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# Characteristics and application potential of municipal solid waste incineration (MSWI) bottom ashes from two waste-to-energy plants



P. Tang <sup>a,\*</sup>, M.V.A. Florea <sup>a</sup>, P. Spiesz <sup>a,b</sup>, H.I.H. Brouwers <sup>a</sup>

<sup>a</sup> Department of the Built Environment, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

#### HIGHLIGHTS

- MSWI bottom ashes properties related to their production time and procedure.
- Environmental impact evaluation of bottom ashes over time.
- MSWI bottom ash application potential in cement clinker production.
- Influence of bottom ash on cement hydration and mortar strengths.

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#### ABSTRACT

This study focuses on municipal solid waste incineration (MSWI) bottom ash characteristics, its heterogeneity, environmental properties, and their stability in time. The physical and chemical characteristics of bottom ashes from two plants were determined over time; results show that their properties are very stable and similar to each other. As an exploratory study, bottom ash was applied in mortars without further treatment, in order to identify the limit conditions of its use as replacement material. Results show that bottom ash fines have a detrimental effect on cement hydration and strength of the mortars.

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#### 1. Introduction

In recent years, the waste-to-energy technology [1] became an efficient method to deal with the increasing amount of municipal solid waste worldwide [2]. The incineration reduces the mass and volume of the solid waste dramatically [3], thus the requirement for landfilling is decreased [4]. Moreover, energy in the form of heat, electricity, etc. can be recovered during incineration [5], together with the recycling of ferrous and non-ferrous metals, paper and plastics from the solid waste [6], which can bring financial benefits. However, there is a considerable amount of solid incineration residues generated after the combustion, typically bottom ash, fly ash, boiler ash, etc., of which bottom ash accounts for about 80% [7].

The bottom ash from municipal solid waste incineration (MSWI) is a stony material that contains stone, brick, ceramic, glass

and unburned organic matter (wood, plastic and fibre etc.) [7] and its properties differ from plant to plant, and country to country [8–11]. Many previous studies were focused on the characteristics of the bottom ash, as well as its disposal or reuse. For instance, Forteza et al. [12] studied the physical, chemical and engineering properties of the bottom ash from MSWI and concluded that the bottom ash has similar properties as natural aggregates and is suitable to be used in road-construction. Olsson et al. [13] and Hjelmar et al. [14] addressed the environmental impact of the bottom ash used in road construction considering the leaching of heavy metals. Others investigated the utilisation of MSWI bottom ash in concrete [15–17], the results of which indicate that the incineration bottom ash is usable as a concrete ingredient. It was also reported that the use of bottom ash as concrete aggregate could affect the durability of concrete, because of the metallic aluminium and glass particles which may contribute to cracks and alkali-silica reaction, respectively [18-21]. Hence, a suitable way to recycle the solid residue in waste-to-energy plants could be by applying it in concrete as a secondary material, but appropriate treatments are needed

<sup>&</sup>lt;sup>b</sup> ENCI HeidelbergCement Benelux, The Netherlands

<sup>\*</sup> Corresponding author. Tel.: +31 0 40 247 5795; fax: +31 0 40 243 8595. *E-mail address*: p.tang@tue.nl (P. Tang).

**Table 1**Maximum leaching of inorganic compounds for building materials (SQD) [30].

Parameter	<sup>a</sup> Shaped [ <sup>d</sup> E <sub>64d</sub> in mg/m <sup>2</sup> ]	<sup>b</sup> Non-shaped [ <sup>e</sup> mg/kg d.m.]	<sup>c</sup> IBC building material [mg/kg d.m.]
Antimony (Sb)	8.7	0.16	0.7
Arsenic (As)	260	0.9	2
Barium (Ba)	1500	22	100
Cadmium (Cd)	3.8	0.04	0.06
Chromium (Cr)	120	0.63	7
Cobalt (Co)	60	0.54	2.4
Copper (Cu)	98	0.9	10
Mercury (Hg)	1.4	0.02	0.08
Lead (Pb)	400	2.3	8.3
Molybdenum (Mo)	144	1	15
Nickel (Ni)	81	0.44	2.1
Selenium (Se)	4.8	0.15	3
Tin (Sn)	50	0.4	2.3
Vanadium (V)	320	1.8	20
Zinc (Zn)	800	4.5	14
Bromide (Br-)	670	20	34
Chloride (Cl <sup>-</sup> )	110,000	616	8800
Fluoride (F-)	2500	55	1500
Sulphate (SO <sub>4</sub> <sup>2-</sup> )	165,000	1730	20,000

<sup>&</sup>lt;sup>a</sup> Building material with a minimum volume unit of at least 50 cm<sup>3</sup>, and has stable shape under normal conditions.

before such an application. Nevertheless, as a combustion residue of many types of solid wastes, bottom ash contains heavy metals, such as lead, copper and antimony etc., as well as some soluble salts. The leaching of these substances from the bottom ash to the environment will result in a potentially negative impact on water resources, human health and environment [22–24]. Thus, the environmental impact of the bottom ash should be evaluated before its application. To estimate the influence of bottom ash on the environment, various pieces of legislation are established on the disposal or reuse of municipal solid waste incineration ashes, which is in consideration of environment protection and reuse/

recycle resources, and based on the conditions in different countries [25–28].

In the Netherlands, there is a national regulation termed the Soil Quality Decree (SQD) [29,30] with the latest version released in 2007. This legislation defines the utilisation of construction materials, considering the protection of the environment from potential risks posed by the leaching of contaminants from construction materials. The goals of this legislation are soil protection and improvement of recycling and reuse of secondary materials. According to this legislation, the emission of the inorganic compounds from the material should be below the limiting values, as shown in Table 1, based on which the materials are divided into three categories: shaped, non-shaped, and IBC materials. The IBC materials refer to materials which can be applied only when an insulation approach is used.

A very important aspect of the application of MSWI bottom ashes is the stability of their chemical and physical properties [31]. It is also known that the properties of the MSWI bottom ashes from different plants and countries vary due to the source of solid waste and incineration process [32,33]. Moreover, the environmental legislation, management and application of the MSWI bottom ashes differ from country to country in consideration of the local situation [34,35]. In the available literature, studies are seldom focused on the stability of the MSWI bottom ashes properties in relation to the different production period and procedure.

The aim of the present study is to identify the stability of the properties of two MSWI bottom ashes over a long time period and to analyze the MSWI bottom ashes in regard to their environmental impact. Additionally, their application potential as a building materials ingredient, such as clinker, cement and sand replacement, etc. is investigated.

