The contribution of life-cycle assessment to environmentally preferable concrete mix selection for breakwater applications

Contribuição da Análise de Ciclo de Vida para informar a seleção de misturas de baixo impacto ambiental para aplicação em estruturas de quebra-mar

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Abstract

ife cycle assessment (LCA) provides a comprehensive framework for positioning low energy and global warming potential alternatives regarding Portland cement and concrete. Published LCA work on alkali-activated cements is, however, relatively limited. In this paper, we illustrate how LCA critically supports concrete technological studies in the search for low impact concrete mixes. Previous research on breakwater applications explored replacing a low-clinker Portland cement and natural aggregates with seven different alkali-activated blast furnace slag (bfs) binder systems and with coarse and granulated bfs aggregates. Its outcome suggested a sodium silicate-activated bfs formulation as the best match between concrete properties and environmental regulation compliance. To validate this outcome through LCA, our cradle to gate assessments followed ISO 14044 (INTERNATIONAL..., 2006b) and used Ecoinvent v.2.2 and CML baseline 2001 v.2.05. We adopted the 'net avoided burden approach' to handle multifunctionality intrinsic to by-product-based AAC. Whilst sodium silicate-activated mixes rivaled the reference regarding GWP, impacts in several categories were increased. LCA highlighted the implications of driving mix selection by focusing on a single environmental impact category.

Keywords: Concrete. Blast furnace slag. Breakwaters. Performance. LCA.

Resumo

A avaliação do ciclo de vida (ACV) oferece uma estrutura abrangente para comparação de impactos ambientais. Entretanto, sua aplicação em estudos sobre cimentos ativados por álcalis é ainda limitada. Neste artigo, ilustramos como ACV pode informar estudos tecnológicos com foco ambiental. Pesquisa anterior sobre estruturas de quebra-mar avaliou misturas de sete variações de ativadores alcalinos e escória de alto-forno (eaf) com agregados de eaf bruta e granulada para substituir cimento de baixo teor de clínquer e agregados naturais em concreto. Uma formulação com silicato de sódio ofereceu a melhor combinação entre propriedades do concreto e conformidade com a legislação ambiental vigente. Objetivando validar esta conclusão da perspectiva de ACV, realizamos estudos do berço ao portão (INTERNATIONAL..., 2006b), utilizando Ecoinvent v.2.2, CML baseline 2001 v.2.05, e 'abordagem do impacto líquido evitado'. Apesar da mistura com silicato de sódio ter apresentado potencial de aquecimento global equivalente ao da mistura de referência, os impactos estimados para as demais categorias aumentaram. A ACV evidenciou as implicações de se orientar a seleção de misturas unicamente por uma categoria de impacto.

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> Recebido em 28/11/16 Aceito em 24/07/17

Palavras-chave: Concreto. Escória de alto-forno. Estruturas de quebra-mar. Desempenho. ACV.

SILVA, M. G. da; GOMES, V.; SAADE, M. R. M. The contribution of life-cycle assessment to environmentally preferable concrete mix selection for breakwater applications. **Ambiente Construído**, Porto Alegre, v. 18, n. 2, p. 413-429, abr./jun. 2018.

ISSN 1678-8621 Associação Nacional de Tecnologia do Ambiente Construído.

Introduction

Clinker Portland is the main contributor to the environmental impact of typical concrete mixes. This awareness has greatly encouraged a number of research efforts, which include searching for reduced clinker content cements; alternative raw materials to replace limestone in kiln feed; and alternative fuel mixtures, as well as enhanced thermal efficiency of the kiln system. From these, replacing clinker by alternative, low impact substitutes is probably the most effective means of significantly reducing global warming potential (DAMTOFT *et al.*, 2008).

The development of new binders has clearly aimed at increasing the proportion of waste materials from other industries, while reducing the environmental impact of concrete and ideally improving its performance. Fly ash, ground granulated blast furnace slag (bfs) and silica fume are aluminosilicates included in the wide variety of waste materials used as supplementary cementitious materials (SCMs), at partial clinker replacement ranging between 10–50%. Interest has been shown to increase this replacement proportion or even create binders entirely made from waste materials (JUENGER *et al.*, 2011).

The literature often indicates sulfo-aluminate clinkers and alkali-activated cements (AAC) among most promising low impact binders, respectively for the reduced kiln temperature requirement (between 1250°C and 1350°C) as compared to that of clinker production (1450°C) (JUENGER et al., 2011), and for using alkali activation from natural, synthetic or industrial waste aluminium-silicate sources (DUXSON et al., 2007). Good mechanical and durability performances make these alternatives particularly appealing (DOUGLAS; BILODEAU; MALHOTRA, 1992; SILVA, 2006a, 2006b; JUENGER et al., 2011; JIMÉNEZ; 2011). SHI: PALOMO, Notwithstanding, few authors have quantified the environmental impact of geopolymers (DAVIDOVITS, 1993, 1999, 2009; DUXSON et al., 2007; WEIL; DOMBROWSKI; BUCHWALD, HABERT; D'ESPINOSE LACAILLERIE; ROUSSEL, 2011; MCLELLAN et al., 2011; TURNER; COLLINS, 2013; HEATH; PAINE; MCMANUS, 2014; DAVIDOVITS, 2015), and published life cycle assessment (LCA) work on AAC is relatively limited.

