cylinders were drilled and crushed before granular material was tested. All monolithic samples were drilled out of the concrete elements, sample size: cylindrical  $\varnothing$ 100 mm with 100 mm in height. The testing of granular materials was performed with the compliance percolation test in accordance with NEN 7383 (2004 NL). In this test, the leaching is measured as a function of the L/S ratio by percolation of the bottom ash. The

**Table 1**  
Mixture design of earth-moist concrete pavement- and curb stones.

Mixture	CEM	Sand 0–4 (%)	Gravel 2–8 (%)	Gravel 8–16 (%)	BGF 2–8 (%)	BGF 8–16 (%)	Water	Plast	Cal. density	W/C ratio
PS-0%	288	51	49		0		104	0.52	2374	0.36
PS-40%	288	51	29		20		104	0.52	2331	0.36
PS-70%	288	51	14		35		104	0.52	2307	0.36
PS-100%	288	50	0		50		104	0.52	2283	0.36
CS-0%	309	50	33	17		0	111	0.43	2354	0.36
CS-40%	309	51	20	9		20	111	0.43	2331	0.36
CS-70%	309	51	11	3		35	111	0.43	2314	0.36
CS-100%	309	49	1	0		50	111	0.43	2295	0.36

All values in kg/m<sup>3</sup>.



**Fig. 3.** Curb stone with BGF 8–16 mm (a) and pavement stone with BGF 2–8 mm (b).

cumulative emission at  $L/S = 10$  L/kg is measured in one cumulative eluate fraction.

Monolithic materials were tested using the tank leaching test in accordance with NEN 7375 (2004 NL). All tested concrete ‘monolithic and granular samples’ were around 35 days old when the tests were performed. Chemical analysis of leachates from leaching tests was performed by inductively coupled plasma atomic emission spectroscopy (ICP-AES). Chloride (Cl), bromide (Br) and sulfate (SO<sub>4</sub>) were analyzed using ion chromatography (IC). Fluoride was analyzed by flow injection analyses with spectroscopic detection.

Batch pH-static leaching experiments were carried out according to EN 14997 on subsamples that were each equilibrated for 48 h at a predetermined pH value between pH 2 and 12. The samples BMF 0–31.5 mm, BGF 2–16 mm (mixture of BGF 2–8 mm and 8–16 mm) and concrete sample PS-100% were used for the pH dependence leaching test. Within this method 15 g of dry bottom ash was suspended in 150 g nanopure demineralized water ( $L/S$  ratio of 10 L/kg) in acid-cleaned 300 mL PTFE vessels, under continuous stirring at 20 °C in contact with the atmosphere. The pH of the different suspensions was controlled using solutions of 1 M HNO<sub>3</sub> and NaOH (analytical grade) and a computerized pH-stat system. After the equilibration period, the suspensions were filtered through 0.45 μm membrane filters. The filtrates were acidified with concentrated HNO<sub>3</sub> (Suprapur<sup>®</sup>) and analyzed by ICP-AES to obtain solution concentrations of a wide spectrum of major and minor elements. It was assumed that total Sulfur (S) as measured by ICP-AES equated to SO<sub>4</sub> (factor 3 to convert S to SO<sub>4</sub>).

### 3. Results and discussion

In this paragraph, two possible granular application routes for the produced BGF fractions are investigated. First, produced BGF was tested for potential application as a granular construction product for open application in road bases or embankments. Second, BGF was tested as natural gravel replacement within earth-moist concrete.

#### 3.1. Bottom ash treatment and application feasibility as a granular construction material

The produced raw, untreated bottom ash (BMF) was subjected to several dry and wet separation steps to upgrade the environ-

mental quality and to produce fractions with desirable physical properties, see Section 2.1. To assess the open application re-use possibilities, the bottom ash input material, which was combined with several fractions at different steps in the process and the final granulates, leaching was tested using the percolation leaching test (NEN 7383). The results in Table 3 show a number of interesting features regarding the treatment processes. The input material (BMF 0–31.5 mm) already had a relatively low pH in comparison with freshly produced bottom ash. This is most likely a result of the natural weathering period of 3 months. Its pH decrease as a result of weathering has been reported by other authors (Arickx et al., 2010; Dijkstra et al., 2006a; Meima and Comans, 1998). The leaching of most regulated substances did already comply with the limit values (“(SQD) Soil Quality Degree,” 2015) for an open application. However, the leaching of antimony (Sb), copper (Cu), chloride (Cl) and sulfate (SO<sub>4</sub>) exceeded their limit values (bromide (Br) is just below the limit value). It is known that the leaching of these elements can be close to the limit values for open application (Dijkstra et al., 2006a), emphasizing the need for quality improvement of dry treated bottom ash. The further dry separation into a fine fraction (BMF 0–2/3 mm) and a coarse fraction (BMF 2/3–31.5 mm) clearly indicates that the relatively mobile substances Mo, Sb, Br, Cl and SO<sub>4</sub> are preferentially concentrated in the fine fraction. It is postulated that these elements are already dissolved mostly in the pore water of the bottom ash and are transferred to the fine fraction together with most of the water from the bottom ash. Furthermore, in the case of sulfate, release is mainly controlled by gypsum solubility (Cornelis et al., 2008; Meima and Comans, 1998). Gypsum, a sulfate source, is believed to be vulnerable to the sieving treatments due to its low mineral brittleness. It is easily crushed during sieving and becomes more concentrated in the fine BMF 0–2/3 mm fraction. As a result, the coarse BMF 2–31.5 mm shows a substantial reduction of sulfate and chloride and, to a somewhat lesser extent, bromide leaching. It is noted that the Mo leaching in the coarse fraction seems to be somewhat higher than the original input material. Which might be explained by the weak pH dependent adsorption of Mo by reactive iron/aluminum (Fe/Al)-(hydr)-oxide surfaces of less reactive coarser-sized bottom ash particles e.g. glasses and ceramics (Cornelis et al., 2008; Dijkstra et al., 2006b). The same mechanism might also explain the leaching of Sb between the fractions. Overall, the results of the dry separation treatment show that this technology

positively affects the leaching of Mo, Sb, chloride and sulfate and to a lesser extent bromide. However, the produced coarse fraction (BMF 2–31.5 mm) does not comply with the criteria of open application (SQD) with respect to the leaching of Cu, Cl and SO<sub>4</sub>. The leaching of Sb and Mo are close to their limit values.

