Cement 13 (2023) 100077

Contents lists available at ScienceDirect

Cement

journal homepage: www.sciencedirect.com/journal/cement

Sustainability of concretes with binary and ternary blended cements considering performance parameters

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ARTICLE INFO

Keywords:
Cement
Concrete
Supplementary cementitious materials
Durability
Life cycle assessment
Sustainability indices

ABSTRACT

This paper examines the sustainability of cementitious materials and concrete. Although the environmental impact of these materials is often evaluated based on their CO_2 emissions per ton of cement or m^3 of concrete, incorporating performance parameters into sustainability indices is crucial for a more comprehensive assessment. This study evaluates the sustainability of concretes with and without supplementary cementitious materials (SCM), considering compressive strength and durability performance as performance parameters. Results show that the most sustainable concretes have the highest compressive strength and best durability performance. Furthermore, the importance of using locally available materials is highlighted, as transporting SCM over long distances can outweigh the benefits of using them as a replacement for Portland cement.

Introduction

The sustainability of cementitious materials can be analyzed from different standpoints in the cement or concrete life cycle, such as from cradle-to-gate, cradle-to-grave, or cradle-to-cradle. The most elementary calculations can be performed considering the unit binder mass or the sum of the impacts of the component materials per unit volume. It is interesting to evaluate the impact when using, for example, supplementary cementitious materials (SCM) or recycled aggregates. However, to fully assess the impact of modifying the binder or concrete composition, it is essential to consider not only the emissions per unit of mass or volume but to include the effect of changing the required material volume when the concrete's compressive strength is modified, as well as the environmental and economic impact if the service life is reduced. Several evaluations have been proposed to perform a Life Cycle Assessment (LCA) considering functional performance parameters or changing the functional unit from unit mass or volume to structural elements or complete structures, or considering a unit of functional performance.

For example, Damineli et al. propose considering the parameters "binder intensity" and "carbon intensity", taking compressive strength as the performance parameter [1]; Gettu et al. propose evaluating the "apathy towards sustainability" of concrete by considering CO_2 equivalent emissions, energy use, and chloride diffusion and carbonation coefficients in the design factors, integrating durability in the evaluation [2]; Miller et al. propose considering four functional units, three of which consider the effect of concrete mix composition variation on the volume of materials required [3]. Panesar et al. also propose four functional units of incremental complexity involving not only mix composition but also compressive strength and durability [4].

Studies analyzing the sustainability of reinforced concrete elements, such as columns [5], beams and columns [3], and slabs [6,7], built with concretes of varying compressive strength and CO_2 emissions per m^3, have shown that the lower the volume of the structural element, the lower the CO_2 emissions, even when the emissions per m^3 of concrete are higher. In this regard, the Building code requirements for structural concrete (ACI 318) includes initiatives to reduce the carbon footprint of concrete, among which is the use of higher compressive strength concretes [8].

However, studies that include durability parameters in the sustainability analysis of concrete are not widely available. Durability is crucial to evaluate within the LCA of concrete since poorer material durability leads to higher maintenance costs [9,10]. On the other hand, a shorter service life implies an increase in the renovation rate and the generation of construction and demolition waste (CDW) [11,12].

Moreover, most of the binder and concrete LCAs have been carried out without considering the emissions related to material transportation. Therefore, it is urgently necessary to conduct studies on

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https://doi.org/10.1016/j.cement.2023.100077
Received 31 October 2022; Received in revised form 7 July 2023; Accepted 13 July 2023
Available online 14 July 2023

locally available materials and to investigate the effect of transporting long distances to incorporate materials that provide better performance.

This paper aims to evaluate the sustainability of concretes with and without supplementary cementitious material (SCM) available in Argentina, considering performance parameters (compressive strength and durability, through the calculation of the indicators proposed by Damineli et al. [1] and Gettu et al. [2], as well as to assess the impact of transporting the SCM from its extraction or production site to the cement plants distributed throughout the country.

Materials

An ordinary Portland cement (OPC) (CEM I 42.5 N, according to EN 197–1), two binary blended cements with calcined clays, and two ternary blended cements with limestone filler and calcined illitic clay/ natural pozzolan were used.

The binary blended cements were produced with 25% of replacement of OPC by two types of calcined clays from different regions of Argentina and 35% of replacement of OPC by ground granulated blast furnace slag (GGBS) from a steel factory located in San Nicolás, province of Buenos Aires, Argentina. The calcined low-grade kaolinitic clay (LKC) was obtained from the calcination of impure kaolin from the Province of Santa Cruz, Patagonia, Argentina. The calcined red illitic clay (RIC) was obtained from the calcination of illitic clay from the Province of Buenos Aires. The calcination temperature was chosen based on the main clay mineral to guarantee the complete dehydroxylation and formation of amorphous material. To achieve this, LKC was calcined at 750 °C [13, 14], and RIC and OIC were calcined at 950 °C [15,16].

The ternary blended cements were produced using a combination of limestone filler (LF) and a further calcined illitic clay (OIC) from a different quarry of RIC, one natural pozzolan from the Province of Mendoza, Argentina, or GGBS. The calcined orange illitic clay (OIC) was calcined at 950 °C and produced at an industrial scale in a cement plant in Olavarría, Province of Buenos Aires. The natural pozzolan (NP) was ground jointly with the LF to be used in the blended cement. The replacement level of ternary blended cements ranged from 22 to 28%.

