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Life cycle assessment of the reuse of fly ash from biomass combustion as secondary cementitious material in cement products

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Abstract

In this study, we performed a life cycle assessment of the reuse of biomass fly ash as secondary cementitious material in cement mortars as alternative to a reference landfill scenario of the ash. Since biomass ash does contain enhanced levels of elements that are of potential concern for the environment or human exposure, the performed Life Cycle Assessment (LCA), in addition to CO₂ savings, takes into account the impact on all non-toxic categories and human toxicity/carcinogenicity during service and second life stages. Results showed that utilization of biomass ash in cement is preferable over landfill for all the non-toxic categories at both cement replacements rates of 20 and 40 wt.%. In detail, the reduction of CO₂-eq. was found to be between 11-26 % when biomass ash was blended with cement instead of being landfilled. The hydraulic activity of biomass ashes was found to be a critical parameter in this scenario, as it had impacts on the global warming potential (and all other investigated non-toxic categories), and it is therefore crucial to consider the uncertainty related to this aspect in LCA studies. Cement containing biomass ash performed better, on average, when compared with the reference landfill scenario regarding the impact to human toxicity (carcinogenic) category. Contrary, only the utilization in cement for one particular ash type (from paper sludge combustion) showed a better performance than the reference scenario for the ecotoxicity (ET) category. The impact to human toxicity carcinogenic (HTc) and ecotoxicity (ET) was mainly dominated by the leaching of Cr from landfilling of pure biomass fly ash (reference scenario)

and the leaching of Ba, Cu, Cr (VI) and Zn from the second life stage of cement products (i.e., reuse of the crushed cement after service life in road base applications). However, this impact was acceptable when emissions are compared to existing EU landfill directive and regulations on the reuse of secondary materials in construction works. The novel LCA approach performed in this study, which includes impacts of leached contaminants during both the service and second life phase of cement, has shown that the reuse of biomass ash as secondary cementitious materials has a beneficial effect on the majority of the impact categories, with no unacceptable leaching risks.

Keywords: Biomass fly ash, Cement, Life cycle assessment (LCA), Leaching, Second life

1. INTRODUCTION

Cement production accounts for about 5-8% of total anthropogenic CO₂ emissions, mainly due to the conversion of CaCO₃ (limestone) into calcium oxide and the combustion of fossil fuel during heating of the raw material mixture. Many efforts have been made to reduce these emissions, including the (partial) substitution of the traditional Portland cement with alternative cementitious materials such as blast furnace slags and fly ash from coal combustion (Barcelo et al., 2013). The growing use of biomass for sustainable energy production and the corresponding large amount of biomass ash produced, has initiated investigations of the substitution of traditional Portland cement with biomass ash residues (Rajamma, 2009; Siddique, 2012). A scenario study has been conducted to determine volumes and characteristics of biomass ashes in the Netherlands for the 2020-2030 timeframe (Boersma, 2011; Saraber and Overhof, 2010). In this study, emphasis has been put on biomass residues from co-firing and stand-alone biomass combustion/gasification technologies. The analysis forecasts an ash volume from co-combustion and stand-alone biomass plants of 2-3 Mton/year in 2030. These biomass conversion plants are estimated to achieve a growth of up to 125 PJ by 2020, which involves around 6 Mton of biomass combusted per year. The utilization of biomass ash as alternative binder in traditional cements has been identified as bulk application with a potential profitable market in a long term perspective (Paris et al., 2016; Pels and Sarabèr, 2011; van Eijk et al., 2012). This application could contribute substantially to the reduction of greenhouse gas emissions from cement production (Imbabi et al., 2012; Meyer, 2009; Rahla et al., 2019). The inclusion of biomass ash in cement would also result in two additional beneficial effects: 1) reduction of the consumption of energy and raw materials by the cement industry, 2) reduction in the direct landfilling of the biomass combustion residues. On the other hand, some potentially toxic elements present in the biomass ash might be released in the environment during the different life stages of the fly ash containing cement (e.g. reuse of the cement aggregates after initial use in structural applications) (Tosti et al., 2019).

Life cycle assessment (LCA) is increasingly being used as a tool to assess the environmental impacts associated with different management options for waste and residues (Laurent et al., 2014) including valorisation of biomass ash residues in cement products (Teixeira et al., 2016). However, the impact assessment of inorganic contaminants released by leaching from ash residues and cement remains a difficult task and is often neglected in LCA studies (Van den Heede and De Belie, 2012). A few studies (mainly using MSWI bottom ash) have addressed the environmental impact of leaching through LCA based on results from field or laboratory leaching tests (Allegrini et al., 2015; Di Gianfilippo et al., 2016; Olsson et al., 2006). Barbosa et al. (2013) concluded that formulations containing biomass ash presented emission levels of chemical species similar to those observed for the reference formulation and reduced ecotoxicological levels. In general, these studies have shown that the leaching behaviour of materials under different conditions may have an important influence on the LCA results and, therefore, should be considered when choosing between different management strategies.