LCA considers potential environmental impacts of a product or system from raw material extraction to end of life, facilitating the comparison among functionally equivalent alternatives or improvement detection opportunities within specific systems. These assessments are guided by ISO 14040 (INTERNATIONAL..., 2006a) and ISO 14044 (INTERNATIONAL..., 2006b). ISO 14025 (INTERNATIONAL..., 2006c) and CSN EN 15804 (EUROPEAN..., 2012) specifically regulate Environmental Product Declarations (EPDs), which are LCA-based Type III environmental declarations adopted by various countries for standardized communication of results.

LCA provides a comprehensive framework for understanding environmental impacts and offers a powerful platform for positioning low impact alternatives regarding Portland cement and concrete. Its consolidation over the past decade has made it a valuable addition to routine laboratory and field tests. If systematically added early on, LCA can enlighten and optimize exploratory feasibility studies and considerably save time and experimental resources.

Previous laboratory and field investigations were driven by two main criteria that have historically guided the search for alternative concrete mixes low non-renewable content and global warming potential (GWP) - and covered the evaluation of mechanical and environmental properties of several AAbfs mixes. Results achieved indicated a sodium silicate-activated bfs (SSAbfs) formulation as the best match between fresh and hardened concrete properties environmental and regulation compliance for breakwater applications (SILVA, 2006a, 2006b; SILVA; SAADE; GOMES, 2017). Our aim is to verify if this conclusion would be validated by LCA outcomes, by examining the environmental implications of replacing a lowclinker Portland cement and natural aggregates with seven different alkali-activated bfs (AAbfs) binder systems and with coarse and granulated bfs. Nine "cradle to gate" indicators were used to encompass the minimum EPD structure and resource use aspects relevant to the concrete and cement industry. We also applied the 'net avoided burden (NAB)' approach to handle the multifunctionality problems intrinsic to by-product-based AAC, and used Ecoinvent v.2.2 and CML baseline 2001 v.2.05 impact assessment method to maintain comparability to our previous studies.

Previous work on the development of breakwaters

The investigation concerning the breakwater elements examined in this paper began in 2002 (SILVA, 2006a) to strategically explore alternatives for the valorization of steelmaking by-products, particularly bfs. Concrete mix-design and component development are traditionally functional

performance-oriented, based on laboratory testing of concrete specimens at different ages. To verify possible effects of by-product-based concrete components exposed to marine environments over their 'use stage', an umbrella R&D research (SILVA, 2006b) went beyond standard practice and included comprehensive field and laboratory monitoring of full-size breakwater elements, detailed in Silva, Saade and Gomes (2017). Despite its international standardization in the mid 1990s, LCA practice in Brazil was very limited at that time and therefore excluded from the initial investigation scope.

Breakwater element design

Breakwater production demands concrete mixes that combine mechanical performance with workability compatible with complex geometry molding. Design characteristics, compliance with bending and twisting internal tensions, and manufacturing constraints defined the non-reinforced concrete breakwater geometry (Figure 1). Element sizing for different design wave heights (Table 1) were calculated using the Hudson Equation (Equation 1, from the U.S. Department of the Army (1984)). This approach allowed for a factor 10+ of material reduction to produce one breakwater element.

$$W = \frac{w_r . H^3}{K_{d} \cdot (S_r - 1)^3 . cot(\alpha)}$$
 Eq. 1

Where:

W is the block's design weight, in kg;

 w_r is the block's specific weight (adopted as 2300 kg/m³);

H is the design wave height at the toe of the structure, in meters;

 K_d is a dimensionless stability coefficient (adopted as 13);

 S_r is the ratio between the densities of the block's material and that of (sea)water; and

 α is the angle of revetment with the horizontal.

Assessment at product and use stages

The experimental program was divided into three phases. Phase 1 (alkali-activator and mix selection) and Phase 2 (detailed physical and mechanical characterization) comprised the 'product stage' assessment, while Phase 3 comprised lab and field environmental characterization and biological

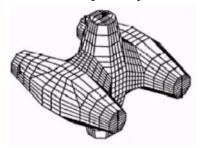
colonization to address the component 'use stage' (SILVA; SAADE; GOMES, 2017).

Firstly, a series of alkali-activators for ground bfs were investigated. Once the most promising activator and corresponding mix were selected to proceed towards a detailed investigation, a thorough physical and mechanical property assessment was carried out. In Phase 2, twelve (100x200) mm-cylindrical specimens (four specimens per testing age) were kept in a wet chamber until testing at 7, 28 or 365 days. Determination of specific gravity, capillary water absorption, shrinkage, air content, void content, modulus of elasticity, tensile strength at bending and splitting tensile strength, resistance to sulfate attack and chloride ions penetration followed procedures standardized by the Brazilian Association of Technical Standards (ABNT).

Finally, ten breakwater elements produced with the selected mix were exposed to marine environments for one year for biological colonization assessment (taxonomic level analysis and systematic ordering), XRD analysis and determination of compressive strength, specific gravity, water absorption, void content, modulus of elasticity, splitting tensile strength and resistance to chloride ions penetration samples extracted from those structures following ABNT procedures. An extra set of specimens and one full-sized breakwater were immersed for one year in a laboratory seawaterfilled tidal movement simulator for environmental characterization of water sampled from the tank. Critical water quality parameter measurements were taken at the beginning and at the end of testing, following the Standard Methods for the and of Water Wastewater Examination (AMERICAN PUBLIC HEALTH ASSOCIATION et al., 1995). Tank water pH and electric conductivity were measured weekly, from a threesample set over the first two weeks and from one sample afterwards. Results were compared to recommendations from the Brazilian National Environment Council (CONAMA) Resolution 357/2005. At the end of the immersion cycle, samples extracted from the blocks were tested in the laboratory.