In this study, the bottom ashes were provided by two waste-toenergy plants of Attero, which are located in Wijster and Moerdijk, in the Netherlands. The plant in Wijster started to be in operation in 1996, and includes three parallel processing lines with an annual capacity of around 800,000 tonnes of solid waste. The collected municipal solid waste firstly goes through a separation line to extract the recyclable materials, such as metals and certain plastics. Then, the modern techniques of film separation and NIR technology (near-infrared detection) together with air separation are applied to separate the plastic packaging materials. After the above pre-separations, the remaining waste is sent to the incineration boiler, the grid temperature of which is around 1000–1300 °C. The heat generated during the incineration converts the water into

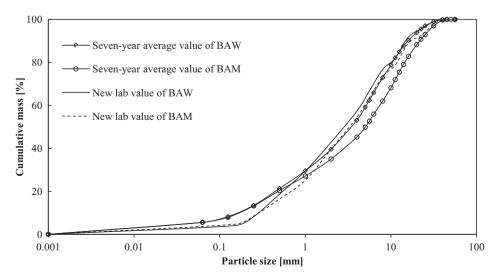


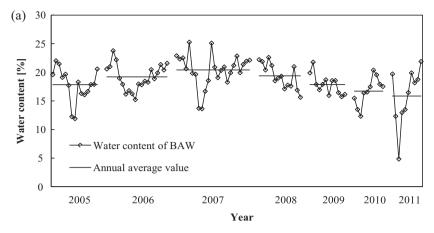
Fig. 1. The particle size distributions of BAW and BAM.

<sup>&</sup>lt;sup>b</sup> Building material that has not been molded.

<sup>&</sup>lt;sup>c</sup> Materials can be used only when isolation is applied.

 $<sup>^{</sup>m d}$  The measured cumulative leaching for a component per unit surface area over 64 days, in mg/m<sup>2</sup>.

e mg elements per kg of dry material.



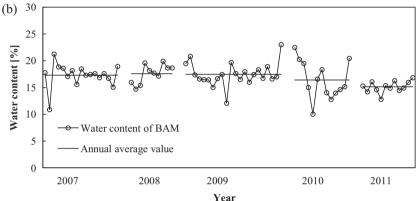


Fig. 2. The water contents of BAW (a) and BAM (b).

**Table 2**The annual average water absorption of BAW and BAM coarse fractions (4–31.5 mm).

24 h water absorption [%]	2006	2007	2008	2010	2011
BAW	-	-	13.23	10.82	11.44
BAM	8.4	7.9	-	8.1	9.8
NA [49]	1.4				
RCA [50,51]	4.1-4.2	6			

NA: natural aggregate, RCA: recycled concrete aggregate.

steam which is then used to generate electricity, which directly supplies the production process of some adjacent industrial companies. The electricity produced from the waste-to-energy plants annually can save more than 70 million cubic metres of natural gas. After the incineration, there is a considerable amount of ash coming out from the incinerator gate which is quenched in the water sink to cool down. Subsequently, the separations of ferrous and non-ferrous metals are conducted on the quenched ash, followed by the sieving out of particles above 31.5 mm. After this, the municipal solid waste incineration (MSWI) bottom ash is ready for further analyses.

The waste-to-energy plant in Moerdijk began to operate in 1997, and has an annual processing capacity of 1 million tonnes. Differently from Wijster, the Moderijk plant does not have the pre-separation of metals, paper and plastic, so the solid waste is mixed and directly incinerated at an average temperature of about 950–1100 °C. The following steps are separation of ferrous and non-ferrous metals.

In order to qualify and classify the bottom ashes from these two plants according to the Dutch legislation [30], and then suggest a possible application, the physical and chemical characteristics of the bottom ashes from Wijster (BAW) and Moerdijk (BAM) were

**Table 3**The densities of OPC, BAW and BAM fractions.

Particle size [mm]	Density [g cm <sup>-3</sup> ]	
	BAW	BAM
0-0.15	2.61	2.58
0.15-0.25	2.66	2.63
0.25-0.50	2.70	2.67
0.5–1	2.70	2.69
1–2	2.72	2.70
2–4	2.72	2.68
4-5.66	2.73	2.75
5.66-8	2.73	2.84
8-11.2	2.75	2.75
11.2-16	2.67	2.67
16–32	2.66	2.61
IBA [53]	$2847 \pm 159 \text{ kg m}^{-3}$	
CEM I 42.5 N	3.10	

IBA: incineration bottom ash.

firstly investigated. The historical test data on BAW and BAM provided by an accredited laboratory was collected and analysed to estimate the change of bottom ash properties with time. The potential of bottom ash fines (<2 mm) to be used as clinker addition and cement replacement after milling was studied here through oxide engineering and isothermal calorimetric measurements. Mortars with bottom ash fines (<2 mm) were prepared and tested to investigate the influence of bottom ash fines on the mortar properties. The environmental impact of the BAW and BAM was studied by a leaching test and 7-year leaching data since 2005 was analysed to determine the stability of leachable matter in the analysed bottom ash. In the end, further possible treatment and application methods were suggested.

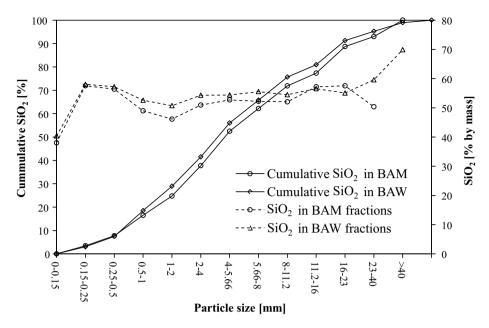


Fig. 3. The amount of SiO<sub>2</sub> in BAW and BAM fractions.

#### 2. Materials and methods

#### 2.1. Materials and their physical properties

The MSWI bottom ashes investigated in this study were provided by the waste-to-energy plants located in Wijster (BAW) and Moderdijk (BAM), as was mentioned before. Since the beginning of the operation of both plants, a periodical bottom ash quality evaluation was conducted. The representative bottom ashes samples were collected from different spots of the large bottom ash piles every 3 weeks and then sealed in plastic buckets before further testing.

The received bottom ash after weighting was dried in the oven at 105 °C until a constant mass was obtained and the water content was determined as described in [36]. The wet sieving test on the bottom ashes was conducted to determine the particle size distribution (PSD) according to EN 933-1 [37]. The sieving test on the oven dried bottom ashes was also performed to compare with the standard test, and then the specific density of the bottom ash particles of each sieved size fraction was measured using a Micrometrics AccuPyc II 1340 helium pycnometer.

The water absorption of the coarse bottom ash particles (4–32 mm) was measured according to EN 1097-6 [38]. The sample was firstly saturated in water at room temperature for 24 h, and then surface dried. After that, the saturated and surface-dried materials were dried further in a ventilated oven at  $110\pm5$  °C until a constant mass was reached. The water absorption was expressed as the mass percentage of the absorbed water per a mass unit of the dry material.

#### 2.2. Chemical properties

The X-ray fluorescence (XRF) analysis was performed using an Epsilon 3 device (PANalytical) with Omnian 3 analysing software, on pressed powder pellets. The X-ray diffraction (XRD) measurement was performed on each fraction of the bottom ash particles using a Rigaku Geigerflex X-ray diffraction spectrometer with Curadiation and a detection angle between 5° and 90°.