The bottom ash was also wet treated with a water separation step to a cumulative L/S ratio 1–2 L/kg. The use of water in this step also implies that bottom ash is washed during separation, to reduce its mobile constituents (Yang et al., 2012). The leaching of the three resulting bottom ash granulate fractions (BGF) were analyzed with the percolation test (Table 3). In general, this treatment step had a very positive effect on the reduction of leaching of the mobile elements Sb, Cu, Mo, Br, Cl, and SO<sub>4</sub>. The dry and wet separation of bottom ash shows potential, reducing the leaching of BGF that than can comply with the SQD leaching criteria for open application; although Cl and SO<sub>4</sub> leaching can sometimes still be critical. However, it is observed that the Cl content is strongly reduced within the BGF and a slight Cl increase in the washed BGF as a function of larger particle size is observed, shown in Table 3. This effect might be explained by the higher porosity of larger particles that can lead to higher (chloride containing) water absorption and, hence, more dissolved Cl in that bottom ash fraction. If necessary, a further reduction of leachable Cl and SO<sub>4</sub> from the BGF fractions can be obtained by increasing the L/S ratio and contact time during washing and/or the water refreshing rate within the process (Stegemann et al., 1995; Yang et al., 2012). An important observation is that during the wet treatment process the Sb leaching is not lowered when comparing input BMF 2–31.5 mm with both washed course BGF fractions. However, this effect is noticeable when comparing BMF 0–2/3 mm input with the washed 0–4 mm BGF fraction where Sb emission is reduced >50%. In addition, in a preliminary pilot washing test a lowering effect on Sb release was also noticed, where the input bottom ash fraction 0–31.5 mm contained about 1.0 mg/kg dm (dm: dry material) and the produced granulate fractions 4–20 mm about 0.25–0.44 mg/kg dm (reduction of about >50%). This effect could be attributed to availability and the washing out of a large part of the soluble state penta antimonite (Sb<sub>2</sub>O<sub>5</sub>), which is likely to be present in weathered bottom ash eluate (Cornelis et al., 2012; Dijkstra et al., 2006a; Meima and Comans, 1998; Paoletti et al., 2001). Due to washing this metal species is transferred to the washing water and the produced sludge residue. However, the Sb release from the granular material can still be at critical limit value for open application. Considering the leaching of all regulated substances in the BMF and BGF samples (Table 3), it is concluded that the wet separation technology strongly improves the overall environmental quality of most of the initial mobile abundant elements (Sb, Ba, Cu, Mo, Cl, SO<sub>4</sub> and Br). Although in the case of the course washed fractions, Sb reducing is less noticeable.

Finally, the results of this study show that both dry and wet separation are favored in order to improve the initial environmental quality of the washed course-size bottom ash fractions. The leaching of Cl and Sb could remain critical to meet the SQD limit values for an open application (using an L/S ratio of 1–2). Therefore, the option to use the BGF as a gravel replacement in concrete was also assessed.

### 3.2. BGF application as a gravel replacement in concrete

Previous research showed that particles shape, distribution, material porosity and density of MSWI bottom ash aggregates differs compared with natural aggregates which, in the end could influence the final fresh and hardened physical properties of a cement based concrete product (Chimenos et al., 1999; Tang et al., 2015). The tested bottom ash granulate fractions within this study are specifically developed to overcome many of the more or less unwanted and till now known properties. To gain optimal and effective replacement properties, the particle-size distribution (PSD) of both BGF fractions (Fig. 4) were modified by adjusting the sieving mesh size within the wet treatment to resemble natural aggregates. When comparing their PSD, the recycled and natural materials do show some difference in the particle shape and to a lesser extent also in particle-size distribution, although the variation within the PSD of the BGF fractions are limited to about 5–15%. The variation fits within the criteria of the NEN-EN 12620 (aggregates for concrete). This also shows that with the wet treatment process is effective with which an adjustable BGF particle distribution can be obtained. Furthermore, the BGF is more rectangularly shaped than the natural round aggregate which mainly originates from the slag, glass and ceramic particles. BGF shows a higher material porosity and related water absorption, mainly caused by the porous slag particles.

The analyzed material properties of the BGF relevant for the requirements to produce concrete are described in Table 2. Parameters such as Loss on ignition (LOI 500), Alkali equivalent, Metallic Al (+Zn) and the sulfur trioxide (SO<sub>3</sub>) content are not measured for natural gravel as these are only relevant for bottom ash fractions that are considered for use in concrete. The requirements for re-use of bottom ash in concrete and the test methods to be used are described in Dutch guidance document (“BRL 2507 AEC granulaat als toeslagmateriaal voor beton,” 2013). Table 2, indicates that all tested BGF parameters meet their limits specified in NEN-EN 12620 and NEN 5905. Therefore, it is concluded that the BGF properties seem to be compatible with the requirements to replace natural gravel with BGF in concrete. Another important parameter that determines the application of BGF as aggregate replacement in concrete is the emission of contaminants from the concrete to soil and ground water. Hence, the tank leaching test was performed on

