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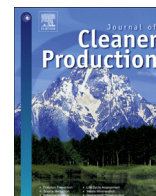
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## Environmental and economic perspective of waste-derived activators on alkali-activated mortars

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## ABSTRACT

Alkali-activated binders have been considered a low carbon alternative to cement and are produced by reacting aluminosilicate precursor with an alkali-activator. However, alkali-activators have been observed to be a major contributor to the environmental burdens of alkali-activated materials regarding various environmental impact categories. Therefore, this study aims to perform an environmental impact assessment using life cycle assessment methodology on alkali-activated mortars produced from chemically modified one- and two-part waste-derived activators (waste glass and rice husk ash) in comparison to conventional one- and two-part alkali-activated mortars, to estimate the influence of activator on environmental impact of mortar. Additionally, a simplified cost analysis of the different mortar compositions was conducted. A sensitivity analysis was performed on the key parameters, and allocating emissions to waste glass and rice husk ash. Results show that waste glass and rice husk ash-derived alkali-activated mortar resulted in up to 62%, 61%, 76% and 56% reduced emission respectively in climate change, fossil depletion, terrestrial acidification and photochemical ozone creation formation when compared to conventional alkali-activated mortar counterpart. Sensitivity analysis indicated that waste glass and rice husk ash are not so sensitive to mass allocation, with a maximum of 5% increased emissions observed in the above-mentioned impact categories. Additionally, sensitivity analysis on sodium hydroxide demonstrated that production from chlorine-alkali electrolysis using technology-mix produced improved environmental performance in comparison to production from brine solution and diaphragm route, respectively. Sensitivity analysis on sodium silicate using an alternative inventory data indicated the emissions can increase regarding one-part or decrease regarding two-part alkali-activated mortars. Results from cost analysis indicated up to 19% cost savings from waste-derived alkali-activated mortar compared to conventional alkali-activated mortar. In conclusion, chemically modified waste-derived activators are a promising alternative in improving environmental performance of alkali-activated materials if their usage also reduces or substitutes the need for conventional alkali-activators.

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### 1. Introduction

Cement production has rapidly grown in recent years with an estimate of 4.7 billion tonnes produced globally in 2016 (CEMBUREAU, 2017), and an estimated contribution between 4% and 8% of global CO<sub>2</sub> emissions (Andrew, 2018). CO<sub>2</sub> emissions from

cement production results respectively from thermally decomposing carbonates (mainly limestone) into oxides and CO<sub>2</sub>, to produce clinker; and during generation of high amount of energy (from combustion of fossil fuels) needed to heat raw materials (over 1000 °C) (Andrew, 2018). There are different types of cement binders categorized from CEM I – V, containing cement clinker in the range of 5%–100% (BS EN 197-1, 2011) with different percentages of industrial by-products and/or wastes materials such as blast furnace slag, silica fume, pozzolana, fly ash, burnt shale and limestone, to partially replace cement binder for an improved environmental performance among other reasons (CEMBUREAU, 2015a; 2015b; 2015c).

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In addition to integrating industrial by-products with clinker for an improved environmental performance of cement, there has been increased research in reducing environmental impact of cement binders through development of alternative types of binders such as alkali-activated binders (AABs) sometimes referred to as geopolymer binders (Provis, 2018). Geopolymers are largely used to describe low calcium alkali-activated aluminosilicate binders, thus, categorized as a sub-group of alkali-activated materials (Heath et al., 2014; Provis, 2018). Davidovits (1994) defined the concept of geopolymers as polycondensation of polymeric aluminosilicates and alkali-silicates, yielding three-dimensional polymeric frameworks. They do not require very high temperatures during production, thus, reducing high fuel consumption (Davidovits, 2015). By and large, AABs are produced by reacting solid aluminosilicate raw materials (precursor) with an alkali-activator to form a hardened binder (Tong et al., 2018; Vinai and Soutsos, 2019). These aluminosilicate precursors can be in the form of virgin raw materials such as metakaolin, or in the form of industrial side-streams with a high Si/Al ratio such as coal fly ash (CFA) and granulated blast furnace slag (GBFS), which have demonstrated good results in production of AABs (Provis, 2018).

AABs can be produced in two pathways namely one-part and two-part pathways (Luukkonen et al., 2018a).

- Two-part AABs as depicted in Fig. 1 are more conventional and are produced by reacting solid aluminosilicate precursor with a concentrated aqueous solution of alkali silicate and hydroxide, and water. They are mainly suitable for precast applications (Luukkonen et al., 2018a). However, with respect to large scale production of two-part AABs, impracticalities of handling large amounts of corrosive, viscous and hazardous alkali solutions led to increased research in the development of one-part AAB (Luukkonen et al., 2018a).
- One-part AAB as depicted in Fig. 2 is produced by reacting solid aluminosilicate precursor with solid alkali silicate, thereby producing a dry mixture binder similarly to cement. One-part AABs are more suited for in situ applications where handling of viscous alkali solutions can be more challenging, making them more scalable in future as it is similar to cement in terms of usage and packaging (Luukkonen et al., 2018a; Provis, 2018).

AABs (especially one-part AABs) can be a potential alternative to Ordinary Portland cement (OPC). Although, they are not likely to fully replace OPC, they can serve as a supplementary binder to OPC thereby reducing CO<sub>2</sub> emissions from the cement industry (Luukkonen et al., 2018a). Some advantages of AABs over OPC include but are not limited to high resistance against acids, high temperature resistance, high strength and high durability (Weil et al., 2005). However, these advantages also depend strongly on mixture composition of materials, curing time and temperature (Weil et al., 2005). Besides, alkali-activated materials provide means of reusing and recycling waste materials as secondary raw

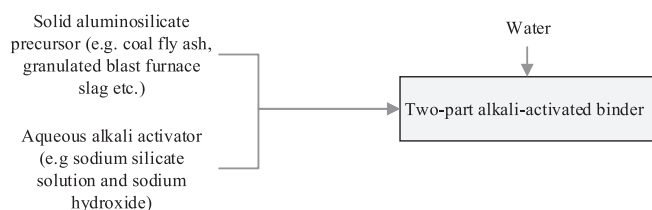


Fig. 1. Two-part alkali-activated binder.