In cement production, the parameters normally used to define clinker characteristics based on the oxide composition are hydraulic ratio (HR), silica ratio (SR), alumina ratio (AR) and lime saturation factor (LSF) [39,40]. To evaluate the potential use of MSWI bottom ashes as a raw material for clinker production, the HR, SR, AR and LSF based on the chemical properties of investigated bottom ash samples were calculated as follows [40]:

$$HR = \frac{CaO}{SiO_2 + Al_2O_3 + Fe_2O_3} \eqno(1)$$

$$SR = \frac{SiO_2}{Al_2O_3 + Fe_2O_3} \tag{2}$$

$$AR = \frac{Al_2O_3}{Fe_2O_3} \tag{3}$$

$$LSF = \frac{CaO}{2.8 \times SiO_2 + 1.18 \times Al_2O_3 + 0.65 \times Fe_2O_3} \times 100$$
 (4)

These parameters calculated for the MSWI bottom ashes were compared with that for cement clinker production, as will be shown later in Table 5.

To analyse the reactivity of the bottom ashes, the cement hydration heat development with bottom ash was studied by isothermal calorimetry (eight-channel TAM Air, Thermometric). The bottom ash sample was milled and blended with OPC (CEM I 42.5 N) at various BAW or BAM ratios (0%, 10%, 20%, 30%, 40% and 100%) and the water to powder ratio was kept constant (w/p = 0.7) for all mixtures. After 5 min of manual mixing, the mixture was carefully transferred into the glass ampoule to avoid loading the sample on the upper and side walls of the ampoule. Then the sealed ampule with the sample was loaded into the isothermal calorimeter and the temperature was kept at 23 °C and the test was stopped after 5 days. The obtained data was analysed and compared with that of plain cement paste.

The bottom ash was heated at  $500\,^{\circ}\text{C}$  for at least 4 h and the mass percentage loss based on the dry mass was used as the loss on ignition (LOI). The ferrous components in the MSWI bottom ash were separated by a magnet, and the loose metallic iron components (such as cans, screws, nuts, nails, etc.) were picked out manually, and the amount was expressed as the mass percentage in the total bottom ash.

To quantify the metallic aluminium content in the bottom ash samples, the test method as described in [41] was applied. According to [42], the separation of metallic Al in coarse bottom ash fraction can be enhanced by using more efficient eddy current separators. However, the enhancement is not sufficient for fine bottom ash particles [43]. Therefore, prior to the application of fine bottom ash particles, the amount of metallic aluminium should be determined. In this study, the bottom ash fines were milled into powder before the measurement and three samples were tested for each bottom ash sample.

The amount of the heavy metals in BAW and BAM was determined periodically by an external accredited laboratory following the procedures given in NEN 6966 [44].

#### 2.3. Application of MSWI bottom ash in mortars

To investigate the influence of the fine bottom ash particles used as sand replacement (<2 mm) on the properties of mortar, the mortars were prepared following EN 196-1 [45]. The OPC:water:sand mass ratio was 2:1:3 for the reference sample, and then the sand was replaced by fine bottom ash particles by 10%, 20% and 30% by mass. The OPC used in this study was CEM I 42.5 N from ENCI (the Netherlands) and the sand is CEN normalised sand 0–2 mm according to EN 196-1 [45]. The water to cement ratio was kept at 0.5. Hardened mortar prisms  $(40\times40\times160~\text{mm}^3)$  were used for the determination of flexural and compressive strengths after 7, 28, and 91 days of water curing.

#### 2.4. Leaching test

To estimate the environmental impact of BAW and BAM resulting from a possible emission of harmful elements, the leaching test following NEN 7383 [46] was performed and the results were compared with the limit leaching values according to the Dutch legislation [30]. In order to evaluate the leaching behaviour of the elements in MSWI bottom ash samples from both plants from different production periods, the leaching data from 2005 to 2012 for both BAW and BAM were collected and analysed. The leaching test in this study was performed on the MSWI bottom

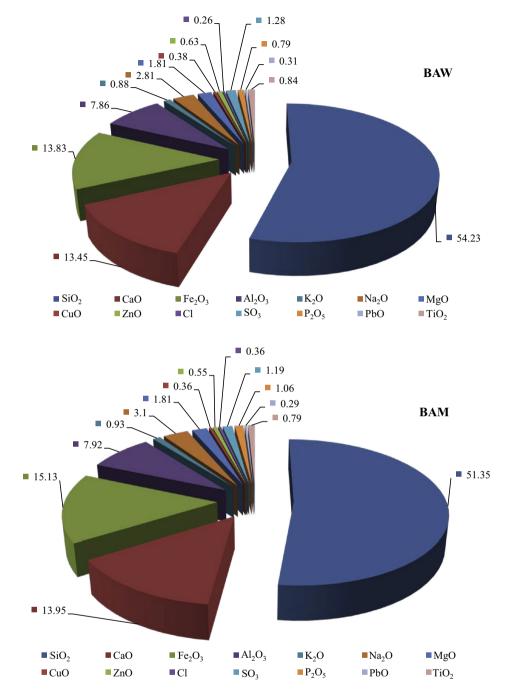


Fig. 4. The chemical compositions of BAW and BAM.

**Table 4**The dominant chemical compositions of bottom ashes and other materials.

Main chemical composition	BAW	BAM	CEM I 42.5 N [56]	SF [57]	WBA [58]	Fly ash [59]	CDW [60]	HCFA [60]
SiO <sub>2</sub>	54.2	51.4	19.4	41.3	49.4	27.2	38.1	33.1
CaO	13.4	13.9	62.1	16.4	14.7	16.4	29.7	36.2
Fe <sub>2</sub> O <sub>3</sub>	13.8	15.1	3.6	13.5	8.4	1.8	3.7	5.4
$Al_2O_3$	7.9	7.9	5.2	9.57	6.6	11.7	6.3	10.8

SF: sand fraction (0.1–2 mm) of MSWI bottom ash, WBA: natural weathered MSWI bottom ash, Fly ash: MSWI fly ash, CDW: construction and demolition waste, HCFA: high calcium fly ash from coal combustion power plant.

**Table 5**The calculated parameters used to define the characteristics of clinker.

Parameters	Recommended range [40]	0–2 mm		0-31.5 mm	
		BAW	BAM	BAW	BAM
HR	1.7-2.4	0.21	0.22	0.18	0.19
SR	1.7-2.7	2.56	2.10	2.50	2.23
AR	1.5-3.5	0.66	0.58	0.57	0.52
LSF	≥93%	9.36	9.98	7.91	8.56

ashes according to NEN 7383 [46], using a column leaching test with a liquid/solid ratio of 10 l/kg. The MSWI bottom ash was crushed to a particle size under 4 mm, and placed in the column. Water was forced to flow through the bottom ash particles from the bottom to top for a certain period. The concentration of chemical elements in the eluate was analysed using the ICP-AES according to NEN 6966 [44], and the amount of chloride and sulphate was determined through HPLC following NEN-EN-ISO 10304-2 [47]. According to the Dutch legislation – Soil Quality Decree (SQD) [30], the leaching of the inorganic elements should comply with the limiting values (Table 1) to protect the environment from the potential pollution from emissions from MSWI bottom ash

#### 3. Results and discussion

#### 3.1. Physical properties

The 7-year average particle size distributions (PSDs) data of BAW and BAM are shown in Fig. 1, together with the newly measured data. The PSD curves of both bottom ashes are relatively stable over a period of 7 years. Despite the different production processes, the PSD of BAW and BAM are very similar. Approximately 70% of the bottom ash particles are in the range

of 0.1–11.2 mm, which resembles the commonly used aggregate in concrete [48].