The aim of this work is i) to compare the potential impacts associated with the current waste management practice for biomass ash (reference scenario, i.e. landfilling) and the potential of reuse as a secondary cementitious material in cement, ii) to identify the critical parameters of the modelled systems (i.e. substance emissions and processes contributing to impacts), and iii) to identify the sensitive parameters in terms of methodological choices, assumptions and data selection with regard to final results. The novelty of this work is that the use of biomass fly ash in cement is not only assessed for the use scenario in cement applications but also in the subsequent life phase when the demolition waste is reused as a sub-base material in road constructions. The results of this study also provide new insights in the methodological setup of LCA scenarios for the assessment of alternative materials to replace cement.

2. MATERIALS AND METHODS

2.1. Biomass fly ash

Three biomass fly ashes (FA1, FA2 and FA3) were investigated in this study. The samples had a particle size <1 mm and were stored dry in the laboratory. Sample FA1 originated from a circulating fluidized bed installation that combusts a mixture of clean wood and cacao husks, molasses or other clean biomass streams that were occasionally added. The sample FA1 was collected from the electrostatic precipitator. The sample FA2 originated from the combustion of wood pellets in a pulverized fuel installation and was collected from the electrostatic precipitator. The sample FA3 was taken from a bubbling fluidized bed incinerator. The fuel consisted of a mixture with an equal share of recovered paper sludge

from the de-inking step of the paper recycling process and recovered waste wood. The fly ash was a mixture collected from the electrostatic filter (90% by mass) and the textile bag filter (10% by mass) cleaning system units. The investigated biomass ashes cover a fairly wide range of biomass fuels and conversion technologies and can, therefore, be considered representative for future biomass ash use in cement products.

2.2. Blended cement mortars

Mortar samples were prepared by dry mixing the Portland Cement CEM I 42.5N with FA in its “as received” form in accordance with the European standard EN 196-1 (2005). Ordinary Portland cement was replaced with both 20 and 40 wt. % of FA1, FA2 or FA3. These combinations resulted in six test samples that are referred to in this paper as 20_FA1, 40_FA1; 20_FA2, 40_FA2 and 20_FA3 and 40_FA3, respectively. In addition, a reference sample of pure Ordinary Portland Cement was also casted (OPC). Prisms (160 x 40 x 40 mm) were cured for 28 days in a controlled temperature and humidity room (at 20 °C and 95% humidity). All specimens were prepared with a water-binder weight ratio (w/b) of 1:2 and a sand-binder ratio of 3:1. After curing the specimens were subjected to compressive strength (EN-196-1, 2005) and leaching tests (see Section 2.3 for more details).

2.3. Leaching tests

Results from standardized leaching tests were used as input for the LCA model in order to estimate the potential release of contaminants during the proposed management scenarios. In particular two different tests were used in this work: tank leaching tests (FprCEN/TS 16637-2, 2013) and parallel batch extraction tests at different liquid to solid (L/S) ratios (EPA, 2012). In short, results from the tank leaching tests were used as input to model the service life of cement (i.e. release from monolithic material), whereas results from the batch tests were used to model the landfill and end of life scenario (i.e. release from granular materials at different L/S). Section 2.5 and Sections 3, 4 and 5 of the Supplementary Material provide for more details on the modelling aspects. A detailed description of the mentioned tests and the obtained results is reported in Tosti et al. (2018).

2.4. LCA methodology

2.4.1. Goal, scope and model

This study was conducted according to the requirements of ISO 14044 (ISO, 2006) and the ILCD Handbook (EC-JRC, 2010). The goal of the study is to compare the potential impacts associated with the current waste management of fly ash from biomass combustion (i.e. landfilling) with its potential reuse as secondary cementitious material (SCM) in cement

formulations as an alternative management strategy. In particular, the analysis aims at verifying whether the expected CO₂ reductions from using biomass ash in cement are also supported by a similar environmental performance as ordinary cement applications throughout the entire life cycle. A reference scenario in which biomass ash is landfilled and traditional Portland cement is used was compared to an alternative scenario in which landfilling is avoided and a blended cement (containing biomass ash) is used. Due to the nature of the alternative scenario, this study has a goal encompassing two services: 1) the handling, treatment and/or use of one ton of biomass ash and 2) the production of cement material (i.e. pure or mixed with biomass ash). Cement replacement with biomass ash was investigated at two levels: 20 and 40 wt. % of total binder weight. Therefore, two functional units are defined for the application of 1 ton of biomass fly ash, as collected from the incineration plant, in the production and use of cement: FU₂₀ for producing, using and re-using a total of 5 ton of cement when biomass ash is used to substitute 20% of cement, and FU₄₀ for 2,5 ton of cement (with 40% substitution). Since these formulations result in a lower compressive strength than that of pure OPC (Tosti et al., 2018), additional OPC was accounted for in order to reach the same compressive strength for all formulations, as further explained below. Biomass fly ash is considered to enter the system without any impacts in agreement with the zero burden assumption (Ekvall et al., 2007). A temporal horizon of 100 years was selected for the analysis and The Netherlands was adopted as reference geographic area for the model assumptions in particular for the landfill scenario. Even though the impact from the recycling of ashes might be higher in a longer time horizon, the fate of products after second life stage and at timeframes of over 100 years is highly uncertain and, therefore, disregarded in this study. The Life cycle impact assessment (LCIA) included the midpoint impact categories recommended by the European Commission in the International Reference Life Cycle Data System (ILCD) Handbook (Hauschild et al., 2013), as outlined in Table S1 of the Supplementary Material. The LCA modelling software EASETECH version 2.3.6 (Clavreul et al., 2014) that was developed at DTU in Denmark was used in this study.