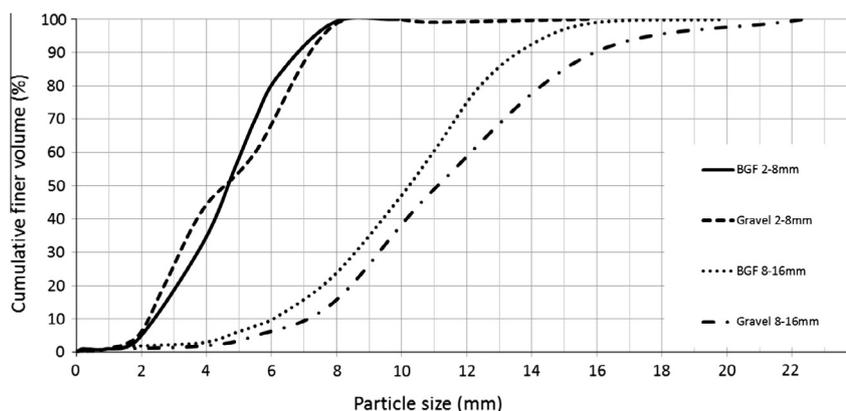


Fig. 4. Particle-size distribution of natural gravel and BGF fractions of both at 2–8 mm and at 8–16 mm size fractions.

**Table 2**  
Material properties of raw materials for concrete production.

Parameter	BGF 2–8	BGF 8–16	Sand 0–4	Gravel 2–8	Gravel 8–16	CEM II	CEM III	Limit <sup>a</sup>
LOI 500 (% mass)	0.95	0.59	n.m.	n.m.	n.m.	n.m.	1.0	≤5.0
Alkali equivalent (% mass)	0.14	0.16	n.m.	n.m.	n.m.	0.59	0.77	≤0.2
Metallic Al (% mass)	0.47	0.29	n.m.	n.m.	n.m.	n.m.	n.m.	
Metallic Al + Zn (% mass)	0.73	0.80	n.m.	n.m.	n.m.	n.m.	n.m.	≤1.0
SO <sub>3</sub> (% mass)	0.15	<0.03	n.m.	n.m.	n.m.	n.m.	n.m.	≤0.8
Anhydrite (% mass)	n.m.	n.m.	n.m.	n.m.	n.m.	2.8	3.3	
Chloride content (% mass)	≤0.03	≤0.03	≤0.003	≤0.003	≤0.003	0.04	0.06	
Los Angeles (LA) coefficient	33	31	>35	>35	>35	n.m.	n.m.	
Density $P_{rd}$ (kg/m <sup>3</sup> )	2400 <sup>b</sup>	2250 <sup>b</sup>	2640	2570	2570	3000	3020	≥2000
Water absorption 24 h (% mass)	6.9	6.8	0.2	1.3	1.3	n.m.	n.m.	

n.m.: not measured.

<sup>a</sup> Limit values in accordance to NEN-EN 12620/NEN 5905.

<sup>b</sup> ±100 kg/m<sup>3</sup>.

**Table 3**  
Total element emission measured with percolation test (NEN 7383) of granular state bottom ash fractions from different treatment processes.

Parameter	BMF 0–31.5	BMF 0–2/3	BMF 2–31.5	BGF 0–4	BGF 2–8	BGF 8–16	Limit granular material
<i>n</i>	3	2	1	1	1	1	
pH	8.41	8.14	9.04	8.46	8.41	8.45	No limit
EC	1681	2495	730	956	696	654	No limit
Antimony	<b>0.55</b>	<b>0.68</b>	0.25	0.30	0.30	0.28	0.32
Arsenic	0.08	<0.10	0.05	<0.10	<0.10	<0.10	0.90
Barium	0.39	0.39	0.50	0.28	0.20	0.17	22.00
Cadmium	<0.0085	<0.01	<0.01	<0.01	<0.01	<0.01	0.04
Chromium	0.05	<0.10	0.05	<0.10	<0.10	<0.10	0.63
Cobalt	0.04	<0.10	0.03	<0.10	<0.10	<0.10	0.54
Copper	<b>1.53</b>	<b>1.45</b>	<b>1.30</b>	<0.15	<0.15	<0.15	0.90
Mercury	<0.0012	0.005	0.00	<0.005	<0.005	<0.005	0.02
Lead	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	2.30
Molybdenum	0.51	<b>1.20</b>	0.84	0.32	0.23	0.20	1.00
Nickel	0.09	<0.10	0.05	<0.10	<0.10	<0.10	0.44
Selenium	0.01	<0.039	0.01	<0.039	<0.039	<0.039	0.15
Tin	0.02	<0.10	0.02	<0.10	<0.10	<0.10	0.40
Vanadium	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	1.80
Zinc	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	4.50
Fluoride	5.6	2.3	2.4	4.0	2.6	2.4	55
Chloride	<b>2725</b>	<b>4950</b>	<b>1700</b>	550	570	<b>687</b>	616
Sulfate	<b>6533</b>	<b>11,000</b>	<b>3200</b>	<b>3367</b>	1833	1393	2,430
Bromide	18.8	<b>25.0</b>	14.0	2.8	3.0	3.5	20

All values in mg/kg dm. Bold: above limit value.

*n*: Sample amount.

concrete curb stones (CS) samples to assess leaching using gravel replacement rates from 0–100%. The results reported in Table 4 show that all BGF containing concrete mixtures comply with the limit values for monolithic construction materials. However, with an increasing replacement rate of BGF, it results in even slightly lower emissions of metals, although it is unclear whether this difference is significant. A possible explanation for this effect, is sorption and or complexation of leachable mobile ‘heavy’ metals on reactive Fe/Al-(hydr)-oxide and glassy surfaces within the porous slag. This would enable an additional barrier for further transport through the cementitious matrix (Cornelis et al., 2008; Piantone et al., 2004; Saffarzadeh et al., 2011; Zevenbergen et al., 1996). Finally, only the leaching of Cl seems to be slightly increased within the sample with 100% gravel replacement with BGF (CS-100). However, the measured Cl release (470 mg/m<sup>2</sup>) is still more than two orders of magnitude below the limit value (110,000 mg/m<sup>2</sup>). Overall, the results of the curb stones with BGF replacement rates between 0% and 100% have obtained an environmental quality which fulfills the requirements specified by the SQD.