Water-to-binder (*w/b*) ratios between 0.4 and 0.6 were used for concrete production. Natural silica sand from the Parana river, Argentina, was used as fine aggregate (FAg), and crushed granite from a quarry at Olavarría, Province of Buenos Aires, as coarse aggregate (CAg). Different chemical admixtures were employed to achieve the desired slump (8-12 cm). A polycarboxylate-based superplasticizer (BASF, Trostberg, Germany) was used in M1-M3, a multifunctional mid-range water reduction (MIRA 353, GCP Applied Technology) was used in M4-M9, and M10-M12 were produced without chemical admixture. The concrete mix composition is shown in Table 1. The notation (*w/b*-TBC-%SCM) indicates the *w/b* ratio, the total binder content (TBC), and the percentage of OPC replacement by SCM by mass.

Methodology

Compressive strength

Concretes were characterized according to their compressive strength at 28 and 90 days of curing. Cylindrical concrete specimens with a diameter of 10 cm and height of 20 cm were used to measure the compressive strength in accordance with ASTM C39. The specimens were demolded after 24 h and cured in water saturated with lime. Table 1 shows the compressive strength results.

Durability parameters

The durability of concrete was measured by assessing the resistance to chloride migration and natural carbonation in a rural environment.

Chloride migration test

Chloride migration (NT Build 492) was determined on concretes with calcined clays and NP, using cylinders of a diameter of 10 cm diameter and a height of 5 cm, cured for 28 and 90 days. After the chloride exposure, the specimens were split and the depth of chloride ingress was measured by spraying them with silver nitrate solution. The chloride migration coefficient (D_{nssm}) was determined according to Eq. (1), where T is the average temperature of the anolyte solution during the test (°C), L is the thickness of the specimen (mm), U is the applied voltage (V), t is the test duration (hour), and x_d is the average value of the penetration depth (mm):

$$D_{nssm} = \frac{0.0239\ (273+T)\ L}{(U-2)\ t}\left(x_d - 0.0238\sqrt{\frac{(273+T)\ L\ x_d}{U-2}}\right) \quad (1)$$

Natural carbonation test

The carbonation coefficient (k_c) was calculated using Eq. (2) [17], where x_c is the carbonation depth (mm) at a given time, and t is the given

Table 1

Concrete mix proportions, compressive strength, and durability-related parameters.

Mix ID	Notation (w/b-TBC-%SCM)	Quantity, kg/m^3 FAg	Compressive strength, MPa CA	Chloride migration coefficient, (D_{nssm}), x10^{-12} m^2/s) Water	Carbonation coefficient, mm/years$^{0.5}$					
					28 days	90 days	28 days	90 days	7 days	28 days
M1	0.50–350–0%SCM	807	1050	175	32.4	41.0	12.7	8.5	1.3	1.0
M2	0.50–350–25%LKC	788	1050	175	37.4	46.6	1.6	1.5	3.2	2.5
M3	0.50–350–25%RIC	805	1050	175	28.3	38.4	21.8	5.2	3.5	3.2
M4	0.4–410–20%LF-7.5% OIC	746	1075	165	47.1	54.4	6.4	5.5	0.8	0.6
M5	0.5–330–20%LF-7.5% OIC	816	1075	165	38.5	41.1	15.0	6.8	2.0	1.3
M6	0.6–270–20%LF-7.5% OIC	873	1075	162	28.0	32.7	26.8	–	4.4	3.2
M7	0.4–410–12.4%LF-12.6%NP	732	1075	165	45.0	51.9	6.0	4.1	0.7	0.5
M8	0.5–330–12.4%LF-12.6%NP	802	1075	165	35.4	38.7	14.7	5.6	1.4	0.8
M9	0.6–270–12.4%LF-12.6%NP	864	1075	162	23.1	27.5	25.6	–	3.2	2.9
M10	0.5–360–6%LF-22% GGBS	900	900	180	32.8	40.9	-	–	1.3	1.6
M11	0.5–360–11%LF-11% GGBS	900	900	180	32.6	-	–	–	0.6	0.6
M12	0.5–352–20%GGBS	840	1000	176	34.1	41.5	-	–	1.6	1.0

time (years).

$$k_c = \frac{x_C}{\sqrt{t}} \tag{2}$$

The carbonation depth was measured after 36 months of exposition in concrete specimens cured for 7 and 28 days, corresponding to good and excellent curing, respectively. Good curing is close to curing a cast-in-place concrete structure, while excellent curing corresponds a precast element. The specimens were prismatic with 100 mm height and 70 mm side length and were placed in a rural area (CO_2 concentration of 350–400 ppm), with the molding and lateral faces exposed to air and rain. The carbonation depth was determined on a sawn section of the concrete prisms using a phenolphthalein solution as a pH indicator. The average of 11 values between 10 and 60 mm from the molding face was taken from the carbonation depth, which corresponds to the maximum penetration depth.

Sustainability assessment

The Life Cycle Assessment (LCA) was performed following the guidelines of ISO 14,040 and ISO 14,044 [18,19], and the free software OpenLCA was used to compile the Life Cycle Inventory (LCI) and perform the impact evaluation.

The aim of the LCA is to determine the CO_2 equivalent emissions and embodied energy per unit of mass and volume of material. Two functional units were set to examine the sustainability of cements and concretes. The first functional unit was one ton of binder for primary analysis, and the second was one cubic meter of concrete to consider performance parameters. The LCA was carried out from cradle to gate. Therefore, the stages of casting, construction, use, deconstruction/demolition of the structure, and recycling and final disposal of concrete were not included.

