Analysis of the environmental impacts of alkali-activated concrete produced with waste glass-derived silicate activator - A LCA study

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1 **Analysis of the environmental impacts of alkali-activated concrete**

2 **produced with waste glass-derived silicate activator – a LCA study.**

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9

10 **ABSTRACT**

11 Concrete is responsible for a significant share of global GHG emissions, which can be

12 mainly ascribed to the production of clinker. Alkali-activated concretes have been

13 investigated in literature as a possible alternative, but the sustainability still appears reduced

14 by the high embodied energy of chemicals typically used for the activation step. This paper

15 investigates concrete belonging to strength classes 35, 50 and 70 MPa and produced with a

16 silicate activator derived from waste glass (AABR). Through a Life Cycle Assessment (LCA),

17 the investigation aims to compare the AABR to Ordinary Portland Cement (OPC) concrete

18 and alkali-activated concrete produced with commercially available chemicals (AABC). The

19 effects produced by the variations of some key parameters (impact allocation of precursors,

20 energy mix, amount of activator in the concrete, distance of procurement of raw materials)

21 over the total impact of the AABR are also investigated. Results show that the adoption of

22 alkali-activated concretes instead of OPC concrete allows a significant reduction in

23 environmental categories of global warming (averagely 64% reduction for AABC and 70% for

24 AABR), acidification potential (averagely 23% for AABC and 35% for AABR), and terrestrial

25 eutrophication (averagely 53% for AABC and 60% for AABR). In addition, the study

26 evidenced that the use of waste glass-based activator allows a significant reduction in every

27 environmental category when compared to the use of commercially available chemicals.

28

1.   INTRODUCTION

1.1. Background

Concrete is the most utilised building material in the world (Scrivener et al., 2018). Most recent figures on Portland cement production in 2018 suggest a yearly global production of 4.1 billion tonnes (Cembureau, 2019), 80% of which is produced in developing countries. Due to the combined effect of limestone decalcination and fuel utilised for heating the kiln up to 1450 °C, Portland cement (PC) production is responsible for about 5 – 7% of the world's carbon dioxide emissions (Meireles et al., 2019, Costa and Ribeiro, 2020, Kotsay and Jaskulski, 2020, Nwankwo et al., 2020).

In order to achieve the target to limit the temperature increase to 1.5°C above pre-industrial levels set by the Paris Agreement, signatory countries committed to undertake ambitious efforts in reducing greenhouse gas emissions (Rogelj et al., 2016). Fulfilling the CO_2 emission reduction targets will require the construction sector to undergo significant changes.

Most common building materials such as steel, glass, bricks or concrete have relatively high embodied energies. Embodied energy of concrete has been calculated between 0.6 and 2 MJ/kg, whereas embodied energy of steel, brick and glass have been calculated equal to 25, 2-5, and 15-25 MJ/kg respectively (Hammond and Jones, 2008, Dixit, 2016). When recycling is taken into account, embodied energy of steel can be reduced to about 9 MJ/kg (Hammond and Jones, 2008), and a similar value was calculated for recycled glass (Morini et al., 2019). Despite the embodied energy per unit mass of steel, bricks or glass can look higher than the one of concrete in a cradle-to-gate analysis (Lehne and Preston, 2018), it is important recalling that direct comparisons should be carried out only for equivalent function (or performance) and utilisation scenarios. The mass of concrete used in construction outweighs by far any other material, and thus concrete has significant environmental impacts in terms

of carbon emissions, as extensively discussed in the scientific literature and brought to the attention of the general public by recent UK media coverages (The Guardian 2019). The problem lies in the production of clinker (i.e. the main constituent of Portland cement), and therefore, through the substitution of clinker with other, less energy intensive materials, the goal of having a versatile, strong and durable material with low carbon emissions can be achieved. Among the candidates for clinker substitution, magnesium oxides derived from magnesium silicates (MOMS) and alkali-activated binders (AAB) were considered the most promising substitutes, leading to a reduction of over 85% of the emissions (Lehne and Preston, 2018).

Due to the carbonation reactions involved in MOMS binder, a 100% emission reduction could be achieved. However, constraints in terms of supply (due to cost or resource availability) have been evidenced (Shi et al., 2019). Furthermore, the reactivity of ultramafic rock-derived MgO still needs to be fully investigated, as for the time being most of the previous research used magnesium oxide from the decarbonation of magnesium carbonate, which is neither carbon neutral nor sustainable (Scrivener et al., 2018).

AAB (sometimes also referred to as 'geopolymers', although the term should be used only for low calcium binding systems) technology exploits the reaction between aluminosilicate materials (called precursors) and alkali chemicals (called activators) to produce a solid, dense binding matrix. Precursors can be sourced among waste/by-product streams to produce concrete with good mechanical properties and good durability against physical and chemical attacks (Shi et al., 2019). Despite a wide range of activators has been investigated to date, the most common activation method still consists of the combined use of alkali hydroxide (sodium or potassium hydroxide) and alkali silicate (sodium or potassium silicate) solutions (Provis, 2018). The alkali hydroxide is required for increasing the pH of the pore solution and thus triggering the dissolution of Si and Al species, whilst the alkali silicate is used for providing free and active silicate to the system, thus fostering the nucleation of the inorganic aluminosilicate polymeric 3D chain.

84   The use of high embodied energy chemicals for the activation step has an impact on the

85   cost of the alkali-activated concrete production, as well as on the environmental performance

86   of the end products. In order to mitigate this problem, research worldwide has been

87   investigating the production of alternative activators that could be obtained with low-energy

88   processes or that can be sourced from waste or by-product streams. However, strong

89   debate has been ongoing in the scientific community whether or not AAB can significantly

90   decrease the carbon emission of the construction industry.

91   Vinai and Soutsos proposed a method for the production of sodium silicate powder by

92   recycling glass waste (Vinai and Soutsos, 2019). Alkali-activated concretes were

93   manufactured by activating blends of fly ash and ground granulated blast furnace slag

94   (GGBS) with a Na_2SiO_3 powder obtained by thermally treating waste glass powder and

95   sodium hydroxide. Concretes with strength ranging from 35 MPa to 70 MPa were

96   investigated, and a preliminary cost analysis suggested that these were 30% to 35%

97   cheaper than those produced with commercially available chemicals, and 4% to 16%

98   cheaper than PC concrete (Vinai and Soutsos, 2019). The method was independently

99   replicated and assessed in another research that confirmed the effectiveness of the process

100   in producing suitable sodium metasilicate for alkali activation (Samarakoon et al., 2020)

101   **1.2.   Contribution and potential impact of this study**

102   This paper analyses the environmental impact of alkali-activated concrete manufactured

103   using waste glass-derived sodium silicate as the activator (AABR) and compares it with

104   Ordinary Portland Cement (OPC) concrete and alkali-activated concrete produced with

105   commercially available chemicals (AABC). The full life cycle analysis recommended for the

106   Environmental Product Declaration in relevant Standard (BS EN 15804, 2012), as well as

107   the assessment of environmental performance of buildings described in relevant Standard

108   (BS EN 15978, 2011), fall outside the scope of this paper. However, the methodology

109   followed in this study made use of LCA technique for providing a quantitative comparison

110   among different activators that can be used for producing AAB, and for benchmarking the

111   results against OPC concrete production (i.e. the *status quo*).

112   To the best of Authors knowledge, this is the first advanced LCA study on waste glass-

113   derived solid sodium silicate activator, as the only other available study on the environmental

114   impact of this activator was carried out by Samakaroon et al. (2020) and was limited to the

115   emission analysis in the context of a wider study. The significance of this paper lies in the

116   provision of objective and detailed data that are expected to help researchers and industry

117   stakeholders to have a more complete picture on AAB concrete and to boost

118   environmentally responsible actions. Furthermore, these results can foster further research

119   in the study of secondary sources for the production of activators, which can – and must –

120   have a primary importance in the scaling-up of AAB concrete technology. This study

121   demonstrated that the emissions related to the activator could be cut by more than 30% by

122   using waste-derived solid sodium silicate instead of commercially available sodium silicate

123   and sodium hydroxide solutions, thus reinforcing the position of AAB as strong candidates

124   for curbing greenhouse gases emissions from the construction industry.

125

126   **2. LITERATURE REVIEW**

127   The discussion on the carbon emissions of alkali-activated binders started in early 1990's.

128   Comparing the emissions of Portland cement production with the possible emissions of

129   geopolymer cement, Davidovits claimed a possible reduction in CO_2 emission by 80% - 90%

130   (Davidovits, 1993). These results boosted the worldwide interest in alkali-activated materials

131   as potential substitute for Portland cement in concrete application, following global

132   environmental concerns raised by events such as the United Nations Conference on

133   Environment and Development (UNCED), also known as the Rio de Janeiro Earth Summit in

134   1992. Fawell et al. (1999) published a paper discussing the life cycle inventories for the

135   production of sodium silicates (Fawer et al., 1999), and data from this work fostered

136   subsequent LCA analysis of alkali-activated concrete.

137   Habert et al. (2011) described the LCA-based environmental assessment carried out on

138   alkali-activated materials, suggesting that, due to the production of sodium silicate and

139   considering allocations of emissions from industrial processes involved in the production of

fly ash or GGBS, geopolymer concrete had an impact on global warming similar to Portland

cement-based concrete (Habert et al., 2011). Results from Turner and Collins (2013) aligned

with these outcomes, indicating that the CO_2 footprint of geopolymer concrete was

approximately 9% less than comparable Portland cement-based concrete (Turner and

Collins, 2013) when all the production stages were taken into account. These findings were

heavily criticised by Davidovits (2015), objecting that data for the mix proportions and from

the silicate production were not reliable and the analyses grossly overestimated the actual

emissions from geopolymer concrete production (Davidovits, 2015).

