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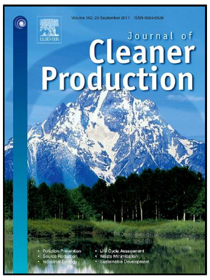

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Evaluation of the potential improvement in the environmental footprint of geopolymers using waste-derived activators

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Abstract

Geopolymers produced from an aluminosilicate precursor and an alkaline activating solution have emerged as low carbon alternative binders which can substitute for Portland cement (PC) in many applications. The presence of soluble silicate in the activating solution of a geopolymer is known to yield a denser and more compact material with higher mechanical strength compared to hydroxide-activated geopolymers. However, these silicate solutions are the most expensive component of geopolymer cements, as well as the highest contributors to their environmental impacts in most life cycle categories. Geopolymers are widely accepted as a more environmental friendly material due to their claimed lower CO_2 emissions due to their synthesis from industrial by-products or wastes, as well the low energy demand during their production. However, the use of alkali-silicate activators can significantly increase other environmental impacts, leading to controversies regarding whether geopolymers can really be considered as a more sustainable material. Thus, this study evaluates the life cycle impacts of a geopolymer produced from a kaolin sludge residue from the Brazilian mining industry. Alkaline solutions derived from sodium hydroxide solutions and two different soluble silica sources were used as activators: a commercial sodium silicate (waterglass), and chemically modified rice husk ash (RHA). The processes which contribute the most to the life cycle impacts of geopolymers are thermal curing, waterglass production, and sodium hydroxide production. The use of RHA-derived sodium silicate may reduce environmental impacts by more than 60% in 6 of the 9 categories assessed, indicating that this is a favourable alternative where RHA is locally available. Although the binders evaluated here have differences in mechanical properties, those using RHA-derived activators exhibit impacts lower than PC for 4 of the 8 categories evaluated, and a reduction of more than 70% in global warming potential. RHA-based activators are identified as a promising alternative for impact reduction in geopolymer production, and more detailed assessments of the performance and reactivity of these activators should be conducted.

Keywords:

Life-Cycle Assessment (LCA); waste valorization; geopolymer; rice husk ash; metakaolin.

1. Introduction

To progress toward a more sustainable construction industry, various alternative cements have been assessed (Scrivener et al., 2016). In this sense, the development of the class of materials known as alkali-activated cements or "geopolymers", as well as calcium sulfoaluminate, belite-rich, and magnesium-oxide based cements, can play an important role in emission reduction strategies and reduce reliance on Portland clinker-based cement (Shi et al., 2011).

Geopolymers are a type of cement derived from the alkali activation of industrial wastes or by-products (usually fly ash or amorphous aluminosilicate minerals). These materials are now being used in large-scale concrete production, and can show technical and environmental advantages compared to PC (Duxson et al., 2007; Van Deventer et al., 2012). A key advantage often quoted in the technical literature is the saving of as much as 40-80% of the GHGs emissions compared to PC, as well as the re-use of wastes and/or industrial by-products as major constituents of the geopolymer binder (Bernal et al., 2016; Davidovits, 2008; Habert et al., 2011; McLellan et al., 2011). The alkali solutions most commonly used as activators in geopolymer synthesis are sodium and potassium silicates. The presence of silica in the activating solution tends to lead to the formation of a denser and more compact binder structure with high mechanical strength (Fernández Jiménez and Palomo, 2005; Yang et al., 2008). However, these silicate solutions represent the most expensive component in geopolymer production, and are the highest contributor to the environmental footprint of these materials (Habert and Ouellet-plamondon, 2016; Habert et al., 2011), as alkali silicates are produced by thermal or hydrothermal routes from silica and alkali carbonates or hydroxides (Fawer et al., 1999).

The environmental impacts of a geopolymer binder can be calculated as the contributions of its two main components - precursor and alkali activator - as well as the energy involved in its manufacture (thermal and mechanical processes, and including any non-ambient curing steps) and raw materials transportation. Life cycle assessment (LCA) reports have identified that the embodied energy and environmental impacts (global warming potential and others) are mainly affected by the type and concentration of alkali-activator solution (Habert et al., 2011; Heath et al., 2014; McLellan et al., 2011; Tempest et al., 2009; Witherspoon et al., 2009). Evaluating the eco-profile of different geopolymers compared to PC concrete, Habert et al. (2011) found that although offering decreased CO_2 emissions, geopolymer production was calculated to increase some other environmental impacts by up to 1000%, such as fresh water ecotoxicity (FAETP), especially due to waterglass production.

The interest in valorising additional industrial wastes and by-products, as well as the need to reduce the production cost of geopolymer binders, has promoted the development of alternative alkali-activators based on the combination of alkalis with waste-derived amorphous silica sources; these can exhibit similar or even higher effectiveness compared to traditional alkali-activator based on silicate soluble (Bernal et al., 2011; Detphan and Chindaprasirt, 2009; Puertas and Torres-Carrasco, 2014; Rodríguez et al., 2013; Zivica, 2004, 2006). Agro-industrial wastes such as rice husk ash (RHA) have a high potential for implementation as a silicate source for the alkali activator (Bernal et al., 2012; Bouzón et al., 2014; Detphan and Chindaprasirt, 2009; Kim et al., 2014; Prasetyoko et al., 2006). RHA is generated by biomass power plants burning rice husks, and is mainly made up of silica (ideally > 90%), potassium (< 5% as K_2O) and phosphorous (<3% as P_2O_5), with variable (but ideally low) unburnt carbon content. RHA can be dissolved into an alkali hydroxide solution for use as a silica source in geopolymer synthesis, yielding similar mechanical properties compared to the use of commercial waterglass (Bernal et al., 2012). Although studies have shown a potential reduction of GHG emissions of 63% when an alternative sodium silicate based on RHA is used (Mellado et al., 2014), the effect on other environmental impact categories has not yet been assessed in detail and can be considered the novelty of the work presented here. Therefore, the assessment of the real and quantified environmental impact related to the alkali-activators in geopolymers and how can be reduced through the use of alternative silicate solutions derived from industrial wastes or by-products reveals new pathways for development of materials with higher sustainable criteria.

The present study aims to assess the life cycle environmental impacts associated with the production of geopolymer binders based on a kaolinitic residue, and specifically the effect of using alternative alkali activators based on RHA chemically modified by caustic solutions. The systems produced using conventional sodium silicate solution were previously assessed by (Longhi et al., 2016). The results are compared to geopolymers synthesized using sodium hydroxide and waterglass as activators, and to traditional Portland cement, to assess whether the application of an alternative sodium silicate may reduce environmental impacts associated with geopolymer production. The document shows a section related to the raw materials used and the subsequent procedure related to the geopolymers production followed by all the information related to LCA data based used. The results of LCA are also shown considering the assumptions of different conditions, such as transport, application of thermal curing and mechanical performance.