2.4.2. Scenarios description, system boundaries and assumptions

Two scenarios were considered in this study (Figure 1).

Figure 1 Flow chart representation of the LCA reference scenario and the LCA scenario for the use of biomass ash in cement. The dashed line indicates the boundary conditions for the LCA models. The reference scenario includes the traditional cement production (i.e. Ordinary Portland Cement OPC), while the BIOCement scenario takes into account the additional cement needed to maintain the compressive strength of the OPC reference. Only transport to the landfill site (i.e. reference scenario) and the cement plant (i.e. BIOCement scenario) is included.

Reference scenario: In the reference scenario, it was assumed that biomass fly ash is disposed in an industrial waste landfill situated in The Netherlands. Biomass fly ash is transported from the energy plant to the landfill site (situated at 100 km distance) where it is disposed. The landfill site was assumed to have a height of 20 meter and to remain open for the first 30 years with a net leachate production rate of 300 mm/year. After the final covering of the landfill the net leachate production rate was assumed to diminish to 5 mm/year throughout the remaining 70 years. These assumptions were used, together with the density of FA1, FA2 and FA3, to calculate the final liquid to solid ratio (L/S) (Table 1) following the approach described in Section 3 of the Supplementary Material. Leaching of inorganic substances from the landfill at the calculated L/S was included in the study as described below in Section 2.5. Part of the generated leachate is sent to the wastewater treatment plant (WWTP) (see Table 1 for collection efficiencies). The electricity consumption in the WWTP was also accounted for. The reference scenario involves, in parallel to the landfiling process, the modelling

of the pure OPC cement production (see Table 2 for process detail), the cement service and second life stages, that would happen anyway when biomass ash is landfilled instead of used as secondary cementitious material (see Figure 1).

Table 1 Dataset used to model the reference scenario. The leachate generation and efficiencies for the removal of individual substances in the WWTP are reported in the Supplementary Material (Table S3).

<i>Reference scenario</i>					<i>Process used in the model</i>
		<i>FA1</i>	<i>FA2</i>	<i>FA3</i>	
Transport	km	100	100	100	Truck, 28t-32t, Euro6, highway_EASETECH Database (Clavreul et al., 2014)
Infiltration rate (30 years)	mm/y	300	300	300	Leachate_Landfill_Treatment _emission to surface water (Olesen and Damgaard, 2014)
Infiltration rate (70 years)	mm/y	5	5	5	
Layer	m	20	20	20	
Density FA	kg/m³	690	750	916	
Leachate collection efficiency* (10 years)	%	95	95	95	
Leachate collection efficiency (90 years)	%	90	90	90	Electricity, high voltage, production mix; NL (Ecoinvent Centre, 2017)
Cumulative liquid to solid ratio (L/S) (100years)	L/kg	0.7	0.6	0.5	
Electricity consumption	kWh/kg	0.000443	0.000443	0.000443	

*Remaining leachate assumed to be stored in the landfill

BIOCement: In this scenario, biomass fly ash is used as secondary cementitious material in cement for outdoor pavement construction, thus considering all benefits derived from avoiding landfill. The BIOCement scenario is composed of three stages: i) mixing biomass FA with traditional OPC cement, ii) service life of cement containing biomass fly ash (50 years), and iii) utilization of crushed cement as road subbase for 50 years (i.e. second life). First, biomass fly ash is transported from the energy plant to the facility (200 km) where it is mixed with traditional cement. This step requires electricity for the mixing and the corresponding electricity consumption was taken from Hewlett (2003). After a pavement service life of 50 years, the cement is crushed, and the aggregates are subsequently reused as a subbase material for road construction. The leaching of inorganic substances during the service and second life phases was also considered.