It became therefore apparent that the assessment of the actual environmental performances

of alkali-activated materials were heavily influenced by local conditions, and no result could

claim general validity. McLellan et al. (2011) developed a LCA analysis of geopolymer

concrete by considering Australian feedstock, and suggested a possible reduction of

greenhouse gas emissions in the range of 44 – 64% (McLellan et al., 2011). Yang et al.

(2013) investigated the production of alkali-activated concrete referring to Korean lifecycle

inventory (LCI) database, suggesting that alkali-activated concrete would allow a possible

CO_2 emission reduction between 55 and 75%, but warning on the crucial effect of type,

concentration, and dosage of activators (Yang et al., 2013).

Heath et al. (2014) focussed on the production of geopolymer concrete from meta-clay

precursors, observing that this can have a lower global warming potential (GWP) than

Portland cement-based binders. However, it was recognised that large reductions in GWP

are unlikely without the substitution of commercially available soluble silicates, as sodium

silicate used for activation is responsible for the greatest emission contribution and thus it

should be targeted for reducing GWP of alkali-activated materials (Heath et al., 2014).

In a further study on the environmental impact of alkali-activated cements, Habert and

Ouellet-Plamondon (2016) revised their results looking into the environmental profiles of

different precursors and activators, raising important issues regarding the allocation of

impact to by-products such as fly ash and GGBS. Outcomes suggested that slag-based AAB

can reduce the GWP by a factor of four, but all the other environmental impacts investigated

by a LCA study gave worst results than OPC-based materials (Habert and Ouellet-Plamondon, 2016). The GWP impact of a thermally treated mix of albite and sodium hydroxide proposed in the literature (Feng et al., 2012) was also examined. The contribution of such obtained 'one part geopolymer' cement was less than 5% of the GWP of 100% OPC cement, and when economic allocations on slag and fly ash were also considered, an 80% reduction in GWP was achieved (Habert and Ouellet-Plamondon, 2016).

Further research focussed on the production of alternative, waste-derived silicate activators and their environmental effects. Passuello et al. (2017) investigated AAB manufactured using calcined kaolin sludge activated with a waste-derived silicate solution obtained through the dissolution of rice husk ash (RHA) in aqueous NaOH as previously proposed by others (Bouzón et al., 2014). Outcomes indicated that AAB activated with waste-derived sodium silicate allowed a 75% reduction of GWP and beneficial effects on acidification potential (AP), eutrophication potential (EP) and photochemical oxidation (POCP), whereas it showed impact higher than OPC in other LCA categories (Passuello et al., 2017). Tong et al. (2018) proposed a hydrothermal method for the production of sodium silicate solution by dissolving rice husk ash (RHA) in sodium hydroxide solution under the following conditions: NaOH solution concentration 3 M, heating temperature 80 °C, process duration 3 hours. Authors claimed that the waste-derived sodium silicate was able to provide suitable activation for alkali-activated binders, with a reduction of 55% in activator costs, and delivering environmental benefit according to SUB-RAW approach proposed in the literature (Tong et al., 2018).

The implications of energy mix and process for production of NaOH were discussed in a recent paper investigating the local conditions in Ecuador (Salas et al., 2018). With a low-carbon energy mix based on solar power and hydropower, as well as through the production of NaCl (for obtaining NaOH) from seawater evaporation, a 64% GWP reduction when comparing AAB with OPC concretes can be achieved. Other considerations on local conditions such as material availability and impacts related to transportation operations can be found in the literature (Sandanayake et al., 2018).

196 Maddalena et al. (2018) investigated a range of novel binders for insulation purposes,

197 including NaOH-activated metakaolin-based alkali-activated materials, and carried out an

198 extensive LCA study focussing on the local conditions and raw material availability in the UK.

199 They concluded that novel binders can have a carbon footprint up to 23-55% lower than

200 Portland cement (Maddalena et al., 2018). Robayo-Salazar et al. (2018) came to similar

201 conclusions comparing the carbon emissions of a AAB concrete obtained from a blend of

202 natural volcanic pozzolan and slag activated with sodium silicate and sodium hydroxide and

203 OPC. Their results suggested that AAB concrete in the Colombian context showed GWP

204 44.7% lower than the one calculated for OPC with same mechanical properties (Robayo-

205 Salazar et al., 2018).

206 According to Scrivener et al., AAB can play a role in the reduction of global CO_2 emissions

207 only if the CO_2 footprint of activators such as sodium silicate can be at least halved

208 (Scrivener et al., 2018).

209 Summarising, available scientific literature showed that:

210   i.   Environmental benefits from substituting OPC with AAB in concrete need to be

211   considered under local conditions.

212   ii.   Activators and in particular alkali silicates are the main contributors to environmental

213   impacts of AAB and thus only their substitution with less harmful activators can

214   deliver significant benefits.

215   iii.   LCA is the best tool for capturing the whole picture, as some impacts are often

216   overlooked, being the main focus on GWP.

217   iv.   Waste-derived activators and thermally treated solid materials can reduce the

218   emissions significantly in comparison to OPC concretes.

219 In the following sections, an LCA study that investigated the environmental impacts of an

220 AAB concrete produced using a novel solid activator developed by thermal treatment of

221 waste glass powder is presented. Obtained LCA indicators are compared to those of OPC

222 concrete and AAB concrete produced with commercially available chemicals having

223   equivalent fresh properties and strength class. Main outcomes from the investigation and

224   their limitations are discussed.

225   **Table 1.** Summary of the findings from literature review.

Reference	Material	Precursors	Activators	Environmental analysis	CO_2 emission reduction compared to OPC
Davidovits, 1993	Concrete	Calcined clays, slag	Alkali silicates	Stoichiometric assessment of active oxides production	80% - 90%
Habert et al. 2011	Concrete	Fly Ash, slags, or metakaolin.	NaOH, sodium silicate	LCA	Small or no reduction when allocation was considered
Turner and Collins, 2013	Concrete	Fly ash	NaOH, sodium silicate	LCA	9%
McLellan et al., 2011	Paste	Fly ash, silica fume, gibbsite	NaOH, sodium silicate	Sum of emissions from production and transportation	44% - 64%
Yang et al., 2013	Concrete	Fly ash, slag, metakaolin	$Ca(OH)_2$ Sodium silicate NaOH	CO_2 contribution from raw materials, transportation, mixing and curing	55% - 75%
Heath et al., 2014	Concrete	Meta-clays	Alkali hydroxides Alkali silicates	LCA	40%
Habert and Ouellet-Plamondon, 2016	Concrete	Fly ash, slag	Alkali hydroxides, alkali silicate	LCA	75%
Feng et al., 2012	Concrete	Thermally treated albite (from literature)	NaOH	LCA	95%
Passuello et al., 2017	Paste	Calcined kaolin sludge	waste-based sodium silicate	LCA	75%
Tong et al., 2018	Concrete	Fly ash, slag	Waste-based sodium silicate	SUB-RAW	n.a.
Salas et al., 2018	Concrete	Natural zeolite	Sea water-derived NaCl for the production of NaOH and low carbon energy mix	LCA	64%
Maddalena et al., 2018	Binder	Metakaolin	NaOH	LCA	23% - 55%
Robayo-Salazar et al., 2018	Concrete	Natural volcanic soil	NaOH, sodium silicate	GWP, GTP	44.7%

226

3. MATERIALS AND METHODS

3.1. Production process of Ordinary Portland Cement-based concrete and Alkali Activated Binder-based concrete

One of the reasons why concrete is the most popular building material lies in the possibility of obtaining almost any desired properties in fresh and hardened state by adjusting the mix proportions and select suitable constituents. This adds a significant complexity in properly evaluating and comparing the environmental impacts arising from different concretes. As there is not a universal and accepted mix proportioning method for OPC concrete, and even less consensus exists for AAB concrete, there is a strong debate in the research community on how a comparison between different concretes can be fair and objective. The mix proportion influences the physical and mechanical properties of concrete, as well as its environmental impact. The choice of the mixes to be compared is therefore essential in ensuring sounds results and avoiding misleading interpretations. In this study, mix proportions from published papers (Rafeet et al., 2017, Vinai and Soutsos, 2019), focussing on the production of concrete with three specified nominal strengths (35, 50 and 70 MPa respectively) and desired fresh properties, were used. Hereafter a brief recall on the mixes and the assumptions and limitations of these data is provided. Full details can be found in the original publications.

- *Concrete specifications:* three concrete mixes with consistency class S2 were investigated, targeting (a) a typical ready mix concrete, with cube compressive strength of 35 MPa; (b) a typical structural concrete with cube compressive strength 50 MPa; (c) a high strength mix for precast concrete applications, with cube compressive strength of 70 MPa (Rafeet et al., 2017).