2. Materials and Methods

2.1. Materials

Calcined kaolin sludge (CKS) was used as the aluminosilicate precursor for geopolymer production in this study. The kaolin sludge was generated in the mining industry of the state of Pará (Brazil) in the process of beneficiation to obtain high purity kaolin. Various wastes are generated during beneficiation; here, the residue generated between centrifugation and filtering steps (which is rejected as impure kaolin, and usually sent to tailings dams) was considered. The high content of kaolinite and moderate amounts of titanium dioxide, ferrite and quartz present in this residue allow the generation of a pozzolanic powder through calcination (Barata and Angélica, 2012). In this study, the kaolin sludge was dried in a filter press, calcined at 750 °C for 1 h, and ball milled for 1 h (Longhi et al., 2016; Souza and Dal Molin, 2005). The chemical composition, mean particle size and specific surface of the CKS is shown in Table 1. Alkali activators derived from analytical grade NaOH (~99%) and two different soluble silicate solutions were used. Sodium silicate solution with 26.5 wt.% SiO_2, 10.6 wt.% Na_2O, and 62.9

wt.% H_2O (Sigma-Aldrich) was used as a reference soluble silica source. The alternative alkali activator was prepared by the dissolution of a rice husk ash (RHA) obtained from the rice industry of southern Brazil, into NaOH solution. The RHA was mainly constituted by SiO_2, as is shown in the Table 1. The RHA-based solution was prepared with analytical NaOH to obtain a SiO_2/Na_2O molar ratio of 2.2, with dissolution carried out at 100 ºC for 1 h in a refluxed system to achieve the highest dissolution degree and avoid loss of water by evaporation (Bouzón et al., 2014). The content of the dissolved silica was calculated and adjusted trough the weight of non-dissolved RHA remaining after the dissolution and subsequent filtration processes. From this information, the composition of the filtered RHA-based solution was calculated to be 19.2 wt.% SiO_2, 8.8 wt.% Na_2O, and 70.6 wt.% H_2O.

Table 1. Chemical composition, mean particle size and specific surface of the raw materials used.

		CKS (Calcined kaolin sludge)	RHA (Rice husk ash)
Chemical composition (wt%)	SiO_2	61.3	97.3
	Al_2O_3	33.8	--
	Fe_2O_3	2.8	--
	TiO_2	2.1	--
	K_2O	--	2.1
	CaO	--	0.6
Mean Particle size (µm)		11.0	9.87
Specific surface ($m^2 \cdot g^{-1}$)		19,7	9.82

For comparative purposes, Portland cement (PC) type II-E (ABNT, 1997), with a 75 wt.% clinker, 20 wt.% slag and 5 wt.% of gypsum, with a mean particle size of 29.8 µm, was used as a reference binder.

2.2. Sample preparation

All geopolymers were formulated with an overall Na_2O/Al_2O_3 molar ratio of 1.0, and the content of soluble silicates (either reference or alternative) was adjusted to obtain an overall SiO_2/Al_2O_3 molar ratio between 3.1 and 4.0, by manipulating the silicate activator content and adding solid NaOH to adjust the overall Na_2O content. Additional water was added to the activation solution in order to achieved a water/solids ratio of 0.40. The pastes were mixed mechanically for 6 min and then cast into 20 mm cubic moulds, vibrated for 1 min, and cured at

50 ºC for 24 h with a high relative humidity (RH >90%). After thermal curing, the samples were stored in a sealed plastic container at room temperature and 90% RH. The mix designs and synthesis conditions are shown in Table 2, where the pairs of systems WG-1/RHA-1 and WG-2/RHA-2 each have equivalent SiO_2/Al_2O_3 molar ratios (i.e. contents of soluble silicates). The quantities reported are adjusted to obtain 1 kg of fresh paste, which comprises the precursor (CKS), anhydrous activator (NaOH and sodium silicate), and water. The PC paste with a water/cement ratio of 0.40 was produced by mechanical mixing for 1 min, and cured at 25 ºC immersed in saturated limewater.

Table 2. Geopolymer formulations (per kg of paste) defined according to their overall molar compositions, for the mixes of calcined kaolin sludge (CKS) activated by sodium hydroxide (NaOH), and sodium hydroxide combined with commercial waterglass (WG) or dissolved rice husk ash (RHA).

Mix ID	Overall molar ratio		Quantity of raw materials (g)				
			Precursor	Source of soluble silicates*			
	SiO_2/Al_2O_3	Na_2O/Al_2O_3	CKS	WG solution	RHA-based solution	NaOH (solid)	Additional water
NaOH	3.1	1.0	568.0	--	--	293.9	139.5
RHA-1	3.5	1.0	504.5	--	217.1	211.2	67.2
RHA-2	4.0	1.0	446.3	--	417.0	135.7	1.0
WG-1	3.5	1.0	537.6	138.8	--	220.2	103.4
WG-2	4.0	1.0	506.0	283.7	--	144.1	66.1

*Source of soluble silicates: Corresponds to the sodium silicate solution either waterglass or RHA-based solution. The values reported correspond to the total amount of $Na_2SiO_3 \cdot nH_2O$.

The compressive strength was evaluated for hardened pastes using a Universal testing machine (EMIC) with a crosshead displacement speed of 0.5 mm/min according to an adaptation of the Brazilian Standard NBR 7215 (Associaçao_Brasileira_de_Normas_Técnicas, 1995) suitable of the specimen type produced here, in order to analyze five 20 mm cubic paste specimens for each mixture. Testing was conducted at 28 days to enable the geopolymers to be compared directly to PC-based systems.

2.3. Life cycle assessment

2.3.1. Goal of the study

The goal of the study is to compare the life cycle environmental impacts of 1 kg of geopolymer paste (CKS+ anhydrous alkali activator + water) derived from different raw materials.

2.3.2. Study scope: system boundaries and functional unit

There are several environmental impacts related to geopolymer production, such as freshwater, marine and terrestrial ecotoxicity, human toxicity, and others, which are mainly associated with waterglass production (Habert et al., 2011; Mellado et al., 2014). Considering that the aim of replacement of this raw material is to decrease these environmental impacts, the study is focused on the assessment of an alternative sodium silicate solution, derived from locally available materials in Brazil.

The system studied considers the commercial production of five geopolymer formulations, from cradle to gate, including raw materials production, transport, and waste beneficiation, and all the associated emissions and energy consumptions (Figure 1). Mixing and molding are not considered within the system boundaries.

The relative reactivities of the precursors used in a geopolymer mix, as well as the material dosages, define the kinetics of the chemical reactions taking place during activation and the subsequent evolution of mechanical strength. Most fly ash-based geopolymer systems require the use of a thermal curing with temperatures up to 80 °C in order to achieve an acceptable strength at early ages. On the other hand, the use of thermal curing can be omitted under many conditions when highly-reactive metakaolin or slag are used as precursors (Mo et al., 2014; Van Deventer et al., 2012). Thermal curing for metakaolin-based geopolymers can provide marked advantages in the mechanical performance at early ages (Heah et al., 2011) and a higher efficiency in the manufacture of precast products at large scale. Therefore, in this study, thermal curing is considered in the LCA according to the conditions followed in the experimental program.

The functional unit is 1 kg of paste (CKS + anhydrous alkali activator + water). This procedure was selected to evaluate binder production, as this product may have multiple specific applications in civil engineering (columns, beams, walls, bridges, pavements, etc.), each requiring a distinct concrete mix design, and so a singular function cannot be selected. It may be assumed, as a starting point, that the materials here are to be used in applications where a similar mass of paste is needed in each case to achieve comparable in-service performance (i.e. subject to minimum cover depth requirements or specified block/element geometries, rather than defining the volume of paste required for a given element strength), which enables an initial comparison to be made on a mass basis rather than requiring normalization to account for different material performance levels. This aspect will be revisited later in the paper. Since RHA and CKS derived from a mining industry are wastes arising from other life cycles, only the impacts incurred in their beneficiation were taken into account in this study.