Inclusion of biomass ash in cement decreases the compressive strength after curing time of 28 days by about 3 to 50% depending on fly ash types and replacement ratios, as shown in Tosti et al., (2018). To obtain comparable results, the performance of cement systems with biomass ash as SCM was compared to an average strength of 48.4 MPa which was the measured average compressive strength of pure OPC. The additional OPC requirement per kg of fly ash was calculated to compensate for the loss in strength and to obtain a comparable basis by adding OPC to the BIOCement until the average strength value of 48.4 MPa is reached (see Table 2 for values and Section 2, Table S2 of the Supplementary Material for calculations). The production of pavement and road construction processes were not included in the

boundaries since these processes are not considered to be affected by the utilization of biomass ash in cement. The inclusion of fly ash in cement could affect the durability of products and consequently the life time and maintenance operations required. However, this aspect is difficult to quantify and there is currently no method available in LCA that relates cement durability to fly ash content. Therefore, the maintenance process was considered to be the same for the reference and BIO Cement scenarios. The model described in this section was also applied to the traditional OPC cement application in the parallel reference scenario (Figure 1). Only different leaching data are used for the calculation of the release of inorganic substances from OPC and BIO Cement. Complete information on the leaching dataset can be found in the Supplementary Material Sections 4 and 5, Tables S4 and S5.

Table 2 Dataset used to model the different BIO Cement scenarios. The complete list of leaching values of elements for the service life and second life phases are reported in the Supplementary Material (Tables S4 and S5).

<i>BIO Cement scenario</i>								<i>Process used in the model</i>
		<i>20_FA1</i>	<i>20_FA2</i>	<i>20_FA3</i>	<i>40_FA1</i>	<i>40_FA2</i>	<i>40_FA3</i>	
Transport Fly ash	km	200	200	200	200	200	200	Truck, 28t-32t, Euro6, highway_EASETECH Database (Clavreul et al., 2014)
Electricity for Additional Blending	kWh/kg FA	0.035	0.035	0.035	0.035	0.035	0.035	Electricity, high voltage, production mix; NL (Ecoinvent Centre, 2017)
Additional OPC*	kg/kg FA	0.04	0.32	0.06	0.36	0.57	0.31	Cement production, Portland; Europe without Switzerland (Ecoinvent Centre, 2017)

*the amount of additional OPC expressed as kg/kg of FA is equivalent to the % of strength lost due to FA addition when compared to the average strength of pure OPC (48.4 MPa as average compressive strength value of 6 measurements performed in the lab). Complete calculation is reported in Section 2, Table S2 of the Supplementary Material.

2.5. Emissions from leaching

Results from laboratory leaching tests were taken as the basis for estimation of potential release during the life cycle phases within the 100 years time frame considered in the two scenarios. The leaching during the service life phase (i.e. monolithic) was calculated as total element mass released following a simplified diffusion model which takes the cumulative release (mg/m^2) measured during the standardized tank test after 64 days into account (Birgisdottir, 2005; Kosson et al., 1996). Complete information is reported in Section 4 of the Supplementary Material.

The assessment of the release of inorganics from the granular materials (i.e. from the landfilled fly ash in the reference scenario, and from the second life phase of the cement aggregates) required an estimation of the expected amount of water in contact with the residues within a given time horizon, i.e. the L/S ratio. The expected L/S ratios of the different scenarios

were estimated following the approach proposed by Kosson et al. (2002) as reported in the Supplementary Material (i.e. Section 5). The measured release from batch tests was then used to calculate the concentration of elements in the leachate from landfilling of biomass ash and the release from cement aggregates during the second life stage at the estimated L/S ratios (see Sections 3 and 5 in the Supplementary Material). For landfilling of biomass ash, the process included in EASETECH (i.e. *Leachate_Landfill_Treatment_emission to surface water*) was used to model the leachate generation at the calculated L/S and elements' removal efficiencies in the waste water treatment plant (Table S3 in the Supplementary Material). All emissions from WWTP were considered to be emitted to surface water.

Leaching emissions from the service life and second life of cement products were inventoried as cumulative emissions to industrial soil. The USEtox model used in the LCA considers two soil types, agricultural and industrial. However, it is not considered common practise that cement is used in agricultural setting. This approach, as reported by Allegrini et al. (2015), set the boundaries between the natural environment and the techno-sphere at the interphase between the material (i.e. cement pavement and layer of crushed cement below the road) and soil. The partitioning of contaminants into the natural environment between water and soil is determined completely by the fate modelling included in the USEtox model (Rosenbaum et al., 2008). This approach was used consistently in both the reference and the BIOCEment scenarios and was considered appropriate for the scope of this work. Chromium release from cement products is considered to be in the oxidation state Cr (VI). This assumption was based on results obtained from pH dependent leaching tests on the same cement products investigated in this study (Tosti et al., 2018) and other literature on construction and demolition waste (Butera et al., 2015).

