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1 (8036 words)

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

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20

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 CO₂ 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.

48 1. Introduction

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

54 Geopolymers are a type of cement derived from the alkali activation of industrial wastes or by-
55 products (usually fly ash or amorphous aluminosilicate minerals). These materials are now
56 being used in large-scale concrete production, and can show technical and environmental
57 advantages compared to PC (Duxson et al., 2007; Van Deventer et al., 2012). A key advantage
58 often quoted in the technical literature is the saving of as much as 40-80% of the GHGs
59 emissions compared to PC, as well as the re-use of wastes and/or industrial by-products as
60 major constituents of the geopolymer binder (Bernal et al., 2016; Davidovits, 2008; Habert et
61 al., 2011; McLellan et al., 2011). The alkali solutions most commonly used as activators in
62 geopolymer synthesis are sodium and potassium silicates. The presence of silica in the
63 activating solution tends to lead to the formation of a denser and more compact binder structure
64 with high mechanical strength (Fernández Jiménez and Palomo, 2005; Yang et al., 2008).
65 However, these silicate solutions represent the most expensive component in geopolymer
66 production, and are the highest contributor to the environmental footprint of these materials
67 (Habert and Ouellet-plamondon, 2016; Habert et al., 2011), as alkali silicates are produced by
68 thermal or hydrothermal routes from silica and alkali carbonates or hydroxides (Fawer et al.,
69 1999).

70 The environmental impacts of a geopolymer binder can be calculated as the contributions of its
71 two main components - precursor and alkali activator - as well as the energy involved in its
72 manufacture (thermal and mechanical processes, and including any non-ambient curing steps)
73 and raw materials transportation. Life cycle assessment (LCA) reports have identified that the
74 embodied energy and environmental impacts (global warming potential and others) are mainly

75 affected by the type and concentration of alkali-activator solution (Habert et al., 2011; Heath et
76 al., 2014; McLellan et al., 2011; Tempest et al., 2009; Witherspoon et al., 2009). Evaluating the
77 eco-profile of different geopolymers compared to PC concrete, Habert et al. (2011) found that
78 although offering decreased CO₂ emissions, geopolymer production was calculated to increase
79 some other environmental impacts by up to 1000%, such as fresh water ecotoxicity (FAETP),
80 especially due to waterglass production.

81 The interest in valorising additional industrial wastes and by-products, as well as the need to
82 reduce the production cost of geopolymer binders, has promoted the development of alternative
83 alkali-activators based on the combination of alkalis with waste-derived amorphous silica
84 sources; these can exhibit similar or even higher effectiveness compared to traditional alkali-
85 activator based on silicate soluble (Bernal et al., 2011; Detphan and Chindaprasirt, 2009;
86 Puertas and Torres-Carrasco, 2014; Rodríguez et al., 2013; Zivica, 2004, 2006). Agro-industrial
87 wastes such as rice husk ash (RHA) have a high potential for implementation as a silicate source
88 for the alkali activator (Bernal et al., 2012; Bouzón et al., 2014; Detphan and Chindaprasirt,
89 2009; Kim et al., 2014; Prasetyoko et al., 2006). RHA is generated by biomass power plants
90 burning rice husks, and is mainly made up of silica (ideally > 90%), potassium (< 5% as K₂O)
91 and phosphorous (<3% as P₂O₅), with variable (but ideally low) unburnt carbon content. RHA
92 can be dissolved into an alkali hydroxide solution for use as a silica source in geopolymer
93 synthesis, yielding similar mechanical properties compared to the use of commercial waterglass
94 (Bernal et al., 2012). Although studies have shown a potential reduction of GHG emissions of
95 63% when an alternative sodium silicate based on RHA is used (Mellado et al., 2014), the effect
96 on other environmental impact categories has not yet been assessed in detail and can be
97 considered the novelty of the work presented here. Therefore, the assessment of the real and
98 quantified environmental impact related to the alkali-activators in geopolymers and how can be
99 reduced through the use of alternative silicate solutions derived from industrial wastes or by-
100 products reveals new pathways for development of materials with higher sustainable criteria.

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

113 2. Materials and Methods

114 2.1. Materials

115 Calcined kaolin sludge (CKS) was used as the aluminosilicate precursor for geopolymer
116 production in this study. The kaolin sludge was generated in the mining industry of the state of
117 Pará (Brazil) in the process of beneficiation to obtain high purity kaolin. Various wastes are
118 generated during beneficiation; here, the residue generated between centrifugation and filtering
119 steps (which is rejected as impure kaolin, and usually sent to tailings dams) was considered. The
120 high content of kaolinite and moderate amounts of titanium dioxide, ferrite and quartz present in
121 this residue allow the generation of a pozzolanic powder through calcination (Barata and
122 Angélica, 2012). In this study, the kaolin sludge was dried in a filter press, calcined at 750 °C
123 for 1 h, and ball milled for 1 h (Longhi et al., 2016; Souza and Dal Molin, 2005). The chemical
124 composition, mean particle size and specific surface of the CKS is shown in Table 1. Alkali
125 activators derived from analytical grade NaOH (~99%) and two different soluble silicate
126 solutions were used. Sodium silicate solution with 26.5 wt.% SiO₂, 10.6 wt.% Na₂O, and 62.9