### 3.3. Fresh and hardened concrete properties with increasing BGF content

During the production at total quantity of >20 ton, fresh earth-moist (zero slump) concrete was produced. The fresh

**Table 4**  
Total element emission measured with the diffusion test (NEN 7375) of monolithic state curb stone concrete with increasing natural gravel replacement with BGF 8–16 mm.

Parameter	CS-0%	CS-40%	CS-100%	Limit monolithic material
pH start	11.6	11.6	9.8	No limit
pH end	11.9	11.9	11.5	No limit
Antimony	0.4	0.4	0.4	8.7
Arsenic	4.0	4.0	2.2	260
Barium	19.0	20.0	3.3	1,500
Cadmium	0.1	0.1	0.04	3.8
Chromium	4.0	4.0	2.2	120
Cobalt	2.4	2.4	1.3	60
Copper	4.0	4.0	2.2	98
Mercury	0.02	0.02	0.02	1.4
Lead	8.0	8.0	4.4	400
Molybdenum	0.8	0.8	0.6	144
Nickel	4.0	4.0	2.2	81
Selenium	0.6	0.6	0.3	4.8
Tin	1.6	1.6	0.9	50
Vanadium	8.0	8.0	4.8	320
Zinc	16.0	16.0	8.7	800
Fluoride	128	96	49	2,500
Chloride	88	80	470	110,000
Sulfate	960	960	1200	165,000
Bromide	2	2	5	670

All values in mg/m<sup>2</sup>/64 days.

material properties did not vary significantly as a result of the BGF (0–100%) replacement. The consistencies and compaction rates of the mixtures, also known as ‘green’ strength, were all approximately equal (visually and manually judged). The additional mixture workability, also known as the open time or setting time, was likewise not influenced by increasing BGF quantities, although it is known that an increase of salts such as chloride and sulfate can accelerate or retard cement hydration (Cheung et al., 2011). The BGF fractions have up to 5 times higher water absorption rates ( $\pm 7\%$ ) compared to natural gravel ( $\pm 1.3\%$ ). This high absorption is mainly related to the porous slag content and could affect the workability properties when it is not fully saturated during usage. However, this high BGF water content may also act as an ‘internal aquifer’, which may further enhance internal cement hydration, resulting in a higher strength developed and matrix densification. This can be advantageous in earth-moist concrete mixtures, where a relative low water to binder (W/B) ratio is usually used, which would result in less efficient cement hydration by too low water contents.

#### 3.4. Concrete strength of pavement stones with BGF 2–8 mm content

The pavement stones were constructed out of two concrete layers, a thin top layer (0.5–1.0 cm) and thick (7.0–7.5 cm) bottom layer. Only the bottom layer comprises of various replacement levels of BGF 2–8 mm. The whole stone specimen was tested on tensile strength development, from 0% up to 100% gravel replacement, of which the data is shown in Fig. 5. The 7 days strengths were stable at around 3.7 N/mm<sup>2</sup>. At 35 days, strength results slightly increase at higher BGF replacement levels (from 4.5 N/mm<sup>2</sup> with 0% BGF up to 5.1 N/mm<sup>2</sup> with 100% BGF). However, this increase of 12% is deemed not to be significant with respect to the standard deviations of the analyses. Finally, all mixtures comply with the strength limit of 3.5 N/mm<sup>2</sup> at 35 days related to the standard. The authors propose several mechanisms to explain the observed increase in strength:

- The improvement of the attachment of inner particles due to the introduction of rougher and more rectangularly shaped BGF. This enables a better mineral resilience between the aggregates and the cement binder in comparison with the round and more smoothly shaped gravel.
- The production of earth-moist concrete and its related low L/B ratio, is known as insufficient in generating an effective cement hydration. The mineral BGF matrix with its relatively high porosity and related high water absorption ( $\pm 7\%$ ) could act as an inner aquifer which slowly releases water over time, resulting in more effective cement hydration and related strength growth.

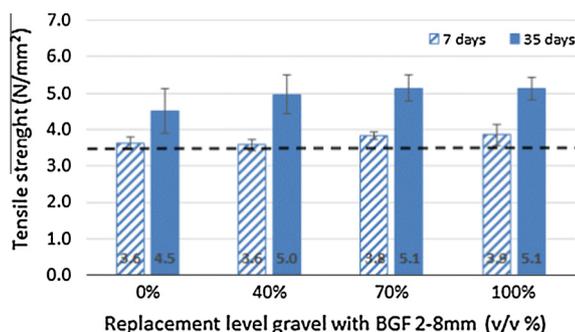


Fig. 5. Tensile strength of pavement stones with increasing replacement levels (0–100%) of BGF 2–8 mm for natural gravel 2–8 mm. Strength limit value at 35 days is 3.5 N/mm<sup>2</sup>.

- The smaller sized BGF contains a relatively high percentage of potential pozzolanic reacting bottom ash slag compared with the larger BGF fraction. This may act as reactive hydraulic aggregate to gain additional matrix densification and reduced porosity (Zhang and Zhao, 2014), resulting in higher strength development at 35 days and beyond (Piantone et al., 2004; Zevenbergen et al., 1996)

To obtain a better understanding of the mechanisms behind these results, further research into the binding mechanism between the cement and the GBF in comparison with the gravel is necessary.