2.6. Sensitivity and uncertainty analysis

Several assumptions were made in this study with regard to the release of elements, ash transportation distances and need for additional OPC. To assess the influence of these assumptions, a sensitivity analysis was performed starting with a perturbation analysis to identify the most sensitive parameters. Sensitivity analysis is subsequently conducted to investigate these sensitive inputs and to analyse the importance of assumptions made in the input model on the final result. The parameters for the perturbation analysis were identified through a contribution analysis to identify the most important processes. A total of 73 parameters for the reference scenario and 33 parameters for the use in cement scenario were tested in the perturbation analysis. This analysis included multiples runs of the model where each of the parameters was augmented by 10% of its default value and the model was run with a singular augmented parameter value. The result was then compared with the default scenario (i.e. all parameters set to their default value) and sensitivity ratios (SR) were then calculated for each parameter to identify the most sensitive parameters. The SR is defined as the ratio between the change

in the result of the particular parameter variation and the default result, divided by the ratio between the change in the parameter value and the default parameter value (see Section 7.2 in Supplementary Material). To compare different sensitivity ratios in each scenario and each impact category, normalized sensitivity ratios (NSR) were calculated. The NSR is defined as the ratio between the sensitivity ratio of one parameter in one impact category and the maximum absolute value among all SRs in the same impact category. All relevant information and equations are reported in Section 7.2 of the Supplementary Material. After the identification of the most sensitive parameters (i.e. 7 parameters for reference scenario and 5 parameters for the biomass ash in cement scenario), an uncertainty analysis was carried out. The analysis was done with a probability distribution set for each parameter value. A Monte Carlo analysis (10000 random variables) was performed subsequently to assess the propagation of the uncertainty through the model.

3. RESULTS AND DISCUSSION

3.1. Contribution analysis

Figure 2 shows the contribution of processes to the total characterized potential impacts for Climate change (Global Warming Potential, GWP_{100}), Human toxicity, cancer effects (HTc), Human toxicity, non-cancer effects (HTnc) and Ecotoxicity freshwater (ET) categories. The impacts on the GWP_{100} category are consistent with those on all the non-toxic categories (Ozone depletion (ODP), Particulate matter (PM), Ionising radiation human health (IR), Photochemical ozone formation (POF), Terrestrial acidification (AP), Eutrophication Terrestrial (TEP), Eutrophication Freshwater (FEP) Eutrophication Marine (MEP) and Depletion of abiotic resources (ADP)) since they all show a comparable trend with that of GWP_{100} . In the remainder of the article, we therefore focus on global warming and toxicity impacts, but in Supplementary Material Section 6, Table S6-S11, all ILCD impact categories are included.

The contributions to potential impacts have been aggregated into five groups: cement production, transport of biomass fly ash, landfill, leaching from service life and second life stages. For illustrative purposes, the discussion in this paragraph focuses on the 20_FA1 scenario, replacement of 20% wt. of OPC with biomass fly ash FA1, since the results in the total impacts are similar for all scenarios investigated including the 40% replacement. Complete information on characterized impact values for all categories and for all scenarios can be found in Section 6 of the Supplementary Material.

The results in Figure 2 show that for both the BIOCEment scenario and the reference scenario the cement production contributes almost entirely (i.e. 98%) to the impact on GWP_{100} . Emission of CO_2 is the main contributor to the impact on GWP_{100} category. The processes responsible of the CO_2 emission are carbonation of limestone and production of thermal

energy required to reach the sintering temperature in the cement kiln as already observed in previous literature (Huntzinger and Eatmon, 2009; Mohammadi and South, 2017; Van den Heede and De Belie, 2012).

The cement production is also contributing to the HTnc category for about 85% of the total impact in the case of the BIO Cement scenario and for 95% in the reference scenario. When taking a closer look to the underlying data (see Table S12, Section 6 of the Supplementary Material), it becomes clear that in this category the emissions of Hg to air from cement production is mainly responsible for the impact on the HTnc.

About 60% of the total impact to the category HTc in the reference scenario is due to the leaching of inorganics from landfill and second life stage (Figure 2). The impact to the HTc category in the 20_FA1 scenario is determined for about 50% by the leaching of inorganics in the second life stage. In particular, the release of Cr to surface water (i.e. during landfill) and industrial soil (i.e. during second life) is contributing almost entirely to the impact on the HTc category. The rest of the impact is determined by cement production for both reference and BIO Cement scenario.

Concerning the impact to the ET category in the BIO Cement scenario, this is determined for more than 80% by the release of Ba, Cu, Cr, Sb and Zn from second life stage. In the reference scenario, the impact to ET category is divided between cement production and release from second life of cement with a relatively smaller contribution (i.e. less than 15%) from service life stage. Percentages of contribution slightly change depending on type of biomass ash and the replacement ratio. It is interesting to note that transport of fly ash provides an almost negligible contribution to all impact categories. Only in the case of impact to HTnc, transport contributes for about 10% of the total impact.

The only observed difference in contribution to potential impacts among fly ash types is observed for the reference scenario when FA3 is assessed (see Section 6, Tables S10 and S11 of the Supplementary Material). Here, the contribution of the landfill process to HTc impact is minor: less than 10% of total impact instead of about 40%. This difference is related to the substantially lower Cr release in comparison to the other fly ash types.