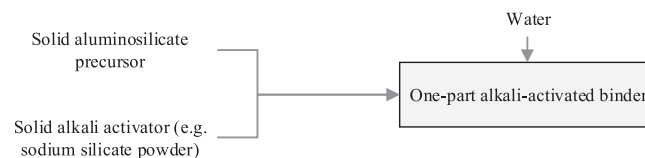


Fig. 2. One-part alkali-activated binder.

materials, to avoid problems of waste disposal and associated environmental burdens. To further promote alkali-activated materials as a feasible and beneficial option, precursors and activators should be locally available to minimise transportation costs and emissions. The downside of AABs is not having a standardised approach when compared to OPC, thus, not making it a solution suitable for all circumstances (Provis, 2018). Additionally, they can be susceptible to cracking, failures and deterioration as a result of their brittleness. This can however be overcome by reinforcement with fibres or other suitable materials (Alomayri, 2017).

The environmental sustainability of alkali-activated materials is highly dependent on alkali-activators (commonly used are sodium hydroxide and sodium silicate). More information on production of these chemicals can be found in the supplementary material. Sodium silicate as an activator is known to produce a more compact and dense material with higher mechanical strength in comparison to hydroxide-activated materials. Thus, a higher quantity of sodium silicate is used when both chemicals are used as activators (Mohseni, 2018; Nematollahi et al., 2015). However, sodium silicate has also been observed to be a major contributor to the environmental burdens of alkali-activated materials regarding various impact categories such as global warming potential (GWP), acidification potential (AP), abiotic depletion potential (ADP), toxicity potentials etc. (Abdulkareem et al., 2019a; Habert et al., 2011; McGuire et al., 2011). These emissions are mainly as a result of the high energy consumption required during their production (Fawer et al., 1999). Furthermore, a study carried out by Abdulkareem et al. (2019b) comparing the environmental assessment of one-part and two-part alkali-activated mortars (AAMs) demonstrated that one-part AAM had higher environmental impact when compared to its two-part counterpart mainly as a result of the additional energy required to produce sodium silicate powder which is required in the production of one-part AAM (Abdulkareem et al., 2019b). Although, the powder activator is a more desirable option when compared to the aqueous activator regarding handling and use, it is however not desirable due to its higher environmental burdens.

In a bid to develop activators with reduced environmental impacts, while retaining high mechanical properties of alkali-activated materials, there has been increased research in the production of sodium silicate from other silica rich waste materials such as rice husk ash (Tchakouté et al., 2016; Tong et al., 2018), sugarcane bagasse ash (Norsuraya et al., 2016; Tchakouté et al., 2017), waste glass (Passuello et al., 2017; Vinai and Soutsos, 2019) and silica fume (Bajza et al., 1998; Rouseková et al., 1997) etc. Most of these studies are however focused on the mechanical properties of alkali-activated materials from these waste derived activators (chemically modified by caustic solution) and not on the environmental impacts. As a result, this study will focus on environmental impact assessment of waste-derived AAMs in comparison to conventional AAMs, to estimate and compare their environmental performance. In addition, a simplified cost analysis of these different mortar compositions will be conducted to assess their economic sustainability.

## 2. Materials and method

### 2.1. Materials

#### 2.1.1. Waste glass (WG) cullet

Glass production has large similarities with sodium silicate production as it involves reaction of silica sand and soda ash at a temperature of more than 1100 °C (EU, 2007). Thus, making silicon dioxide the main component of glass. WG cullet is a potential source of silicate with amorphous silica content of 70–75% (Vinai and Soutsos, 2019). Glass production is quite extensive which means that there is a high amount of WG available for open and closed-loop recycling. However, composition, colour and contamination of WG can reduce the amount of glass that can be reused in closed-loop recycling of glass (Vinai and Soutsos, 2019). Over 12 million tons of glass bottles and jars are collected and recycled in Europe with an average glass recycling rate of 74% (FEVE, 2019).

Alkali-activator from WG can be produced using hydrothermal or fusion methods. Hydrothermal method involves heating of WG in an alkaline solution while fusion method involves heating WG and NaOH powder at a very high temperature. Hydrothermal method produces aqueous alkali-activator while fusion methods produce powder alkali-activator (Vinai and Soutsos, 2019). Hence, making possible production of one-part waste-derived alkali activator. The environmental impacts of producing powder activator from WG cullet using the fusion method was assessed following the production process reported by Vinai and Soutsos (2019). WG cullet was milled by means of Retsch PM 400 ball mill for 10 min and then reacted with NaOH powder by heating in an electric furnace at 150 °C for 1 h. The mass proportion of WG cullet and NaOH was 48% and 52% respectively. The advantage of this procedure is that it produces a powder activator as opposed to a liquid activator. Mortar produced from WG-derived activator is prepared by mixing required amounts of sand, CFA, GBFS and activator. The mixtures were cured at room temperature (Vinai and Soutsos, 2019). From this mixture, a one-part waste-derived AAM is produced.

#### 2.1.2. Rice husk ash

Rice husk ash (RHA) is an agricultural waste material resulting from combustion of rice husk, which is a residual stream from the pod of rice grains (Gursel et al., 2016). Rice husk is generated while milling paddy and has a number of applications such as in fertilizer and substrate, raw material for brick production and renewable energy source from biomass etc. (Tong et al., 2018). Gasification or controlled burning of rice husks in kilns or power plants is an effective way of disposing rice husks thereby producing high quality RHA suitable as a supplementary cementitious material (SCM) (Gursel et al., 2016; Meryman, 2009). RHA is rich in silica, approximately 90%, similarly to glass (Norsuraya et al., 2016; Tong et al., 2018) and has been considered a potential source of silicates for alkali-activated binders (Ghosh, 2013; Kamseu et al., 2017; Liu et al., 2016; Ma et al., 2012; Tong et al., 2018). Temperature and duration of combustion process are important during combustion of rice husk, as shorter duration and higher temperatures are expected to maximize the amorphous content of RHA and induce formation of crystalline SiO<sub>2</sub>. The RHA share of rice husk is 18–22% of dry content by weight. Market availability of amorphous RHA with silica content of about 95% makes industrial production of silicate from RHA possible (Tong et al., 2018).