#### 3.5. Concrete strength of curb stones with BGF 8–16 mm content

The curb stone elements were similar to the pavement stones, produced out of two layers of concrete and BGF 8–16 mm was added only in the bottom layer. Their observed overall flexural strength development is somewhat different compared to the tensile strength of the pavement stones. At 7 days, a stable flexural strength (6.0 N/mm<sup>2</sup>) is gained at 0–40% of BGF contents. However, strengths slightly decreased to 5.2 N/mm<sup>2</sup> at 70–100% of BGF contents, of which the data is shown in Fig. 6. This overall effect is also reproduced at the 35 days strength development. At 0–40% BGF addition, the 35 days strength reached an average of 6.6 N/mm<sup>2</sup> and decreased to an average of 5.8 N/mm<sup>2</sup> at higher than 40% BGF contents. This 5.8 N/mm<sup>2</sup> strength means about 12% loss compared to the reference, although it is not significant to the level of BGF added. In summary, a stable and one time strength loss at both 7 and 35 days was observed when more than 40% of BGF was added. However, all curb stone mixtures comply with the 35 days strengths limit of 5.0 N/mm<sup>2</sup>, in accordance with the standard. The authors propose the following mechanisms for the observed strength decrease:

- Strength is influenced by the decline of particle's hardness/abrasion (also known as Los Angeles coefficient). The hardness of the BGF 8–16 mm (value  $\geq 31$ ) is slightly lower compared to the BGF 2–8 mm ( $\geq 33$ ) and both moderately lower compared to the natural aggregates (value  $\geq 35$ ), which results in a potential weaker matrix.
- The smoothness of particles can be of influence, as the BGF 8–16 mm contains a relatively high content of large sized ceramics and glass particles compared to the BGF 2–8 mm. The poor shape of these coarse glass and ceramic aggregates causes a decrease in adhesive strength between the BGF aggregate and the cement paste, resulting in a decreased flexural strength (Jani and Hogland, 2014). Overall, when using only the BGF 2–8 mm instead of 8–16 mm fractions, as with the pavement stones, the observed strength loss may be further minimized.

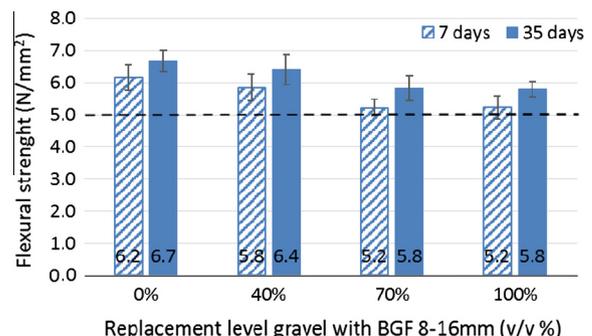


Fig. 6. Flexural strength of curb stones with increasing replacement levels (0–100%) of BGF 8–16 mm for natural gravel 8–16 mm. Strength limit value at 35 days is 5.0 N/mm<sup>2</sup>.

- The course BGF, which has a lower specific surface area, contains much less potential pozzolanic slag particles compared with the fine BGF, which results in suppressed later age (35 days) strength development.

In consistency with the results of pavement stone strengths, further research on the aggregate/cement binding mechanism is necessary.

### 3.6. Freeze-thaw deicing salt resistance of BGF containing pavement stones

An average Northern European climate requires specific freeze-thaw deicing salt resistance properties of outdoor concrete elements. In this study, these measurements were performed by determining the surface mass loss/m<sup>2</sup> of the bottom layers of pavement stones that contain the BGF 2–8 mm. The top layers of the pavement stones were not tested, as they are composed of only primary aggregates in combination with a higher binder content. As shown in Fig. 7, the obtained mass loss results of the BGF containing concrete showed no significant increase with higher BGF contents. An average of ±0.220 kg/m<sup>2</sup> is obtained with no significant differences between the 0% reference and 100% BGF replacement. All measured values are well below the prescribed limit of 1.0 kg/m<sup>2</sup>. To summarize, even at higher BGF contents, and therefore higher porosity and potentially higher liquid adsorption, no performance loss was observed. This indicates that the performance of BGF related to freeze-thaw resistance is also in compliance, similarly to the natural gravels that are nowadays used as high performance top layer aggregates.

### 3.7. Potential emissions of bottom ash and concrete products in multiple life phases

#### 3.7.1. Second life emissions of crushed and demolished pavement stones

Although the CPR indicates that emissions of construction products should not have an exceedingly high impact over the entire life cycle, there is currently no regulatory framework to assess this property. Therefore, additional experiments were performed indicating potential trends in material behavior in a second or multiple life phase. As a first step, it can be imagined that the BGF containing concrete ends as crushed construction and demolition product. This material can be used as recycled aggregate (second life) suitable as concrete aggregate, road base material or other granular construction applications and should be tested with a percolation leaching test according to Dutch regulations. Leaching data shown in Table 5, is obtained from crushed concrete with 0%, 40% and

100% of BGF, replacing the gravel content. The results shows that the overall leaching of all crushed pavement stones is in compliance with the limit values for an open (granular) application used within a second life. Nevertheless, there seems to be a slight increase of only salts release at higher BGF replacement levels e.g. Cl, SO<sub>4</sub> and Br leaching. However these emissions remained far below the limit values.

#### 3.7.2. Multiple life emissions of crushed and demolished pavement stones

It is known that alkaline materials have a tendency to carbonate, take up CO<sub>2</sub> from the atmosphere during their life time (Dijkstra et al., 2006a; Meima and Comans, 1998; Meima et al., 2002). The extent to which this process occurs and how it affects the material properties, is dependent on the material performance, application scenario and time. The potential endpoint of the Portland cement based carbonation process is the equilibrium with the mineral calcite (CaCO<sub>3</sub>), resulting in a pH of about 8.5.