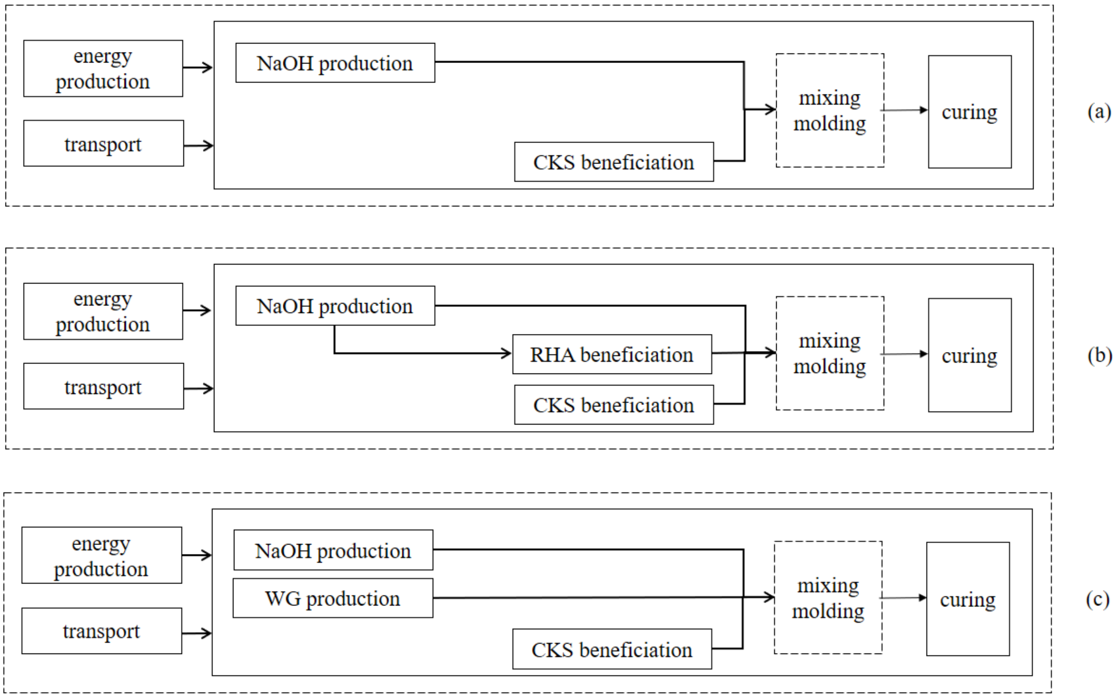

Figure 1. Schematic description of system boundaries for the geopolymers (a) NaOH-based geopolymer (without sodium silicate); (b) RHA-1 and RHA-2; and (c) WG-1 and WG-2.

Although the synthesis conditions of the geopolymers were defined at laboratory scale, their environmental profile calculation considered production at industrial scale (except for thermal curing), considering mean energy consumption values informed by local providers. In this sense, the beneficiation process for the kaolin sludge includes the power consumption of filter presses (drying), rotary kilns (calcination) and industrial grinders (milling). The values for the equipment consumption were collected considering the equipment currently commercially available in Brazil. The total amount of energy spent in the beneficiation of 1 kg of kaolin sludge is 0.0034 kWh for drying, 0.0046 kWh for thermal processing, and 0.0190 kWh for milling. The assessments of energy involved during the beneficiation processes were carried out assuming a sludge with initially 80 wt.% water which was dewatered, and a further mass loss of 17% due to dehydroxylation via thermal processing of the clay minerals. Other potential outputs such as particulate matter released in milling, and the emissions from burning were not considered. These values are expected to be about 20 times lower than those reported by other studies (Habert and Ouellet-Plamondon, 2016; Heath et al., 2014), since these authors considered the whole metakaolin production chain, while the present study is based on the beneficiation of a residue (kaolin sludge).

RHA beneficiation processes include the electricity consumption of a reflux condensation reactor (0.0084 kWh per kg of alternative sodium silicate solution) to digest RHA in NaOH solution, with 16% loss of weight during this process.

The composition of sodium silicate is generally expressed as $Na_2O \cdot nSiO_2$, where n corresponds to the molar ratio. Commercial waterglass solutions can be available with SiO_2/Na_2O molar ratios between 1.6 and 3.3, produced by thermal (furnace) or hydrothermal processes. At higher SiO_2/Na_2O molar ratios, the content of soluble silicates is increased and the subsequent quantity of solution required for a specific alkaline activation can be also modified. According to Zah and Hischier (2007), in the furnace process, waterglass is produced directly by melting pure silicon sand and soda at temperatures around 1100 - 1200 °C, where a material with high SiO_2/Na_2O molar ratio is generated. This sodium silicate can be used in a solid form, but is usually dissolved and made available as an aqueous solution (~37 wt.%). In the hydrothermal process, sand is dissolved in sodium hydroxide solution within an autoclave at temperatures between 180 – 300 °C (Trabzuni et al., 2011). Zah and Hischier (2007) report that, after filtration, sodium silicate with a lower SiO_2/Na_2O molar ratio is obtained as a 48 wt.% solution. This solution may be later spray-dried in order to obtain a powder.

In this work, sodium silicate 48 wt.% hydrothermal liquor was selected for LCA calculations, since it is the product in the Ecoinvent database (Ecoinvent Centre, 2014) closest in composition to that which is commercially available in Brazil (as used in the experimental design). The difference in water content of the sodium silicate (37 vs. 48% solids) was corrected proportionally for the LCA calculations. It is important to note that sodium silicate production by the hydrothermal liquid process is expected to consume about 10 times less energy than the furnace process, as reported by Fawer et al. (1999). The results presented in the following sections are valid for this specific situation. The results for geopolymers derived from other types of sodium silicate may vary, as also stated by Provis (2014).

Sodium hydroxide is produced mainly by the chlor-alkali process, where chlorine and alkali are produced by electrolysis of a salt solution, through three main technologies: diaphragm cell, membrane cell, and mercury cell (Althaus et al., 2007). Although the mercury cell was the first deployed at industrial scale, and is still the most used in some parts of the world, in Brazil the dominant technology is the diaphragm cell (63%), followed by the membrane cell (23%), and the mercury cell (14%) (ABICLOR, 2012). In this study, the Brazilian NaOH production mix was considered, yielding a 50 wt.% NaOH aqueous solution. Ecoinvent data were adapted for the chemical factory construction, not considering the module "building, multi-storey", assuming that large storage areas are not needed for sodium hydroxide production, and considering that the database instructs that this module shall not be used if its relative importance would be high in a certain environmental inventory, as is the case here (Kellenberger et al., 2007).

Assessing the contribution of transport by truck, which is the most used mode for cargo transportation in Brazil, a distance of 100 km from the raw material producers to the mix plant was considered as an indicative case. However, the cost is higher as the distance is increased.

Finally, for comparison purposes, Ecoinvent data for Portland cement (cement production, blast furnace slag 5-25%, US only), was adapted to the Brazilian production profile, considering one of the most currently used cements in Brazil, with 75 wt.% clinker, 20 wt.% ground-granulated blast-furnace slag (GBFS) and 5 wt.% gypsum. The LCA for the PC systems was carried out on the basis of 1 kg of cement paste (clinker + gypsum + GBFS + water, with a w/c ratio of 0.40).

Thermal curing of geopolymer materials was assessed considering a heat curing chamber, lab scale, with a consumption of 0.54 kWh per kg of paste. It is noted that this energy consumption is based on a small laboratory oven and so is a very conservative value when compared to larger industrial processes which can operate at higher energy efficiency, either through the use of recovered waste heat or simply through reduced losses per unit mass of product in a much larger operation. Considering that thermal curing is not widely applied to PC-based materials in Brazil, it was not considered within their LCA.