- *OPC concrete mixes:* mix proportions suitable for the production of concrete having the desired properties were developed using the BRE method (Marsh et al., 1997) by Rafeet et al. (2017). Authors claimed that mixes were obtained without considering the use of admixtures such as superplasticizers, which would have reduced the water content and

254 therefore the cement content. This might lead to some overestimation of the impact of the 70

255 MPa strength concrete.

256 *- AAB concrete produced with commercially available chemicals (AABC):* mix proportions

257 were developed using the method proposed by Rafeet et al. (2017). Concretes were

258 produced with blends of fly ash and GGBS (the higher the GGBS content, the stronger the

259 concrete was), namely 80%-20%, 70%-30%, and 30%-70% fly ash-GGBS blends for 35, 50

260 and 70 MPa concrete respectively. Binders were activated with NaOH and sodium silicate

261 solutions, the former purchased in prills then dissolved in water, the latter procured from

262 Fisher Scientific and having chemical composition 25.5% SiO_2, 12.8% Na_2O, 61.7% water.

263 Dosage of chemicals was controlled by two parameters, namely the alkali modulus AM (i.e.

264 the mass ratio Na_2O/SiO_2) and the alkali dosage M+ (i.e. the mass ratio Na_2O/binder).

265 Declared values were AM = 1.25 and M+ = 7.5% (Rafeet et al., 2017).

266 *- AAB concrete produced with waste-derived activator (AABR):* the three mixes were

267 proposed by Vinai and Soutsos (2019), where the use of commercially available chemical

268 solutions for the activation was replaced by the inclusion in the mix of a solid powder of

269 sodium metasilicate (Na_2SiO_3) obtained by processing waste glass powder and NaOH (mass

270 ratio 11:10 glass powder:NaOH) in oven at temperature ranging from 150°C to 330°C.

271 Authors demonstrated the suitability of the waste-derived sodium metasilicate in activating

272 blends of fly ash-GGBS mortars using M+ = 7.5% and AM = 1 as chemical dosages (this

273 latter due to the chemical nature of the sodium metasilicate). They then provided mix

274 proportions derived from Rafeet et al. (2017) by substituting the amount of chemicals (NaOH

275 and sodium silicate solution) with a suitable amount of novel activating powder, which

276 resulted being about 18.8% of the binder mass (Vinai and Soutsos, 2019). The Authors did

277 not provide an experimental validation of the proposed mixes, and thus the assessment of

278 the required quantity of activating powder should cover a range rather than as a fixed value.

279 For this reason, in this paper a sensitivity analysis was carried out on the powder content, in

280 the range 15% - 25% of the binder mass.

281 The processes for the production of OPC, AABC and AABR concretes are not dissimilar. For

282 OPC concrete, aggregate (i.e. sand and gravel) is mixed with OPC powder, and then water

283 is added (sometimes aggregate fraction is pre-mixed with a certain amount of water in order

284 to ensure its saturation). For AABC concrete, aggregate and binder blend (i.e. fly ash and

285 GGBS) are mixed, the liquid fraction (i.e. sodium silicate solution, NaOH and water) is

286 prepared and then added in the mixer. For AABR concrete, aggregate, binder blend and

287 activating powder are mixed, then water is added. The same layout of equipment for the

288 production of concrete can be assumed in the three cases. No special need for curing was

289 assumed in the analysis.

290

291   **3.2.   Goal and scope of concrete LCA**

292 The goal of this study is to evaluate if the AAB concrete with activators from recycled waste

293 glass (AABR concrete) could represent a valid solution to mitigate the environmental impacts

294 of concrete. Specifically, AABR concrete is compared to OPC and to AAB concrete with

295 activators from commercial sources (AABC concrete). To enable an objective comparison,

296 the related assessments have the same system boundaries (see Figure 1), which

297 comprehend the raw material extraction and processing, the processing of secondary

298 material input, the transport to the manufacturer. No cut-off rule is applied for the calculation

299 since the entire production chain of each raw and secondary materials is included in the

300 study. In line with previous studies on concrete, the functional unit of the study is 1 m^3 of

301 concrete with a specified compressive strength (concrete with 35, 50 and 70 MPa cube

302 compressive strength have been evaluated). Only functionally equivalent concretes have

303 been compared, i.e. concretes from the same strength class having thus same functional

304 unit and same function, production, operation and use scenarios. It has been assumed that

305 concrete mixes from different binding systems would have the same durability. The detailed

306 analysis of concrete durability is outside the scope of this paper, although there is general

307 consensus that AAB concrete can have better durability than OPC concrete under harsh

308 environmental conditions, thus the obtained results should be conservative in respect of

309 OPC concrete and not misleadingly in favour of AAB concrete in this regard. Primary data

310 have been preferably used for the inventory. When primary data were not available, good

311 quality secondary data from Ecoinvent 3.4 Cut-off database have been employed, with

312 particular attention to the technological, time and geographical representativeness.

313 As underlined by the previous literature, results can be highly affected by the approach used

314 to deal with waste and by-products. This LCA study has been developed with a cut-off

315 approach (Wernet et al., 2016) and considers GGBS and fly ash as allocable by-products,

316 since they currently have a market value. Specifically, fly ash is a by-product from the coal

317 combustion in thermal power stations and GGBS is a by-product from blast furnaces

318 producing iron. Therefore these materials bear some burdens from their production, which

319 are allocated on an economic basis against the reference flow. The physical allocation has

320 been avoided because of the different unit of measure between fly ash (kg) and energy (MJ)

321 and because of the relatively high mass of GGBS against iron, as discussed by Chen et al.

322 (2010).

323 The glass powder used for producing the activator in AABR concrete can be derived from

324 two different processes, both of them considered in the LCA of this study. Firstly it is

325 considered the glass powder comes as an unintended residue from the glass recycling

326 process. In this case, the glass powder enters the concrete production process as a burden-

327 free flow. Secondly, it is considered the case of production from the grinding of waste glass.

328 In this case, in line with the study of Passuello et al. (2017), the glass powder only bears

329 impacts due to its beneficiation.

330 Inventory and impact calculations have been supported by SimaPro 8 software. The

331 software allows the user to select the method for impact assessment among more than 30

332 methods proposed by international bodies or literature (PRé, 2020). The method chosen in

333 this study for the impact assessment was the International Reference Life Cycle Data

334 System (ILCD) Midpoint+ (version 1.0.9), developed under the coordination of the European

335 Commission and backed by most LCA practitioners. The ILCD method harmonises the

336 existing practices in line with ISO 14040 and 14044:2006. A broad documentation has been

337   developed by the European Commission Joint Research Centre - Institute for Environment

338   and Sustainability, providing guidance for the application of the method and the choice of

339   characterisation factors. These have been detailed in handbook (European Commission,

340   2011) and technical note (European Commission, 2012), where the quality of the indicators

341   and the relevant literature backing their calculation have been thoroughly discussed.

342   Interested readers are referred to these publications for the details. This paper shows the

343   impact results for the categories of global warming potential (GWP), acidification potential

344   (AP), Terrestrial Eutrophication (TE), Freshwater ecotoxicity (FE), Particulate matter (PM),

345   Mineral, fossil & renewable resource depletion (RD). All these categories are recommended

346   with quality levels between I and III in the ILCD supporting information (European

347   Commission, 2012), and have been chosen because of their relevance for the construction

348   sector, and/or because they are in line with literature studies.

349   Contribution analyses have also been developed for the two AAB products to understand

350   which processes of the supply chain have the major influence to the different impact

351   categories and to identify if there is still room for improvement. Finally, the AABR production

352   chain has been further investigated through a sensitivity analysis on some key variables that

353   are significantly uncertain. The robustness of the results has been verified and discussed

354   through the introduction of flexible parameters on the economic allocation factors, the energy

355   mix used for the processes of glass powder production, the raw materials ratios and the

356   transport distances. Eventually, some simplifications have been included in the analysis

357   when the complexity and the uncertainty of some parameters determining the scenarios

358   would have introduced excessive variability in the results, making a general comparison

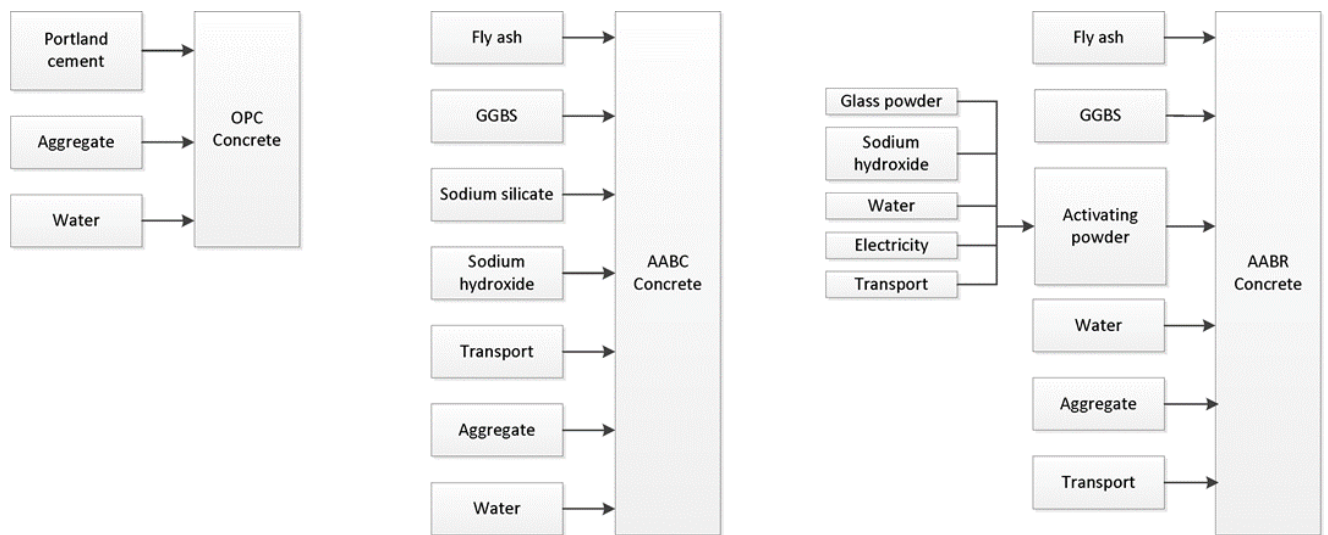359   ineffective and meaningless. Section 4.4 details the limitations of this study.