127 wt.% H₂O (Sigma-Aldrich) was used as a reference soluble silica source. The alternative alkali
 128 activator was prepared by the dissolution of a rice husk ash (RHA) obtained from the rice
 129 industry of southern Brazil, into NaOH solution. The RHA was mainly constituted by SiO₂, as is
 130 shown in the Table 1. The RHA-based solution was prepared with analytical NaOH to obtain a
 131 SiO₂/Na₂O molar ratio of 2.2, with dissolution carried out at 100 °C for 1 h in a refluxed system
 132 to achieve the highest dissolution degree and avoid loss of water by evaporation (Bouzon et al.,
 133 2014). The content of the dissolved silica was calculated and adjusted through the weight of non-
 134 dissolved RHA remaining after the dissolution and subsequent filtration processes. From this
 135 information, the composition of the filtered RHA-based solution was calculated to be 19.2 wt.%
 136 SiO₂, 8.8 wt.% Na₂O, and 70.6 wt.% H₂O.

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

		CKS (Calcined kaolin sludge)	RHA (Rice husk ash)
Chemical composition (wt%)	SiO ₂	61.3	97.3
	Al ₂ O ₃	33.8	--
	Fe ₂ O ₃	2.8	--
	TiO ₂	2.1	--
	K ₂ O	--	2.1
	CaO	--	0.6
Mean Particle size (µm)		11.0	9.87
Specific surface (m ² ·g ⁻¹)		19,7	9.82

139

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

143 2.2. Sample preparation

144 All geopolymers were formulated with an overall Na₂O/Al₂O₃ molar ratio of 1.0, and the
 145 content of soluble silicates (either reference or alternative) was adjusted to obtain an overall
 146 SiO₂/Al₂O₃ molar ratio between 3.1 and 4.0, by manipulating the silicate activator content and
 147 adding solid NaOH to adjust the overall Na₂O content. Additional water was added to the
 148 activation solution in order to achieved a water/solids ratio of 0.40. The pastes were mixed
 149 mechanically for 6 min and then cast into 20 mm cubic moulds, vibrated for 1 min, and cured at

150 50 °C for 24 h with a high relative humidity (RH >90%). After thermal curing, the samples were
 151 stored in a sealed plastic container at room temperature and 90% RH. The mix designs and
 152 synthesis conditions are shown in Table 2, where the pairs of systems WG-1/RHA-1 and WG-
 153 2/RHA-2 each have equivalent SiO₂/Al₂O₃ molar ratios (i.e. contents of soluble silicates). The
 154 quantities reported are adjusted to obtain 1 kg of fresh paste, which comprises the precursor
 155 (CKS), anhydrous activator (NaOH and sodium silicate), and water. The PC paste with a
 156 water/cement ratio of 0.40 was produced by mechanical mixing for 1 min, and cured at 25 °C
 157 immersed in saturated limewater.

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

Mix ID	Overall molar ratio		Quantity of raw materials (g)				
			Precursor		Source of soluble silicates*		
	SiO ₂ / Al ₂ O ₃	Na ₂ O/ Al ₂ O 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

162 *Source of soluble silicates: Corresponds to the sodium silicate solution either waterglass or RHA-based solution. The values
 163 reported correspond to the total amount of Na₂SiO₃·nH₂O.

164

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

171 2.3. Life cycle assessment

172 2.3.1. Goal of the study

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

175 2.3.2. Study scope: system boundaries and functional unit

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

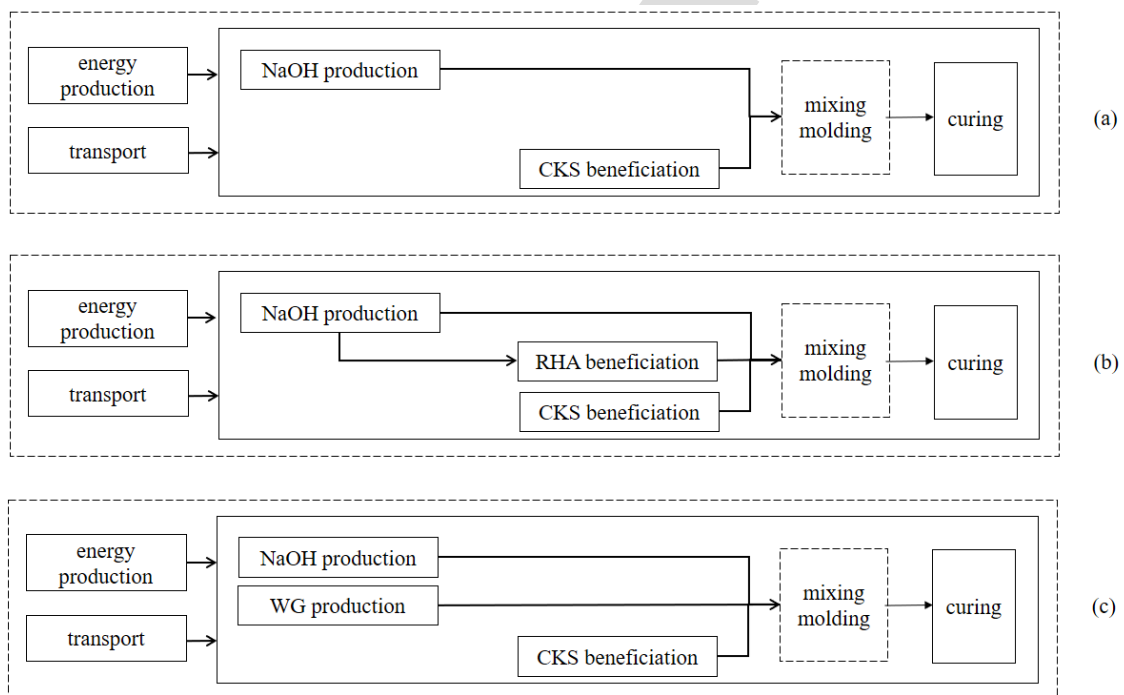
182 The system studied considers the commercial production of five geopolymer formulations, from
183 cradle to gate, including raw materials production, transport, and waste beneficiation, and all the
184 associated emissions and energy consumptions (Figure 1). Mixing and molding are not
185 considered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