2.3.3. Inventory analysis

Data from raw materials production, transport emissions, and energy production were extracted from the Ecoinvent database version 3.1 (Ecoinvent Centre, 2014), and adapted to the Brazilian context, considering the Brazilian energy grid mix (Table 3).

Table 3. Brazilian energy grid mix, used for all foreground processes

Energy source	Share (%)
Hydropower	83.76
Natural gas	5.87
Biomass	3.44
Oil	2.89
Nuclear	2.46
Hard coal	1.56
Wind	0.01

The cut-off procedure was applied to the residues, and the impacts associated with their generation were not allocated in this study.

2.3.4. Life cycle impact assessment

The CML-01 method (Guinée et al., 2000) was applied for impact calculation in the following categories: acidification potential – generic (AP), global warming potential –100yr (GWP), eutrophication potential – generic (EP), freshwater aquatic ecotoxicity – 100yr (FAETP), human toxicity potential – 100yr (HTP), marine aquatic ecotoxicity – 100 yr (MAETP), photochemical oxidation (summer smog) – low NO_X (POCP), stratospheric ozone depletion – steady state (ODP), and terrestrial ecotoxicity – 100 yr (TAETP). Inventory and impact calculations were performed in the software OpenLCA (GreenDelta, 2013).

3. Results and Discussion

3.1. Environmental analysis of the binders produced

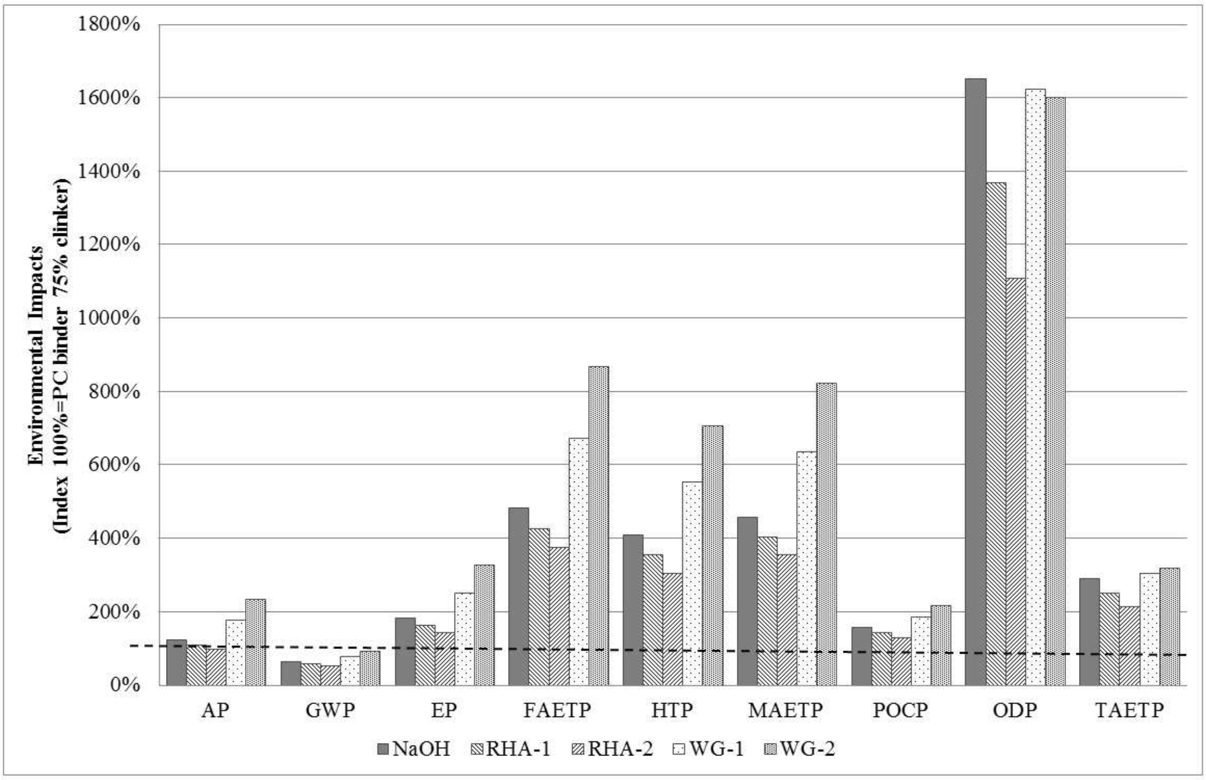

Figure shows the environmental impacts related to the production of 1 kg of geopolymer binder, in comparison with Portland slag cement with 75% clinker, based on pastes with the same cement content. It is important to note that for GP pastes thermal curing was applied, while for PC paste no thermal treatment was considered. The results show different trends according to the impact category. In comparison to Portland cement, the geopolymers present lower global warming potential (GWP) impacts: reductions between 7 and 22% for the binders based on waterglass (WG-1 and WG-2), between 41 and 47% for those based on RHA, and 35% for the binder derived from an alkali activator based on NaOH. These achievements are lower than those reported by Mellado et al., (2014), who calculated reductions of 50% for a WG-based geopolymer and 63% for RHA geopolymer. However, these authors considered PC mortar with an emission factor of 1 kg CO_2 per kg cement, which is higher than the one applied to this study, 0.73 kg of CO_2 per kg cement. The reductions compared to Portland cement are also less than the headline claims of many studies in the literature as summarized in the Introduction section of this paper, and this will be examined in detail below.

For the categories acidification potential (AP), eutrophication potential (EP) and photochemical oxidation (POCP), the geopolymer blends based on rice husk ash (RHA-1, RHA-2) have impacts up to 62% higher than PC. For the remaining categories (freshwater aquatic ecotoxicity - FAETP, human toxicity potential - HTP, marine aquatic ecotoxicity - MAETP, stratospheric ozone depletion - ODP, terrestrial ecotoxicity - TAETP), each of the geopolymer binders assessed show potential impacts higher than PC, with values up to 17 times higher for ozone layer depletion (ODP). These trends are in accordance with those reported by Habert et al. (2011), for fly ash based concrete geopolymer in comparison with PC concrete (70% clinker, 30% fly ash), except for FAETP and ODP. Although these trends are similar, the comparison is only illustrative, because Habert et al. (2011) compare FA geopolymer concretes and PC concrete with same mechanical strength, while this study compares PC and geopolymer binders with different mechanical properties.

It is interesting to note that in the case of ODP, the values for the GP binders are between 11 and 17 times higher than PC. However, the emissions associated with 1000 kg of PC paste (1.34×10^{-5} kg CFC-11-eq) are similar to those emitted by a single incandescent lamp generating 20 million lumen-hours of light (i.e. a typical 800-lumen household lamp operating for 2.85 years; 1.1×10^{-5} kg CFC-11-eq) (Scholand and Dillon, 2012), so it is evident that the overall impact in this category is rather low in the context of other societal activities. The ecotoxicity (FAETP, MAETP, TAETP) and human toxicity (HTP) characteristics of the geopolymer formulations also appear unfavorable due particularly to the use of NaOH. However, with continuing improvement in the emissions profile of the chlor-alkali industry and the reduction in reliance on mercury cells for alkali production worldwide, this should also be considered a conservative calculation when assessing future materials production. Binders produced using an alternative silica source based on chemically modified rice husk ash (RHA-1, RHA-2) have lower impacts than those based on commercial waterglass for all the categories evaluated. This agrees with the reports in other studies (Habert et al., 2011; McGuire et al., 2011; Mellado et al., 2014; Provis, 2014; Weil et al., 2009), that relate the majority of geopolymer impacts to activator production, and highlights the importance of the development of alternative sourcing pathways for activators as discussed here.