Mix design followed the American Concrete Institute (ACI) method and aimed at reaching compressive strength compatible with breakwater structures, established as 46.6 MPa at 28 days (40 MPa-fck, with a 4 MPa-standard deviation). Water/binder ratio (0.45) and aggregate proportion, in mass, were kept constant and ensured comparability of the LCA results, as suggested by Habert and Ouellet-Plamondon (2016).

Figure 1 - Breakwater structure geometry





Source: Silva (2006a).

Table 1 - Breakwater structure sizing for varied design wave heights

Wave height (m)	Breakwater element height (m)
0.5	0.23
1	0.46
2	0.91
3	1.36
4	1.81
5	2.26

Source: Silva (2006a).

CP III-32 RS contained 66% of granulated bfs, in mass. The vitrification degree and refractive index of the basic granulated bfs used were 96% and 1.65, respectively. Hydrated lime CH I, gypsum (fineness below 0.075mm) and sodium silicate (silica module of 1.62 and solids content of 47.78%) were selected in previous studies (COELHO *et al.*, 2004a, 2004b, 2004c, 2005a, 2005b, 2009; SILVA, 2006a) and tested as activators. Table 2 summarizes the materials used and Table 3 shows the 28-day compressive strength values registered for the studied AAbfs mixes and corresponding descriptive statistics.

Clinker – (mix A); clinker and lime – (mixes B and C); lime and gypsum – (mix E) and sodium silicate activated slag (SSAbfs) D3 mixes did not comply with breakwaters strength requirement (Table 3). Poor performance achieved by D3 mix (3% of Na₂O) probably resulted from the limited activation capacity of the low sodium content available. From these formulations, Mix A has possibly the highest potential to be modified for increased resistance; it would not completely exclude clinker from the formulation, but could significantly reduce its use.

Distinguished compressive strength achieved by SSAbfs mixes D4 and D5 at 28 days suggested sodium silicate as the activator choice. Strength values registered for these two mixes were very similar. Mix D4 then proceeded towards environmental characterization and biological colonization analysis, due to its lowest activator content.

Environmental characterization of water sampled from D4 block immersion tank showed that cadmium, lead, copper, nickel, zinc and manganese concentrations exceeded CONAMA's limit; however only lead, nickel and manganese showed a growth rate compared to the experiment. Water pH variation was not significant over time. The initial pH growth trend observed was probably connected to measurement uncertainties rather than to dissolution of substances in the monitored breakwaters. Concerning biological colonization, taxa found in Mix D4 elements after a one-year immersion cycle characterizes them as 'artificial reefs'.

Multifunctionality modelling challenge in life cycle assessment

For the sake of efficiency and practicality, LCA is divided into four main stages:

- (a) goal and scope definition, in which the analysis purposes and comprehension are defined;
- (b) inventory analysis, which compiles and quantifies inputs and outputs for a product throughout its life cycle;
- (c) impact assessment, aimed at understanding and evaluating the magnitude and significance of the potential environmental impacts for a product system throughout the life cycle of the product; and
- (d) interpretation, when the results of the evaluation are presented.

Table 2 - Materials used in the different AAbfs mixes, in mass (per 40kg of binders and water/binder ratio 0.45) - added water accounted for water in the sodium silicate solution - mixes compliant with breakwater compressive strength requirements are highlighted

Mix	CP III - 32 RS	Groun d slag	Lime	Sodium Silicate	Gypsum	Granulated bfs (fine aggregate)	Coarse bfs size B1 (9,5- 19 mm)	Coarse bfs size B2 (19- 25 mm)	Water
Clinker-activated bfs									
A	20.00	20.00	-	-	-	41.76	24.76	37.00	18.00
Clinker/lime-ac	Clinker/lime-activated bfs								<u>.</u>
В	9.41	27.39	3.20	-	-	41.76	24.76	37.00	18.00
С	11.76	28.24	4.00	-	1	41.76	24.76	37.00	19.80
Sodium Silicate	Sodium Silicate-activated bfs								<u>.</u>
D3 (3% Na ₂ O)	-	31.41	2.00	6.59	1	41.76	24.76	37.00	14.55
D4 (4% Na ₂ O)	-	28.58	2.00	9.42		41.76	24.76	37.00	12.60
D5 (5% Na ₂ O)	-	27.01	2.00	10.99	•	41.76	24.76	37.00	12.26
Lime/gypsum-activated bfs									
Е	-	36.80	0.80	=.	2.40	41.76	24.76	37.00	18.00

Source: Silva, Saade and Gomes (2017).

Table 3 - 28-day compressive strength registered for the AAbfs concrete mixes, in MPa. Variance analysis ANOVA (at 5% significance) showed significant differences among the averages (p-value = 0.000) - the Duncan test (also at 5% significance) identified five homogeneous groups [E; D3; B+C; A and D4+D5]

AAbfs concrete Mixes	Activator	Compressive strength at 28 days (MPa)	n	Min (MPa)	Max (MPa)	Standard deviation (MPa)
A	Clinker	36.1	4	1.05	1.05	1.05
В	Clinker-lime	25.1	5	0.91	0.91	0.91
C	Clinker-lime	24.8	5	0.41	0.41	0.41
D3	Sodium silicate (3% Na ₂ O)	22.1	5	0.82	0.82	0.82
D4	Sodium silicate (4% Na ₂ O)	48.4	5	3.35	3.35	3.35
D5	Sodium silicate (5% Na ₂ O)	49.7	4	1.00	1.00	1.00
Е	Lime-gypsum	16.0	5	0.57	0.57	0.57

Source: Silva, Saade and Gomes (2017).