The environmental impacts of producing alkali-activator from RHA using the hydrothermal method was assessed following the production process reported by Tong et al. (2018). RHA was milled for 15 min by means of Retsch PM400 to produce a fine RHA powder. RHA powder was dissolved in a NaOH solution, heated and kept under magnetic stirring. The composition of the solution is

NaOH (8.7%), RHA (24%) and water (67.3%) in mass (Gursel et al., 2016; Tong et al., 2018). To produce alkali-activated mortar from RHA-derived activator, fly ash and GBFS were blended manually and sand was subsequently added to the mix. Afterward, the activator was added to the mix and subsequently cured at room temperature (Tong et al., 2018). From this mixture, a two-part waste-derived AAM is produced.

### 2.2. Life cycle assessment methodology

Life cycle assessment (LCA) addresses “environment aspects and potential environmental impacts throughout a products’ life cycle from raw material acquisition through production, use, end-of-life treatment, recycling and final disposal” (EN ISO 14040, 2006; EN ISO 14044, 2006). LCA framework is actualised in four phases which are: goal and scope definition, life cycle inventory (LCI) phase, life cycle impact assessment (LCIA) phase and interpretation phase. Additionally, to establish confidence in LCA results, it is recommended to conduct assessment procedures such as sensitivity analysis, consistency check and completeness (EN ISO 14040, 2006).

#### 2.2.1. Goal and scope of study

The goal of this study is to estimate and compare environmental impacts of chemically modified waste-derived alkali-activated mortar (one- and two-part) in comparison to conventional alkali-activated mortars (one- and two-part), to estimate the influence of activator on environmental impact of mortar. The results are compared to assess if waste-derived activators improve environmental performance of AAMs. In addition, a simplified cost analysis was carried to compare the cost of these different mortars to assess their economic sustainability. The functional unit is defined as the environmental burdens generated due to the activities involved in the production of 1 m<sup>3</sup> of AAM with compressive strengths between 52 MPa–60 MPa at 28 days. It is assumed that the scenarios will have similar applications with similar service life.

The system boundary (Fig. 3) utilized in this study contains the main production steps of cradle-to-gate AAM including raw material production, waste treatment and mixing of constituents in which the associated emissions and energy consumptions are calculated. Transportation is excluded from this study as it is assumed all raw materials considered have similar transportation distances. The use phase and end-of-life phase are also excluded from this study as it is assumed that comparable and similar impacts are expected from these phases.

#### 2.2.2. Scenario description

Four scenarios were considered in this study namely.

- S1 – one-part mortar (conventional powder activator)
- S2 – two-part mortar (conventional aqueous activator)
- S3 – one-part mortar (WG-NaOH derived powder activator)
- S4 – two-part mortar (RHA-NaOH derived aqueous activator)

S1 and S2 represents reference scenarios for conventional alkali-activators in the production of one-part and two-part AAM respectively. S3 and S4 represent scenarios derived from chemically modified WG and RHA alkali-activators to produce one-part and two-part AAMs respectively. The synthesis conditions for the AAMs are defined at laboratory scale. Table 1 illustrates mix proportions of these scenarios. All mortars have similar quantities of constituents and equivalent magnitude of compressive strengths at 28 days to aid comparison. All mortars were cured at room temperature, hence, avoiding energy required for thermal curing. WG and RHA are considered as wastes in this study and allocated no

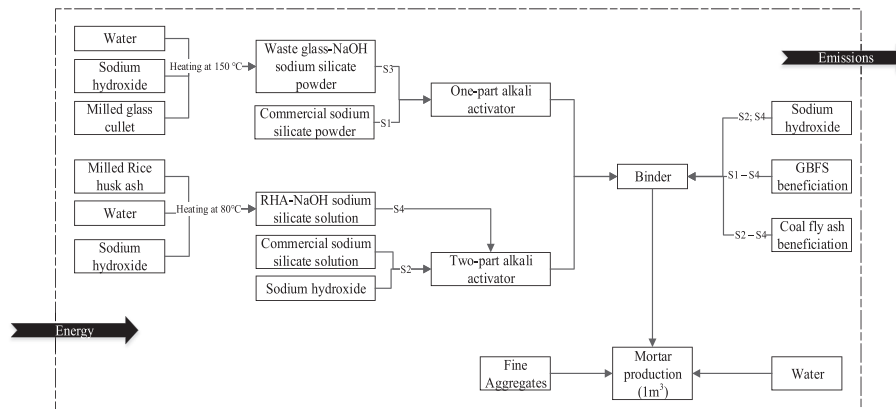


Fig. 3. System boundary depicting production of alkali-activated mortar.

Table 1

Mix proportions of mortars.

Constituent (kg/m <sup>3</sup> )	S1 52 MPa <sup>a</sup>	S2 58 MPa <sup>b</sup>	S3 58 MPa <sup>c</sup>	S4 60 MPa <sup>b</sup>
Coal fly ash	0	321	313	326
GBFS	373	214	208	217
Sand	1445	1471	1432	1493
WG-NaOH powder	0	0	126	0
RHA-NaOH solution	0	0	0	196
Sodium silicate	109 <sup>d</sup>	157 <sup>e</sup>	0	0
Sodium hydroxide pellets	0	26	0	36
Water	241	131	231	99

WG – waste glass. RHA – Rice husk ash. NaOH - Sodium hydroxide. GBFS - Granulated blast furnace slag.

<sup>a</sup> Yang et al. (2010).

<sup>b</sup> Tong et al. (2018).

<sup>c</sup> Vinai and Soutsos (2019).

<sup>d</sup> Sodium silicate powder.

<sup>e</sup> Sodium silicate solution.

environmental burdens based on the EU directive that states “a substance is considered as by-product and not waste if it fulfils certain criteria which are 1) further use of the substance is certain; 2) the substance can be used directly without any further processing other than normal industrial practice; 3) the substance is produced as an integral part of a production process; 4) further use is lawful (European Parliament and Council, 2008)”. As a result, only impacts incurred during treatment of WG and RHA will be considered.