To approximate and compare potential effects of pH neutralization of the concrete pavement stones and to study this behavior, various bottom ash materials from this study were tested with the pH dependent leaching test e.g. untreated bottom ash (BMF 0–31.5 mm), treated bottom ash granulate (BGF 2–16 mm) and crushed pavement stone concrete (PS-100%). The test results (Fig. 8) indicate trends in the emission of substances considering pH changes over the multiple life cycles of the material. This approach has also been used several times to identify release controlling phases and can assist in the development of treatment technologies to improve the leaching behavior (Cornelis et al., 2012; Dijkstra et al., 2006b; Gougar et al., 1996; Meima and Comans, 1999; Vítková et al., 2009). The results of the pH dependence tests for Ba, Cu, Sb, SO<sub>4</sub> and chromium (Cr) are presented in Fig. 8. The leaching of barium from the pavement stones, in the relevant pH domain (pH 8–12), is substantially higher than both bottom ash materials (untreated BMF and the treated BGF materials in Table 3). These observations are also consistent with the percolation test results in Table 5, showing already a high Ba release from the reference concrete with 100% gravel (PS-0%). This observation might be explained by a lower amount of carbonate in the concrete in comparison with the bottom ash. Between

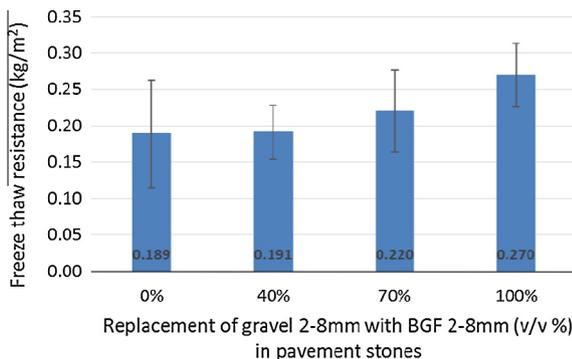


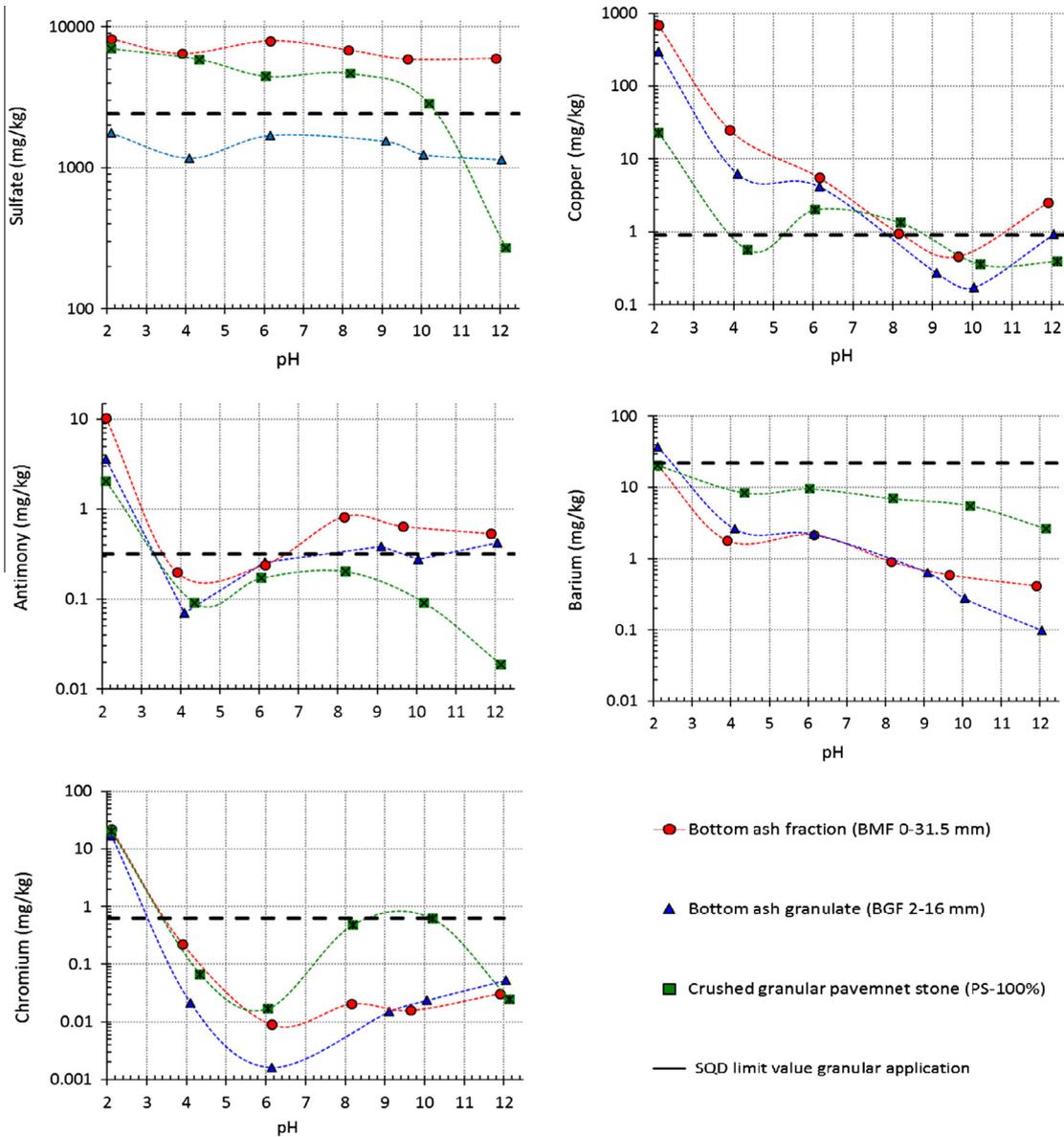
Fig. 7. Freeze-thaw resistance of pavement stones with increasing replacement levels (0–100%) of BGF 2–8 mm for natural gravel 2–8 mm. Limit value is 1 kg/m<sup>2</sup>.

Table 5

Total element emission of granular state pavement stone concrete with increasing natural gravel replacement with BGF 2–8 mm.