Meaningful application of this technique relies on a number of crucial methodological choices regarding, for example, system boundaries (TILLMAN et al., 1994), functional units, characterization factors (HUIJBREGTS, 1998), (allocation) methods to solve multifunctionality problems (BELTRAN et al., 2016), and selection of impact assessment methods background/foreground data sources. Assumptions to support such choices join data inaccuracy and model imprecisions, limitations simplifications, spatial and temporal variability, among others, to become unavoidable uncertainty sources in LCAs (BJÖRKLUND, 2002; REAP et al., 2008, BELTRAN et al., 2016).

The vague nature of ISO 14044 (INTERNATIONAL..., 2006b) proposed guidelines, combined with a growing desire to follow a 'life cycle approach', without a clear notion of what it means, has led to confusion

regarding LCA's suitability and strategic contribution (CURRAN, 2014). As a result, it is common to find conflicting LCA studies due to flawed methodological approaches, transparency and lack of consensus in critical practitioners' choices.

From those choices, impact distribution in multifunctional processes is one of LCA's most controversial methodological issues, which highly influences a study's final result. A multifunctional process is an activity that fulfills more than one function: a production process generating more than one product, a waste management process with more than one waste flow, or a recycling process providing waste management and material production (EKVALL; FINNVEDEN, 2001). Most common clinker replacement alternatives used as SCMs or in AAC consist of multifunctional process outputs. To solve the issue of sharing and distributing material and energy flows across

ISO 14044 multiple functions, 2006b) (INTERNATIONAL..., suggests stepwise procedure. First, allocation should be avoided "wherever possible", either by dividing the multifunctional process into sub-processes or by expanding the product system to include the additional functions related to the by-products. Second, when allocation cannot be avoided, the system inputs and outputs should be divided based on the "underlying physical relationships between them". Finally, if those physical relationships are not easily identified, other relationships between the products and functions, such as their economic value, should be used.

The European Waste Framework Directive (EUROPEAN..., 2008) is a major turning point regarding LCA approaches in clinker replacement investigations. In early studies, industrial 'waste' inputs – for example, SCMs like bfs - were typically considered as impact-free consequences of those processes (SAADE; SILVA; GOMES, 2015). By establishing 'end-of-waste criteria', which specify when certain waste ceases to be waste and obtains a status of a by-product (or a secondary raw material), the EU Directive changed the impact accountancy dynamics and incremented an already vigorous discussion on impact distribution methods and criteria.

A previous literature review (SAADE; SILVA; GOMES, 2015) showed a lack of consensus concerning appropriateness of allocation methods; insufficient arguments to favor a single distribution method; and sector-specific approaches, such as Habert's (2013) proposal for the cement industry. On one hand, most by-product-based SCMs are produced in large amounts. Mass allocation, therefore, attributes to them considerable portions of the corresponding multifunctional process impacts (CHEN et al., 2010). On the other hand, economic allocation considers market information, inherently flawed to account for ecological goods, within an assessment that is supposed to represent environmental implications based on physical interactions (PELLETIER; TYEDMERS, 2011).

Allocation simply transports part of the multifunctional process' impact to the product system that incorporates the by-product. This fails to consider the benefit of returning material content to the economic cycle via a recycling operation instead of depleting virgin raw material and further effects on its scarcity. Avoidance of by-product's end of life potential impacts is not accounted for either.

The 'avoided burden' approach includes this benefit in product life cycle modeling. However, it fails to distribute it fairly. The avoided impact is discounted only from the multifunctional process that generated the by-product, and results not adapted to the waste user, since the benefit is attributed solely to the waste generator (CHEN *et al.*, 2010). The industry that uses the by-product not only does not receive the benefit, but also absorbs its processing impacts.

Aiming at improving multifunctional modelling, whilst better distributing the benefits across the players in the recycling chain dynamics, Saade, Silva and Gomes (2016) proposed the 'net avoided burden (NAB) approach'. Firstly, the NAB approach computes into the joint system (e.g. steel/cement) all loads that are caused or prevented by raw material replacement (Equation 2 and Figure 2). Secondly, it identifies the environmental challenges faced by the partnering industries and distributes impact/benefits by assigning to each partner the (positive or negative) consequences of avoiding those given problems.

$$\begin{split} I_{net} &= I_{subs.prod.} - [I_{byprod.process.} - I_{byprod.EOL\ loads} + ... + \\ I_{other\ loads}] \end{split}$$
 Eq. 2

Where:

 I_{net} is the net avoided burden;

I_{subs.prod.} is the avoided impact, associated to the substituted product;

 $I_{byprod.process.}$ is the by-product processing impact;

 $I_{byprod.EOLloads}$ is the impact related to by-product end-of-life (EOL) - if not used; and

 $I_{other loads}$ are any other loads related to by-product use that may arise in different cases (e.g. transport loads).