Fig. 2 depicts that the water content of BAW varies from 13% to 25% and the one of BAM from 12% to 23% in a period of 5–7 years. The various water contents of both bottom ashes result from the water quenching and open-air storage.

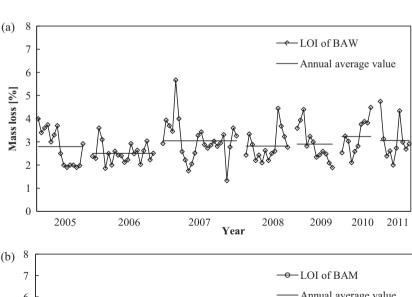
The annual average water absorption values for both bottom ashes are presented in Table 2. The data shows that BAW has a higher water absorption than BAM, which could be caused by the higher porosity of BAW or higher content of organic matter [12]. Compared with natural aggregates [49] and recycled aggregates [50,51] as shown in Table 2, the bottom ashes have higher water absorption value.

It can be observed from Table 3 that the densities of different particle size fractions of BAW and BAM are very comparable. The densities vary between 2.58 and 2.84 g cm<sup>-3</sup>, which are lower than that of cement and comparable to the densities of natural aggregates [52]. The particle density of each bottom ash fraction is slightly lower than the incineration bottom ash presented in [53], and the variation of the density is very small. The densities of particles between 0.5 and 11.2 mm are higher than other fractions; the possible explanation is the high amount of remaining metals, which is in line with [53].

#### 3.2. Chemical properties

#### 3.2.1. Chemical compositions

An example of the  $SiO_2$  distribution in different BAW and BAM size fractions is shown in Fig. 3. Other oxides (CaO, Al $_2O_3$ , Fe $_2O_3$ , CuO,  $SO_3$ ) and Cl content in the analysed bottom ashes are



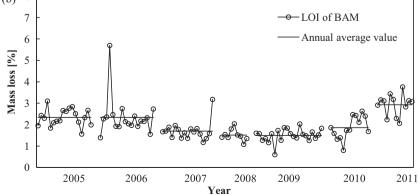


Fig. 5. The loss on ignition (LOI) of BAW (a) and BAM (b) at 500 °C.

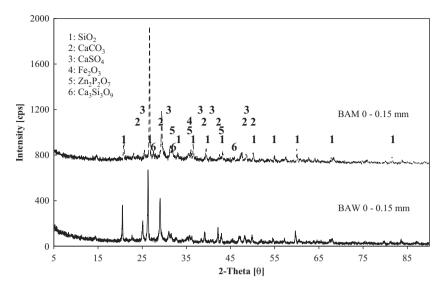


Fig. 6. The XRD patterns of BAW and BAM particles under 0.15 mm.

**Table 6**The crystalline phases in BAW fractions (\* represent the relative intensity of peaks).

Particles size [mm]	SiO <sub>2</sub>	CaCO <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	Ca <sub>3</sub> Si <sub>3</sub> O <sub>9</sub>	$Zn_2P_2O_7$	CaSO <sub>4</sub>
BAW 0-0.15 mm	****	***	*	_	***	***
BAW 0.15-0.25 mm	****	*	*	***	*	***
BAW 0.25-0.5 mm	****	*	*	**	*	***
BAW 0.5-1 mm	****	**	*	*	*	**
BAW 1-2 mm	****	**	*	*	*	**
BAW 2-4 mm	****	*	*	*	*	**
BAW 4-5.66 mm	****	*	*	**	**	*
BAW 5.66-8 mm	****	*	**	***	**	***
BAW 8-11.2 mm	****	*	**	*	*	*
BAW 11.2-16 mm	****	_	**	*	*	*
BAW 16-23 mm	****	*	**	*	*	*
BAW 23-31.5 mm	****	*	**	*	*	**

(\*\*\*\*: very high amount; \*\*\*: high amount; \*\*: moderate amount; \*: low amount).

**Table 7**The crystalline phases in BAM fractions (\* represent the relative intensity of peaks).

Particles size [mm]	SiO <sub>2</sub>	CaCO <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	Ca <sub>3</sub> Si <sub>3</sub> O <sub>9</sub>	Zn <sub>2</sub> P <sub>2</sub> O <sub>7</sub>	CaSO <sub>4</sub>
BAM 0-0.15 mm	****	***	*	***	***	***
BAM 0.15-0.25 mm	****	***	*	***	***	***
BAM 0.25-0.5 mm	****	**	*	*	**	***
BAM 0.5-1 mm	****	**	*	**	**	***
BAM 1-2 mm	****	***	*	***	***	***
BAM 2-4 mm	****	*	**	*	*	*
BAM 4-5.66 mm	****	*	*	**	***	**
BAM 5.66-8 mm	****	**	**	*	***	**
BAM 8-11.2 mm	****	**	*	*	**	**
BAM 11.2-16 mm	****	**	*	*	*	**
BAM 16-23 mm	****	*	*	*	*	*
BAM 23-31.5 mm	****	**	*	***	***	**

(\*\*\*\*: very high amount; \*\*\*: high amount; \*\*: moderate amount; \*: low amount).

presented in Appendix (Fig. A1). The amount of SiO<sub>2</sub> in each bottom ash particle size fraction was measured and calculated to the proportion of the mass of bottom ash fraction, and also calculated to the total amount of all the bottom ash fractions. In Fig. 3, the solid curves represent the cumulative distribution of the oxide in the bottom ash fractions (left *y*-axis), while the dashed curves show the content of SiO<sub>2</sub> in bottom ash fractions (right *y*-axis). The total chemical compositions of BAW and BAM are summarized in Fig. 4. It can be observed that the dominant oxides in both

bottom ashes are SiO<sub>2</sub>, CaO, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub>, which cumulatively account for around 88% of the bottom ash, which is in line with [54,55]. The finer particles contain higher amounts of CaO, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> and a lower amount of SiO<sub>2</sub>. Additionally, the distribution between the main oxides of BAW and BAM is very similar. This indicates that the main chemical composition is relatively stable, and is mostly independent of the raw solid waste before the incineration process. Compared with the dominant oxides in OPC [56] and other waste or secondary materials [57–60] as shown in Table 4, the bottom ashes in this study contain higher amounts of SiO<sub>2</sub> and lower amounts of CaO.

Based on the chemical properties of the bottom ash, the parameters for defining clinker characteristics were calculated and the results are shown in Table 5. It can be seen that the parameters HR, AR, LSF (defined previously in Section 2.2) of the bottom ash particles under 2 mm, as well the complete bottom ash PSD are under the required values (shown in Table 5), except for SR. Therefore, the bottom ash analysed in this study is not appropriate to be used as raw material for clinker production. However, the high amount of SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> indicates its potential to be used as a correction material [40] for the clinker production. Further research would be needed in order to prove if this potential can be fulfilled; a detailed study of the calcination process in the kiln is necessary in order to verify this possibility.