Figure 2 Percentage of contribution of processes to the total impact for the reference scenario (i.e. FA1) and BIOCement scenario (i.e. 20_FA1). The results are considered representative for all other scenarios.

3.2. Comparison of scenarios

In Figure 3, the internally normalized results from the modelling of scenarios are shown for GWP₁₀₀, HTc, HTnc and ET impact categories. Generally, the BIOCement scenario has lower average potential impact values compared to the reference when GWP₁₀₀ and all other non-toxic categories (ODP, PM, IR, POF, AP, TEP, FEP, MEP, ADP) are considered. As observed during the contribution analysis, the cement production process is dominating the impacts to GWP₁₀₀ and, therefore, the observed differences between the reference and BIOCement scenarios in terms of non-toxic impacts are consistent with the lower amount of OPC cement used in the latter scenario. The total amount of cement in the BIOCement scenario is determined by the hydraulic activity of the biomass fly ash. The differences are small, but FA2 shows the lowest hydraulic activity and, therefore, requires a relatively high amount of additional cement (see Table 2) to maintain the strength development properties of the reference system. Thus, FA2 has also a slightly higher impact in the GWP₁₀₀ category. This observation is particularly evident in the 40_FA2 scenario in which replacing 40% of cement with FA2 resulted in a limited (only 15%) reduction of the impact on GWP₁₀₀ (and other non-toxic categories) compared to the reference scenario. When FA3 is used in the 40% formulation the impact in the same category is 26% lower than the reference.

The ability of biomass ash to develop strength is a crucial parameter that should be measured and accounted for as outlined in Table 2. This approach allows for a more uniform basis in LCA studies for the possible re-use of different biomass ash types in cement.

The impacts of FA1 and FA2 (both for 20% and 40% replacement) on HTc are substantially lower than the reference scenario, with FA2 performing better than FA1. Contrary, FA3 shows a higher impact on HTc than the reference scenario. The magnitude of impacts is related to the different release of elements from pure FA during landfill. In particular, as previously observed in the contribution analysis, the release of Cr to fresh water from the landfill (after WWTP) process has a significant influence on the HTc impact results of the reference system. Therefore, a cleaner ash will give a higher relative impact than a more contaminated ash, as the difference to the landfill scenario becomes lower. Leaching of Cr from the biomass ash is very important in determining the impact on HTc of the reference scenario (see also Section 3.3). When biomass ash is added to cement, the leaching of chromium is controlled by the cement matrix and results in a more consistent release (Tosti et al., 2018). The different relative impacts on HTc that are observed for the different BIOCement scenarios (Figure 3) are, therefore, predominantly determined by the leaching properties of the landfilled ashes in the reference scenario. In particular, pure FA2 shows a relatively high leaching, which reduces its relative impact (compared to the other ash types) when applying it in cement, due to the mitigating effect of the cement matrix on the leaching of biomass ash (Tosti et al., 2018). Contrary, FA3 is a relatively “clean” ash (particularly regarding Cr release) with a Chromium concentration in the leachate generated during landfill of FA3 of around 0.06 mg/L. This low concentration implies a very small cumulative Cr released to fresh water after WWTP. As result, the cumulative leaching during second life of 20 and 40_FA3 specimens makes the BIOCement scenario performs worse than reference in the HTc category.

As was also observed for GWP₁₀₀ category, the different impacts to HTnc are consistent with the lower amount of cement used when fly ash is added to cement due to Hg emissions to air from cement production process. Hence, the impacts to HTnc are mainly determined by the lower use of OPC cement in the BIOCement system rather than by the direct impact from the added fly ash.

The higher impact to ET category when BIOCement is compared to reference systems for FA1 and FA2 is due to different detection limits obtained for the analysis of dissolved Zn in leachates from pure cement (no ash addition) and from FA1 and FA2 containing cement at both replacement ratios (Tosti et al., 2018). We have, therefore, conservatively used the detection limit value as input for this LCA study, which has resulted in higher than reference impacts for FA1 and FA2.

Figure 3 Comparison of results expressed as percentage of the corresponding reference scenario (internal normalization).

3.3. Sensitivity analysis and uncertainty analysis

Table 3 summarizes the parameters in the reference and BIOCEment scenarios for which the model is most sensitive (i.e. with a NSR > 0.8) in relation to the impact categories GWP₁₀₀, HTc, HTnc and ET. Tables with all normalized sensitivity ratios (NSR) values are reported in Section 7.2 of the Supplementary Material. It is important to stress that during perturbation analysis, the cement production process (and the corresponding emissions), was not entered as a parameter in the model and, therefore, no NSR was calculated for that process. This decision was based on the fact that the goal of the study was to investigate the influence of biomass fly ash addition to cement as alternative option to landfilling and, therefore, only the “additional OPC” production needed to maintain the same cement strength in the case of biomass ash inclusion was entered as a parameter in the model.