Parameter	PS-0%	PS-40%	PS-100%	Limit granular material
pH end	12.4	12.2	12.1	No limit
Antimony	0.01	0.01	0.02	0.32
Arsenic	<0.05	<0.05	<0.05	0.90
Barium	5.00	4.40	5.00	22.00
Cadmium	<0.001	<0.001	<0.001	0.04
Chromium	<0.05	<0.05	<0.05	0.63
Cobalt	<0.03	<0.03	<0.03	0.54
Copper	<0.05	0.10	0.20	0.90
Mercury	<0.0004	<0.0004	<0.0004	0.02
Lead	<0.10	0.26	<0.10	2.30
Molybdenum	0.07	0.07	0.11	1.00
Nickel	<0.05	<0.05	<0.05	0.44
Selenium	<0.007	<0.007	<0.007	0.15
Tin	<0.02	<0.02	<0.02	0.40
Vanadium	<0.10	<0.10	<0.10	1.80
Zinc	<0.20	<0.20	<0.20	4.50
Fluoride	1.2	1.2	1.4	55
Chloride	60	89	160	616
Sulfate	38	54	66	2.430
Bromide	1.0	1.1	1.6	20

All values in mg/kg ds.



**Fig. 8.** pH Dependency leaching of elements Sb, Ba, Cu, SO<sub>4</sub> and Cr form various bottom ash materials (red dots: bottom ash fraction BMF 0–31.5 mm/blue triangle: bottom ash granulate BGF 2–16 mm/green cubes :crushed granular pavement stone PS-100%). The dashed line is the element limit in the SQD. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

pH 12–10, the increased Ba leaching may also be affected by the dissolution of ettringite from the cementitious matrix, stimulating solubility. Hence, at lower pH <10, barium release further increases and it is assumed that the leaching of Ba is controlled by the solubility of barium sulfate (Sloot et al., 2001; Vollpracht and Brameshuber, 2015).

As for copper, the leaching pattern for all three materials is more or less similar, albeit that the availability of Cu is substantially lower in the concrete in comparison with the bottom ash. The leaching of Cu from the bottom ash fractions (BMF and BGF), between pH 6 and 10, is mainly controlled by the formation of copper hydroxide (Cu(OH)<sub>2</sub>, Klemm, 1998) and the complexation to dissolved organic matter (Impellitteri et al., 2002; Meima et al., 2002; Zomeren and Comans, 2004). In addition, the BGF containing concrete seems to indicate a somewhat elevated Cu release between pH 6 and 10, which is the typical pH range where the binding of Cu to organic ligands is important. Possibly, the

increased Cu leaching could be caused by the addition of the organic fatty acid based super plasticizer (SP) during the concrete production process. This SP contains reactive organic molecules with probably a high proportion of negatively charged carboxylic groups. Copper has a high affinity for organic ligand complexes with these carboxylic groups (fulvic acids). Overall, the results show that Cu leaching of BGF is significantly lower than the regulatory limit value and is strongly reduced compared with that of BMF. This effect is caused by the washing step where a substantial fraction of the dissolved organic carbon (DOC) fraction (humic and fulvic acids) was removed. Pilot results show that DOC reduces more than 50% from 100–130 mg/kg dm from initial BMF input (2–31.5 mm) to 25–50 mg/kg dm in the washed BGF 2–16 mm (mixture of BGF 2–8 mm and 8–16 mm).

The leaching of Sb from BGF as a function of pH reflects the typical behavior of an oxyanion with relatively pH independent leaching at neutral (pH 7) to alkaline pH (12), which is opposite of most

cat-ions (Dijkstra et al., 2006a). Antimony leaching from bottom ash is not yet completely understood. However, multiple possible reaction mechanisms affecting Sb leaching are described (Cornelis et al., 2012; Dijkstra et al., 2006a) and the extent to which these properties are dominant will most likely determine the net leaching behavior. Dissolved Sb is assumed to be predominantly Sb(V) in weathered ashes (pH range 8–10) and Sb(III) in fresh produced ashes at >pH 10 (Cornelis et al., 2012). These forms of Sb behave differently with respect to conditions in the bottom ash, e.g. pH, DOC, Ca and Fe/Al containing minerals/oxides (Cornelis et al., 2012; Dijkstra et al., 2006a; Meima and Comans, 1998). Additionally, pH-dependence data for the treated BGF fraction (blue triangle data points) show that the washing and wet separation step strongly reduces Sb leaching at the natural to moderately high pH of the bottom ash granulate (pH 8.5–12). This result indicates that part of the abundant Sb in the ash is present in soluble form that is removable by washing techniques. It has been suggested that Sb (III) can form complexes with DOC in bottom ash (Cornelis et al., 2012; Wilson et al., 2010). It should be noted that the leaching at pH 8.5 can remain critical after wet processing of the bottom ash into BGF.

The leaching of Sb in concrete seems to be significantly further reduced by cement use. This effect is partly caused by dilution of 50% of total aggregate replacement. Furthermore, the relatively low Sb leaching at high >pH 11 in concrete seems to indicate that Sb is incorporated in ettringite or possibly romeite, which is typical for cementitious matrices (Baur et al., 2004; Cornelis et al., 2012; Klemm and Bhatti, 2002; Vollpracht and Brameshuber, 2015). In addition, ettringite and romeite stability is related to pH and start to dissolve towards lower (pH <10.5). This process can initiate Sb release (Cornelis et al., 2012; Dijkstra et al., 2006a; Klemm and Bhatti, 2002; Klemm, 1998; Sloot et al., 2001) as seems to be consistent with the results shown in Fig. 8c. In summary, the Sb leaching of BGF is still critical towards the limit values of the SQD, although the emission from crushed concrete is significantly lower than the regulatory limit value.