Methodological approach

Reference mix design

For the LCA study, we introduced a reference mix (REF mix), designed based on mix design curves that relate compressive strength to water/binder ratio, cement content and total coarse aggregates. The estimated compressive strength at 28 days (52 MPa) complies with the established criterion for breakwater application (40 MPa-fck, with a 4 MPastandard deviation). The Brazilian equivalent to OPC (CP I) corresponds to less than 4% of national production and requires special ordering. Thus, REFmix reflects current regional availability and used reduced-clinker CP III-32 RS cement (~30% clinker) and natural fine and coarse aggregates (1:2.25:2.25:0.45, in mass of cement: gravel B1: gravel B2: water). Water/binder ratio mirrored the AAbfs mixes (Table 2).

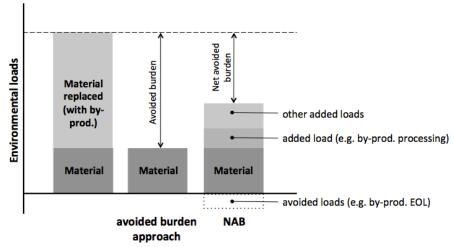


Figure 2 - 'Net avoided burden (NAB)' vs. traditional avoided burden approach

Source: Saade, Silva and Gomes (2016).

LCA of breakwater concrete mixes

System boundaries and functional unit

For selecting concrete mix designs appropriate for breakwater application, we are interested in the impacts *embodied* in the components ('product' stage). In such cases, 'cradle to gate' assessments are the most adequate, and were performed consistently with published LCA studies on concrete mix designs (HABERT; OUELLET-PLAMONDON, 2016). Such boundary choices limit the system boundaries to the production of components and the appropriate mix design, and is justified as all concretes have similar life cycle ends (from gate-to-grave), regardless of their composition (HABERT, 2013).

Our cradle to gate LCAs followed ISO 14044 (INTERNATIONAL..., 2006b) guidelines and adopted 'one unit of characteristic compressive strength (1MPa)' as the functional unit. Bfs multifunctionality modelling within the joint steel/cement production process (Figure 3) used the NAB approach (SAADE; SILVA; GOMES, 2016): the problem avoided by the steelmaking industry is the final disposal of slags; while the cement and concrete industry avoided increased resource depletion for extracting, producing or processing energy and carbon-intensive clinker and natural aggregates.

LCI and LCIA indicator calculations

Table 4 shows the data sources used for mix modelling. When no specific, local data were available, the Ecoinvent v. 2.2 database (GOEDKOOP; SCHRYVER; OELE, 2008) was used by replacing the original energy grid by the Brazilian matrix.

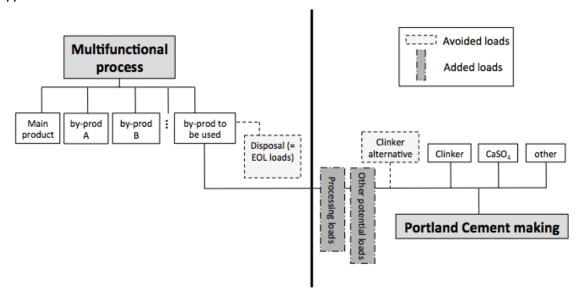
For each concrete mix, nine impact indicators were calculated per functional unit:

- (a) renewable energy (E_{ren}, in MJ/MPa);
- (b) non-renewable energy (E_{nren}, in MJ/MPa);
- (c) global warming potential (GWP, in kg CO_{2e}/MPa);
- (d) acidification potential (AP, in kg SO_{2e}/MPa);
- (e) eutrophication potential (EP, in kg SO_{2e}/MPa);
- (f) photochemical ozone creation potential (POCP, in kg C₂H_{4e}/MPa);
- (g) ozone layer depletion potential (ODP, in kg CFC-11_e/MPa);
- (h) blue water footprint (bWF, in m³/MPa); and
- (i) non-renewable raw material consumption (NRC, in kg/MPa).

The first seven indicators comprise the minimum environmental impact categories structure for environmental product declarations (EPD) defined by CEN EN 15804 (EUROPEAN..., 2012). The last two indicators, although rarely documented in EPDs, report information relevant to construction materials in general, and concrete, in particular.

Blue water footprint (bWF) and non-renewable raw material (NRc) were calculated directly from the product's life cycle inventory by filtering and computing identified inputs. Primary renewable (E_{ren}) and non-renewable (E_{nren}) energy and the remaining indicators advanced to impact analysis procedure using, respectively, the Cumulative Energy Demand (CED) and CML baseline 2001 v.2.05 methods. Both methods are included in the SimaPro 7.3 platform impact assessment tool list.

Figure 3 - Joint system boundary for bfs modeling according to the 'net avoided burden (NAB)' approach



Source: Saade, Silva and Gomes (2016).

Table 4 - Data sources for breakwater LCA modeling

Material	Source
Blast furnace slag (bfs)	Steelmaking industry reports ³
CP III-32 cement	Silva (2006b)
Gypsum	Raw material database from SimaPro 7.3 ^b
Hydrated lime	
Water	Ecoinvent v.2.2
Sodium silicate	Econivent v.2.2
Granite ^c	

Note:

The blue water footprint (bWF) was calculated as proposed by Hoekstra *et al.* (2011), then divided by the functional unit (Equation 3).

$$bWF = (\sum_{i=1}^{n} bW_i \text{ per functional unit})$$
 Eq. 3 Where:

bWF is the blue water footprint, in m³/MPa;

 bW_i is the quantity of each (surface or underground) blue water input identified in the product's life cycle inventory; and

n is the number of water source inputs identified in the product's life cycle inventory.