Selected scenarios focus on AAM recipes with comparable compressive strength. Data for these scenarios were collected from literature studies. It was ensured that these data were closely related with respect to system boundaries and masses of substances to ease comparison and consistency. Additional scenarios were not taken into consideration as there are limited information on waste-derived AAMs with comparable compressive strength within the scope of this study. For instance, one-part RHA-NaOH derived mortar studied by Luukkonen et al. (2018b), obtained a compressive strength of 32 MPa at 28 days which is high enough, however, not as high as compressive strength of mortars in the scope of this study. Also, Puertas and Torres-Carrasco (2014) and Torres-Carrasco and Puertas (2015) carried out a study on two-part WG-derived activator focusing on paste preparation and not mortar, which is out of scope of this study. Thus, the limitations of this study include the limited number of scenarios for waste-derived AAMs.

### 2.2.3. Inventory analysis

Life cycle inventory (LCI) is the phase where inputs and outputs as well as produced emission of all unit processes included in the

system boundary are quantified. LCI data for processes such as NaOH, sand, electricity and water were sourced from GaBi database. LCI for sodium silicate solution and sodium silicate powder were sourced from Ecoinvent database while LCI for CFA, GBFS, RHA and WG were collected from literature. Table 2 shows data sources for the different processes considered in this study. In this study, pedigree matrix (see supplementary material for more details) is applied to assess the quality of data utilized in this study. Data sources are evaluated based on five independent characteristics namely, reliability, completeness, temporal correlation, geographical correlation, and further technological correlation. Each independent characteristic is scored between 1 and 5 based on 5 quality levels (Weidema et al., 2013).

RHA beneficiation process include electricity for milling (0.0432 MJ/kg) and electricity of 0.74 MJ/kg to dissolve RHA in NaOH by heating at 80 °C for 3 h (Tong et al., 2018). Due to simplifications in the analysis of heat required to dissolved RHA in NaOH, the estimated energy is more than actual energy needed (Tong et al., 2018). WG cullet beneficiation include electricity consumption for milling glass (0.18 MJ/kg) (Vinai and Soutsos, 2019). Electricity required to heat glass powder with NaOH was 0.072 MJ/kg. This value is according to technical specifications obtained for industrial ovens with required capacity (Vinai and Soutsos, 2019). The synthesis conditions for some of the alkali-activated mortars are defined at laboratory scale. Thus, energy consumed might be more conservative when compared to industrial scale production. For GBFS and CFA, they are assumed to come into the system boundary with no environmental burdens, since they are considered wastes. However, GBFS which is used as a supplementary cementitious material (SCM), goes through the processes of

**Table 2**  
Data sources with data quality indexes.

Type of data	Source	Data quality indexes Pedigree matrix
Sodium hydroxide	GaBi database 2019 – EU-28: Sodium hydroxide (caustic soda mix, 100%)	(3,3,2,2,2)
Sodium silicate solution	Ecoinvent database – EU-28: Sodium silicate production, hydrothermal liquor, product in 48% solution state	(2,2,5,1,1)
Sodium silicate powder	Ecoinvent database – EU-28: Sodium silicate production, spray powder, 80%	(2,2,5,1,1)
Sand	GaBi database 2019 – EU-28: Sand 0/2	(3,3,4,3,4)
Water	GaBi database 2019 – EU-28: tap water	(3,3,4,4,3)
Electricity	GaBi database 2019 – FI: electricity grid mix	(3,3,4,3,4)
Coal fly ash	Coal fly ash beneficiation (Kawai et al., 2005)	(4,4,5,5,2)
GBFS	GBFS beneficiation (Marceau and VanGeem, 2003)	(2,3,5,4,1)
Waste glass	Waste glass beneficiation (Vinai and Soutsos, 2019)	(2,4,1,3,1)
Rice husk ash	Rice husk ash beneficiation (Tong et al., 2018)	(2,4,2,3,1)

GBFS – granulated blast furnace slag.

granulation, drying, crushing and grinding (Marceau et al., 2007). Thus, materials and energy required to process GBFS were modelled according to data from Marceau and VanGeem (2003). With respect to CFA, emission associated with treatment process was adopted from Kawai et al. (2005). Environmental impacts of capital good such as trucks, equipment, buildings were not considered in this study. Although, capital goods can have a significant influence on the total results (Liikanen et al., 2017), however, scope of this study is limited to materials and energy needed to produce alkali-activated mortars.

#### 2.2.4. Life cycle impact assessment

The environmental impact categories that will be employed in assigning LCI results to specific environmental issues will be based on four impact categories: climate change, terrestrial acidification, fossil depletion and Photochemical ozone creation (POC) (human health). This is because, cement binder production is associated with environmental issues such as consumption of energy, raw materials and emissions to air, land, and water. Also, fuel handling and storage can be a potential source of soil and groundwater contamination (Stajanca and Estokova, 2012). The associated impact categories to these environmental issues will therefore be relevant for assessment of emissions from binder and mortar production processes (Chen et al., 2010; Van Den Heede and De Belie, 2012). Results of other environmental impact categories (fine particulate matter formation, freshwater consumption, freshwater ecotoxicity, freshwater eutrophication; human toxicity, ionizing radiation, land use, marine ecotoxicity, metal depletion, stratospheric ozone depletion, terrestrial ecotoxicity) are given in the supplementary material. Environmental performance modelling was conducted using GaBi 9.1.0.53 software and selected method was ReCiPe 2016 v1.1 (midpoint hierarchist time-frame) method (Thinkstep, 2019). ReCiPe indicators provide information on the environmental issues associated with inputs and outputs of the product system (EN ISO 14040, 2006). It is a widely adopted method due to its robustness (Hischier et al., 2010).

#### 2.2.5. Sensitivity analysis

Sensitivity analysis “is a procedure to determine how changes in data and methodological choices affect life cycle assessment results” (EN ISO 14040, 2006). Sensitivity analysis is conducted to 1) identify key parameters or factors influencing a product system and the effect of different conditions on the product system 2) study uncertainty in a model input and apportioning of different sources of uncertainty in a model input (Guo and Murphy, 2012). For this study, sensitivity analysis will be based on the first option of how different factors influence a product system.

### 3. Results

#### 3.1. Environmental assessment of mortars

The results generated as presented in Fig. 4 are based on the environmental assessment of mix proportions of different mortars (Table 1). They illustrate the studied environmental impacts and contribution of different input materials and energy to respective impact categories. Here, S3 and S4 are compared to S1 and S2 (see section 2.2.2 for a detailed description of scenarios).