A general observation from Table 3 (and S14-S19 for details) is that the model is sensitive to just a few parameters for every impact category, and that these parameters are consistent among different ash types. For instance, the release of Ba and Cr from the second life stage of cement are sensitive parameters for all fly ashes with respect to the HTnc and HTc categories in the reference and ash utilization in cement scenario, respectively. The transport and the additional OPC parameters influence the results on GWP₁₀₀ (and all other non-toxic categories), independently from the biomass ash type.

Table 3 Parameters resulting from perturbation analysis with a normalized sensitivity ratio (NSR) higher than 0.8. GWP_{100} is reported as representative of all non-toxic impact categories. The complete list of NSR values for all parameters is reported in Section 7.2 of the Supplementary Material.

		FA1	FA2	FA3
Reference	GWP_{100}	Transport	Transport	Transport
	HTc	Infiltration Rate, Layer, Density, Leaching from landfill (Cr)	Infiltration Rate, Layer, Density, Leaching from landfill (Cr)	Leaching from second life (Cr) of traditional cement
	$HTnc$	Leaching from second life (Ba) of traditional cement	Leaching from second life (Ba) of traditional cement	Leaching from second life (Ba) of traditional cement
	ET	Leaching from second life (Ba) of traditional cement	Infiltration Rate, Layer, Density, Leaching from second life (Ba) of traditional cement	Leaching from second life (Ba) of traditional cement
BIOCement (20 and 40%)	GWP_{100}	Production of additional OPC	Production of additional OPC	Production of additional OPC
	HTc	Leaching from second life (Cr)	Leaching from second life (Cr)	Leaching from second life (Cr)
	$HTnc$	Leaching from second life (Zn)	Production of additional OPC (Hg emission)	Leaching from second life (Ba)
	ET	Leaching from second life (Cu), Leaching from second life (Zn)	Leaching from second life (Zn)	Leaching from second life (Cr)

However, some differences are observed in the sensitive parameters among biomass ash types and the corresponding cements replacements. The HTc category of the reference scenario for FA1 and FA2 is sensitive to landfill parameters (i.e. infiltration rate, FA height layer, bulk density of FA. See Section 7.1 Table S13 of the Supplementary Material for the description and values of parameters) and leaching of Cr from the pure biomass ash. A difference in the sensitivity of the model to parameters is observed also in the BIOCement scenario for the $HTnc$ category. In this case, the low hydraulic activity of FA2 required a relatively high amount of additional cement, which influences the impact on the $HTnc$ (mainly due to the emission of Hg to air associated with additional cement production). This is not the case for FA1 and FA3 where the variation of leaching values of Zn and Ba from the second life stage of cement (i.e. of cement aggregates in road base) has the largest influence on the resulting impact to $HTnc$. The leaching of Cu and Zn from the second life stage are the most sensitive parameters that affect the impact to the ET category in the BIOCement scenarios. Values of NSR are reported in Section 7.2 of the Supplementary Material.

Since sensitivity of parameters does not necessarily confer uncertainty of results, the uncertainty of the model results was evaluated by assigning probability distributions to the most sensitive parameters shown in Table 3 and, subsequently, quantifying the error propagation by means of Monte Carlo analysis (details are provided in Section 7.3 of the Supplementary Material). The results of the uncertainty analysis are shown in Figure 4 as normalized values expressed in person equivalent (PE).

In this type of data representation, the characterized results obtained from the analysis are divided by a normalization value that is called Person Equivalent (PE), which is a quantification of the environmental impact in a specific category caused annually by the activities of an inhabitant of the affected area (EC-JRC, 2010). In the presented study, Europe was selected as area of interest. The normalization values are reported in Table S1 of the Supplementary Material.

The identified uncertainty, as reflected in the error bars (95% confidence interval as calculated by the EASETECH software) in Figure 4, is relatively small for the three biomass ashes for the categories GWP_{100} and all the other non-toxic categories. Hence, the conclusions from the comparison between the reference and BIO Cement scenarios do not change, i.e. that the use of fly ash in cement is beneficial. This observation can be explained by the fact that the most sensitive parameter (i.e. additional OPC, see Table 3) affecting GWP_{100} and all the other non-toxic categories show a small deviation around the average value in this study (see Table S20 of the Supplementary Material).

The uncertainty is relatively large for the category HTc and is mainly related to release of Cr from the second life stage of cement. Nevertheless, the difference in HTc impact between these two scenarios is still significant and the uncertainty bars do not overlap when comparing these scenarios (except for 40_FA1 and 40_FA3 where the uncertainty bars overlap between both scenarios).