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

197 The functional unit is 1 kg of paste (CKS + anhydrous alkali activator + water). This procedure
198 was selected to evaluate binder production, as this product may have multiple specific

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

209



210

211 **Figure 1.** Schematic description of system boundaries for the geopolymers (a) NaOH-based

212 geopolymer (without sodium silicate); (b) RHA-1 and RHA-2; and (c) WG-1 and WG-2.

213

214 Although the synthesis conditions of the geopolymers were defined at laboratory scale, their

215 environmental profile calculation considered production at industrial scale (except for thermal

216 curing), considering mean energy consumption values informed by local providers. In this
217 sense, the beneficiation process for the kaolin sludge includes the power consumption of filter
218 presses (drying), rotary kilns (calcination) and industrial grinders (milling). The values for the
219 equipment consumption were collected considering the equipment currently commercially
220 available in Brazil. The total amount of energy spent in the beneficiation of 1 kg of kaolin
221 sludge is 0.0034 kWh for drying, 0.0046 kWh for thermal processing, and 0.0190 kWh for
222 milling. The assessments of energy involved during the beneficiation processes were carried out
223 assuming a sludge with initially 80 wt.% water which was dewatered, and a further mass loss of
224 17% due to dehydroxylation via thermal processing of the clay minerals. Other potential outputs
225 such as particulate matter released in milling, and the emissions from burning were not
226 considered. These values are expected to be about 20 times lower than those reported by other
227 studies (Habert and Ouellet-Plamondon, 2016; Heath et al., 2014), since these authors
228 considered the whole metakaolin production chain, while the present study is based on the
229 beneficiation of a residue (kaolin sludge).

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

233 The composition of sodium silicate is generally expressed as $\text{Na}_2\text{O} \cdot n\text{SiO}_2$, where n corresponds
234 to the molar ratio. Commercial waterglass solutions can be available with $\text{SiO}_2/\text{Na}_2\text{O}$ molar
235 ratios between 1.6 and 3.3, produced by thermal (furnace) or hydrothermal processes. At higher
236 $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratios, the content of soluble silicates is increased and the subsequent quantity
237 of solution required for a specific alkaline activation can be also modified. According to Zah
238 and Hischer (2007), in the furnace process, waterglass is produced directly by melting pure
239 silicon sand and soda at temperatures around 1100 - 1200 °C, where a material with high
240 $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio is generated. This sodium silicate can be used in a solid form, but is
241 usually dissolved and made available as an aqueous solution (~37 wt.%). In the hydrothermal
242 process, sand is dissolved in sodium hydroxide solution within an autoclave at temperatures

243 between 180 – 300 °C (Trabzuni et al., 2011). Zah and Hirschier (2007) report that, after
244 filtration, sodium silicate with a lower $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio is obtained as a 48 wt.% solution.
245 This solution may be later spray-dried in order to obtain a powder.

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

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

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

270 Finally, for comparison purposes, Ecoinvent data for Portland cement (cement production, blast
 271 furnace slag 5-25%, US only), was adapted to the Brazilian production profile, considering one
 272 of the most currently used cements in Brazil, with 75 wt.% clinker, 20 wt.% ground-granulated
 273 blast-furnace slag (GBFS) and 5 wt.% gypsum. The LCA for the PC systems was carried out on
 274 the basis of 1 kg of cement paste (clinker + gypsum + GBFS + water, with a w/c ratio of 0.40).
 275 Thermal curing of geopolymer materials was assessed considering a heat curing chamber, lab
 276 scale, with a consumption of 0.54 kWh per kg of paste. It is noted that this energy consumption
 277 is based on a small laboratory oven and so is a very conservative value when compared to larger
 278 industrial processes which can operate at higher energy efficiency, either through the use of
 279 recovered waste heat or simply through reduced losses per unit mass of product in a much larger
 280 operation. Considering that thermal curing is not widely applied to PC-based materials in Brazil,
 281 it was not considered within their LCA.