Part of the database used in the LCA was generated based on local data because Argentina's energy matrix differs from that of other countries, and natural gas being a significant fraction of the fuels burned in the clinker kilns. To validate the database, it was compared with the ICE database (Inventory of Carbon and Energy) [20]. CO_2 equivalent emissions and embodied energy of the ordinary Portland cement, calcined illitic clays and coarse aggregates were estimated using data from a cement plant and a quarry located in Olavarría, Buenos Aires, Argentina. Bibliographical data was consulted for natural silica sand, calcined kaolinitic clay, limestone filler, natural pozzolan, ground blast furnace slag, tap water, and chemical admixture. The sources are referenced in Table 2.

The Life Cycle Impact Assessment (LCIA) was conducted considering two impact categories: the CO_2 equivalent emissions (ECO_2eq) and the embodied energy per ton of material or m^3 of concrete. ECO_2eq/t material was estimated using Eq. (3), proposed by the Intergovernmental Panel on Climate Change (IPCC) [21]. There, the ECO_2eq are the CO_2 equivalent emissions, CO_2, CH_4, and N_2O correspond to the amount of carbon dioxide, methane gas, and nitrous oxide emitted during the process under study.

$$ECO_2eq\ [kg] = CO_2[kg] + 25 * CH_4 + 298 * N_2O \tag{3}$$

The calculation considered emissions from the use of electrical power (based on the Argentinian energy matrix), fossil fuel burned during the production of the materials, CO_2 released from the decarbonization of limestone, and those related to the transportation of the materials within the production/extraction plants. However, emissions from the transportation of materials from production/extraction plants to the ready-mix concrete plant or casting site were not included. Table 2 shows the concrete component materials' emission and energy use factors. The emission factors of ECO_2eq and embodied energy were not accounted in the calculation due to the dispersion of the values depending on the type of chemical admixture and the lack of local information for the estimation according to the own database.

A customized method was developed in OpenLCA to calculate the unitary ECO_2eq and energy consumption of blended cements and concretes based on the emission and embodied energy factors presented in Table 2.

Table 2
Emission and energy factors for the concrete compound materials.

Material	ECO CO_2eq (kg)/t of material	Embodied energy (MJ)/ t of material
Ordinary Portland Cement (OPC)	896	4864
Crushing coarse aggregate (CA)	46	532
Natural silica sand (FAg)*	14	356
Low-grade calcined kaolinitic clay**	193	2481
Calcined illitic clay	258	3754
Limestone filler (LF)***	35	832
Natural pozzolan (NP)***	25	610
Granulated blast furnace slag (GBFS)***	47	1124
Tap water****	9	205

Value taken from *[22], **[23], ***[24], ****[25].

Unitary ECO_2eq per ton of material and concrete

ECO_2eq per ton of binder was estimated considering the OPC content and the type and ratio of SCM used (Tables 1 and 2). Additionally, ECO_2eq per m^3 of concrete was calculated for each mix by considering their proportions.

Influence of SCM transportation on the ECO_2eq of blended cements

Although the construction industry generally employs local materials due to the large volume consumed for housing and infrastructure construction, the recommendations for using SCM are often based on the mechanical and durability properties they provide, regardless of the location of the extraction site and the transport distance to the market.

To analyze the impact of transporting SCM from the extraction site to different regions of the country, an additional ECO_2eq was quantified for each concrete. For M1, which does not have SCM, a transport distance of 0 km is considered. For LF, a distance of 0 km is also assumed since it is extracted from the Portland cement plant's quarry. For M2 to M12, only the transport of the required amount of SCM (LKC, RIC, OIC, GGBS) per m^3 of concrete is considered. For example, for M2 and M3, it is considered the transport of 87.50 kg of LKC and RIC, respectively, and for M4, that of 30.75 kg of OIC.

Given the territorial extension of Argentina and the geographic distribution of cement plants and SCM extraction sites, four distance intervals are proposed to evaluate the influence of transportation. For each interval, the average distance between the SCM extraction site and the cement plants was calculated: 333 km is adopted for <500 km, 1100 km for 501–1500 km, 1948 km for 1501-2500 km, and 3105 km for >2500 km, which is the distance for transporting LKC from the south of the country to the northernmost cement plant.

Trucking is the only mode of transport considered due to the poor railroad network connection in the country, and an emission factor of 0.124 kg ECO_2eq/t*km is applied [26].

Afterward, the impact assessment is made based on four different indicators:

Energy and carbon intensity parameters

The measurement of ECO_2eq in a unit of mass or volume of concrete is limited, as it does not involve any mechanical or durability properties of concrete. To provide a more comprehensive assessment of sustainability, indices proposed by Damineli et al. [1] and Gettu et al. [2] have been used to measure the CO_2 and energy intensity of concrete, respectively.

The carbon intensity (*ci*) can be calculated according to Eq. (4), where ECO_2eq represents the emissions associated with producing one m^3 of concrete and p is the analyzed performance [1]. In this study, ci is determined by using p as the compressive strength (cs) at 28 and 90 days of curing (ci_{cs}).

$$ci = \frac{ECO_2eq}{p} \quad (4)$$

Additionally, the energy intensity (ei_{cs}) is studied at 28 and 90 days of curing using Eq. (5). The embodied energy of concrete is a crucial parameter as clinker, and some artificial pozzolans are produced mainly using thermal energy, which requires a large amount of fossil fuels [27]. Thus, minimizing ei_{cs} would lead to further environmental benefits, such as reducing global warming and environmental pollution [28].

$$ei = \frac{ECO_2eq}{cs} \quad (5)$$

Sustainability assessment based on durability-related parameters

Gettu et al. [2] proposed a decision support framework considering three parameters: embodied energy, compressive strength, ECO_2eq/m^3 of concrete, along with chloride diffusion and carbonation coefficients. This decision support framework involves two indices: the ei_{cs} and the *A-indices*. To calculate ei_{cs}, they suggest considering the compressive strength at 365 days, as it is more representative of concrete's mechanical and durability performance during its service life than at earlier ages [2]. However, as data on the compressive strength at 365 days of concretes under study is not available, $ei_{cs,90}$ is adopted for this paper.