The loss on ignition (LOI) of BAW and BAM was measured at 500 °C to evaluate the amount of unburned organic matter residuals and the results are depicted in Fig. 5. It can be noticed that the LOI of BAM is more stable over time than that of BAW. Moreover, BAW has a higher annual average LOI value than BAM, which indicates less organic or burnable matter left in BAM. Interestingly, a pre-separation of paper and plastic from municipal solid waste was conducted in the waste-to-energy plant of Wijster, so it was expected that BAW should have a lower amount of unburned matter. This discrepancy could be explained by the contribution of paper and plastic to reach a higher incineration temperature and better contact between the heat source and the incinerated mass, which eventually decomposes more organic matter.

#### 3.2.2. Mineralogical properties

The minerals identified in bottom ashes by XRD in literature were mainly quartz  $(SiO_2)$ , calcite  $(CaCO_3)$ , anhydrite  $(CaSO_4)$ , gehlenite  $(Ca_2Al_2SiO_7)$  and magnetite or hematite  $(Fe_2O_3)$ , with quartz and calcite being the major crystalline phases [57,61]. In

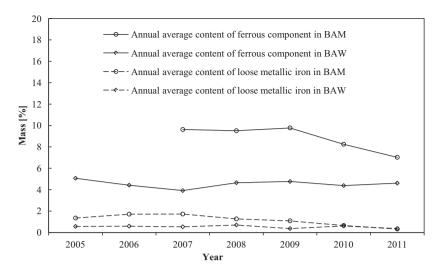


Fig. 7. The amount of ferrous metals and loose metallic iron in BAW and BAM.

**Table 8**The amount of metallic aluminium in BAW and BAM particles under 2 mm.

	Mass	Volume of H <sub>2</sub>	Amount of	metallic Al
	[g]	[ml]	[mg]	[% wt.]
BAW	5.30	44.80	38.64	0.73
	5.54	45.90	39.58	0.71
	6.20	50.30	43.38	0.70
BAM	5.01	21.50	18.54	0.37
	5.48	23.00	19.84	0.36
	5.65	24.50	21.09	0.37

this study, the diffractograms of bottom ash particles under 0.15 mm of BAW and BAM are shown in Fig. 6 as an example and the relative intensities of the relevant peaks of other bottom ashes fractions are shown qualitatively in Tables 6 and 7.

The analysis of the XRD results of BAW shows that the main crystalline phases are SiO<sub>2</sub>, CaCO<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, Zn<sub>2</sub>P<sub>2</sub>O<sub>7</sub> and CaSO<sub>4</sub>. In all the fractions, SiO<sub>2</sub> is the main crystalline phase. The particles under 0.15 mm of BAW contain relatively high amounts of CaCO<sub>3</sub>; Fe<sub>2</sub>O<sub>3</sub> can be found in all the fractions of BAW and particles above 5.66 mm contain relatively more Fe<sub>2</sub>O<sub>3</sub>, while the particles under 2 mm have a higher amount of CaSO<sub>4</sub>. The main crystalline

phases for BAM fractions are  $SiO_2$ ,  $CaCO_3$ ,  $Fe_2O_3$ ,  $Ca_3Si_3O_9$ ,  $Zn_2P_2O_7$  and  $CaSO_4$ . The particles under 2 mm of BAM seem to contain large amounts of  $CaCO_3$  and  $CaSO_4$ . The variation could be attributed to the hardness of the crystalline phases. For instance, the  $CaSO_4$  and  $CaCO_3$  are softer than  $SiO_2$  and  $Fe_2O_3$ , therefore the coarse fractions contain higher amounts of  $SiO_2$  and  $Fe_2O_3$ , while  $CaSO_4$  and  $CaCO_3$  can be crushed during storage and transport and will be found mainly in the finer fractions.

#### 3.2.3. The content of metallic matter

The amounts of the ferrous components and loose metallic iron in the investigated bottom ashes are shown in Fig. 7. It can be noticed that the amount of the ferrous component and loose metallic iron in both BAW and BAM is quite stable in time. The BAM contains a higher amount of both ferrous metals and loose metallic iron than BAW. This result is in line with the chemical compositions as shown in Section 3.2.1 – BAW has a lower amount of Fe<sub>2</sub>O<sub>3</sub> than BAM. The above mentioned differences between BAW and BAM could be explained as follows: the pre-separation of recyclable material before incineration in Wijster increases the recycling efficiency of metals. Hence, an enhancement of iron recycling in Moerdijk plant could also be necessary for improving the quality of the bottom ash.

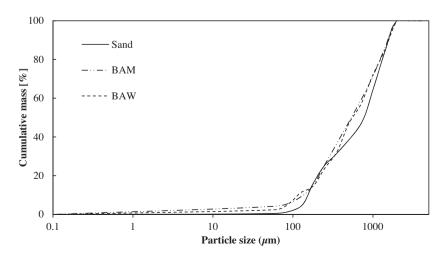
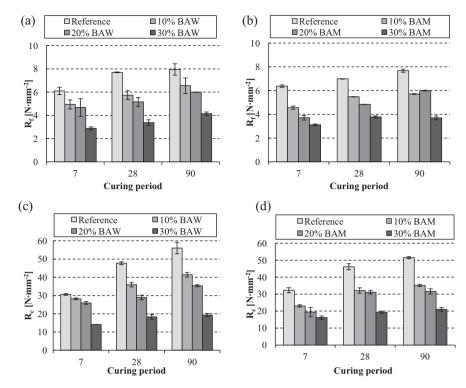
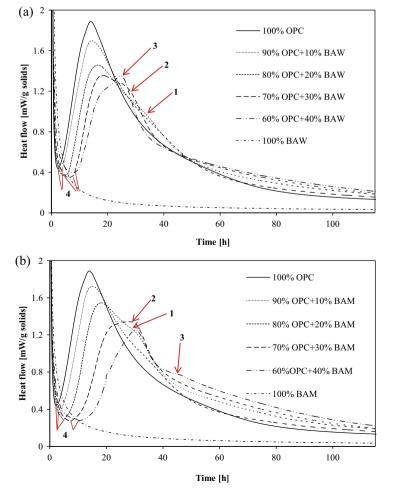


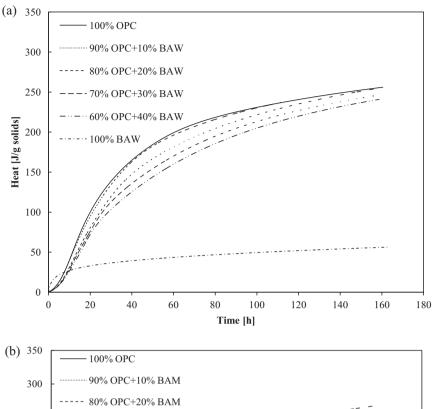
Fig. 8. The particle size distribution (PSD) of standard sand and fine bottom ashes (<2 mm).