282 2.3.3. Inventory analysis

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

286 **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

287

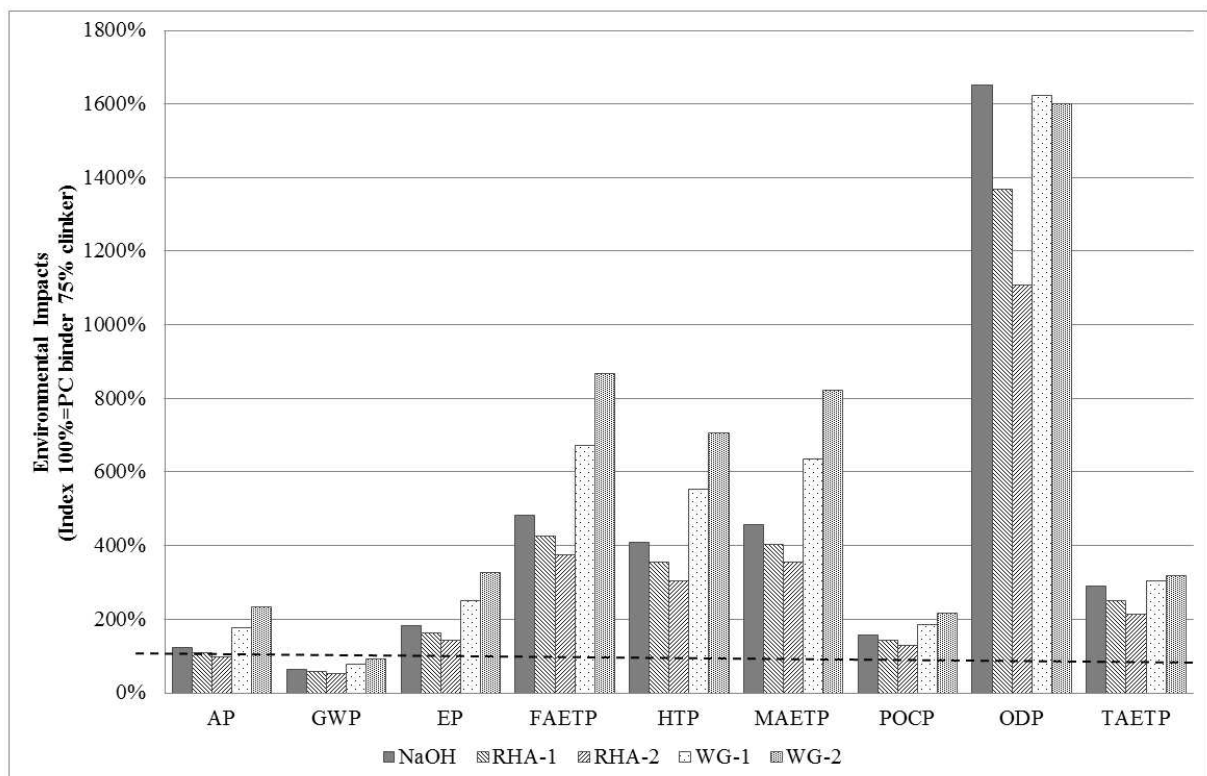
288 The cut-off procedure was applied to the residues, and the impacts associated with their
 289 generation were not allocated in this study.

290 2.3.4. Life cycle impact assessment

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

298 3. Results and Discussion

299 3.1. Environmental analysis of the binders produced



300

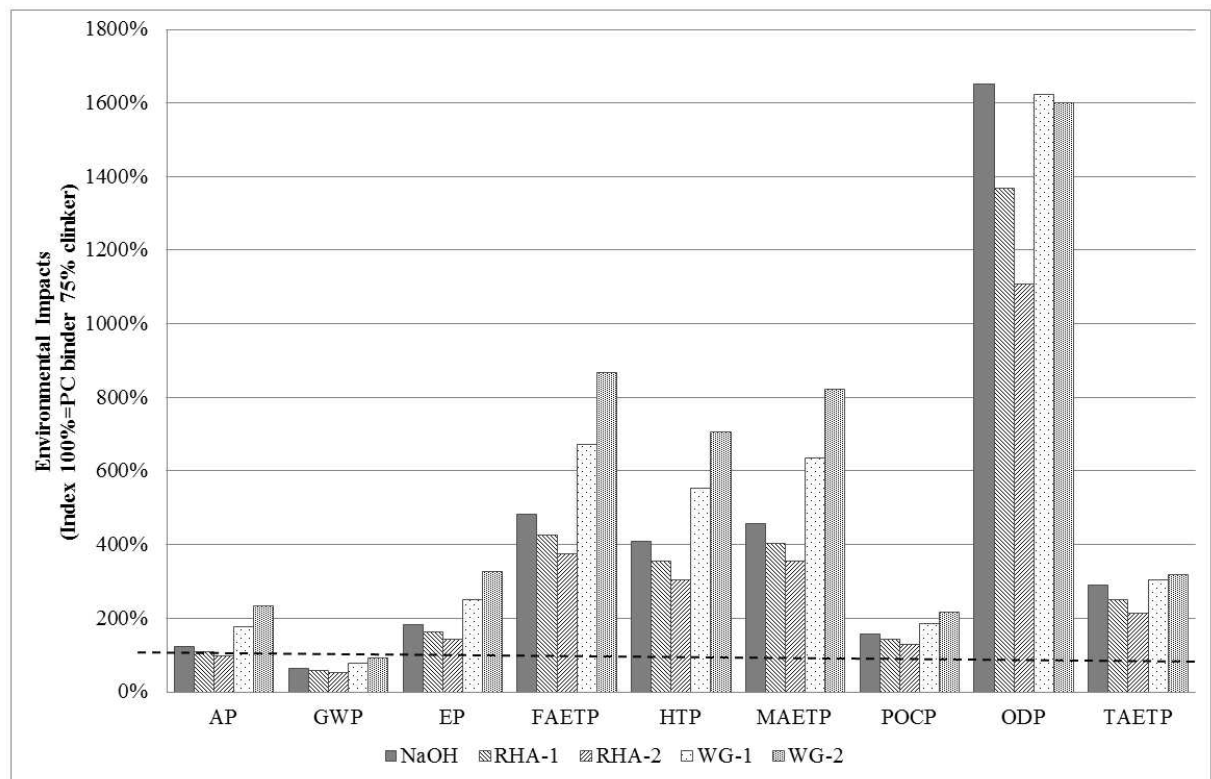
301 Figure shows the environmental impacts related to the production of 1 kg of geopolymer
 302 binder, in comparison with Portland slag cement with 75% clinker, based on pastes with the
 303 same cement content. It is important to note that for GP pastes thermal curing was applied,
 304 while for PC paste no thermal treatment was considered. The results show different trends