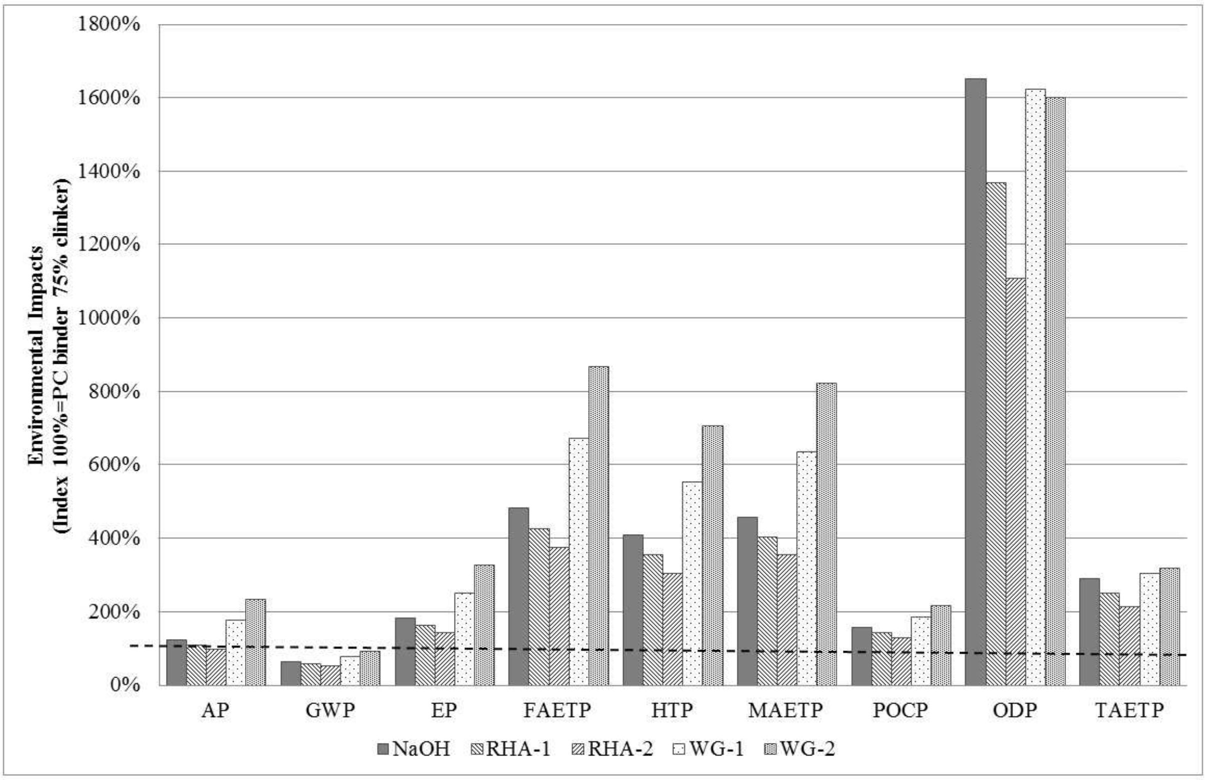

Figure 2. Comparison of the impacts of the different alternatives, considering PC paste (20% GBFS, w/c 0.40) as a base scenario (100%, dashed line). Impact categories are: acidification potential – generic (AP), global warming potential -100yr (GWP), eutrophication potential – generic (EP), freshwater aquatic ecotoxicity – 100yr (FAETP), human toxicity potential – 100yr (HTP), marine aquatic ecotoxicity – 100 yr (MAETP), photochemical oxidation (summer smog)

– low NO_X (POCP), stratospheric ozone depletion – steady state (ODP), terrestrial ecotoxicity – 100 yr (TAETP).

3.2. Participation of each unit process in geopolymer environmental impacts

Figure shows the participation of each unit process in the environmental impacts of the geopolymer systems assessed here. As indicated above, the environmental impact related to the production of the sodium hydroxide and commercial sodium silicate (waterglass) makes the highest contribution to the life cycle of geopolymers. The quantities of sodium silicate solution used for the production of WG-2 has higher contributions (between 44 and 64%), attributed to the high dose of alkali-activator in this binder formulation. Sodium hydroxide production also contributes important environmental impacts in all the mixtures, especially for the NaOH-activated geopolymer, where these values are higher than 51% for all the evaluated impacts, and up to 95% for ODP. Residue beneficiation processes do show some participation, especially in the case of RHA, with contributions between 7 and 12% of all assessed impacts for RHA-1, and between 13 and 27% for RHA-2. However, these figures are lower than those for commercial waterglass, for both dosages evaluated (WG-1 and WG-2).