The *A-indices* combine CO_2 emissions with concrete durability and are defined as the ECO_2eq/m^3 of concrete divided by a material durability parameter. In this framework, if a concrete with a higher *A-index* is preferred, it indicates "apathy" towards sustainability [2]. The indices they propose are the *A-indices* for exposure to chlorides and carbonation, as these are the conditions that lead to corrosion and can be correlated to service life. On the other hand, to avoid introducing structural design considerations, which could narrow the sustainability evaluation based on durability parameters, and to ensure that the *A-indices* reflect only the concrete properties, the calculation is based only on material characteristics [2].

Therefore, they propose two factors that represent the resistance off the concrete to chloride ingress (F_{chlor}, Eq. (6)) and carbonation (F_{carb}, Eq. (7)). Both parameters are correlated to the rate of progression of chloride and carbonation fronts within the concrete and, therefore, reflect the service life of the reinforcement until the onset of corrosion. Gettu et al. chose the numerators of the Eqs. (6) and 7 to give values of the order of unity to F_{chlor} and F_{carb} [2].

$$F_{chlor} = exp\left(\frac{10^{-6}}{\sqrt{D_{cl}}}\right) \quad (6)$$

$$F_{carb} = \left(\frac{5}{k_c}\right)^2 \quad (7)$$

There D_{cl} is the chloride diffusion coefficient (m^2/s), and k_c is the carbonation coefficient ($mm/year^{0.5}$). D_{cl} can be estimated from the D_{nssm} following the procedure of fib Bulletin 34 [2,29] (Eq. (8)).

$$D_{cl} = k_e * D_{nssm} \quad (8)$$

Here, k_e is the environmental transfer variable (Eq. (9)), and it is estimated considering the standard temperature, T_{ref} [K] (taken as 293.15 K), and the ambient air temperature, T_{real} [K] (taken as 287.15).

$$k_e = exp\left(4800 * \left[\frac{1}{T_{ref}} - \frac{1}{T_{real}}\right]\right) \quad (9)$$

Thus, the chloride diffusion coefficient is determined as D_{cl}=0.71*D_{nssm}

Finally, *A-indices* for chloride exposure and carbonation are obtained with Eq. (10) and Eq. (11), respectively:

$$Ai_{chlor} = \frac{ECO_2eq\ per\ m^3\ of\ concrete}{F_{chlor}} \quad (10)$$

$$Ai_{carb} = \frac{ECO_2eq\ per\ m^3\ of\ concrete}{F_{carb}} \quad (11)$$

Table 3 presents the ECO_2eq and embodied energy per m^3 of concrete, $ei_{cs,90}$, chloride and carbonation resistance factors and *A-indices*.

Results

Concrete characterization

The concretes with $w/b = 0.50$ exhibited a compressive strength of 34±4 MPa, with the minimum and maximum values recorded for M3 (28.3 MPa) and M5 (38.5 MPa), respectively. On the other hand, the compressive strength of the concretes with a $w/b = 0.4$ was higher (46.0 ± 1.0 MPa), which can be attributed to their lower porosity and the higher TBC. In contrast, the concrete mixes with a $w/b = 0.60$ had the lowest compressive strength values (25.6 ± 2.5 MPa) due to the increased porosity of the material and the lower TBC (Table 1).

Furthermore, the compressive strength of all concretes increased between 28 and 90 days to varying extents depending on the type of binder used. Notably, the concretes made with binders with highly active SCM, such as LKC, RIC, NP, and GGBS, exhibited high increases in compressive strength (22 to 36%). Conversely, the concretes containing LF showed the lowest increases in compressive strength (9 to 19%).

Durability-related parameters

Chloride migration coefficient

The resistance of concrete to chloride ingress is related to the pore size and pore network connectivity [30–33] and to the ability of the hydrated phases to bind chlorides, either physically (adsorption) or chemically (by Friedel's salt formation) [30,34–37].

The results of the chloride migration test (Table 1) revealed that increasing the w/b ratio for similar concretes (M4<M5<M6 and M7<M8<M9) resulted in an increase in D_{nssm}, with a 3.2 times increase in both cases. Conversely, extending the curing time from 28 to 90 days reduced D_{nssm} for all concretes.

Notably, a more pronounced reduction was observed for M3 (with 25% RIC) and M7-M9 (with 12.6% NP) as compared to M4-M6 (with 20% LF, hence the maximum LF content of all concretes). This reduction was attributed to the pore size reduction and increased formation of hydrated phases during the pozzolanic reaction of RIC and NP, which contributed to the physical binding of chlorides [35,38,39]. These effects were absent when LF was used as a replacement.

Carbonation coefficient

Table 1 presents the carbonation coefficients (k_c) of concretes that were cured for 7 and 28 days. The results show that k_c ranged from 0.6 to 4.4 $mm/year^{0.5}$ for concretes cured for 7 days and from 0.5 to 3.2 $mm/year^{0.5}$ for those cured for 28 days.

Porosity is the primary factor that governs primarily by porosity [40,41]. This study shows the influence of porosity by comparing M4 to M6 and M7 to M9 concretes. An increase in the w/b ratio from 0.40 to 0.60 resulted in an approximately fivefold increase in k_c. Furthermore, prolonged curing decreased k_c in all concretes.