305 according to the impact category. In comparison to Portland cement, the geopolymers present
306 lower global warming potential (GWP) impacts: reductions between 7 and 22% for the binders
307 based on waterglass (WG-1 and WG-2), between 41 and 47% for those based on RHA, and
308 35% for the binder derived from an alkali activator based on NaOH. These achievements are
309 lower than those reported by Mellado et al., (2014), who calculated reductions of 50% for a
310 WG-based geopolymer and 63% for RHA geopolymer. However, these authors considered PC
311 mortar with an emission factor of 1 kg CO₂ per kg cement, which is higher than the one applied
312 to this study, 0.73 kg of CO₂ per kg cement. The reductions compared to Portland cement are
313 also less than the headline claims of many studies in the literature as summarized in the
314 Introduction section of this paper, and this will be examined in detail below.

315 For the categories acidification potential (AP), eutrophication potential (EP) and photochemical
316 oxidation (POCP), the geopolymer blends based on rice husk ash (RHA-1, RHA-2) have
317 impacts up to 62% higher than PC. For the remaining categories (freshwater aquatic ecotoxicity
318 - FAETP, human toxicity potential - HTP, marine aquatic ecotoxicity - MAETP, stratospheric
319 ozone depletion - ODP, terrestrial ecotoxicity - TAETP), each of the geopolymer binders
320 assessed show potential impacts higher than PC, with values up to 17 times higher for ozone
321 layer depletion (ODP). These trends are in accordance with those reported by Habert et al.
322 (2011), for fly ash based concrete geopolymer in comparison with PC concrete (70% clinker,
323 30% fly ash), except for FAETP and ODP. Although these trends are similar, the comparison is
324 only illustrative, because Habert et al. (2011) compare FA geopolymer concretes and PC
325 concrete with same mechanical strength, while this study compares PC and geopolymer binders
326 with different mechanical properties.

327 It is interesting to note that in the case of ODP, the values for the GP binders are between 11
328 and 17 times higher than PC. However, the emissions associated with 1000 kg of PC paste
329 (1.34×10^{-5} kg CFC-11-eq) are similar to those emitted by a single incandescent lamp generating
330 20 million lumen-hours of light (i.e. a typical 800-lumen household lamp operating for 2.85
331 years; 1.1×10^{-5} kg CFC-11-eq) (Scholand and Dillon, 2012), so it is evident that the overall

332 impact in this category is rather low in the context of other societal activities. The ecotoxicity
 333 (FAETP, MAETP, TAETP) and human toxicity (HTP) characteristics of the geopolymer
 334 formulations also appear unfavorable due particularly to the use of NaOH. However, with
 335 continuing improvement in the emissions profile of the chlor-alkali industry and the reduction in
 336 reliance on mercury cells for alkali production worldwide, this should also be considered a
 337 conservative calculation when assessing future materials production. Binders produced using an
 338 alternative silica source based on chemically modified rice husk ash (RHA-1, RHA-2) have
 339 lower impacts than those based on commercial watrglass for all the categories evaluated. This
 340 agrees with the reports in other studies (Habert et al., 2011; McGuire et al., 2011; Mellado et al.,
 341 2014; Provis, 2014; Weil et al., 2009), that relate the majority of geopolymer impacts to
 342 activator production, and highlights the importance of the development of alternative sourcing
 343 pathways for activators as discussed here.