360

361

362   **Figure 1.** Flow charts of the analysed mix designs of concrete.

363

364   **3.3.   Life cycle inventory**

365   This section gives details on the Life Cycle Inventory of the different types of concrete that

366   have been investigated. This allows the reader to clearly understand and replicate the LCA

367   model, eventually modify some parameters and calculate the environmental impacts.

368   Nine different concrete combinations have been considered: OPC, AABC and AABR

369   concretes, each one having nominal compressive strengths of 35, 50 and 70MPa.

370   Table 2 summarises the components and quantities for each of the 9 cases. Quantities of

371   input flows are based on previous studies (Vinai and Soutsos, 2019) and adapted to conform

372   to the nature and chemistry of LCA database entries.

373   Specifically, OPC concrete is composed by Portland cement, water and aggregates. For the

374   Portland cement, a dataset of Ecoinvent 3.4 database has been used. This dataset was

375   chosen as it represents the average situation in Europe, it is focussed on the main current

376   technologies and it is modelled with reference to the year 2017. Transportation of Portland

377   cement from the cement factory to the mixing plant was not included in the analysis, please

378   refer to section 4.4 for further discussion on possible limitations due to this parameter.

379

380

381   **Table 2.** Mix design of the 9 analysed types of concrete, data for 1 m^3 of concrete.

	35MPa			50MPa			70MPa		
	AABC	AABR	OPC	AABC	AABR	OPC	AABC	AABR	OPC
Portland Cement (kg)	-	-	355	-	-	440	-	-	550
Fly Ash (kg)	283.2	283.2	-	258.3	258.3	-	99	99	-
GGBS (kg)	70.8	70.8	-	110.7	110.7	-	231	231	-
Sodium Silicate Solid (kg)	27.7	-	-	28.8	-	-	25.8	-	-
Activating Powder (kg)	-	66.6	-	-	69.4	-	-	62.1	-
Sodium Hydroxide (kg)	26.0	-	-	27.1	-	-	24.2	-	-
Water (kg)	157.3	158.8	220	155.6	157	220	169.1	170.8	220
Aggregate (kg)	1897.5	1897.5	1832	1897.5	1897.5	1750	1925	1925	1668
Total (kg)	2462.4	2476.9	2407	2478.0	2492.9	2410	2474.1	2487.9	2438

382

383   The AABC concrete does not contain any Portland cement and the binder is composed by

384   fly ash, ground-granulated blast-furnace slag (GGBS), sodium hydroxide and sodium

385   silicate. Particular attention was paid on the choice of the proxy dataset for the activators, as

386   these are responsible for the highest share of embodied energy, global warming potential

387   and other environmental impact. As previously described, the dosage of chemicals in the

388   reference literature is controlled by the alkali modulus AM (mass ratio Na_2O/SiO_2) and the

389   alkali dosage M+ (mass ratio Na_2O/binder). The values of AM = 1.25 and M+ = 7.5% in the

390   concrete mixes were claimed to be achieved through the inclusion of a blend of sodium

391   silicate and sodium hydroxide solutions. In order to ensure that the quantities of chemicals

392   were correctly computed in the LCA model, the compositions of the sodium silicate and

393   sodium hydroxide recorded in the Ecoinvent database were checked and equivalent masses

394   of chemicals were calculated. From the value of alkali dosage M+ and the mass of binder

395   obtained from the targeted literature, it was possible to calculate the required Na_2O. The

396 following calculations pertain to the 35 MPa AABC mix, but data for other mixes can be

397 calculated in analogy:

398   Mass of binder = mass of Fly Ash + mass of GGBS = 283.2 + 70.8 = 354 kg   (1)

399   Mass of Na_2O = (mass of binder) × (M+) = 354 × 0.075 = 26.5 kg   (2)

400   Mass of SiO_2 = (mass of Na_2O) / (AM) = 26.5 / 1.25 = 21.2 kg   (3)

401 According to the information provided by Ecoinvent database version 3, chemical products

402 are always expressed in 100% active substance, whereas the descriptions hint to the most

403 common forms in which the chemicals are used (Ecoinvent, 2013). In the case of sodium

404 silicate solution, the description is "Sodium silicate, without water, in 37% solution state". In

405 such dataset there is an input of "sodium silicate production, solid", which in turn is

406 composed by 0.772 kg silica sand and 0.4 kg soda ash (for 1 kg of sodium silicate).

407 According to the chemical composition of silica sand (100% SiO_2) and soda ash (Na_2CO_3),

408 the resulting sodium silicate would be composed by about 77% SiO_2 and 23% Na_2O.

409 Therefore, in order to incorporate in the concrete mix 21.2 kg of SiO_2 calculated in (3), the

410 equivalent mass of sodium silicate having such composition that needs to be added is:

411   Mass of sodium silicate = 21.2 / 0.77 = 27.7 kg.   (4)

412 A share of this mass of sodium silicate is represented by Na_2O:

413   Mass of Na_2O = (mass of sodium silicate) × 0.23 = 6.4 kg.   (5)

414 In order to achieve the desired 26.5 kg of Na_2O calculated in (2), some 20.1 kg of extra

415 Na_2O needs to be added from NaOH. The database entry for NaOH is "Sodium hydroxide,

416 without water, in 50% solution state", whereas it actually refers to the impacts of 100% solid

417 sodium hydroxide. Knowing that Na_2O represents 77.5% of NaOH, the quantity of NaOH

418 requested for achieving the target amount of Na_2O is:

419   Mass of NaOH = (mass of Na_2O) / 0.775 = 20.1 / 0.775 = 26 kg.   (6)

420 The quantities of sodium silicate and sodium hydroxide calculated in (4) and (6) were

421 therefore used in AABC mix proportions in order to ensure that the required quantities of

422 SiO_2 and Na_2O for the activation matched the quantities provided by Vinai and Soutsos

423 (2019). The amount of water in the mixes was then adjusted by calculating the water from

424   the sodium silicate solution used in the mix from the literature and adding it into the total

425   water.

426   As far as the fly ash is concerned, in line with previous literature (Seto, 2017, Babbitt, 2005),

427   it has been assumed that the production of 1 MWh of electricity from hard coal fuelled plants

428   produces 29.8 kg of fly ash as by-product. A market price equal to 0.1173 €/kWh has been

429   used for the electricity for the economic allocation of impacts, since that is the EU-27

430   average price for non-household consumers recorded in the second half of 2019, according

431   to Eurostat (2019). The fly ash price has been estimated from a market analysis carried out

432   using prices published on the Alibaba web platform by different retailers. The average value

433   (used for the default allocation) is 0.024 €/kg, while minimum (0.009 €/kg) and maximum

434   (0.055 €/kg) values have been adopted in the sensitivity analysis. A similar procedure was

435   used to obtain values for the economic allocation between the pig iron and the by-product of

436   iron slag (precursor of GGBS). Specifically, the market price of pig iron has been calculated

437   as the average price of pig iron exports from Brazil, Russia and Ukraine (331 $/t)

438   (Steelonthenet.com, 2020), while the iron slag average, minimum and maximum values have

439   been obtained from Curry (2020) and from a web research of retailers. The adopted values

440   are respectively 27.5 $/t, 10 $/t and 60 $/t. Transportation of fly ash and GGBS was included

441   in the analysis by considering a distance of 100 km from the available raw materials and the

442   mixing plant. Please refer to section 4.4 for further discussion on possible limitations due to

443   this parameter.

444   The production process of the AABR concrete involves manufacturing an activating powder

445   from recycled glass and sodium hydroxide. This powder is then used to in the concrete

446   mixes. As discussed in section 3.2, glass powder can come both as a residue from the glass

447   recycling (case 1) and from a grinding process of glass waste (case 2). In both cases, in

448   order to produce 1 kg of activating powder it can be estimated an energy consumption equal

449   to 0.072 MJ (in the form of electricity), based on an oven with power consumption of 10kW,

450   running for 2 hours to produce 1000 kg of powder. The mass ratio of glass dust to sodium

451   hydroxide has been fixed equal to 11:10, according to the literature (Vinai and Soutsos,

452   2019). The water added has been estimated equal to 0.1 kg per kg of powder, while

453   transportation of glass waste has been assumed equal to 100 km on a truck, please refer to

454   section 4.4 for further discussion on possible limitations due to this parameter. Table 3

455   summarises the inputs for the production of 1 kg of activating powder in the two cases.

456

457   **Table 3.** Inputs for the production of 1 kg of activating powder

	Input quantities - CASE 1	Input quantities - CASE 2
Glass powder from glass recycling process (burden-free)	0.52 kg	/
Transport of waste glass powder	100 km	/
Glass waste (burden-free)	/	0.52 kg
Transport of waste glass	/	100 km
Electricity (to grind waste glass)	/	0.0072 MJ
Electricity for oven	0.072 MJ	
Sodium hydroxide	0.48 kg	
Water	0.1 kg	

458

459

460   The AABR concrete inventory includes the activation powder, as well as GGBS, fly ash,

461   water and aggregates.