The availability of sulfate within the bottom ash fractions is substantially reduced by the wet separation step (BGF) and to a lesser extent by dry (BMF) treatment of bottom ash. The sulfate leaching is almost pH independent over a wide range of pH values. It has been found that the leaching of sulfate from bottom ash is primarily controlled by the solubility of gypsum (Cornelis et al., 2008; Dijkstra et al., 2006a; Saffarzadeh et al., 2011; Sorlini et al., 2011) and the results in Table 3 are consistent with these observations. Especially for sulfate, the leaching behavior of the concrete differs substantially compared with that of bottom ash. It is assumed that the leaching of sulfate in the concrete is controlled by increasing decomposition of ettringite (between pH 10 and 12) and the observed solubility increase results in the release of mono-sulfate (Baur et al., 2004; Christensen et al., 2004; Klemm, 1998; Sloot et al., 2001).

The results of the pH dependent leaching tests also clearly indicate that the enhanced leaching of sulfate from the bottom ash containing concrete is primarily caused by the concrete and the use of cement (related to anhydrite content of cement used as setting retarder Table 2). However, further quality improvement to reduce SO<sub>4</sub> leaching from crushed concrete should aim specifically at the cement, since this material is the main source of sulfate.

The same observation is made for the pH dependent leaching of chromium (Fig. 8). Untreated and treated bottom ash fractions show a different Cr leaching pattern, in the pH range from about 7–11, in comparison with the BGF containing concrete. The results indicate that Cr(VI) may be present in the concrete, as was also shown by earlier work on the characterization of cement mortars (Sloot et al., 2001). From this study it was also found that Cr leaching emissions levels, are related to the cement type that was used.

Pure Portland cement shows the highest Cr leaching, while blended Portland cements (CEM II and CEM III containing supplementary cementitious materials) show decreasing Cr levels, all in the pH range of 7–11 (Sloot et al., 2001). However, the results of the analyzed BGF containing concrete cannot be completely explained based on the results of this research. In this study, a CEM III with a composition of moderate 40–60% clinker and moderate 60–40% GGBS was used. This type of cement mortar has shown a relatively low to moderate Cr leaching ( $\pm 0.1$  to <0.01 mg/kg at pH 5–10), where concentration is related to the clinker content (Sloot et al., 2001). Possibly, the bottom ash can exhibit some oxidizing properties causing the conversion of Cr(III) to Cr(VI) in the products. This potential effect should be studied in further work.

To finalize, the leaching behavior as a function of pH does also indicate possible expected changes in the emission of contaminants in multiple life phases, as the pH of alkaline materials tends to neutralize as a result of carbonation. This process will proceed much faster when the surface area of the material is increased in the case of granular demolition material. The pH test can be used as a basis for judgement of potential long-term weathering effects on the emission of contaminants. Further research is necessary to assess the possible scenarios in multiple life phases of construction materials and the expected degree and rate of weathering that will occur. It should be noted that, apart from the assessment of emissions to soil and groundwater, the physical properties of construction materials are also important and can as well contribute to decisions on future applications in multiple life phase.

#### 4. Conclusions

A novel treatment process of MSWI bottom ash is developed to gain a higher level of its re-use as secondary granulate material (BGF) within earth-moist cement mixtures. A comprehensive overview demonstrates the production, design and characterization of BGF, e.g. their physical and chemical properties and related performance within a granular (open) and monolithic (concrete) application. This study leads to the following conclusions:

- The design and development of an MSWI bottom ash treatment process should focus fully on the desired granulate (end-product) properties and its final application, gaining a reliable and optimal performing granulate.
- A wet (water washing) treatment in addition to or compared with dry treatment enables the significant removal of unwanted substances within bottom ash, e.g. soluble salts (chloride and sulfate), heavy metals and organic structures as well as unwanted fine and unburned particles. Additionally, the wet treatment is an ideal process for effectively tuning and adjusting needed BGF particle-size distribution.
- BGF fractions are in compliance with the standard EN 12620 (aggregates for concrete) and do not show large variations when compared with natural aggregates. The physical and chemical properties of BGF differ slightly compared to natural gravel (e.g. lower density material  $\pm 2400$  kg/m<sup>3</sup> instead of 2600 kg/m<sup>3</sup>, higher water absorption; 6.8% instead of 1.3%, and lower crushing resistance of 31–33 instead of >35 (LA index).
- The leaching of BGF 2–8 mm and 8–16 mm were not fully in compliance with the open (granular) application criteria of the Dutch Soil Quality Degree (SQD). Release of chloride and antimony are just above or just below their limit values. An additional wet treatment optimization, e.g. at increased L/S ratio (extracting more soluble salts) and/or the addition of specific additives could further improve their environmental quality to comply with the SQD granular limit value (Comans et al., 2000; Yang et al., 2012).

- In two cement based earth-moist concrete mixtures (pavement- and curb stones), coarse natural gravel fractions were replaced up to 100% with bottom ash granulate. Both mixtures, with increasing BGF contents, maintained necessary fresh and hardened physical and chemical performances, e.g. workability and strength, freeze-thaw limits. A moderate tensile strength increase was observed with higher BGF content within the pavement stone mixtures. Especially the BGF 2–8 mm showed promising results, where the smaller size particles have the tendency to give an overall better performance compared to the larger size fraction size 8–16 mm.
- Replacing BGF up to 100% for natural gravel did not affect the overall emission and leaching behavior of the monolithic state concrete. The leaching behavior and analyses of granular state, crushed BGF containing concrete, within a second life phase, shows that all measured elements are far below their limit values in accordance with the SQD for granular (open) application.
- An indication of the potential emission of crushed BGF containing concrete within multiple-life phases indicated that the emissions might change in a following life phase due to weathering and carbonation. Rising sulfate (SO<sub>4</sub>), barium (Ba) and chromium (Cr) release (by mineral dissolution and oxidation) were observed at  $\leq$ pH 10.5. Where Cr can be attributed to the cement type and SO<sub>4</sub> and Ba to overall cement use; the elements are not related to the BGF content.

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**ECN**

Westerduinweg 3  
1755 LE Petten  
The Netherlands

P.O. Box 1  
1755 ZG Petten  
The Netherlands

T +31 88 515 4949  
F +31 88 515 8338  
info@ecn.nl  
www.ecn.nl