Analogously, the calculation of non-renewable raw material (NRc) per functional unit (Equation 4) added mineral non-renewable raw material inputs identified in the product's life cycle inventory.

$$NRc = (\sum_{i=1}^{n} NR_i \text{ per functional unit})$$
 Eq. 4

Where:

NRc is the consumption of non-renewable raw material, in kg/MPa;

 NR_i is non-renewable primary material input identified in the life cycle inventory; and

n is the number of non-renewable raw materials inputs identified.

The calculations of primary renewable (E_{ren}) and non-renewable (E_{nren}) energy per functional unit followed Equation 6. SimaPro enables automatic calculation by selecting CED as the impact assessment method.

$$E = (\sum_{i=1}^{n} E_i \text{ per functional unit})$$
 Eq. 5

Where:

E is the primary embodied energy of concrete mixtures, in MJ/MPa, divided into renewable (E_{ren}) and non-renewable (E_{nren}) sources;

^a data for steel slags production was extracted from reports directed to the local environmental agency;

^b SimaPro 7.3 has a built-in database for raw material without any industrial processing; and

^c granite gravel data was adapted from Ecoinvent basaltic gravel data.

 E_i is the (renewable/non-renewable) primary energy input identified in the life cycle inventory; and

n is the number of primary energy inputs identified.

Potentials for global warming, acidification, eutrophication, photochemical ozone creation and ozone layer depletion were obtained by multiplying the mass of each substance by its equivalence factor - concerning the reference substance - provided by CML baseline 2001 v.2.05. Global warming potential (GWP), expressed as the mass of carbon dioxide equivalent per functional unit (CO_{2e}/MPa), was calculated using Equation 6.

$$GWP = (\sum_{i=1}^{n} GWP_i \times m_i \text{ per functional unit})$$

Eq. 6

Where:

GWP represents the global warming potential, in kg of CO_{2e}/MPa;

 GWP_i is the CO_{2e} -equivalence factor for each greenhouse gas (GHG) considered by CML 2001 v.2.05;

 m_i is the mass of each corresponding GHG emission; and

n is the number of GHG considered.

The acidification potential (AP) is expressed in mass of sulfur dioxide-equivalent per functional unit (SO_{2e}/MPa), as in Equation 7.

$$AP = (\sum_{i=1}^{n} AP_i \times m_i \text{ per functional unit})$$

Eq. 7

Where:

AP represents the acidification potential, in kg of SO_{2e}/Mpa;

 AP_i is the SO₂-equivalence factor for each acid producer considered by CML 2001 v.2.05;

 m_i is the mass of the acid producer; and

n is the number of acid producers considered.

The eutrophication potential (EP) is expressed in mass of phosphate-equivalent per functional unit (PO_{4e}/MPa), as in Equation 8.

$$EP = (\sum_{i=1}^{n} EP_i \times m_i per functional unit)$$

Eq. 8

Where:

EP represents the eutrophication potential, in kg of PO_{4e}/MPa:

 EP_i is the PO₄-equivalence factor of each eutrophication substance considered by CML 2001 v.2.05;

 m_i is the mass of the eutrophication substance; and n is the number of eutrophication substances considered.

The potential for forming atmospheric oxidants or POCP (Photochemical Ozone Creation potential) of a hydrocarbon is a relative measure of how much the ozone concentration measured at a single location varies if its emission is altered by the same amount as that of ethylene, set as a reference. PCOP is expressed in mass of ethylene-equivalent per functional unit (C_2H_{4e}/MPa) (Equation +).

$$POCP = (\sum_{i=1}^{n} POCP_i \times m_i \ per \ functional \ unit)$$
 Eq. 9

Where

POCP represents the photochemical ozone generation potential, in kg of C₂H_{4e}/MPa;

 $POCP_i$ is the C₂H₄-equivalence factor of each reactive substance considered by CML 2001 v.2.05;

 m_i is the mass of the reactive substance; and

n is the number of reactive substances considered.

The ozone depletion potential (ODP) calculation follows Equation 10. The World Meteorological Organization (WMO) characterization model expresses ODP of each gas in mass of chlorofluorocarbon-equivalent per functional unit (CFC-11_e/MPa), as in Equation 10.

$$ODP = (\sum_{i=1}^{n} ODP_i \times m_i \text{ per functional unit})$$

Eq. 10

Where:

ODP represents the depletion potential of the ozone layer, in kg of CFC-11_e/MPa;

 ODP_i is the CFC-11-equivalence factor of each gas considered by CML 2001 v.2.05;

 m_i is the mass of the gas; and

n is the number of gases considered.

Contribution and sensitivity analyses

Contribution and sensitivity analyses were carried out to:

- (a) identify the key contributors to the impact values estimated; and
- (b) to assess how sensitive those values were to different impact distribution methods.

Both analyses were applied to the impact categories calculated through CML 2001 v.2.05 (AP, EP, GWP, ODP and POCP).

Results and discussion

Our LCA results clearly distinguished breakwater-compliant mixes from non-compliant ones (Table 3). Besides the obvious reduction in non-renewable content (NRc), basically three performance profiles were noticed for mixes unsuitable for breakwater application (Figure 4). Mixes A, B and C performed very homogeneously and slightly (AP, EP, GWP, ODP and Eren) or noticeably better (POCP, NRc and bWF) than the reference. Mix E clearly outperformed POCP, NRc, bWF and E_{nren}. Despite registering the lowest compressive strength in the group, its impact/strength ratio suggests potential for low strength concrete applications. Finally, Mix D3 showed a completely distinct profile shape,

which is adherent to those exhibited by strength-compliant SSAbfs mixes D4 and D5.