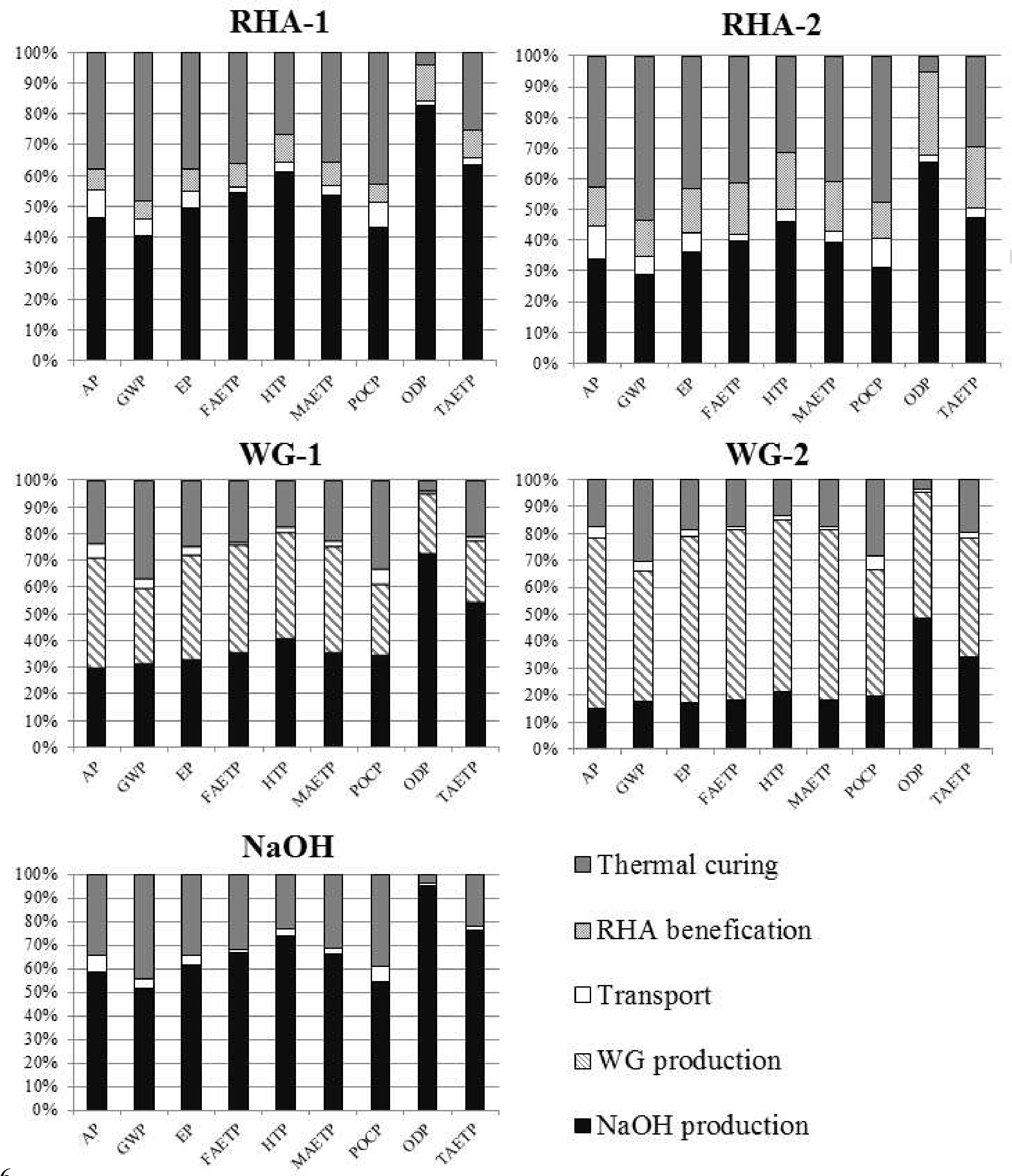

Figure 3. Contribution of each unit process to binder life cycle environmental impacts, for geopolymers activated by different activators as marked.

Transport has minor contributions (less than 10%) for the assessed impacts in the mixtures. It is worth mentioning that a transport distance of 100 km between raw materials production and geopolymer central mix plant was assumed, and in the case of higher distances from suppliers, the impacts related to transport would also increase, as reported in other studies (McLellan et al., 2011). That point is of special concern in the case of Brazil, where large distances between providers and consumers may apply, particularly when beneficiating residues from mining or agricultural processes which are not necessarily located close to areas of high consumer demand for building materials.

Finally, thermal curing presents a pronounced participation in most impacts, except for ozone layer depletion (ODP). This is important because that process is far from indispensable for many geopolymer formulations (Provis, 2014), and avoiding the application of heat during the curing may reduce these impacts significantly; up to 50% in the case of GWP for RHA-2. This aspect will be explored in detail in the subsequent section.

3.3. Effects of WG substitution and thermal curing

Considering that thermal curing plays an important role in defining the environmental profile of geopolymers, and that the use of an alternative silica source for the activator may reduce impacts, Figure 4 shows the relative impacts of PC and of geopolymers produced without thermal curing, taking WG-2 (produced with the higher amount of commercial sodium silicate among the mixes considered here) as the base scenario. Binders produced with conventional sodium silicate (WG-1, WG-2) present the higher environmental impacts among the five formulations in most cases here, with the exception of global warming potential where higher impacts are associated to binders based on PC. Compared to WG-2 without heat curing, PC now shows a GWP 55% higher.

The use of an alternative silicate solution based on dissolved rice rusk ash (RHA-1, RHA-2) can be considered as a potential activator option with lower environmental impacts, in comparison to the geopolymers produced with alkali activators based on NaOH and commercial waterglass. For the case of GWP, the mixes using RHA have the lowest environmental impacts, and mixture RHA-2 shows a GHG reduction of 61% against WG-2 as a ‘baseline’ geopolymer, and ~75% against PC. The blends based on RHA have lower impacts than PC for four (AP, GWP, EP, and POCP) of the nine categories evaluated.

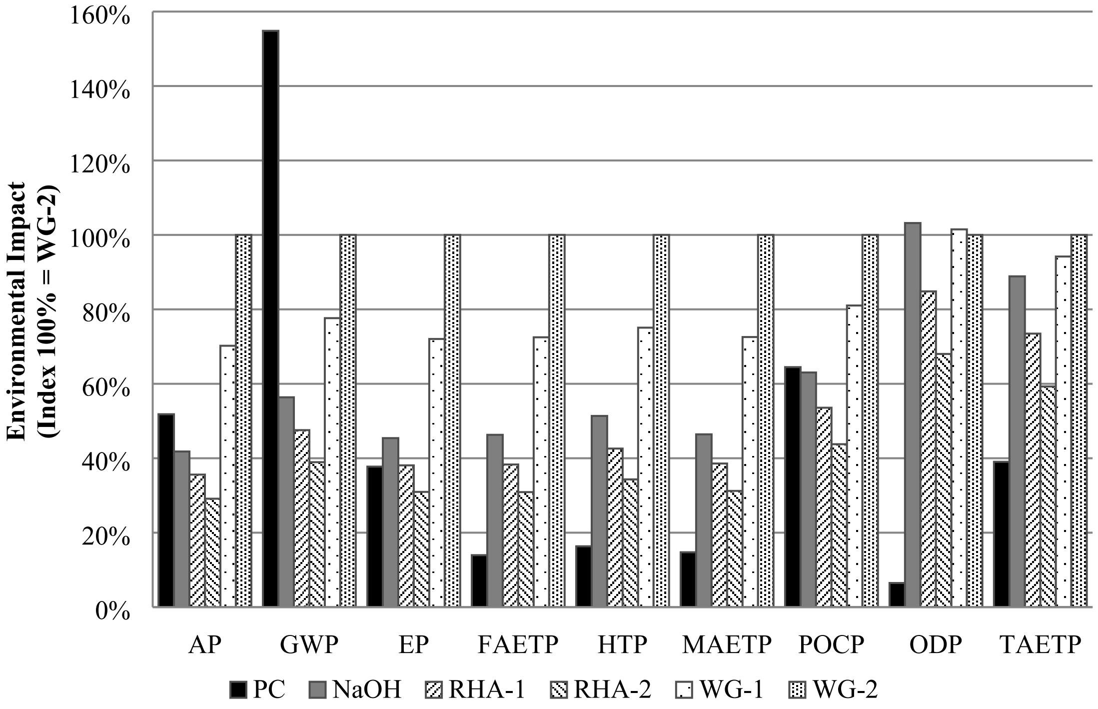

Figure 4. Comparison of the impacts of the different alternatives, produced without thermal curing, considering WG-2 as a base scenario.

Considering that geopolymers produced using sodium silicate activators derived from RHA or waste glass, and a range of aluminosilicate precursors, have been shown to have similar properties to comparable mix designs using commercial waterglass (Bernal et al., 2012; Puertas and Torres-Carrasco, 2014), Figure 5 shows the impact reduction of RHA-1 compared to WG-1, and RHA-2 compared to WG-2. Comparing WG-1 and RHA-1, the impacts decrease by between 16% and 49%, with the highest reduction values related to acidification potential, eutrophication potential, freshwater aquatic toxicity, marine aquatic ecotoxicity, and human toxicity potential (all reduced by 40-50%). When compared to WG-2, RHA-2 presents impact reductions between 32% (ODP) and 71% (AP), and most notably in the same categories listed above. Pronounced reductions are reached in the toxicity categories (FAETP, HTP, MAETP, TAETP), where geopolymer concretes are known to have higher impacts in comparison to PC concrete (Habert et al., 2011).