Nevertheless, the type and amount of carbonatable material significantly affect the carbonation rate. This is because the reaction of CO_2 with the hydrated products modifies the pore structure over time [41]. Increased availability of $Ca(OH)_2$ favors the precipitation of $CaCO_3$, which leads to the segmentation of pore structure and a decrease in the carbonation rate. On the other hand, in the presence of less $Ca(OH)_2$, CO_2 reacts with the AFm phases and the C-S-H, causing carbonation shrinkage [41]. This increases the connectivity of the pore structure and

Table 3
ECO_2eq and embodied energy per m^3 of concrete, $ei_{cs,90}$, chloride, and carbonation resistance factors and *A-indices*.

Mix ID	ECO_2eq (kg)/ m^3 concrete	Embodied energy (MJ)/m^3 concrete	Energy intensity, $ei_{cs,90}$ (MJ/m^3)/ MPa	Chloride resistance factor, F_{chlor}			Carbonation resistance factor, F_{carb}			Ai_{chlor}	Ai_{carb}
				28 days	90 days	7 days	28 days	28 days	90 days	7 days	28 days
M1	399.8	2376.4	58.0	1.4	1.5	15.5	23.1	286.5	266.1	25.8	17.3
M2	350.2	2223.5	47.7	2.6	2.6	2.4	3.9	137.0	132.9	145.2	90.4
M3	355.8	2334.9	60.9	1.3	1.7	2.1	2.4	276.0	211.5	173.4	147.6
M4	378.6	2379.1	43.7	1.6	1.7	36.3	82.6	236.6	228.7	10.4	4.6
M5	322.0	2049.4	49.9	1.4	1.6	6.6	14.1	236.9	204.2	49.0	22.8
M6	279.5	1801.5	55.1	1.3	–	1.3	2.4	222.2	–	217.4	115.9
M7	380.6	2321.2	44.7	1.6	1.8	59.2	108.5	234.6	211.2	6.4	3.5
M8	323.5	2002.8	51.8	1.4	1.7	12.4	40.1	237.5	195.5	26.1	8.1
M9	280.7	1763.4	64.1	1.3	–	2.5	3.0	222.0	–	111.4	92.5
M10	322.6	2008.2	49.1	–	–	15.7	10.0	–	–	20.5	32.2
M11	341.7	2088.3	64.1	–	–	63.0	63.0	–	–	5.4	5.4
M12	350.5	2153.0	51.9	–	–	10.0	27.7	–	–	35.0	12.7

leads to a relatively high carbonation rate over time [42,43]. This study observed this effect by comparing concretes with active SCM, low CaO content, and $w/b = 0.50$ (M2 and M3) with respect to M1, containing 100% OPC as binder and $w/b = 0.50$.

Sustainability assessment

Unitary ECO_2eq per ton of material and concrete

Fig. 1 shows the ECO_2eq of binders used in the different concrete mixes. The results show that OPC (M1) had the highest ECO_2eq (895.7 kg ECO_2eq/t binder). Blended cements (M2-M12) had a lower ECO_2eq/t binder ranging from 770.3 to 689.9 kg ECO_2eq/t cement, reducing the ECO_2eq of the blended cements by 14 to 23% with respect to the OPC.

The reduction in ECO_2eq was found to be dependent on the replacement level and type of SCM used. The higher the replacement level, the greater the reduction. Additionally, the embodied energy of SCM was also found to influence the ECO_2eq of the binders. For example, calcined illitic clays were calcined at a high temperature using fossil fuels and ground to provide pozzolanic properties, so the ECO_2eq of this type of SCM is higher than for LF, which only requires grinding.

Fig. 2 shows the ECO_2eq (rhombus) and embodied energy (circles), both measured per m^3 of concrete. The parameters exhibit similar behavior, indicating that a significant proportion of concrete's ECO_2eq is attributed to its embodied energy, which arises from the burning of fossil fuels and the generation of electricity. ECO_2eq ranged from 279.5 kg ECO_2eq/m^3 (M6) to 399.8 kg ECO_2eq/m^3 (M1), while embodied energy ranged from 1763 MJ/m^3 (M9) to 2376 MJ/m^3 (M1).

The concrete with the highest ECO_2eq and embodied energy was M1, made with a TBC of 350 kg/m^3 of concrete and OPC only – the binder with the highest ECO_2eq/t. Among concretes made with blended cements (M2-M12), it is possible to achieve reductions in ECO_2eq between 5 and 30% with respect to M1, depending mainly on the TBC per m^3 and the ECO_2eq of blended cement. The lowest ECO_2eq and embodied energy per m^3 were obtained for M6 (279.5 kg of ECO_2eq/m^3 and 1801.5 MJ/m^3) and M9 (280.7 kg ECO_2eq/m^3 and 1763.4 MJ/m^3) due to their low TBC (270 kg of binder/m^3), the reduced ECO_2eq of the binders with respect to OPC, and the fact that 74.6% of the ECO_2eq of concrete is associated with binder production [44].

In contrast, M4 and M7 have the highest ECO_2eq/m^3 among those made with blended cements. The reduction of ECO_2eq/m^3 of concrete is only 5% compared to M1, associated with their high TBC (410 kg of binder/m^3). Despite the 22% reduction in ECO_2eq of blended cements with respect to OPC, the additional 60 kg of cement/m^3 of concrete significantly raises the ECO_2eq of the concrete.

Therefore, and in agreement with the literature [5,22,45], the ECO_2eq of a concrete mix is primarily determined by the binder used and the TBC. The lower the ECO_2eq/t of cement, the lower the ECO_2eq/m^3 of concrete, and the lower the TBC per m^3 of concrete, the lower the ECO_2eq/m^3 of concrete.