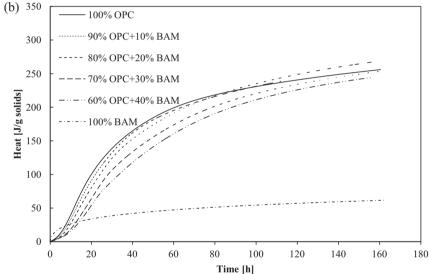


 $\textbf{Fig. 9.} \ \ \textbf{The flexural} \ (R_f) \ \text{and compressive} \ (R_c) \ \text{strengths of mortars with BAW (a and c) and BAM (b and d) as sand replacement.}$ 



**Fig. 10.** Heat flow normalised to the mass of solid at 23 °C with a w/b ratio of 0.7.





**Fig. 11.** Released heat normalised to the mass of solid at 23 °C with a w/b ratio of 0.7.

The amount of metallic aluminium in bottom ash particles under 2 mm of BAW and BAM is depicted in Table 8. The particles under 2 mm of BAW contain more metallic aluminium, around twice as much compared to BAM. However, the XRF results show that BAW and BAM have quite similar amounts of equivalent Al<sub>2</sub>O<sub>3</sub> (Fig. 4 and Appendix). Thus, it can be concluded that the aluminium in BAW particles under 2 mm has a lower level of oxidation. Previous research addressed that the oxidation level of metallic aluminium in MSWI bottom ash and the small size of non-ferrous scraps limit the recycling of aluminium; hence, the production of energy from H<sub>2</sub> generated by the chemical reaction between metallic aluminium with a high pH solution is more beneficial [62,63]. Therefore, how to recover the metallic aluminium from the fine bottom ashes in the form of hydrogen in a practical way is more important and interesting than enhancing the separation of metallic aluminium by eddy current separator.

#### 3.3. Properties of hardened mortars

The bottom ash particles under 2 mm have a very similar particle size distribution (PSD) and density with sand as shown in Fig. 8 and Table 3. Therefore, in this study the bottom ash particles under 2 mm were applied in mortar as sand replacement to investigate their effect on the mortar properties. The flexural and compressive strengths of mortar with various ratios of BAW or BAM after several periods of curing are shown in Fig. 9.

It can be observed from Fig. 9 that the use of both bottom ashes decreases the flexural and compressive strengths, and the reduction of the strengths increase with an increasing amount of the bottom ash, as shown in Fig. 9a. The decrease of the flexural and compressive strengths of mortars with bottom ashes is about 20–50%, depending on their dosage. Similar results were reported in [18,64]. No visible cracks or swelling were observed in the samples in this study, which indicates that the decrease of the strength may

**Table 9**Total heat released per gram of solid after 111.4 h of hydration. The sample of pure OPC is used as reference for all the other samples.

-	Solid	OPC in sample/ OPC in	Cumulative heat after 111.4 h [J/g solids]	Ratio of reference [%]
-		reference fatio		
	OPC	1	236.65	100.00
	10% BAW	0.9	236.98	100.14
	20% BAW	0.8	229.80	97.10
	30% BAW	0.7	221.51	93.60
	40% BAW	0.6	214.02	90.44
	100% BAW	0	51.00	21.55
	10% BAM	0.9	238.31	100.70
	20% BAM	0.8	243.27	102.80
	30% BAM	0.7	228.70	96.64
	40% BAM	0.6	220.03	92.98
	100% BAM	0	56.01	23.67

**Table 10**The heavy metal concentration of BAW and BAM.

Element	BAW		BAM				
	Range of metal concentration (2005–2011)	Average	Range of metal concentration (2005–2007)	Average			
	mg/kg d.m.		mg/kg d.m.	mg/kg d.m.			
Antimony (Sb)	44-160	71.3	25–56	42.8			
Arsenic (As)	5.6-10	8.55	4.3-8.3	6.68			
Beryllium (Be)	<2	<2	<2	<2			
Cadmium (Cd)	4.6-43	11.46	5.6-74	21.7			
Chromium (Cr)	130-250	175	140-300	186			
Cobalt (Co)	15-53	22.2	14-28	21			
Copper (Cu)	2600-11,000	4620	2700-3900	3280			
Mercury (Hg)	<0.05	< 0.05	<0.05	< 0.05			
Lead (Pb)	750-2200	1374	990-2500	1438			
Manganese (Mn)	370–1600	744	550-990	782.5			
Molybdenum (Mo)	5.5-9.3	7.8	8–11	9.24			
Nickel (Ni)	120-190	149	110-210	162			
Selenium (Se)	<2	<2	<2	<2			
Vanadium (V)	21-27	23.7	21-30	24.2			
Zinc (Zn)	2300-5200	3160	1800-3200	2480			

not be caused by the metallic aluminium in bottom ash. The possible explanation of this phenomenon is that during the mixing of the mortar, part of the water was absorbed by the bottom ash particles due to their high water absorption (as shown in Fig. 8 the BAW and BAM have more fine particles under 125  $\mu m$ ). Thus, the cement hydration progressed less, resulting in the reduction of the strength [65].

## 3.4. The influence of milled BAW and BAM fines (<2 mm) on cement hydration

Fig. 10 shows the development of the heat flow during the hydration of OPC, for OPC with various replacement ratios of BAW and BAM, normalised to the total powder content.

As described in [66], the hydration of OPC can be divided into four stages: the initial, the induction, the acceleration, and the deceleration stage. Fig. 10a and b show that the plain BAW and BAM did not show typical hydration curves as OPC, and their heat release rates tend to be stable after 40 h. This indicates that the BAW and BAM have lower hydraulic activities; this result is in line

with the very low HR value shown in Table 5. The low hydraulic property of both bottom ashes is due to the existence of high amount of crystalline phases, such as SiO<sub>2</sub> and CaCO<sub>3</sub> (Fig. 6) [57].

Fig. 10 shows that the profiles of the heat release rate during cement hydration of samples with cement and BAW or BAM are very similar to each other. For the cement samples mixed with BAW or BAM the last three stages of cement hydration are retarded and each reaction stage is extended. The more BAW or BAM is added, the longer the retardation effect is observed on cement hydration. For samples with 20%, 30% and 40% BAW or BAM, there is a heat release peak observed after about 26 h of hydration, as shown in Fig. 10, position 1, 2, 3 on different curves in the deceleration stage of cement hydration. According to [66,67], these peaks refer to the dissolution of  $C_3A$  and the precipitation of ettringite, which is defined as "sulphate depletion peak" because of the participation of sulphate to the reaction. Therefore, it can be concluded that adding the bottom ash enhances the reaction of sulphate.

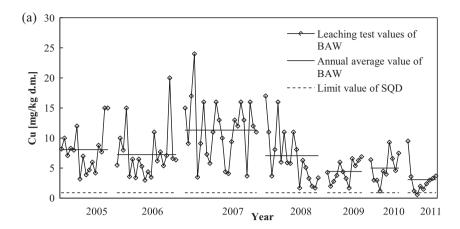
The observed influence of bottom ashes on the cement hydration can be explained as follows: (a) the heavy metals such as Cu, Pb, Zn, etc. may coat the cement particles, which disturbs the cement hydration [68]; (b) the bottom ash contains a certain amount of organic matter, mainly fulvic and humic acids as reported by Arickx et al. [69], which may have a retardation effect on cement hydration; (c) the high amount of CaSO<sub>4</sub> present in bottom ashes enhances the sulphate participation in the deceleration stage of cement hydration.

Fig. 11 shows the cumulative heat released by the samples with BAW or BAM, compared with pure OPC. The cumulative heat generated by the samples was computed starting from the inflexion point when the heat flow starts to rise again after the initial drop, in order to reduce the influence of sample preparation [70] (in Fig. 10, position 4).