462   Table 4 lists the proxy datasets used for each element. Datasets were chosen from

463   Ecoinvent 3.4 database and are considered representative of the materials used in the case

464   study here discussed.

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469

470   **Table 4.** Correspondence between flows and datasets in Ecoinvent 3.4

Input	Proxy dataset
Portland Cement	Cement, Portland {Europe without Switzerland}	market for	Cut-off, S
Fly Ash	Built from "Heat, district or industrial, other than natural gas {Europe without Switzerland}	heat production, at hard coal industrial furnace 1-10MW	Cut-off, U" (allocated)
GGBS	Built from "Pig iron {GLO}| production	Cut-off, S" (allocated) + Ground granulated blast furnace slag {GLO}| production	Cut-off, U
Sodium Silicate Solution	Sodium silicate, without water, in 37% solution state {RER}	sodium silicate production, furnace liquor, product in 37% solution state	Cut-off, S
Sodium Hydroxide	Sodium hydroxide, without water, in 50% solution state {GLO}	market for	Cut-off, S
Water	Tap water {GLO}	market group for	Cut-off, S
Aggregate	Gravel, crushed {CH}	market for gravel, crushed	Cut-off, S
Electricity	Electricity, medium voltage {RER}	market for	Cut-off, S
Transport	Transport, freight, lorry 16-32 metric ton, EURO6 {RER}	transport, freight, lorry 16-32 metric ton, EURO6	Cut-off, S

471

472   Some variables have also been introduced in the LCA model to enable the development of

473   sensitivity analyses. Table 5 summarises the variables and the related values that have

474   been set for the analysis. In particular, for the evaluation of the effect of the electricity mix

475   used to produce the activating powder on its environmental impacts (parameter named

476   "Electr_activ_powder"), it has been chosen to perform the sensitivity analysis using the

477   average European grid mix as a benchmark, the Swedish mix for a low carbon option

478   (having a high share of renewables) and the Polish mix for a carbon intensive option (having

479   a high share of energy from coal). The ratio of the activating powder in the AABR mix design

480   (variable named "activating_powder_quantity") has been varied from 15% to 25% because it

481   is a mixing parameter that has not been optimised yet and, as a consequence, future

482   variation of the quantity of this constituent in the concrete mix could be possible. As

483   previously discussed, the economic allocations of fly ash and iron slag (parameters named

484   "alloc_flyash" and "alloc_slag") consider the variation in market prices of the two by-

485   products. Finally, the parameter "distance" has been introduced to evaluate how a local or

486   national provision of binder raw materials might affect the results.

487

488   **Table 5.** Parameters for the sensitivity analysis of impact results.

Name of variable	Description	Base case (default value)	Scenarios	
Electr_activ_powder	Electricity mix used to produce 1 kg of activating power	market group for electricity, medium voltage Europe without Switzerland	• market for electricity, medium voltage PL • market for electricity, medium voltage SE	
activating_powder_quantity	Quantity of activating powder (% of the binder mass) in the mix design of AABR concrete	18.8%	MIN: 15%	MAX: 25%
alloc_flyash	Economic allocation for the outputs of electricity and fly ash from hard coal furnace	99.4% electricity; 0.6% fly ash	MIN: 99.8% electricity; 0.2% fly ash	MAX: 98.6% electricity; 1.4% fly ash
alloc_slag	Economic allocation for the outputs of pig iron and iron slag (precursor of GGBS)	97.6% pig iron; 2.4% iron slag	MIN: 99.1% pig iron; 0.9% iron slag	MAX: 94.9% pig iron; 5.1% iron slag
distance	Distance for the transportation of binder raw materials (fly ash, GGBS, sodium hydroxide, sodium silicate, glass powder)	100 km	500 km	

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4. RESULTS AND DISCUSSION

4.1. Life cycle impact assessment and interpretation

Main results obtained through the LCA analysis are detailed in this section. Environmental impacts of OPC, AABC and AABR concretes within the same strength class are compared for the impact categories of GWP, AP, TE, FE, PM and RD, as indicated in section 3.2. Results for AABR concrete were obtained under the case 1 scenario (activating powder produced from residues of glass recycling), which can be considered relevant also for case 2 (activating powder produced from a grinding process of glass waste), since outputs differ only by 0.1% for all impact categories. Table 6 shows the absolute values of the obtained potential impacts, while Figure 2 provides a graphical representation of relative results, where the 100% is set for the highest value reached for each impact category in each strength class. As it can be noticed, the potential impact on climate change of OPC concrete is significantly higher (about three times) than both alkali-activated concretes for each strength class. AABR concrete results showed the lowest GWP impact, while AABC concrete is averagely 16% more impactful. OPC concrete shows the highest potential impacts also for the TE and the AP categories. On the other hand, values for indicators FE and RD were comparable between OPC and AABC, although results for OPC were slightly lower. In particular, the main contributor in the RD impact category for all the investigated concrete mixes was the consumption of gravel, followed by the consumption of Portland cement (for OPC), sodium silicate (for AABC and AABR), and, to a lesser extent, sodium hydroxide (for AABC). As far as the PM indicator is concerned, lowest impacts are obtained by OPC, but it has to be underlined that, in absolute terms, the impact is rather low for all the analysed materials. Results on PM normalised per person according to the EC-JRC Global method, see for example (Crenna et al., 2019), show therefore that 1 m^3 of all the analysed types of concrete are in the range 0.01 to 0.02.

When comparing AABR to AABC, it was noticed that impact indicators were always lower for the former.

522 **Table 6.** Potential impacts of OPC, AABC and AABR concretes for the strength classes of
523 35, 50 and 70 MPa.

Impact category	Unit	35 MPa			50 MPa			70 MPa		
		OPC	AABC	AABR	OPC	AABC	AABR	OPC	AABC	AABR
Climate change	kg CO_2 eq	333	123	100	409	142	118	507	181	160
Particulate matter	kg PM2.5 eq	0.055	0.095	0.076	0.0655	0.105	0.087	0.079	0.122	0.105
Acidification	molc H+ eq	0.845	0.705	0.542	1.020	0.777	0.608	1.250	0.875	0.724
Terrestrial eutrophication	molc N eq	2.660	1.400	1.090	3.190	1.510	1.190	3.890	1.640	1.360
Freshwater ecotoxicity	CTUe	579	965	672	682	1040	738	817	1100	827
Mineral, fossil & ren resource depletion	kg Sb eq	0.0075	0.0112	0.0084	0.0078	0.0116	0.0086	0.0083	0.0116	0.0089

524

525 Analysing the results obtained for OPC concrete, it can be observed that Portland cement

526 bears the highest share of impacts: its contribution varies from 96% to 99% for all the impact

527 categories, except for RD, for which it accounts for 79%. The contribution analysis of AABC

528 and AABR concretes are shown in Figures 3 and 4 respectively. As it can be noticed, GGBS,

529 sodium silicate and sodium hydroxide are the main contributors in almost all impact

530 categories for AABC concrete, while fly ash accounts averagely for 6% of impacts and gravel

531 gives significant contributions only for RD (48%) and TE (24%) indicators. On the other

532 hand, the activating powder is the most significant contributors in the analysis of the AABR

533 concrete, accounting for most of the impact in all the analysed categories, apart from RD.

534 Specifically, 96% to 98% of the impacts from the activating powder are due to the sodium

535 hydroxide used in the activator production process. An important share of impacts is borne

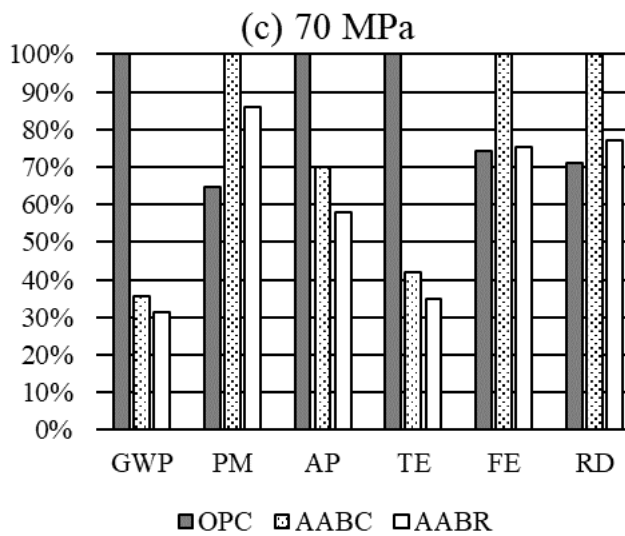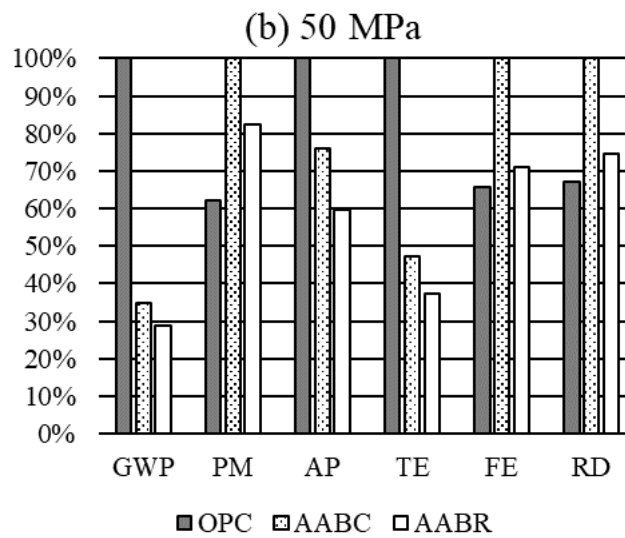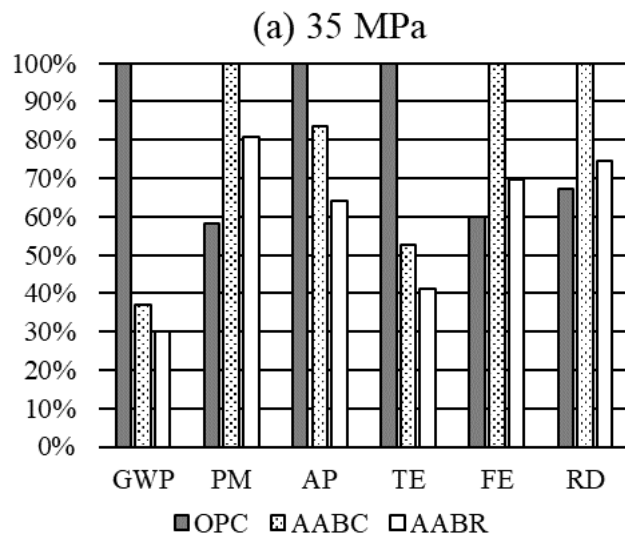536 by GGBS as well, whilst fly ash averagely accounts for 8% of impacts.