Influence of SCM transportation on the ECO_2eq of blended cements

Argentina is a vast country spanning over 2791,820 km^2 with a maximum width of 1408 km and a distance of 3694 km between its North and South points [46]. Given these distances, it is essential to consider the impact of transporting building materials. To assess whether it is necessary to transport large volumes of materials to provide better-performing materials or to produce concretes with local materials and improve mechanical and durability performance by means such as w/b ratio reduction and curing assurance, it is necessary to examine mechanical and durability performance in conjunction the impact of transportation.

Fig. 3 displays a map of Argentina, showing the location of the

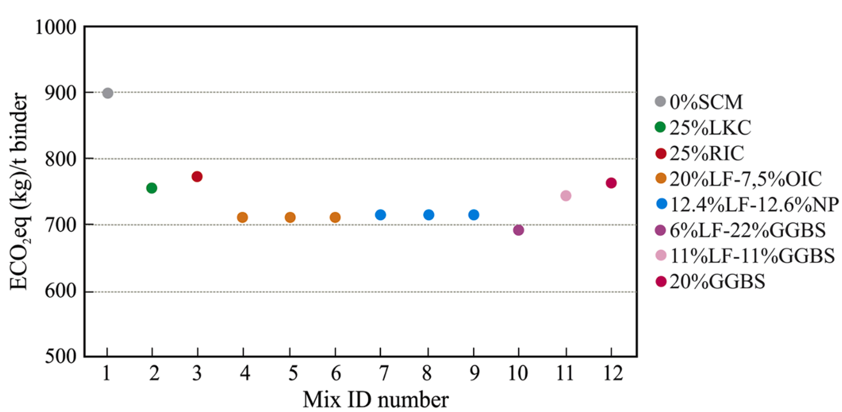

Fig. 1. ECO_2eq (kg)/ton of binder.

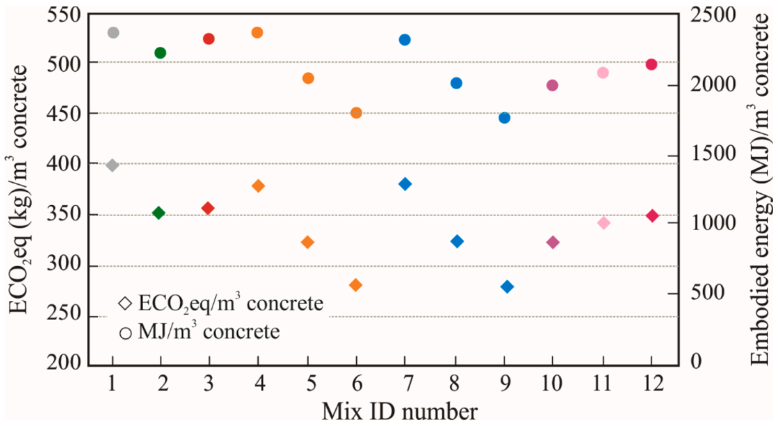

Fig. 2. ECO_2eq (kg) and embodied energy (MJ) per m^3 of concrete.

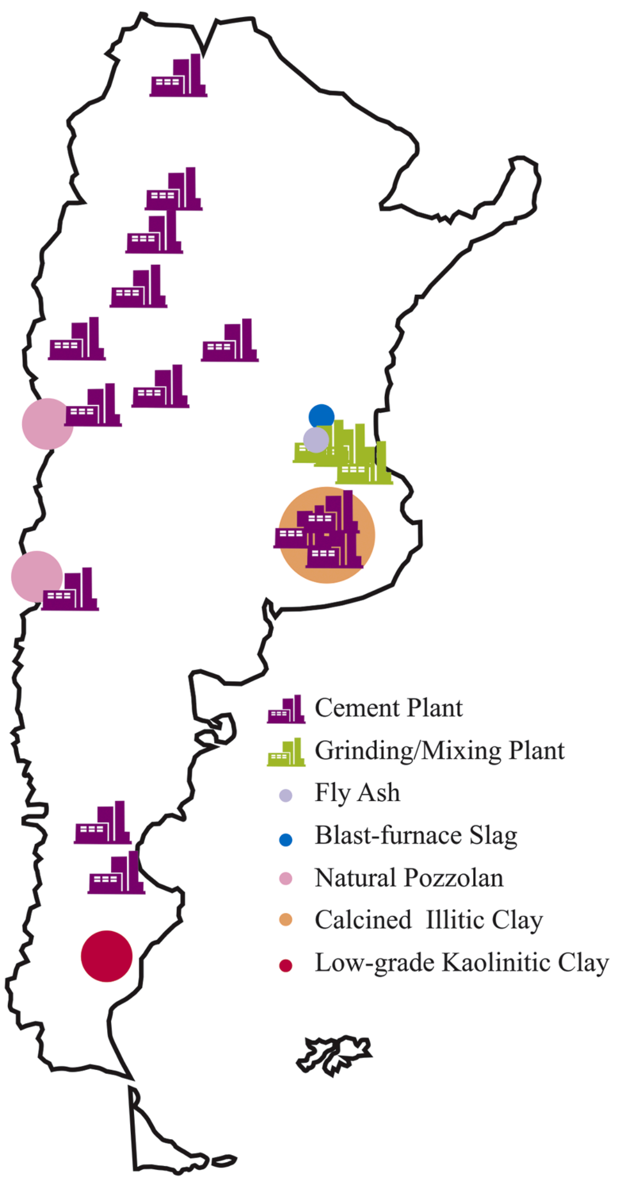

Fig. 3. Map of the Argentinian territory and geographical location of cement plants and SCM's extraction sites.

cement plants (purple factory), the mixing/grinding plants (green factory), where cement/SCM are mixed or grounded with the cement, and different SCM sources. Cement plants are spread across the country, and SCM sources are geographically scattered. In many cases, they are distant from large urban centers. For instance, FA (light purple circle) and GGBS (light blue circle) are close to the Metropolitan Region of Buenos Aires, where 31% of the country's population resides. However, they are not abundantly available in the country, limiting their share of the Portland cement market [44]. In contrast, illitic clays (light orange circle) are widely available in the Buenos Aires Province [47], and their deposits are typically located near cement plants [39]. For this reason, a ternary cement with calcined illite clay and LF has been marketed in the country since 2019 [48,49].