Regarding the toxic categories, it is important to also quantify the actual risks associated to those emissions, which is determined by concentrations and not the aggregated cumulative release as used in an LCA study. Therefore, in this study, a first check was performed by comparing the leaching properties of biomass ash and biomass ash containing cement with regulatory limits as specified in the EU Landfill Directive (European Commission, 1999) and the Dutch Soil Quality Decree (Bodemkwaliteit, 2007), respectively. All three biomass ashes and the corresponding blended cements comply with the criteria for landfilling of hazardous material (Table S21 of the Supplementary Material) and application as monolithic and/or granular material in construction (Tosti et al., 2018), respectively. Based on these findings, we conclude that for those cases where the impacts to toxic categories is overlapping (e.g. HTc in 40_FA1, and HTc in 40_FA3, see Figure 4) no ranking can be made between the reference and ash utilization in cement scenarios, but that no unacceptable risk is associated with that impact.

Figure 4 Normalized results from uncertainty analysis, error bars are the 95% confidence intervals. The results are expressed as person equivalent per functional units. Since the amount of cement used to produce 20 or 40% fly ash containing mixtures changes, the results can be compared only for identical (i.e. 20% or 40%) replacement ratios.

4. Conclusions

In this study, the potential environmental impacts associated with the reuse of biomass fly ash as secondary cementitious material, as an alternative management option to landfilling, were evaluated by means of a life cycle assessment (LCA). Both the service life phase (cement application) as well as the second life phase of cement products

(use of cement aggregates in road base) were taken into account, with focus on the cumulative release of potentially toxic elements during a life cycle of 100 years.

Effects on non-toxic impact categories

Life cycle assessment modelling of management options for 1 ton of biomass ash has demonstrated that the utilization of biomass ash into cement is preferable over landfilling for all non-toxic impact categories considered: Global warming potential (GWP), Ozone depletion (ODP), Particulate matter (PM), Ionising radiation human health (IR), Photochemical ozone formation (POF), Terrestrial acidification (AP), Eutrophication Terrestrial (TEP), Eutrophication Freshwater (FEP), Eutrophication Marine (MEP) and Depletion of abiotic resources (ADP). This preference was demonstrated for all types of biomass ash investigated at cement replacement ratios of 20 and 40%. Cement production was the most important process for all non-toxic impact categories accounting for about 85 to 98 % of total impacts. The hydraulic activity of biomass ash in cement determines the total amount of traditional (OPC) cement needed to obtain the same compressive strength between the reference and BIOCement scenarios and, therefore, the lower impact to those categories determined by the cement production process. Biomass ash from the combustion of clean wood in a circulating fluidized bed installation (i.e. FA1), and biomass ash from the combustion of paper sludge and recovered waste wood in a bubbling fluidized bed incinerator (i.e. FA3) were found to perform better than biomass ash originating from the combustion of wood pellets in a pulverized fuel installation (i.e. FA2). The lower impact to global warming was found to be between 11 (i.e. FA2) and 18% (i.e. FA1) of the reference scenario when biomass ash replaces 20% of cement, and between 15 (i.e. FA2) and 26% (i.e. FA3) at a replacement rate of 40%. Uncertainty related to variability of the most sensitive parameters did not appear to be critical for the resulting impacts on all non-toxic categories investigated. Based on these conclusions we can recommend that using a higher amount of fly ash is beneficial for the non-toxic categories and this amount will in practice be limited by the physical strength requirements for the application.

Effects on toxic impact categories

The impact to human toxicity carcinogenic (HTc) and ecotoxicity (ET) was determined by the leaching of metals whereas the impact to human toxicity non carcinogenic (HTnc) was determined by cement production as for non-toxic categories.

Comparison between the reference and the BIOCement scenarios showed that cements containing FA1 and FA2 perform better, on average, than blends containing FA3 regarding the impact to HTc category. Contrary, only BIOCement scenario containing FA3 showed a better performance than the reference for the ecotoxicity (ET) category. These observed performance differences between the reference and BIOCement scenarios are mainly determined by the leaching

properties of biomass ash itself. Leaching from biomass fly ash determines the concentrations of contaminants in the leachate generated during landfilling and, therefore, the impact on human toxicity of the reference scenario.

Consideration of the uncertainty of leaching data from pure biomass ash and all the investigated cement specimens resulted in a different ranking of scenarios particularly when the human toxicity carcinogenic category was assessed. However, a higher impact on the toxic categories does not necessarily imply an actual risk associated with the investigated scenarios in this work. Comparison of leaching values with regulatory criteria for soil quality and landfill leachate is strongly advised to assess whether actual risks may occur.

General conclusion

Methodologically, this study has demonstrated the importance of considering the hydraulic activity of fly ash and the strength development of the blended cement, as well as the leaching of metals from the alternatively landfilled fly ash, in LCA modelling of the reuse of biomass fly ash as secondary cementitious material. For the selected range of broadly representative types of biomass ashes that were evaluated in this study, we have shown that the use of biomass ash in cement was beneficial with regard to the majority of the impact categories, while not conferring any additional risk.

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