537

538

Figure 2. Relative environmental impacts of OPC, AABR and AABC concretes for the strength class of (a) 35 MPa, (b) 50 MPa, and (c) 70 MPa.
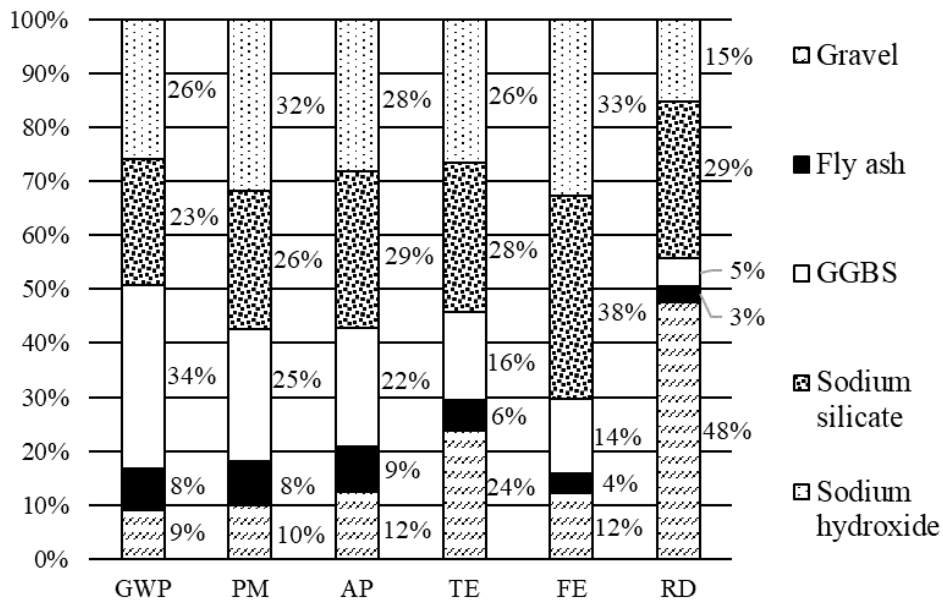
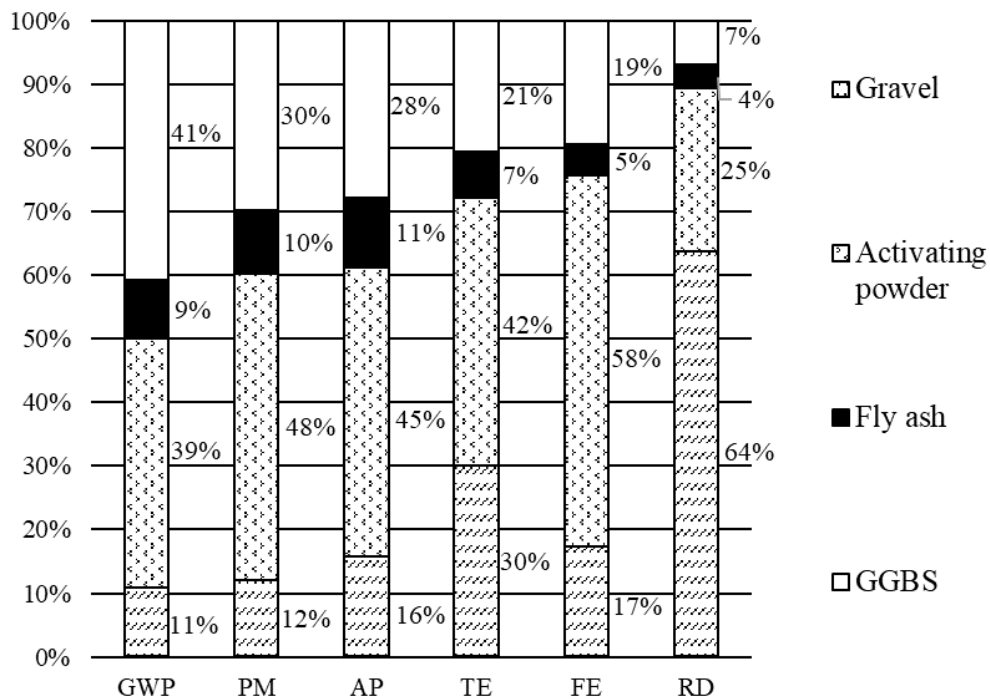**Figure 3.** Impact contribution analysis of 50 MPa AABC concrete.

Figure 4. Impact contribution analysis of 50 MPa AABR concrete.

4.2. Sensitivity analysis

As discussed in section 3.2, some variables have been introduced in the LCA model to

develop a sensitivity analysis on the impact results. Specifically, it was felt relevant to

25

550 understand how assumptions on economic allocations of fly ash and iron slag, as well as

551 transport distance of binder elements, could affect the results for AABC and AABR

552 concretes. For the fly ash, minimum (0.009 €/kg) and maximum (0.055 €/kg) market prices

553 have been used for the analysis. Similarly, the price of iron slag (precursor of GGBS) has

554 been varied from 10 $/t (minimum value) to 60 $/t (maximum value). Transport distance has

555 been increased to 500 km for fly ash, GGBS, sodium hydroxide and glass powder to take

556 into account an average transportation that would be necessary for national provision of

557 materials in typical European countries. The ranges of parameters that were adopted in the

558 sensitivity analysis are shown in Table 5. Figure 5 shows how the total impact on climate

559 change of 50 MPa AABC and AABR concretes varies when varying these parameters. As it

560 can be noticed, the variation of fly ash allocation values does not affect significantly the

561 results (-3%/+7%). Assumptions on slag allocation resulted in slightly higher variation (-

562 7%/+14%), while the highest effect can be observed for the increase in transport distance

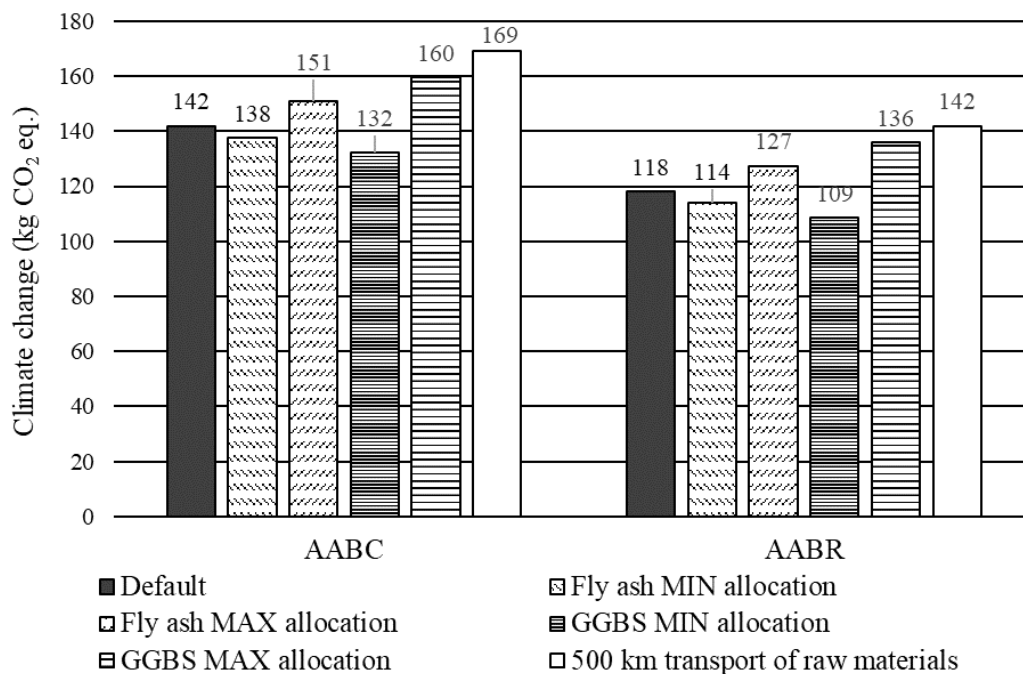563 (+20%).