In addition, NP (light pink circle) is located in the Andean region near cement plants in Zapala, Mendoza, and San Luis. These plants are near to urban centers such as Neuquén, Mendoza, and San Luis. LKC (dark salmon circle), on the other hand, is only available in the southern part of Argentina (Patagonia). Patagonia is sparsely populated, and only 5% of the country's Portland cement is marketed there [50]. The minimum transportation distance between cement plants based in Patagonia and urban centers in the central region of the country is 1856 km. As a result, the market for LKC in the construction industry is limited.

Fig. 4 shows the increase in ECO_2eq/m^3 of concrete for the different mixes studied due to the transport of SCM. The analysis reveals that:

- For distances less than 500 km, the increase in ECO_2eq/m^3 is only between 0.40 to 1.12%.
- For distances between 501 and 1500 km, the increase in ECO_2eq/ m^3 ranges from 1.01 to 5.00%.
- For distances between 1501 and 2500 km, the increase in ECO_2eq/ m^3 ranges from 1.55 to 6.94%.
- For distances longer than 2500 km, the increase in ECO_2eq/m^3 could be as high as 9.62%.

The results indicate that the impact of transport on ECO_2eq/m^3 is negligible for short distances, even when SCM replaces 25% of OPC. The impact remains low for distances ranging from 501 to 1500 km, which covers transportation from the farthest east and west points of the country, especially for replacement levels below 20%. For distances between 1501 and 2500 km, which would involve transportation from the central region of the country to the Pania, the northern part of the country, or vice versa, the increase in ECO_2eq/m^3 of concrete exceeds 5% for mixes for a replacement of more than 20%, making it more significant. Distances exceeding 2500 km would only be feasible under an unrealistic scenario of transporting LKC from the southern region to the northernmost cement plant in the province of Jujuy. Nonetheless, this shows that the increase in ECO_2/m^3 of concrete due to the transportation of the SCM becomes considerably significant when non-locally available SCM are used.

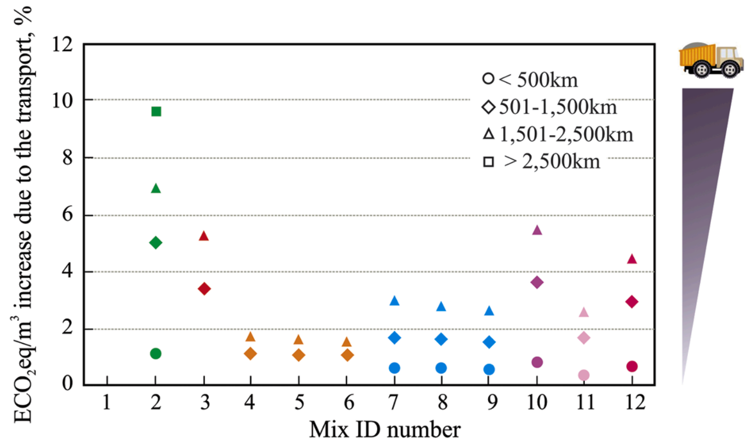

Fig. 4. Increase in ECO_2eq (kg) due to transport of the SCM.

Sustainability assessment based on energy and carbon intensity parameters

Fig. 5 illustrates the energy intensity (ei_{cs}) (Fig. 5a) and carbon intensity (ci_{cs}) (Fig. 5b) of concretes that were cured for 28 (◀) and 90 (▶) days. The behavior of ei_{cs} and ci_{cs} differs from that of the embodied energy and ECO_2eq per m^3 of concrete due to the influence of compressive strength.

For a design compressive strength of 28 days, M3 exhibited the highest $ei_{cs,28}$, and $ci_{cs,28}$. Although M3 allowed an ~2% reduction in embodied energy and a ~6% reduction in ECO_2eq per m^3 of concrete compared to M1, the 28 days compressive strength was relatively low, with $ei_{cs,28}$, and $ci_{cs,28}$ being ~13% and ~2% higher than M1, respectively. Conversely, the minimum $ei_{cs,28}$ (~31% lower than for M1), and $ci_{cs,28}$ (~35% lower than that of M1) were both achieved for M4, which exhibited almost the same embodied energy and an ECO_2eq ~5% lower than the corresponding to M1. This is due to the relatively high compressive strength of M4 (Table 1), which improved concrete efficiency according to mechanical strength criteria.

The use of SCM, mainly when they are non-active or when the reaction occurs over a relatively long term (28 days or more), can impair the design compressive strength up to 28 due to the dilution effect [49, 51]. M9 showed the lowest embodied energy, which was ~26% lower than that of M1, and one of the lowest ECO_2eq/m^3, which was ~30% lower than that of M1. However, the $ei_{cs,28}$ was ~4% higher than that corresponding to M1, and the $ci_{cs,28}$ was only 1% lower than that of M1.