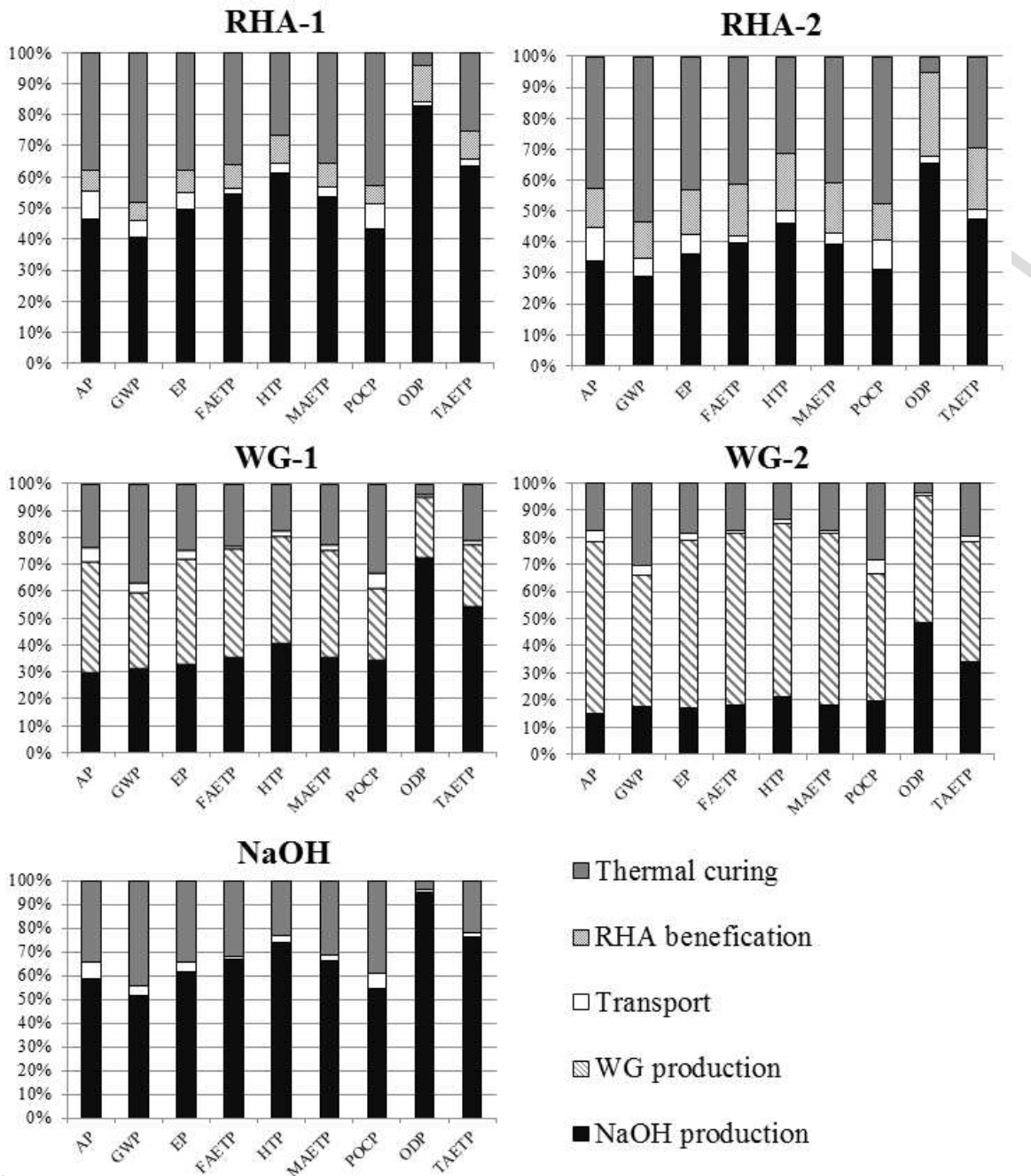
344

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

350 – low NO_x (POCP), stratospheric ozone depletion – steady state (ODP), terrestrial ecotoxicity –
351 100 yr (TAETP).

352 3.2. Participation of each unit process in geopolymer environmental impacts

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



36

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

368

369 Transport has minor contributions (less than 10%) for the assessed impacts in the mixtures. It is
 370 worth mentioning that a transport distance of 100 km between raw materials production and
 371 geopolymer central mix plant was assumed, and in the case of higher distances from suppliers,
 372 the impacts related to transport would also increase, as reported in other studies (McLellan et

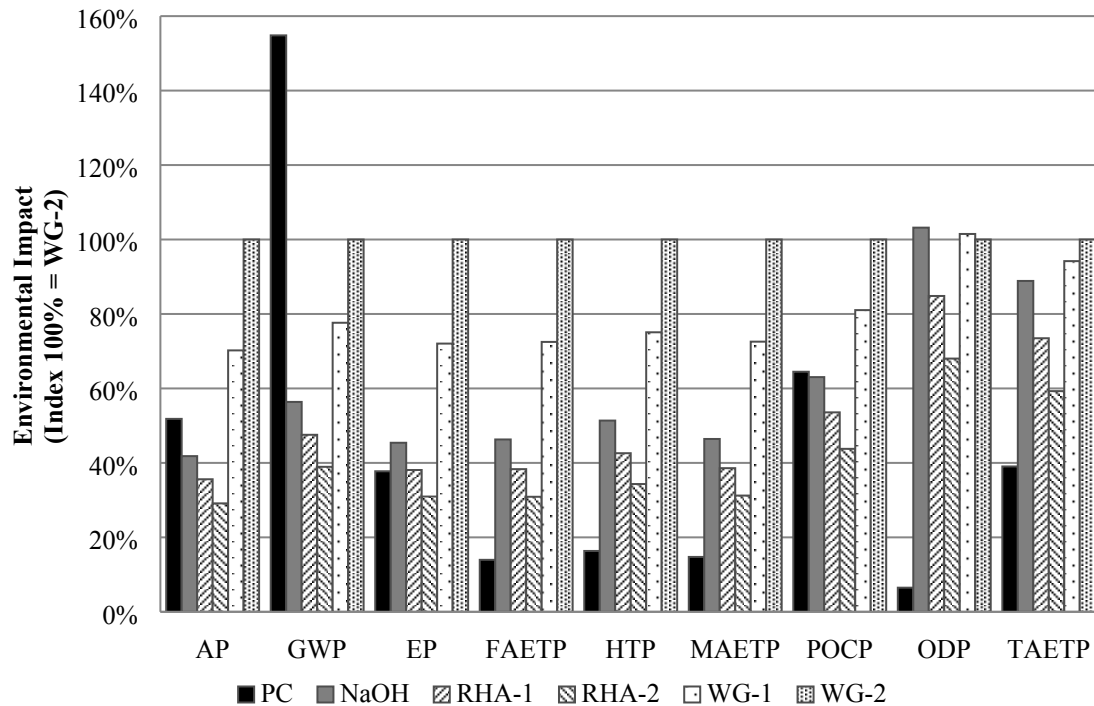
373 al., 2011). That point is of special concern in the case of Brazil, where large distances between
374 providers and consumers may apply, particularly when beneficiating residues from mining or
375 agricultural processes which are not necessarily located close to areas of high consumer demand
376 for building materials.