As was noted above, the underlying inventory data used for NaOH production (and similarly waterglass production) are likely to pre-date modern environmental protection regulations which restrict the use of e.g. mercury-based processes, and many of the other direct emissions to air or water which were historically common practice in the bulk chemicals industry. The greater reliance of geopolymer production on components produced as bulk chemical products, rather than direct production of Portland cement in a kiln, means that the assessment of these materials will be much more prone to such factors. Future improvements in bulk chemical process efficiency and environmental impact (including improved compliance with existing regulations and best-practice worldwide) will therefore be expected to disproportionately improve the calculated footprint of geopolymers and similar materials in an assessment such as this.

This also highlights the importance of considering optimized geopolymer mixes and processes in such assessments; as can be noted from Figure 4, when thermal curing is not needed, RHA-1 and RHA-2 present impacts which are up to 174% higher than PC for these categories, but still much lower than those reported in previous studies (Habert et al., 2011), that reported FAETP impacts more than 10 times higher for metakaolin-based geopolymers than for PC.

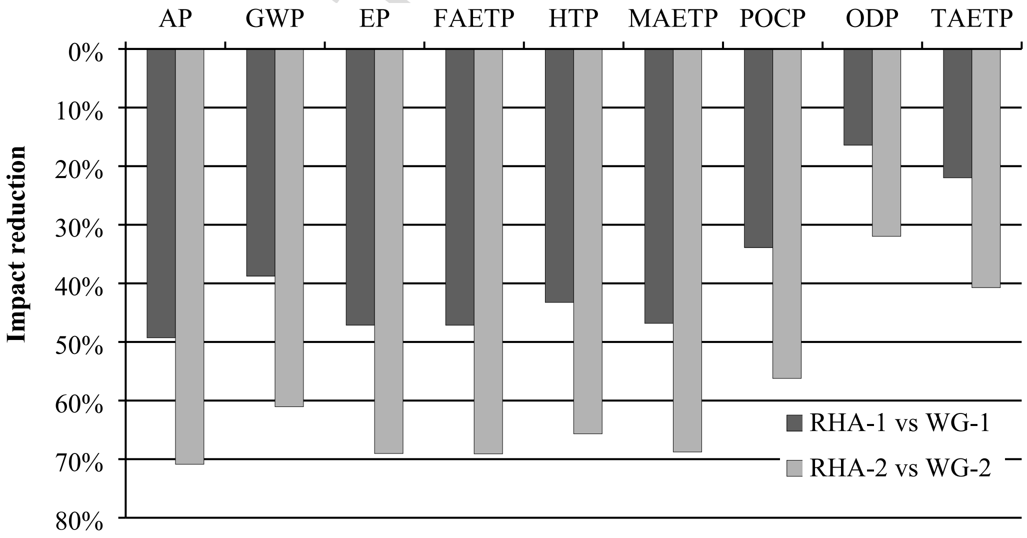

Figure 5. Impact reduction of the geopolymers produced without thermal curing with alternative sodium silicate (RHA) in comparison to those based on waterglass (WG).

3.4. Comparing impacts with compressive strength

The compressive strengths at 28 days of the evaluated materials is given on Table 4, where the geopolymer materials exhibited compressive strengths up to 42.5% higher than the PC control depending on the mix design, although the NaOH activator gave low strength. The presence of soluble silicates in the alkali activator particularly increased the mechanical performance of these binders at earlier ages of curing, as reported in Longhi et al. (2016), which is desirable for precasting of concrete units.

At lower contents of soluble silicate (RHA-1, WG-1: SiO_2/Al_2O_3 molar ratio of 3.5, Table 2), the alternative alkali activator based on dissolved RHA (RHA-1) gave a comparable compressive strength (<5% difference) to the corresponding system produced with commercial sodium silicate (WG-1). However, as the content of silicate is raised (RHA-2, WG-2) to achieve a higher SiO_2/Al_2O_3 molar ratio, the ability to dissolve the RHA into the NaOH solution becomes a limiting factor, the reactivity of the RHA-based solution is lower, more unburnt carbon is present, and a consequent reduction in the mechanical performance is identified. This behavior contrasts with several previous reports, where significant differences between a commercial sodium silicate and a chemically dissolved source of SiO_2 were not observed (Bernal et al., 2012; Bouzón et al., 2014; Rodríguez et al., 2013; Zivica, 2006, 2004). Therefore, the lower reactivity degree identified to the RHA-based activator has a stronger effect at higher values of SiO_2/Al_2O_3 molar ratio (RHA-2). This does become an important caveat in the design of waste-based activators for geopolymers, which was identified above as being potentially important in overall impact reduction: when specifying a waste-derived material, quality control and the ability to ensure good performance becomes imperative.

Table 4. Compressive strength values of the evaluated binders at 28 days. Standard deviations among 5 replicate tests are in some cases rather high due to the use of small (20 mm) paste cube specimens for testing.

Binder	PC	NaOH	RHA-1	RHA-2	WG-1	WG-2
Compressive strength (MPa)	51.8	19.1	68.4	46.9	72.3	71.1
Standard deviation (MPa)	12.4	0.5	5.3	3.8	4.7	6.5

In order to compare the impact values of all the mixes, relating the environmental profile with material function, Figure 6 presents the relation between impacts and compressive strength, taking PC (75% of clinker, 20% GBFS, 5% gypsum) as the base case (100% value).

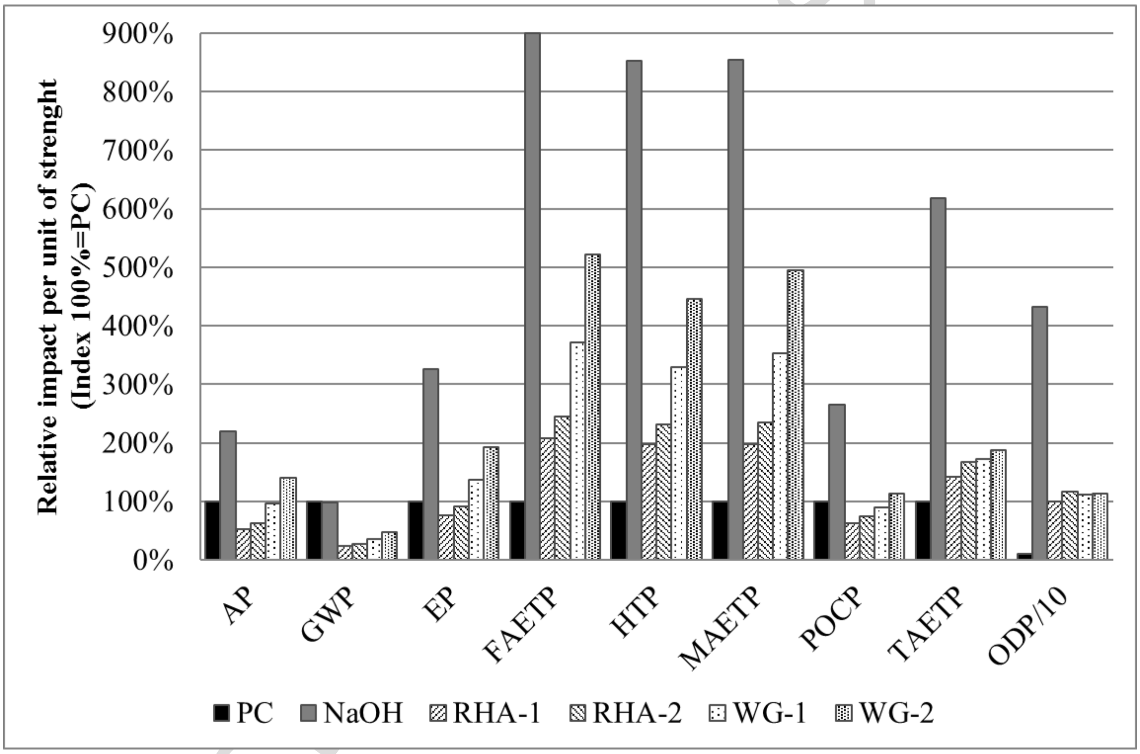

Figure 6. Eco-profile of the evaluated geopolymers in comparison to Portland cement, considering environmental impacts per unit of compressive strength (MPa). Note ODP values are divided per 10.