Only SSAbfs mixes D4 and D5 (Figure 5) were technically suitable for breakwater application. The 1% decrease in activator content in D4 had a negligible effect and both mixes showed equivalent environmental profiles, except for the higher water volume embodied in D5. The technological assessment which originally suggested mix D4 as the most suitable environmental alternative to the low-clinker REFmix was confirmed by most indicators analyzed: GWP, AP, EP, ODP, POCP and NRc results were similar to the reference, whilst energy and bWF were clearly affected by the sodium-silicate use.

Figure 4 - Cradle to gate results for breakwater non-compliant mixes A, B, C, E and D3 (compressive strength ≤40 MPa-fck, with a 4 MPa-standard deviation), relatively to REFmix

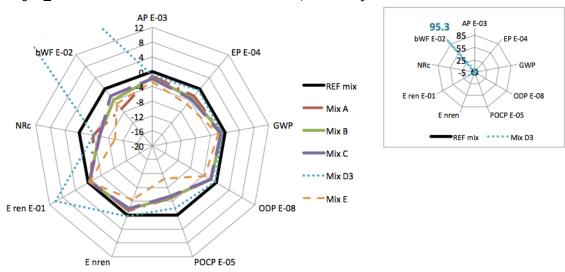
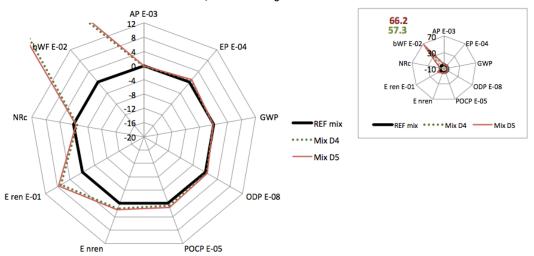


Figure 5 - Cradle to gate results for breakwater-compliant mixes D4 and D5 (compressive strength \geq 40 MPa-fck, with a 4 MPa-standard deviation), concerning REFmix



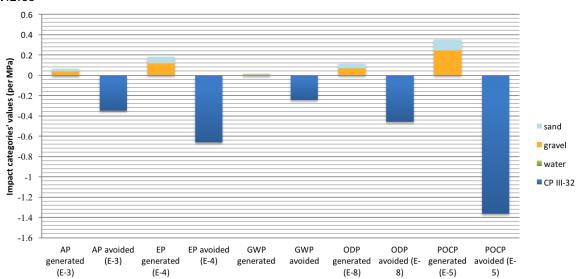
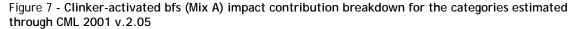
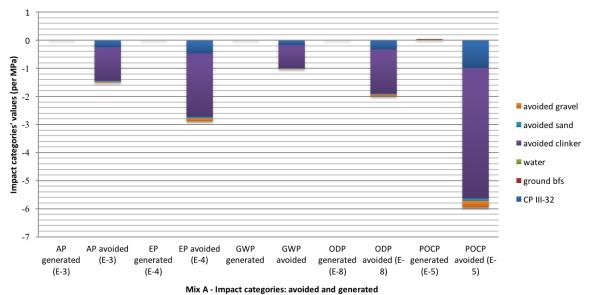


Figure 6 - REFmix impact contribution breakdown for the categories estimated through CML 2001 v.2.05

REFMix - Impact categories: avoided and generated



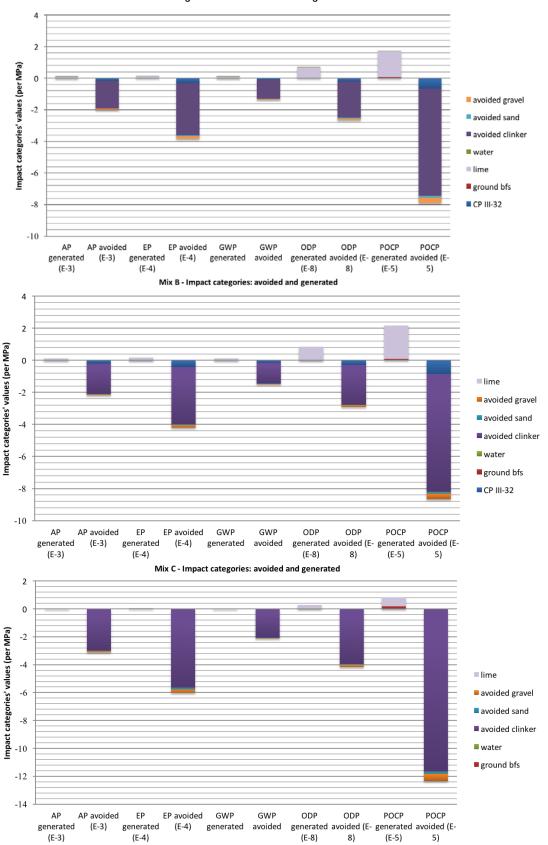


This A mipute categories avoided and generate

The contribution analysis showed that, for the REFmix (Figure 6), generated impacts referred to coarse and fine aggregate, while bfs substitution for clinker in CP III-32 RS cement manufacturing avoided over three times the loads created in all categories estimated through CML 2001 v.2.05.