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

382 3.3. Effects of WG substitution and thermal curing

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

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



399

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

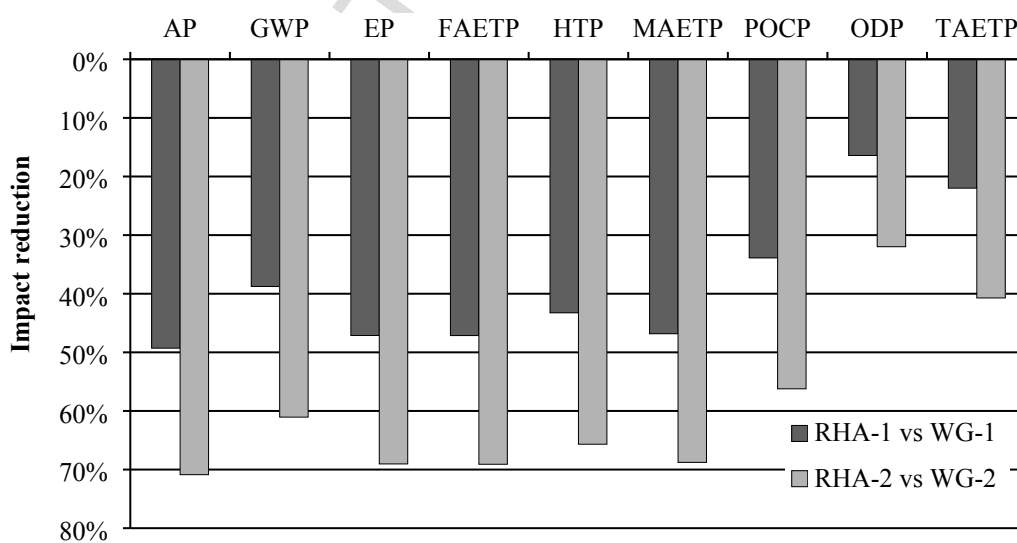
402

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

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

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

431



432

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

435

436 3.4. Comparing impacts with compressive strength

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

443 At lower contents of soluble silicate (RHA-1, WG-1: $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 3.5, Table 2),
444 the alternative alkali activator based on dissolved RHA (RHA-1) gave a comparable
445 compressive strength (<5% difference) to the corresponding system produced with commercial
446 sodium silicate (WG-1). However, as the content of silicate is raised (RHA-2, WG-2) to achieve
447 a higher $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio, the ability to dissolve the RHA into the NaOH solution
448 becomes a limiting factor, the reactivity of the RHA-based solution is lower, more unburnt
449 carbon is present, and a consequent reduction in the mechanical performance is identified. This
450 behavior contrasts with several previous reports, where significant differences between a
451 commercial sodium silicate and a chemically dissolved source of SiO_2 were not observed
452 (Bernal et al., 2012; Bouzón et al., 2014; Rodríguez et al., 2013; Zivica, 2006, 2004). Therefore,
453 the lower reactivity degree identified to the RHA-based activator has a stronger effect at higher
454 values of $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio (RHA-2). This does become an important caveat in the design
455 of waste-based activators for geopolymers, which was identified above as being potentially
456 important in overall impact reduction: when specifying a waste-derived material, quality control
457 and the ability to ensure good performance becomes imperative.