As seen in Fig. 4, the most contributing materials to S1 and S2 are sodium silicate powder and sodium silicate solution respectively. While WG-NaOH activator contribute the most to S3 and RHA-NaOH activator and sodium hydroxide are the most contributing materials to S4. All the other materials had minimal contribution of less than 10% in the respective impact categories, except for S3 and S4 where GBFS contributed 14% in fossil depletion impact category and in photochemical ozone creation (POC) formation where sand contribute 14% to S4. Overall, chemically modified waste-derived mortars in S3 and S4 have shown to have lower impacts than conventional mortars in S1 and S2. This is because of substituting conventional activator with waste-derived activator. This is also in agreement with other reported studies (Heath et al., 2014; McLellan et al., 2011; Passuello et al., 2017; Tong et al., 2018).

As seen in Fig. 4, with respect to climate change (excluding biogenic carbon), S3 and S4 both had 62% lower emissions when compared to S1; 58% and 59% reduced emissions when compared to S2 respectively leading to a potential reduction in greenhouse gases thereby reducing global mean temperature. This is a result of WG-NaOH and RHA-NaOH as alkali-activators respectively in S3 and S4. Environmental impact of the waste-derived activators mainly come from the treatment process and reaction of the wastes with NaOH to produced sodium silicate. NaOH is the major contributor to the waste-derived activators, contributing 98% of emissions in WG-NaOH activator and 69% emissions in RHA-NaOH activator. More information of environmental assessment of the activators can be found in the supplementary material.

In the fossil depletion impact category, S3 had reduced fossil fuel extraction by 61% and 53% respectively, when compared to S1 and S2. While S4 had reduced fossil fuel extraction by 59% and 51% respectively, when compared to S1 and S2. High fossil fuel extraction from S1 and S2 are as a result of sodium silicate powder and sodium silicate solution consuming 1 MJ/kg and 0.6 MJ/kg of fossil fuel respectively, during production (Fawer et al., 1999). Fossil depletion of WG-NaOH and RHA-NaOH mainly comes from sodium hydroxide used during the reaction process to produce alkali-activator signifying lower fossil fuel extraction.

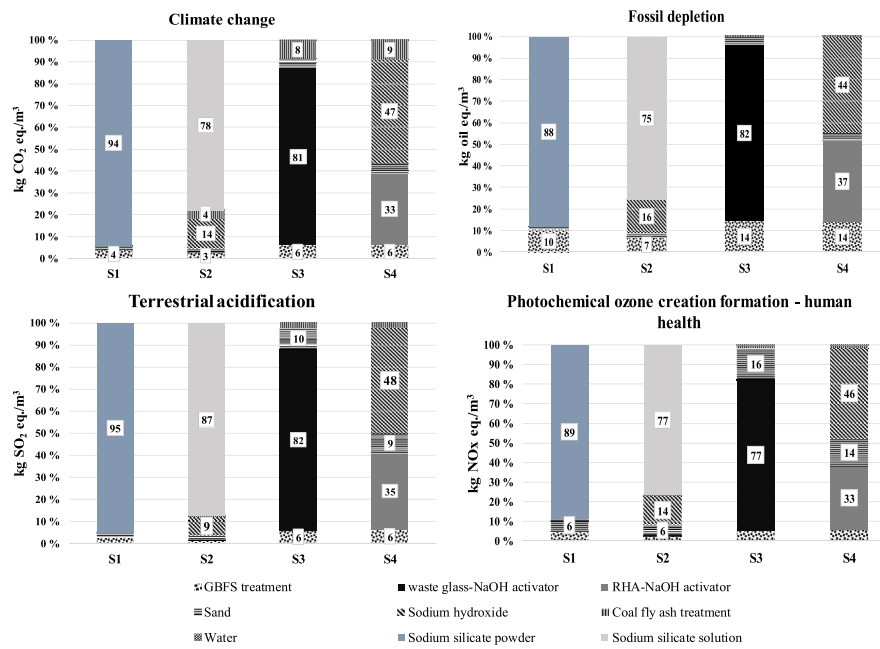


Fig. 4. LCIA results for the different scenarios.

With respect to terrestrial acidification, S3 and S4 had 75% lower emissions respectively when compared to S1 and S2, thereby reducing potential acidity of plant species. While, with respect to POC, S3 and S4 had reduced POC by 51% and 52% respectively when compared to S1, and reduced POC by 56% and 57% respectively, when compared to S2, reducing ozone creations leading to reduced frequency and severity of respiratory diseases in human health. As discussed in earlier impact categories, the major contributors are the activators and the avoided burden gained from using waste materials thereby lowering emissions from S3 and S4. Results showing all impact categories can be found in the supplementary material. This shows that waste-derived activators can completely substitute conventional alkali-activators while also producing materials of equivalent strengths. This is also in line with other reported studies (Passuello et al., 2017). Furthermore, this construes the fact that majority of environmental impacts from alkali-activated materials are associated with the type of alkali-activator used (Abdulkareem et al., 2019a, 2019b) and a waste-derived activator can significantly increase their environmental performance.

### 3.2. Sensitivity analysis

Sensitivity analysis is conducted on the key parameters influencing the system. Based on the results, sodium silicate and NaOH are the major contributing materials to the environmental burdens of AAMs. Consequently, sensitivity analysis will be performed on these chemicals. Additionally, influence of allocation is determined for WG and RHA respectively.

#### 3.2.1. Sensitivity analysis on sodium hydroxide using different production methods

High amounts of sodium hydroxide are required to chemically modify waste glass (S3) and RHA (S4) to produce alkali-activators. Also, additional sodium hydroxide is required in the development of aqueous alkali-activated mortar in S2 and S4 (see Table 1). Thus, the sensitivity analysis was mainly between S2, S3 and S4, using different production methods. The main production method used

in this study for sodium hydroxide is “EU NaOH from chlorine-alkali electrolysis, 100% NaOH, using membrane, diaphragm and amalgam technology mix”. The alternative production methods used for sensitivity analysis include “RER NaOH from brine solution, 100% NaOH” and “DE NaOH using diaphragm route, 100% NaOH”. These are all collected from GaBi database. More information on the emission factors of these different LCI can be found in the supplementary material.