Fig. 11 also reveals that the total heat release from the samples with BAW or BAM after the same hydration time is lower than pure OPC and decreases with increasing amount of bottom ash. The total heat released per gram of blended solid is shown in Table 9 after around 111.4 h hydration. Samples with plain BAW or BAM have the lowest total generated heat. It is known that the reaction of metallic aluminium sample with water or other solution with auxiliary alkali generates 50–70 kJ/mol of heat [71]. Therefore, it can be calculated that the metallic aluminium can produce 13–18 J or 7–9 J of heat per gram of BAW or BAM, respectively. Thus, there is about 25–35% and 13–16% of the total heat released from samples of plain BAW or BAM, respectively, due to the reaction of aluminium. The other part the total heat released can be attributed to the hydraulic reaction of BAW or BAM.

It can be observed in Table 9 that the samples with 10% BAW or BAM and 20% BAM achieve higher cumulative heat than pure OPC, which indicates the contribution of bottom ashes to cement hydration. For samples with 30% and 40% BAW or BAM, the generated heat decreases. However, compared with the replacement level, the reduction of cumulative heat is less (e.g. 20% BAW with 80% OPC generated relative 97.10% heat of the reference, instead of 80%), which indicates the contribution of bottom ashes.

The calorimetric results show that the BAW and BAM have low hydraulic activities, and their addition to cement leads to the retardation of cement hydration. 10% replacement of cement by bottom ashes can still have comparable or even higher total heat release, hence, future studies can focus on its application in concrete. However, for higher replacement levels, the activity of bottom ashes should be increased in order to reduce its influence on cement hydration. Moreover, proper treatments should be considered in order to reduce the retarding effect of bottom ashes on cement hydration.



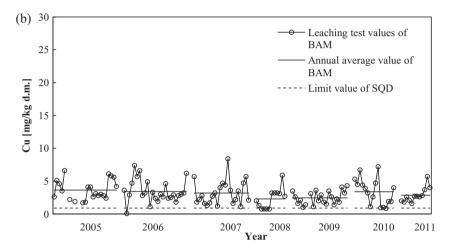


Fig. 12. The leaching data of copper of BAW and BAM from 2005 to 2011.

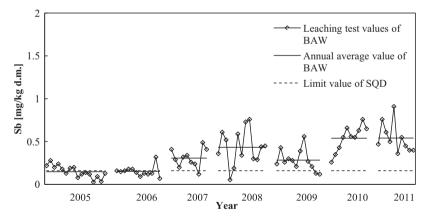


Fig. 13. The leaching data of antimony of BAW from 2005 to 2011.

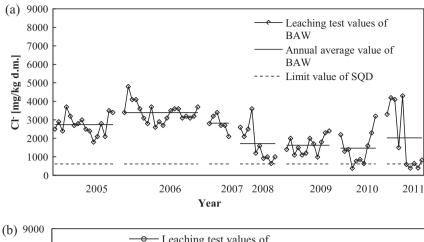
#### 3.5. Leaching behaviour

The amounts of heavy metals in BAW and BAM are shown in Table 10. The concentration of the heavy metals in BAW and BAM is relatively similar, while BAW has a slightly higher content of antimony and copper than BAM.

The comparison of the leaching data with the limit values (in Table 1) from SQD [30] indicates that the BAW and BAM are well under the IBC building material requirements. Hence, both bottom ashes could be potentially used as roadbase material with insulation and control measures. Nevertheless, when compared

with non-shaped material emission limit values, the emission of the heavy metals (shown in Table 1) are all well under the limit value of the non-shaped category, except antimony and copper for BAW and copper for BAM. In addition, the leaching of chloride and sulphate in both BAW and BAM exceed the relevant limit value. Hereby, the leaching data of these components are illustrated in Figs. 12–15.

The maximum copper leaching value for non-shaped building materials is 0.9 mg/kg dry matter (d.m.) (Table 1). It can be seen in Fig. 12 that the leaching of copper from BAW and BAM varies and generally exceeds the limit value most of the time.



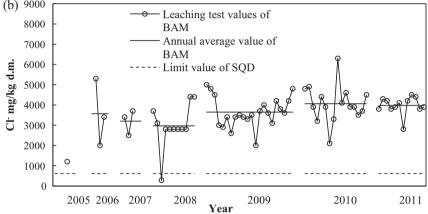


Fig. 14. The leaching data of chloride of BAW and BAM from 2005 to 2011.

Furthermore, the copper leaching from BAW is relatively higher than that from BAM. The annual average leaching value of Cu in BAW is approximately 3–12 times the limit value, while for BAM is 2.5–4 times the limit value.

Fig. 13 shows the antimony emission from BAW in the column leaching test. The antimony leaching for BAM is always under the limit emission value, hence, only the leaching of BAW is shown here. It can be seen that the antimony leaching in BAW fluctuates along with time and the annual average value has a slight increasing trend since 2005 (around 1–3.4 times the limit value).

The leaching of chloride from BAW frequently exceeds the limit value (Fig. 14a). The annual average emission value is about 2.4–5.5 times the limit value, and since 2008 the value is lower than in previous years. Compared with BAW, BAM has a higher leaching value of Cl<sup>-</sup> and the annual average value is around 4.8–6.6 times the limit value (Fig. 14b).

The leaching of sulphate from BAW and BAM is shown in Fig. 15a and b, respectively. The sulphate emission from BAW always exceeds the limit level, and has an increasing trend since 2006. The annual average value is approximately 1.7–5.7 times the limit value. The sulphate leaching of BAM seems lower than that of BAW, and the maximum annual average value is 3.6 times the limit.

It can be noticed from the above analysis that, for using the bottom ashes as non-shaped material, the problematic leaching elements are very similar. Moreover, the emission level of copper, antimony, sulphate of BAW is higher, while BAM has higher chloride emission levels.

To understand the factors contributing to the above mentioned leaching differences between BAW and BAM, and initially provide the potential treatments, the leachability of the problematic contaminants were determined based on their total amount in bottom ashes and the amount released in the leachate (Table 11).

It is described in Table 11 that BAW contains a higher amount of Sb, and its average value is also higher than that of BAM. The leachability of antimony in BAW is 1.8 times that in BAM, which indicates that antimony in BAW is more leachable than in BAM. Hence, if a water washing treatment is applied to reduce the amount of Sb in bottom ash, it could be expected that BAW would have a better washing efficiency. According to previous research, the leaching of copper from MSWI bottom ash is related to the organic matter content [72]. Hence, the slightly higher leachability of copper in BAW than in BAM could be explained as a result of a higher amount of organic matter. This is further proved by the higher LOI value of BAW than BAM in this study (Fig. 5).

The available data of the total amount of chloride and sulphate in BAW and BAM is limited. However, the data shown in Table 11 briefly indicates the extremely high leachability of the chloride and sulphate in both bottom ashes. Hence, the washing efficiency could be expected to be the highest among other leachable elements in bottom ash. This is in line with the findings presented in other studies [73].