In comparison to PC, the mixes based on RHA have lower impacts in 4 categories (AP, GWP, EP, POCP) and up to 150% higher impact values in 4 categories, confirming that the use of an alternative sodium silicate is very promising to reduce binder environmental impacts. In contrast, since NaOH-based geopolymer reached lower compressive strength values, this system figures as the worst option in all categories. The results shown in Figure 6 confirm the need to consider materials function when performing LCA of innovative construction materials (Habert and Ouellet-plamondon, 2016; Van den Heede and De Belie, 2012).

4. Discussion and perspectives

Among the geopolymers studied here, RHA-2 presents the lower environmental impacts per kilogram, in most categories, and has a compressive strength similar to that of PC. RHA-1, WG-1 and WG-2 are suitable for applications that need higher compressive strength values. In this case, RHA-1 impacts show similar trends to RHA-2, being slightly higher due to the higher proportions of sodium hydroxide consumed. However, when compressive strength is considered, RHA-1 performs slightly better than RHA-2 (Figure 6).

Sodium silicate based on RHA emerges as a promising alternative for the production of geopolymers with decreased environmental impacts in most of the categories evaluated. Further reductions of these impacts are related to the diminution of sodium hydroxide consumption, or improvements in its production. The results are in accordance with those reported in the literature (Habert and Ouellet-plamondon, 2016; Habert et al., 2011), that state that reductions on life cycle impacts of geopolymers may be achieved by the substitution of commercial waterglass by an alternative sodium silicate.

Although the assessment conduced here is related exclusively to the use of thermally treated kaolinitic residue as the key geopolymer precursor, the benefits of partial or total substitution of the commercial sodium silicate solution in the activator by dissolved silica sourced from an industrial waste can be also applied for other types of alkali-activated systems. The LCA of both alkali-activated systems (using commercial and non-conventional sodium silicates) highlights the relevance of investigating more options for the potential valorization of industrial wastes, as well as the development of environmentally friendly materials with more competitive cost (Provis, 2017). Assuming that the raw materials are locally available, the synthesis of this alternative activator will also have a positive effect on the total cost of production, as sodium silicate solution represents the raw material with the highest economic value within an alkali-activated binder.

Fly ash and blast furnace slag are the most common by-products assessed in the context of alkali activation technology, but are also in high demand for use in Portland cement blends in many parts of the world. Thus, it is likely that the overall environmental benefit related to the use of aluminosilicate precursors with alkali-activators in place of Portland cement can best be reduced through the use of other types of materials that do not require preconditioning treatments (such as high temperature processes), or other residues without any commercial value. Therefore, along with the use of raw materials from available and high volume waste streams to provide environmental benefits, the cost of production can be also reduced (Jamieson et al., 2015).

However, it is important to note that the reactivity of the precursor plays an important role in the dosage required for the alkali-activation, and more activator solution is needed to activate less-reactive aluminosilicates, which also will represent higher cost and higher environmental impacts. The re-use and valorization of a kaolinitic-rich waste from the mining industry as a raw material for the production of geopolymer might satisfy a potential demand for these materials in the construction industry, without competition from any other applications for the necessary raw material. Geopolymers based on clays are probably more suitable for precast and masonry unit production as their flow characteristics can make them less suitable for general concreting applications, although desirable high early strength can be obtained (Heath et al., 2014).

Since the alkali activators are the most important contributors to the environmental impacts of geopolymers, their dosages, compositions, as well as their type and origin must be considered and carefully assessed in order to further reduce potential environmental impacts. The quantity of alkali activator required can be adjusted based on the reactivity of the precursors (which is in turn influenced by amorphous content, particle size distribution, and SiO_2 and Al_2O_3 content) and the synthesis conditions (alkali and soluble silicate content) to achieve a specific performance level. The open literature reports a wide number of aluminosilicate sources, as well as different dosage criteria, used in formulation of alkali-activated binders. Therefore, detailed assessments are needed when different raw materials, sources or suppliers are being considered (Bernal et al., 2016).

Most of the LCA studies of geopolymers reported in the open literature have been focused on the question of GWP. However, the true environmental profile of these materials extends far beyond GWP, and will define the real effects of this technology in the development of a sustainable construction industry. The choice of a problem oriented impact assessment method (CML-01) here allowed the comparison of the results with other geopolymer LCA studies, with less related uncertainties than damage oriented methods (Van den Heede and De Belie, 2012). However, problem oriented methods may be more difficult to understand than damage oriented methods, as direct comparisons between categories cannot be made. Future studies should consider also damage oriented approaches, to define strategies for impact reductions based on a comparison of normalized damages related to each impact category.

When comparing different concrete compositions, the LCA functional unit should incorporate differences in strength, durability and service life (Van den Heede and De Belie, 2012). This is a key issue when evaluating innovative construction materials. In the present study, the materials assessed do not all present similar technical properties. However, the comparison of environmental impacts and compressive strength (impacts per MPa) enhances the understanding of the materials environmental performance, considering that the environmental benefit of a potential 'green' concrete can be related to its reduced cement content and strength governed structure dimensions in comparison with the reference (Van den Heede and De Belie, 2012).

5. Conclusions

Geopolymers are widely considered a low carbon alternative to Portland cement. However, environmental impacts other than global warming potential are important in the life cycle of this type of binder. In this study, LCA was applied to evaluate the impacts related to geopolymer production, considering different activators, for the specific case study of materials developed from a Brazilian kaolin mining waste. The geopolymers samples were compared to binders based on PC (Portland slag cement with 75% clinker). Some conclusions of this study can be pointed out:

- Binders produced with conventional sodium silicate and thermal curing presented the higher environmental impacts among the five formulations studied here, apart from global warming potential where higher impacts are associated with binders based on PC;
- Thermal curing presented a pronounced participation in most impacts, except for ozone layer depletion (ODP). This is important because that process is far from indispensable for many geopolymer formulations, and avoiding the application of heat during the curing may reduce these impacts significantly; in some cases up to 50%;
- At lower contents of soluble silicate, the alternative alkali activator based on dissolved RHA gave a comparable compressive strength (<5% difference) to the corresponding system produced with commercial sodium silicate;
- In comparison to PC, the mixes based on alkali activator derived from a chemically modified rice husk ash (RHA) have lower impacts in LCA per unit of compressive strength (MPa) in 4 of the categories analyzed (AP, GWP, EP, POCP), and up to 150% higher impact values in 4 other categories.

The study highlights the importance of using alternative activators, for example, based on residues such as rice husk ash (RHA), to produce these binders, since significant impact reductions may be achieved. Future studies should consider durability and service life of the cementitious materials based on RHA activation, and assess potential environmental impacts through damage oriented methods, to improve the understanding of decision makers regarding the overall environmental impacts of this innovative construction material.

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Highlights

- Life cycle impacts of a geopolymer based on kaolin sludge residue are evaluated.
- Chemically modified rice husk ash applied as alternative to sodium silicate.
- RHA-based activators are an alternative for impact reduction in GP production.