The highest impact was observed in RER NaOH in all impact categories, while EU NaOH had the least emissions. It can be deduced that producing sodium hydroxide from brine solution (RER NaOH) and diaphragm route (DE NaOH) respectively, caused more environmental impacts compared to using a technology mix of membrane, diaphragm and amalgam production method (EU NaOH) (Fig. 5). To further reduce the environmental footprints of sodium hydroxide, more efficient production processes can be practiced which in overall improves the environmental footprint of alkali-activated materials.

#### 3.2.2. Sensitivity analysis on sodium silicate using different LCI data

Sodium silicate is a significant contributing material to the overall LCA results. Sodium silicate powder and sodium silicate solution was utilized in S1 and S2, respectively. As a result, sensitivity analysis in this section was mainly between S1 and S2, using different LCI data. The original LCI data used is from Ecoinvent database while the alternative LCI data employed is from Fawer et al. (1999).

With respect to climate change and POC, Fawer et al. (1999) exhibited higher impacts when compared to Ecoinvent in S1, at an estimate of 48% and 71% respectively. However, results were contrariwise for S2 with decreased emissions at 13% and 12% respectively. In fossil depletion category, higher environmental impact was observed in Fawer when compared to Ecoinvent for both S1 and S2 at 63% and 3% respectively, while results were the other way round with respect to terrestrial acidification with decreased emissions of 15% and 51% respectively. These results as shown in Fig. 6, show the interconnectedness of LCI data from both

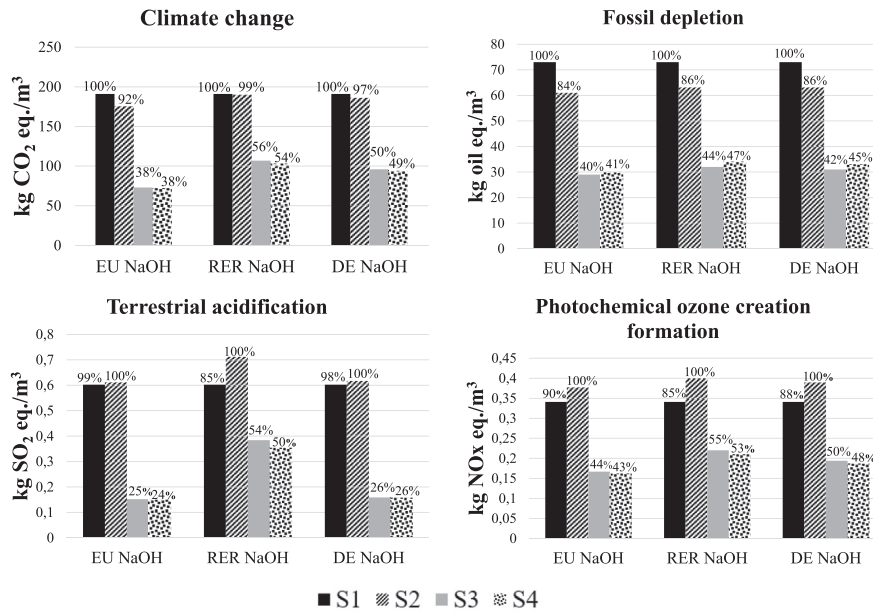


Fig. 5. Sensitivity analysis using different production methods for sodium hydroxide.

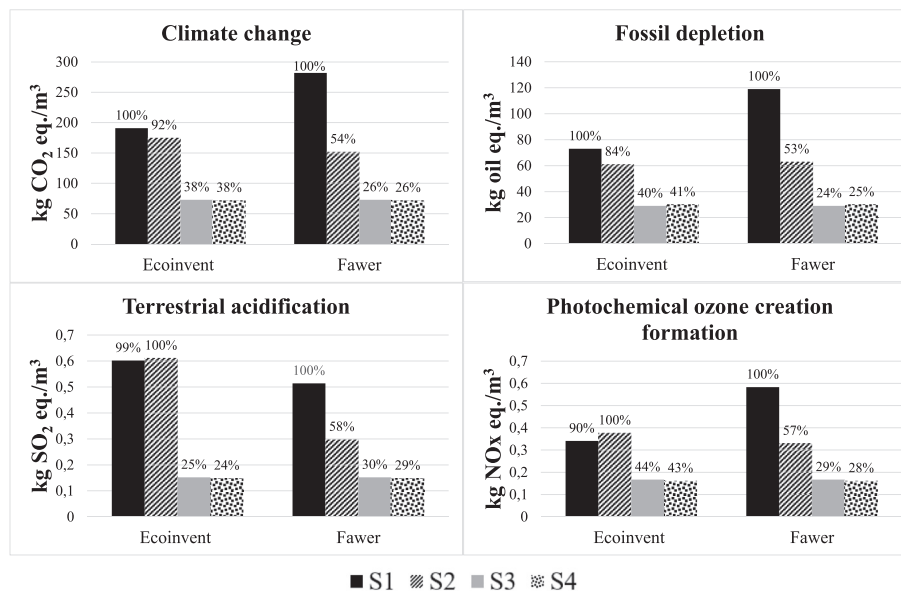


Fig. 6. Sensitivity analysis using alternative LCI data sources for sodium silicate.

Fawer et al. (1999) and Ecoinvent database. Conversely, a study carried out by European Commission on best available techniques (BAT) for manufacture of inorganic chemicals (EU, 2007), detailing inputs and outputs on sodium silicate solution, demonstrated that between 33% and 67% reduced environmental impact can be actualised for the studied impact categories when compared to Fawer et al. (1999) and Ecoinvent database (see supplementary material). This is because of reduced energy consumption and usage of more efficient technologies as document by BAT (EU, 2007). However, the study did not have detailed information for sodium silicate powder. Nonetheless, it can be assumed that reduced emissions are also achieved.

### 3.2.3. Sensitivity analysis on waste glass and RHA

Waste glass (WG) and RHA were assumed to be waste in this study and only associated emissions from the treatment processes were considered. Based on this assumption, a sensitivity analysis was performed to determine how mass allocation of these waste materials influence overall results of the study. Data for WG and rice were collected from Ecoinvent. Allocation of RHA was estimated from rice (non-basmati) production. As discussed earlier, RHA share in rice husk is 18–22% of dry content by weight while 20% of rice husk is generated from rice paddy, which makes it approximately 4% of RHA from rice paddy. Thus, an estimate of 4% of emissions from production of rice (non-basmati) is allocated to RHA (more details can be found in the supplementary material). Fig. 7 presents the influence of allocating emissions to WG and RHA.