The study on the leaching behaviour of BAW and BAM along with time shows that the leaching of most elements are well under the limit value of the legislation, only copper, antimony, chloride and sulphate exceed the limit. For both BAW and BAM, despite the different raw solid waste and different incineration method, the leaching contaminants are the same. Hence, future studies should focus on the investigation of influential factors on copper, antimony, chloride and sulphate leaching from the bottom ash, and then potential treatments can be suggested.

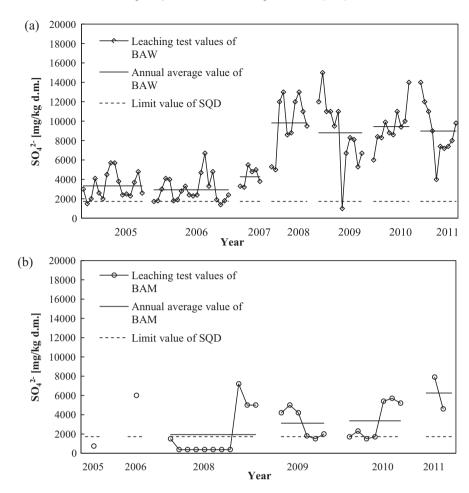


Fig. 15. The leaching data of sulphate of BAW and BAM from 2005 to 2011.

Table 11 The leachability of Sb, Cu,  $Cl^-$  and  $SO_4^{2-}$  in BAW and BAM.

Components [mg/kg d.m.]	Sb		Cu		Cl <sup>-</sup>		$\mathrm{SO}_4^{2-}$	
	BAW	BAM	BAW	BAM	BAW	BAM	BAW	BAM
Total amount	44-160	25-56	2600-11,000	2700-3900	2600/(2400 <sup>a</sup> )	3600	5100/(6100 <sup>a</sup> )	4700
Average value	71.30	42.80	4620	3280	_	_	=	_
Leaching value	0.055-0.76	0.08-0.16	1.2-20	0.99 - 8.4	380-4300/(3000 <sup>a</sup> )	280-6300	$1000-15,000/(5700^{a})$	380-7900
Average value	0.37	0.12	7.37	4.18	-	_	_	_
Leachability [%]	0.11-1.33	0.19-0.52	0.03-0.54	0.03-0.22	$-/(1^a)$	_	$-/(0.9^{a})$	_
Average value	0.59	0.33	0.18	0.12	_	_		_

<sup>&</sup>lt;sup>a</sup> The data obtained.

#### 4. Conclusions and recommendations

This paper presents the characteristics of MSWI bottom ashes from two waste-to-energy plants with different facilities in the Netherlands. The leaching behaviour of both bottom ashes was investigated by analysing 7 years of leaching data. The application potential of the bottom ash fines (<2 mm) was studied experimentally. From the results presented in this study, the following conclusions can be formulated:

- The physical and chemical properties of the bottom ashes from both waste-to-energy plants were stable over time and were very similar with each other, despite different production processes. The coarse bottom ash particles (4–32 mm) had higher water absorption than natural aggregate and recycled concrete aggregate.
- The dominant oxides found in the bottom ashes are SiO<sub>2</sub>, CaO, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub>. Based on the calculations of relevant parameters, these bottom ashes could possibly be used as correction material for clinker production. This possibility needs further research.
- The calorimetric results confirmed that the bottom ashes had very low hydraulic activity and their addition to cement led to the retardation of cement hydration. 10% cement replacement by bottom ashes can reach comparable cumulative heat, while more than 20% replacement led to the decrease of total heat release. However, the addition of bottom ashes contributes to the total heat release of cement hydration.
- $\bullet$  The MSWI bottom ash has a high amount of fine particles (<125  $\mu m)$  which leads to higher water absorption and eventually reduces the amount of water available to react with cement and metallic aluminium in mortar. Hence, the bottom

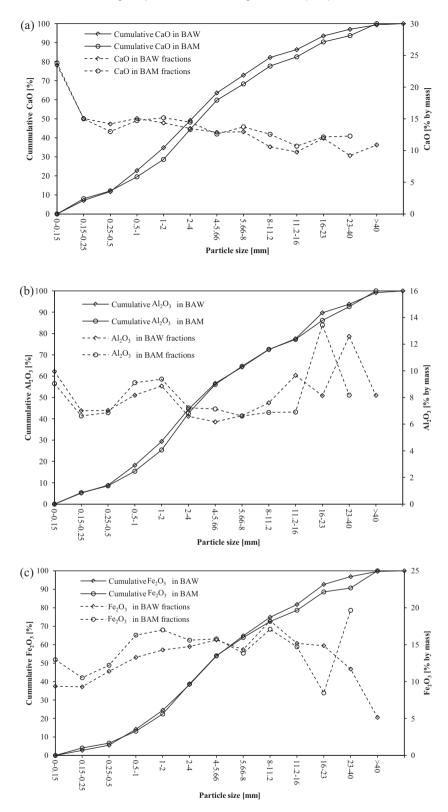


Fig. A1. The amount of CaO, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, CuO, SO<sub>3</sub>, Cl in BAW and BAM fractions.

ashes fines (<2 mm) used as sand replacement reduce the strength of mortars, but with no visible cracks on the mortar. Thus, zero-slump or roller-compacted concrete with low w/c ratio and higher porosity can be an option for the use of bottom ash as aggregate to avoid cracks.

• The leaching behaviour of bottom ashes was very stable over time. The leaching contaminants which exceed the legislative limit were copper, antimony, chloride and sulphate. Thus, a future study will be focused on the reduction of these specific contaminants from the investigated bottom ashes.

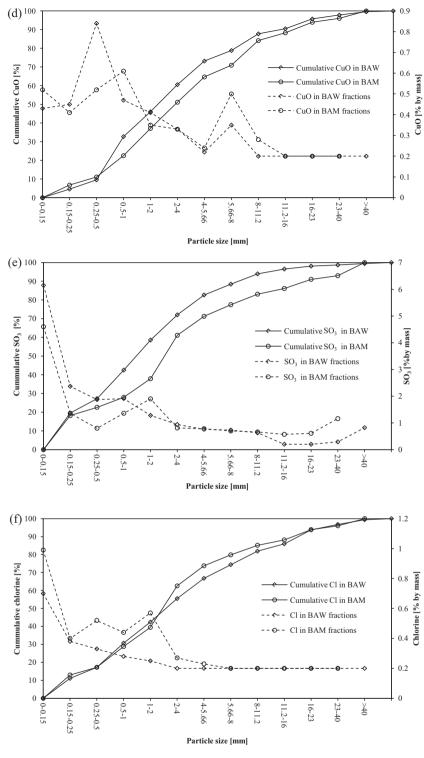


Fig. A1 (continued)

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Benelux, Kijlstra Betonmortel, Struyk Verwo, ENCI HeidelbergCement Benelux, Rijkswaterstaat Zee en Delta-District Noord, Van Gansewinkel Minerals, BTE, V.d. Bosch Beton, Selor, GMB, Geochem Research, Icopal, BN International, Eltomation, Knauf Gips, Hess AAC Systems, Kronos, Joma, CRH Europe Sustainable Concrete Centre, Cement&BetonCentrum, Heros, Inashco (in chronological order of joining).

#### Appendix A

See Fig. A1

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