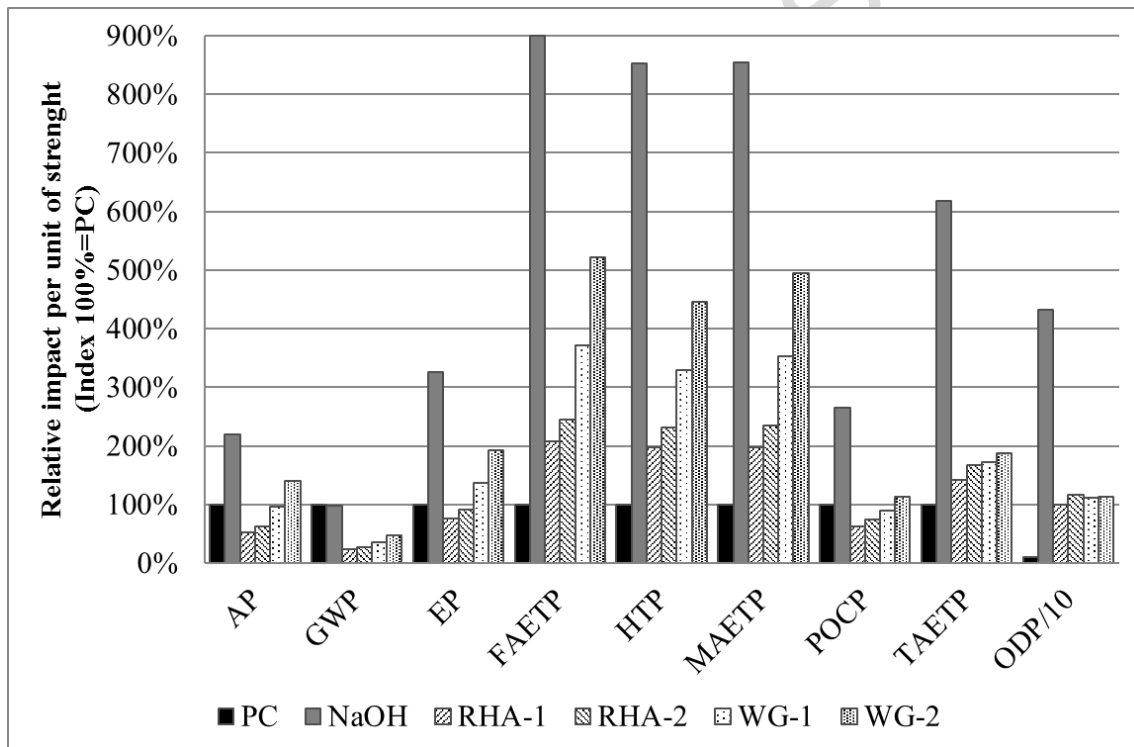
458 **Table 4.** Compressive strength values of the evaluated binders at 28 days. Standard deviations
459 among 5 replicate tests are in some cases rather high due to the use of small (20 mm) paste cube
460 specimens for testing.

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

461

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

465



466

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

470

471 In comparison to PC, the mixes based on RHA have lower impacts in 4 categories (AP, GWP,
 472 EP, POCP) and up to 150% higher impact values in 4 categories, confirming that the use of an
 473 alternative sodium silicate is very promising to reduce binder environmental impacts. In
 474 contrast, since NaOH-based geopolymer reached lower compressive strength values, this system

475 figures as the worst option in all categories. The results shown in Figure 6 confirm the need to
476 consider materials function when performing LCA of innovative construction materials (Habert
477 and Ouellet-plamondon, 2016; Van den Heede and De Belie, 2012).

478 4. Discussion and perspectives

479 Among the geopolymers studied here, RHA-2 presents the lower environmental impacts per
480 kilogram, in most categories, and has a compressive strength similar to that of PC. RHA-1, WG-
481 1 and WG-2 are suitable for applications that need higher compressive strength values. In this
482 case, RHA-1 impacts show similar trends to RHA-2, being slightly higher due to the higher
483 proportions of sodium hydroxide consumed. However, when compressive strength is
484 considered, RHA-1 performs slightly better than RHA-2 (Figure 6).

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

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

501 silicate solution represents the raw material with the highest economic value within an alkali-
502 activated binder.

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

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

521 Since the alkali activators are the most important contributors to the environmental impacts of
522 geopolymers, their dosages, compositions, as well as their type and origin must be considered
523 and carefully assessed in order to further reduce potential environmental impacts. The quantity
524 of alkali activator required can be adjusted based on the reactivity of the precursors (which is in
525 turn influenced by amorphous content, particle size distribution, and SiO_2 and Al_2O_3 content)
526 and the synthesis conditions (alkali and soluble silicate content) to achieve a specific
527 performance level. The open literature reports a wide number of aluminosilicate sources, as well

528 as different dosage criteria, used in formulation of alkali-activated binders. Therefore, detailed
529 assessments are needed when different raw materials, sources or suppliers are being considered
530 (Bernal et al., 2016).

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

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

549 5. Conclusions

550 Geopolymers are widely considered a low carbon alternative to Portland cement. However,
551 environmental impacts other than global warming potential are important in the life cycle of this
552 type of binder. In this study, LCA was applied to evaluate the impacts related to geopolymer
553 production, considering different activators, for the specific case study of materials developed

554 from a Brazilian kaolin mining waste. The geopolymers samples were compared to binders
555 based on PC (Portland slag cement with 75% clinker). Some conclusions of this study can be
556 pointed out:

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

573 The study highlights the importance of using alternative activators, for example, based on
574 residues such as rice husk ash (RHA), to produce these binders, since significant impact
575 reductions may be achieved. Future studies should consider durability and service life of the
576 cementitious materials based on RHA activation, and assess potential environmental impacts
577 through damage oriented methods, to improve the understanding of decision makers regarding
578 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.