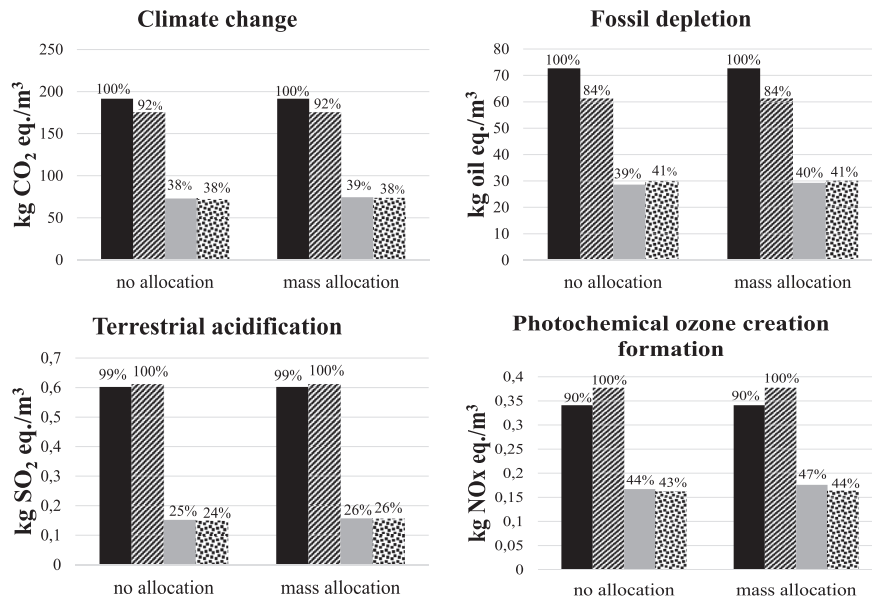


Fig. 7. Sensitivity analysis based on mass allocation of RHA and waste glass.

Scenarios S3 and S4 are particularly focused on since they contain these respective wastes materials. According to Fig. 7, it can be seen that WG and RHA are not so sensitive to the allocation procedure in all the analyzed impact categories, thus, overall, making S3 and S4 a more environmentally friendly option than S1 and S2.

### 3.3. Cost analysis of mortars

To determine the viability of the AAMs considered in this study, financial cost comparison is required. Thus, this study examines a simplified cost of materials needed to generate the cost of a given formulation of mortar. Sodium silicate provides a significant reactive part of silica in combination with NaOH to produce conventional alkali-activator. From data available on industrial trading websites (alibaba, 2019), conventional sodium silicate powder and solution was estimated to cost an average of 414 €/ton and 203 €/ton respectively. NaOH pellets was estimated to cost an average 414 €/ton. The cost associated with production of the waste-derived alkali-activator are as follows:

a) Cost of one-part WG-NaOH alkali-activator: Based on European statistics, material price for recyclates such as waste glass is 55 €/ton (eurostat, 2019). Thus, considering the mass proportion of WG needed for alkali-activator production, cost of WG was estimated to be 29 €/ton of activator. Additionally, based on mass proportion of NaOH pellet required to chemically modify WG powder, cost of NaOH pellets was estimated to be 199 €/ton of activator. Since it is assumed in this study that all wastes are locally available, negligible transportation costs have been considered. Electricity required for milling glass cullet to glass powder is estimated to be 50 kWh/ton (Vinai and Soutsos, 2019). Thus, 26 kWh/ton is estimated for milling of glass cullet. With market price of electricity for Finland estimated to be 0.064 €/kWh as of 2018, 1.7 €/ton is required to mill glass and energy cost for producing alkali powder from waste glass has been estimated to be 38 €/ton. In total 268 €/ton has been estimated to produce powder alkali-activator from WG, of which 74% of the cost is a result of cost of NaOH.

b) Cost of two-part RHA-NaOH alkali-activator: Price of RHA was collected from industrial trading sites (alibaba, 2019). Considering the required mass proportion for RHA required for alkali-activator production, average cost of RHA required for activator is 16 €/ton. Considering mass proportion of NaOH pellet required to chemically modify RHA, cost of NaOH pellet was estimated to be 36 €/ton of activator. Market price for electricity in Finland was estimated 0.064 €/kWh as at 2018, thus, electricity cost for producing alkali-activator from RHA has been estimated to be 14 €/ton. In total, 69 €/ton has been estimated to produce aqueous alkali-activator of which 52% of the cost is as a result of NaOH.

Prices of CFA and GBFS were collected from industrial trading sites with an average price of 35 €/ton and 43 €/ton respectively (alibaba, 2019). Price of water and sand were based on Finnish market prices, at an average price of 5 €/ton and 8 €/ton, respectively. Based on mass proportion of the different input materials,

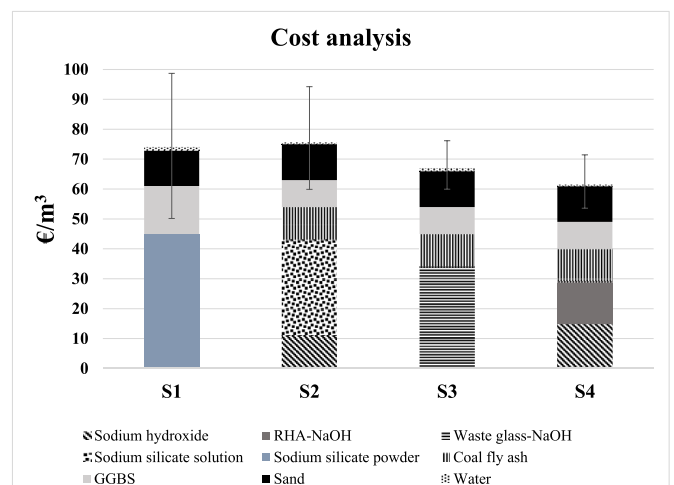


Fig. 8. Simplified cost analysis for the different alkali-activated mortars.

cost of the different mortars was calculated as presented in Fig. 8. Error bars were used to depict the price range of the different mortars. S3 and S4 AAM are slightly cheaper than S1 and S2 AAM, however, there is also possibility of S1 and S2 being cheaper if the lower values of the error bars are considered. These prices may vary quite much because there are no standardised prices for waste materials (RHA, CFA and GBFS) as compared to OPC. With industrially produced AAMs and standardised prices, cheaper price of production can be achieved.