Non-SSAbfs mixes had either a negligible generated impact (Mix A, in Figure 7) or the created

ODP and POCP loads were mostly lime-driven (Mixes B, C and E, in Figure 8). In all cases, avoided impacts particularly due to bfs substitution for clinker and – to a lesser extent - activator (CP III-32 RS) and aggregates substantially prevailed and more than sufficed to neutralize the activators' effects.



Mix E - Impact categories: avoided and generated

Figure 8 - Clinker-and-lime (Mix B and C) and lime-and-gypsum-activated bfs (Mix E) impact contribution breakdown for the categories estimated through CML 2001 v.2.05

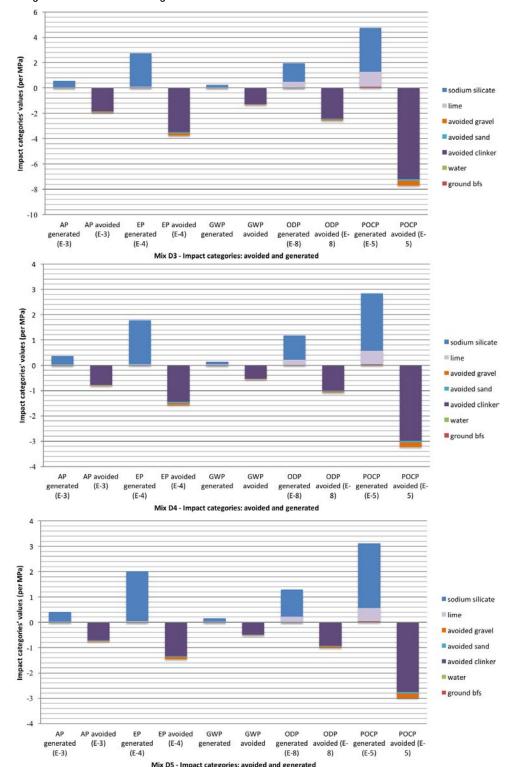


Figure 9 - Sodium silicate activated bfs (SSAbfs) Mixes D3, D4 and D5 impact contribution breakdown for the categories estimated through CML 2001 v.2.05

The contribution analysis revealed a similar impact pattern for all SSAbfs (Figure 9), regardless of their compliance with breakwater applications. EP, ODP and POCP show the most significant impacts per functional unit, and were clearly dominated by

sodium silicate (EP, ODP and PCOP) and lime (ODP and PCOP). In all cases, environmental load avoidance due to bfs substitution by clinker and – to a lesser extent – natural aggregates was considerable, but at different levels depending on

the mixtures. For Mix D3, avoided impacts more than sufficed to counterbalance the activators' effects in all categories. For Mix D4, generated impacts were overcome for AP, GWP and PCOP, while Mix D5 showed environmental favourable balances for AP and GWP only. Overall, sodium silicate, lime, bfs and clinker data should be as accurate as possible, since they were the most relevant contributors to impacts registered across the studied mixes.

The sensitivity analysis (Figure 10) elucidated how results varied for the impact distribution approaches considered. The reduced by-product content in REFmix explains the slight variation detected for all categories but POCP. The bfs generation process is less GWP and ODP intensive. These categories were, therefore, consistently the least sensitive to environmental load partition variation for all the studied mixes. Nevertheless, the AP, EP and POCP indicators did not follow such a pattern, due to the high bfs content (Mixes E and D3) and/or chemical activators' - clinker (Mix A), sodium silicate (Mixes D4 and D5) and lime (Mixes B and C) - in these categories, particularly in the latter.

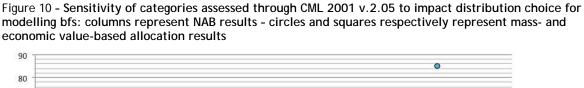
Although the results of the SSbfs mixes D4 and D5 for GWP (and ODP) were close to the results of the REFmix, the focus restricted to only these two categories would clearly influence decision making, which confirms the need to consider the full spectrum of LCA categories and to conduct a careful sensitivity analysis when exploring potential alternative concrete formulations.

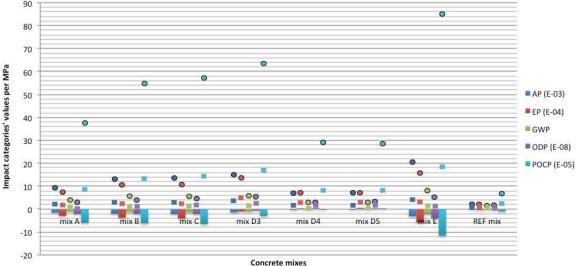
Conclusions

LCA provides a broad environmental setting to fully understand the environmental implications of using clinker substitutes. This paper illustrates how LCA can provide information about concrete technological studies in the search for low impact concrete mixes.

Previous laboratory and field investigations suggested sodium silicate-activated bfs Mix D4 as the best match between fresh and hardened concrete properties and environmental regulation compliance for breakwater applications. Our LCA study showed the most relevant contributors to impacts generated or avoided, for which corresponding data quality is key and should be strategically pursued. The technological assessment outcome which suggested Mix D4 as the most suitable choice would only be corroborated from the GWP perspective. Our study also confirmed the risks of shaping environmentally-oriented mix selection choices solely after GWP.

The consolidation of LCA over the past decade now enables it to be included in routine laboratory and field tests. Regardless of the chosen research path and approach, systematically adding LCA as a screening tool early on in feasibility studies can save considerable time and experimental resources, while holistically developing sustainable materials and components.





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Acknowledgement

The authors acknowledge FAPESP, AMT, FINEP and CNPq for their generous support.

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