564

565

566 **Figure 5.** Sensitivity analysis on climate change results for 50 MPa AABC and AABR

567 concretes. The parameters that have been made vary are the fly ash allocation, the slag

568 allocation and the transport distance for binder elements.

569   A further sensitivity analysis has been developed for the AABR concrete to assess the

570   effects of the quantity of activating powder (ratio of activating powder in the mix design, i.e.

571   activating powder mass per m^3 of concrete) and of the energy used for producing it on the

572   selected environmental impact indicators.

573   Figure 6 shows the percentage comparison between the default mix design (18.8% of

574   activating powder) and mix designs having respectively the minimum (15%) and maximum

575   (25%) share of activating powder. As it can be noticed, the variation is similar for all the

576   analysed impact categories, averagely ranging between -9% (for a minimum ratio of

577   activating powder) and +14% (for the maximum ratio of activating powder). It is worth noting

578   that even in the worst scenario, impacts of AABR result significantly lower than impacts of

579   AABC for all the analysed categories (these latter shown as solid lines in Figure 6).

580   The change in energy mix for producing the activating powder did not significantly affect

581   impact results, which averagely decrease of 1% with the energy mix of Sweden (i.e. high

582   share of renewable energy sources) and increase of 1% with the energy mix of Poland (i.e.

583   high share of coal-based energy). The reason of this high stability is related to the relatively

584   small amount of electricity required for the production of activating powder.
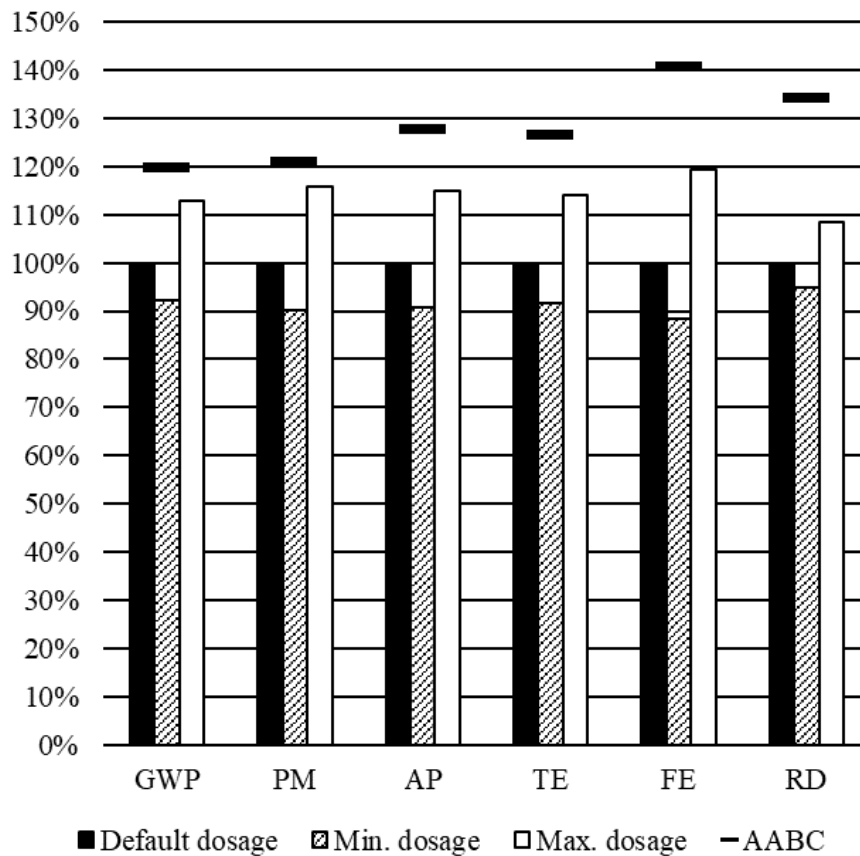
585

Figure 6. Percentage change of impacts in relation to the ratio of activating powder in 50 MPa AABR mix design. For each impact category, the line represents the impact of 50 MPa AABC concrete.

589

4.3. Discussion

The main objective of this study was to assess quantitatively the environmental impact of a waste-based activator for AAB production. There is large consensus in the literature on the use of LCA as a tool for objective and quantitative assessment of the life cycle impact of products, production methods or global processes. Nonetheless, the discussion on LCA of alkali activated binders is very much debated as the number of possible variables in the system is high and different assumptions would lead to significantly diverging results. The main complication in comparing different results lies on the wide variety of mixing approaches (use of chemicals as solid or in solution, quantification of the added chemicals as solid ratios, liquid to solid ratios, molarity of the solution and so on) and the difficulties in ensuring that datasets from LCA database conveniently reflect the actual mix proportions,

601 particularly as far as activators are concerned. Furthermore, there is not such a thing as a

602 "typical concrete" to be used for comparison purposes, due to the large variability of

603 technical requirements and thus of concrete compositions, and therefore a meaningful

604 comparison 'alike for alike' from literature data is very difficult. This research aimed at: (a)

605 providing an assessment of environmental impacts of different classes of concrete; (b)

606 providing a fair and sound comparison between a waste-based activator and commercially

607 available chemicals; (c) determining the effects of variations of some key parameters over

608 the total impact of the AABR.

609 Results from this study were compared to published outcomes from the literature, in order to

610 benchmark the expected values and to confirm the robustness of the methodology that was

611 followed. Data collected from significant available publications are shown in Table 7 and

612 plotted in Figure 7 against results obtained from this study. It can be appreciated that

613 obtained results sits well in this dataset, confirming the robustness of the methodology.

614

615 **Table 7.** CO_2 emissions per cubic meter of concretes, data from literature

Source	Strength class (MPa)	CO_2-eq (kg/m^3)		
		OPC	AABC	AABR
(Habert et al., 2011)	52.5	306	168	-
(Turner and Collins, 2013)	40	354	320	-
(Yang et al.,2013)	24	323	110	-
	40	509	122	-
	70	568	256	-
(Salas et al., 2018)	15	302	110	-
(Samarakoon et al., 2020)	40	-	-	191
	65	-	284	-
This study	35	333	123	100
	50	409	142	118
	70	507	181	160

616

617 The assessment of the environmental impacts of different classes of concrete confirmed

618 current trends in LCA studies on concrete. Alkali activated concretes can significantly reduce

619 most of the impacts identified from the analysis. Climate change indicator can be reduced by

620 63%-65% with the adoption of AABC, and in the range of 68%-71% when using waste-

621 derived activators. The acidification indicator is similarly reduced by 17%-30%, whereas the

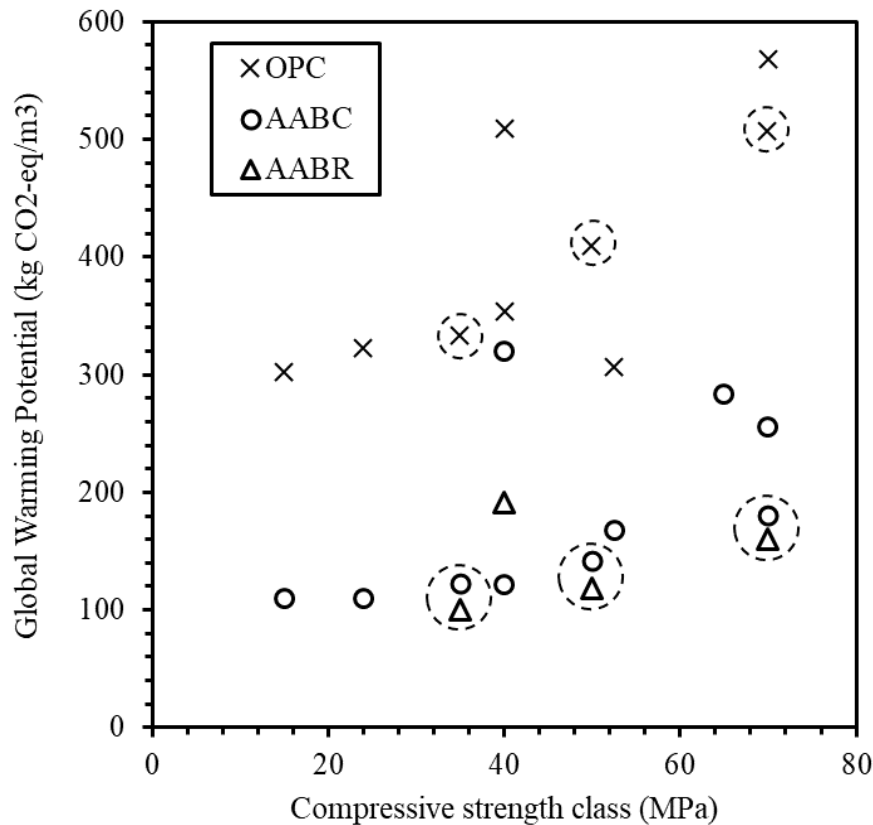622 use of waste glass-based activator allows reductions of 29%-40% of this parameter.

623

624

625 **Figure 7**. Equivalent carbon emissions per cubic metre of concrete versus compressive

626 strength. Circled (dashed line): results from this study.

627

628 Another significant reduction was achieved in terms of terrestrial eutrophication, for which

629 the adoption of AABC concrete could allow a reduction of 47%-58%, whereas reductions in

630 excess of 57%-63% were obtained with use of the waste glass-based activator.