#### 4. Discussion and perspectives

Alkali-activated mortars are considered a low carbon alternative to cement mortars. However, producing a more compact and dense material with higher mechanical strength highly depends on the quantity of alkali-activator. Alkali-activators have also been highlighted as the most contributing material to the environmental impact of alkali-activated materials, with many studies recommending using waste-derived alkali-activators as a substitute to conventional alkali-activators. Furthermore, reduced emissions from sodium silicate can be achieved when fossil fuels are replaced with renewable energy (Abdulkareem et al., 2019a). Despite this being a sustainable alternative in producing alkali-activator, virgin raw materials are still consumed and a high energy consumption in production. WG and RHA have high volumes globally, containing significant shares of silicon oxide which is one of the major elements required in developing alkali-activators. Hence, producing alkali-activator from these waste materials, will mean avoidance of virgin materials consumption and enhancing circularity of wastes.

It should be noted RHA is predominantly produced in rainfed upland regions where rice is grown and incinerated. Therefore, RHA might not be as available in some countries. On the other hand, other silica rich agro-wastes such as straw can be used to replace RHA for countries where rice is not locally produced. WG is available almost everywhere, but its production is more distributed regionally where only large cities produce significant amounts. Therefore, collection from larger areas to the place of utilisation will be needed. Furthermore, material availability and quality of industrial side-streams such as CFA and GBFS, are crucial to the development of AAMs. These industrial side-streams are dependent on production quantity and method of production which can lead to variability in their chemical compositions (Provis, 2018). GBFS is increasingly used in the cement industry, which can reduce its availability for alkali-activated materials. Also, the adoption of renewable energy will limit the availability of CFA, which is a product of energy production from coal combustion. Consequently, research can investigate more materials that do not require much beneficiation (grinding, granulating, and high temperature processes) and has zero to low conventional value for sustainable material productions. Thus, further studies will investigate the scale of supply and demand of these materials and their proposed application.

To further increase the environmental sustainability of waste-derived activators and industrial side-streams, it is important, they are locally available. However, this usually is not the case, and in some instances, raw materials must be transported from one region to another. Transportation implies added cost in production and additional emissions. Depending on where the waste is sourced and where it is produced, transportation burdens might lower net environmental and economic benefits of using wastes. Additionally, increased environmental performance can be actualised by utilising waste heat or renewable energy in the production of waste-derived alkali-activators and beneficiation processes of side-streams.

#### 5. Conclusions

This study highlights the relevance of using waste-based activators to produce alkali-activated mortars in developing environmentally friendly materials. This study lays emphasis on one-part (powder) WG-derived alkali-activated mortar and two-part (aqueous) RHA-derived alkali-activated mortar. In addition, a simplified cost analysis was carried out to compare the cost of these different mortars to assess their economic sustainability. Conclusions from this study include:

- Waste glass and rice husk ash derived alkali-activated mortar resulted in up to 62%, 61%, 76% and 56% lower emissions respectively, in all the four studied impact categories (climate change, fossil depletion, terrestrial acidification and photochemical ozone creation formation) when compared to conventional one- and two-part alkali-activated mortar. Additionally, one-part WG-derived alkali-activated mortar without additional sodium hydroxide during mortar production, exhibits possibility of producing mortar almost completely from side-streams (waste glass, fly ash and granulated blast furnace slag) while maintaining equivalent compressive strength as conventional alkali-activated mortars.
- Waste glass and rice husk ash demonstrated little sensitivity to mass allocation procedure, with increased emissions from 0.2% to 5% in the above-mentioned impact categories, thus, making the waste-derived alkali-activated mortar a more environmentally friendly option.
- Sodium hydroxide was consistent in this study in chemically modifying waste glass and RHA to produce alkali-activators and in the development of aqueous alkali-activated mortar. Thus, a sensitivity analysis was carried out on sodium hydroxide using data from different production methods. The default production method used in this study is "EU-28 sodium hydroxide from technology-mix" while the alternative production method for sensitivity analysis includes "RER sodium hydroxide from brine solution" and "DE sodium hydroxide using diaphragm route". Results illustrated that increased emissions between 1% and 153% was observed from the alternative production routes. Thus, sodium hydroxide production from technology-mix, demonstrated improved environmental performance than sodium hydroxide from brine solution and diaphragm route, respectively. However, with continuous improvement in the emissions profile of the chlor-alkali industry, lower emissions can be expected when assessing sodium hydroxide production in future.
- Sensitivity analysis on sodium silicate using an alternative LCI data (Fawer et al., 1999) showed that with respect to S1, 48%, 63%, and 71% increased impact was observed respectively, for climate change, fossil depletion, and POC and 15% reduced impact in acidification category. With respect to S2, between 12% and 51% of reduced impact is observed for all impact categories except fossil depletion which had a 3% increase. This is all realised when alternative LCI data is compared to the original LCI data.
- With respect to costs of producing waste-derived alkali-activated mortars, analysis show that, a range of 9%–19% of cost savings can be achieved from using waste-derived alkali-activated mortars compared to its conventional counterpart. Additionally, local availability of primary and secondary raw materials needed for production of alkali-activated materials will impact positively on the total cost of production.

It can be concluded that significant environmental impact reductions are not only achieved from waste-derived alkali-activated

mortars, but also provides a useful alternative to utilising waste residues. This study is based on literature studies and quantitatively the results are valid only for those. We can however generalize quite safely that usage of waste glass or rice husk ash as an activator reduces environmental impacts of alkali-activated mortars if their usage also reduces or substitutes the need for conventional alkali-activators in alkali-activated mortars.

### CRedit authorship contribution statement

**Mariam Abdulkareem:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Visualization, Writing - original draft, Writing - review & editing. **Jouni Havukainen:** Conceptualization, Data curation, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Writing - review & editing. **Jutta Nuortila-Jokinen:** Conceptualization, Funding acquisition, Project administration, Resources, Visualization. **Mika Horttanainen:** Conceptualization, Funding acquisition, Methodology, Resources, Supervision, Validation, Visualization, Writing - review & editing.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

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