631 A less sharp outcome was obtained for indicators such as particulate matter, freshwater

632 ecotoxicity and resource depletion. The freshwater ecotoxicity indicator worsen significantly

633 with the adoption of AAB, essentially due to the use of activators. Similar results were found

634 in the literature (Passuello et al., 2017, Di Maria et al., 2018). The particulate matter indicator

635 showed higher values for AAB, due to both the use of GGBS and the use of chemicals for

636 activation, although in absolute terms did not raise particular concern. The natural resource

637   depletion indicator also increased for AAB, which was ascribed to metal depletion in the

638   literature (Matheu et al., 2015). However, with the adoption of AABC the increase is

639   relatively low (7% to 12%), while it is slightly higher for AABC (40% to 49%).

640   It can be noticed that impacts of the three concretes classes always increase at the

641   increasing of the strength class, due to the need to increase the binder content (for OPC) on

642   one hand or the GGBS content (for AAB) on the other hand.

643   The analysis allowed comparison between commercially available chemicals for activation to

644   a waste glass-derived activator, and the study demonstrated that the use of this latter

645   consistently improved the environmental performances of the concrete. Whilst the use of

646   commercially available activators accounts for about 50% on the total climate change

647   indicator, adoption of waste-based activator reduces this burden by 10%. The consequence

648   is that the CO_2 eq. emissions directly allocated to the activators are reduced on average by

649   30%-32% when using the glass-waste derived activator, which is near to the 50% cut

650   claimed by other scholars (Scrivener et al., 2018) for alkali-activated binders to play a

651   significant role in reducing the global warming potential of built environment. It was therefore

652   demonstrated that the development of alternative, waste-derived activators is a key strategy

653   in tacking the environmental impact of concrete in construction. The results can be

654   considered precautionary, as the avoided impacts of non-optimal management of glass

655   cullet waste were not included in the analysis.

656   Eventually, a sensitivity analysis allowed to investigate the impacts of selected key

657   parameters on the environmental performances of AAB concretes. The most significant

658   impacts were observed from assumptions on the GGBS impact allocation, similarly to other

659   works (Habert et al., 2011), and from the transportation distance of raw materials, this latter

660   being the most significant parameter, with increases in the range of 20% of climate change

661   indicator. Similar results were also found in the literature (Peys et al., 2018, Petrillo et al.,

662   2016). The energy mix of electricity and the assumptions on fly ash allocation resulted in

663   minor impacts.

664 The optimisation of the quantity of activating powder in the mix has a significant effect on the

665 environmental performances: a decrease of its dosage (mass ratio of activator/binder) of

666 about 4% (i.e. passing from 18.8% to 15%) led to a reduction of 9% on average, whereas

667 the increase of its dosage of about 6% (i.e. passing from 18.8% to 25%) resulted in a

668 general increase in impact indicator of about 14%. There is therefore a significant potential

669 for further reducing the environmental impacts of AABR concretes through the optimisation

670 of the mix proportions and activation.

671

672 **4.4. Limitations of the study**

673 The objective of this research was to assess the environmental impact of a novel, waste-

674 based activator, and to compare it to alkali-activated concrete produced with commercial

675 chemicals and to OPC concrete. Some choices and assumptions have been made about the

676 boundary of the study, and therefore some limitations need to be pointed out:

677 (1) The system boundaries of the study included the energy consumption for the production

678 of the materials composing the concrete, but did not include the energy for mixing and

679 casting the concrete. The reasons for this choice are as follows:

680 o Concrete batching can be carried out following different methods and equipment

681 according to the typology of the production (precast or onsite placement) and the

682 end-products, these factors can further vary between different construction sites.

683 This huge variability would affect the impact results and would require a focussed

684 study, limited to specific context.

685 o As concluded by Flower and Sanjayan (2007), concrete-batching and placement

686 activities contributed to a minor share of CO_2 to total concrete emissions. However,

687 if this was true for OPC concrete, this contribution can be as high as 15% when

688 considering low-carbon binders such as the materials investigated in the present

689 study. Yang et al. (2013) suggested emissions equal to 0.008 CO_2 kg/kg of concrete

690 due to the concrete production, while Flower and Sanjayan (2007) calculated

691 emissions of 12.3 kg CO_2/m^3 for the concrete-batching and placement activities.

692	Since the bulk density of the concrete mixes summarised in Table 2 is on average
693	2459 kg/m^3 with small fluctuations, the CO_2 emissions related to the concrete
694	production phase can be estimated about between 12 and 20 kg/m^3 and results
695	would therefore need to be offset by this amount. This estimation is in line with
696	values declared in the literature of about 17 kg/m^3 (Salas et al., 2018).
697	(2) Similarly, the study did not attempt at covering the end of life stage for the three
698	investigated concretes, since the reusing/recycling/recovering options would strongly
699	depend on the engineering application, concrete end-product and site-specific
700	strategies, and would anyway be similar for the three mixes. From a comparison
701	perspective, this further analysis would not have added information to the objectives of
702	the study. A further investigation could be carried out assessing likely scenarios for the
703	end of life stage as recommended for Environmental Product Declaration (EPD)
704	preparation (BS EN 15804, 2012).
705	(3) The generation of waste during concrete production was not included in the analysis.
706	This choice was due to the following considerations:
707	o The huge variability of the waste generation in concrete production, which relates to
708	the different construction methods and approaches. Whilst wastage of concrete
709	during on-site civil engineering works can be very high, precast production can
710	reduce the waste concrete by 50% to 60% (Tam et al., 2005), and a WRAP report
711	suggested even further benefits such as negligible wastage and 100% re-use of
712	material (WRAP, 2019). Due to the large variability of this parameter, its detailed
713	investigation was considered to fall outside the scope of this research.
714	o Due to the similarities in the production processes of the three concrete mixes, the
715	assessment of the emissions due to the waste production would have been more or
716	less the same when talking into account the same context, and thus it would only
717	offset the emission amounts, without affecting the comparison.
718	(4) The transportation of raw materials (Portland cement, fly ash, GGBS, waste glass) from
719	the production units to the mixing plant was modelled under some assumptions:

720 o The transportation of Portland cement was not included in the analysis. It is

721   commonly assumed that the relevant geographic market for Portland cement is

722   between 150 – 250 km. However, several precast units on mixing plants are located

723   nearby the cement production factory, since, due to the relatively low economic

724   margins of some precast concrete products, the typical market radius is about 50

725   km (Soutsos et al., 2019), and transportation costs may make the business

726   unprofitable. As this study did not focus on a specific plant but rather on a

727   comparison among concretes of different nature, it was decided not to include the

728   transportation distance in order to avoid this further complication that would have

729   affected the result variability.

730 o Similarly, the availability of fly ash and GGBS is a complex issue that cannot be

731   easily modelled, e.g. the current UK construction market sources GGBS from local

732   production, import from Europe and import from China. Furthermore, due to the

733   industrial trends in steel production and coal-fuelled power stations, future

734   scenarios and availability of GGBS and fly ash are even more uncertain. However,

735   acknowledging that fly ash and GGBS are typically sourced farther away than

736   Portland cement, a transportation distance of 100 km was included in the

737   calculation. Having neglected the transportation distance in the analysis of Portland

738   cement-based concrete, such distance of 250 km can be considered an "offset" in

739   the comparison between mixes. The "extra" 100 km added for fly ash and GGBS

740   would therefore implicitly represent a scenario where these materials are procured

741   some 350 km away from the concrete mixing plant, which is considered reasonable

742   for most of the real cases. In order to assess the impact of transportation, the

743   analysis of AABR production included a sensitivity step in which these materials

744   (including glass waste and sodium hydroxide) were transported with lorries over a

745   distance of 500 km (i.e. 750 km when considering the offset discussed for Portland

746   cement).

747

748   **CONCLUSIONS**

749   The environmental impacts associated to the use of Portland cement in concrete are a

750   growing concern worldwide. Alkali activated binders have been proposed for a low carbon,

751   low impact alternative to Portland cement. However, a debate exists on the actual

752   environmental profile of alkali activated binders, mainly due to the high impacts associated

753   with the use of chemicals for the activation. This paper described the outcomes from a

754   research investigating the environmental impacts of three compressive strength classes of

755   concretes when produced with Portland cement, with alkali activated binders using

756   commercially available chemicals, and with a novel, waste glass-based activator recently

757   described in the literature.

758   The main outcomes from the study were:

759   • The adoption of AAB concrete allows a significant reduction in several environmental

760   indicators such as global warming (63%-65% reduction), acidification potential (17%-

761   30% reduction), and terrestrial eutrophication (47%-58% reduction).

762   • The use of a waste glass-derived activators allowed a further reduction of the

763   aforementioned impacts i.e. global warming (68%-71% reduction), acidification

764   potential (29%-40% reduction), and terrestrial eutrophication (57%-63% reduction).

765   • AAB concrete production has a negative impacts on freshwater ecotoxicity,

766   particulate matter and resource depletion.

767   • The use of waste glass-based activator allowed a consistent reduction in every

768   environmental indicator when compared to the use of commercially available

769   chemicals. CO_2 eq. emissions directly allocated to the activators were found to be

770   30%-32% lower.

771   • The parameters with the highest impacts on the environmental performances of the

772   waste glass-based alkali activated concretes were the GGBS allocation, the

773   transportation of raw materials and the amount of activating powder in the mix. The

774   energy mix and the fly ash allocations were found to be insignificant, as well as

775   milling glass cullet for the production of glass powder.

776 Further research should concentrate on the optimisation of the mix proportions in order to

777 minimise the use of activators in concrete. The investigation of other Si-rich waste streams

778 for the production of waste-derived solid activator is another promising research direction.

779

784

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