At 90 days, the compressive strength gain of concrete containing 20–25% reactive SCM, such as M2, M3, M10, and M12, resulted in a reduction in both $ei_{cs,90}$ and $ci_{cs,90}$. M3 reduced ei_{cs} and ci_{cs} by 26% between 28 and 90 days, achieving an $ei_{cs,90}$ that was 5% higher, and a $ci_{cs,90}$ that was 5% lower than M1. Likewise, M2, M10, and M12 reduced ei_{cs} and ci_{cs} by 18–20% between 28 and 90 days. For the latter, $ei_{cs,90}$ was 11–18% lower than that of M1, and $ci_{cs,90}$ was 13–27% lower than that of M1. In contrast, M4-M6, with 20% replacement of OPC by LF and 7.5% of pozzolanic SCM, showed a reduced ei_{cs} and ci_{cs} of 6–13% between 28 and 90 days. These results support the notion that the use of active SCM in concrete leads to the most significant in the long run, as their reactivity contributes to mechanical performance at a slow rate.

Sustainability assessment based on durability-related parameters

Table 3 shows the Ai_{chlor} coefficients of concretes M1 to M9 cured for both 28 and 90 days. $Ai_{chlor,28}$ ranged from 137.0 to 286.5, and $Ai_{chlor,90}$ was between 132.9 and 266.1, with M2 exhibiting the lowest indices and M1 the highest. Although M1 had a lower D_{nssm} at 28 days than M3, M6, and M9 (Table 1), the ECO_2eq/m^3 of concretes with SCM were between 12 and 30% lower than that of M1 (Fig. 2). Additionally, M2 had the lowest D_{nssm} and allowed a 12% reduction in ECO_2eq/m^3 of concrete compared to M1.

Fig. 6 presents the support framework for sustainability assessment for the service limited by chloride ingress scenario for concretes cured for 28 (Fig. 6a) and 90 days (Fig. 6b), according to the approach proposed by Gettu et al. [2]. The concretes closest to the lower left corner are considered the most sustainable. Thus, M2 was identified as the most sustainable concrete in the scenario of service limited by chloride ingress. The concrete with LKC had a higher Al_2O_3 content than the other concretes and showed reactivity before 28 days, improving the chloride binding and reducing the pore size and connectivity [39].

Towards 90 days of curing, concretes with SCM showed a clear

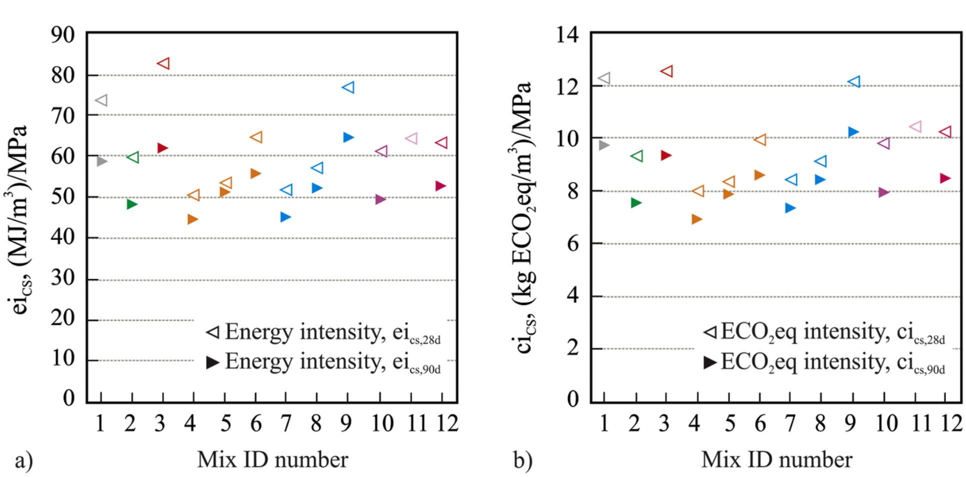

Fig. 5. a) Energy intensity and b) carbon intensity of concretes cured for 28 and 90 days.

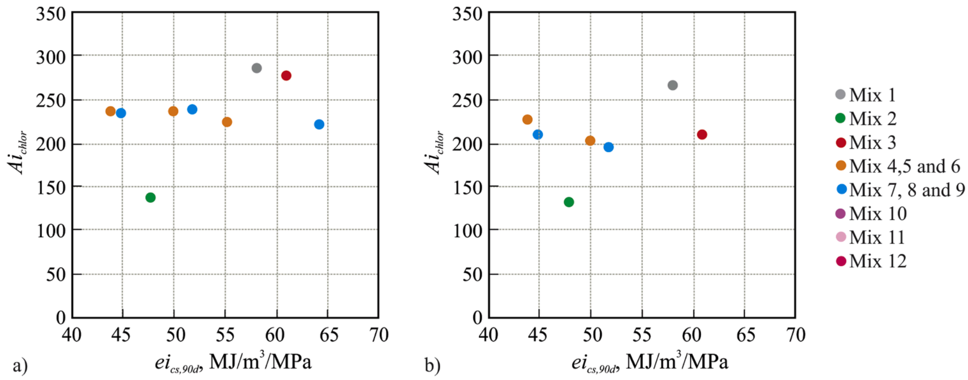

Fig. 6. Decision framework based on the *A-index* and energy intensity ($ei_{cs,90}$) for chloride exposure of concretes cured for a) 28 days and b) 90 days.

improvement. The hydration progress increased the volume of hydrated phases, improved the ability of physical chloride combination, and reduced the D_{nssm}. Therefore, concretes with SCM subjected to prolonged curing were identified as the most sustainable concretes for the scenario of service limited by chloride exposure.

The Ai_{carb} coefficients of concretes cured for 7 and 28 days are given in Table 3. They exhibited widely dissimilar values, depending on the type of binder and the w/b ratio of the concrete. The $Ai_{carb,7}$ ranged between 5.4 (M11) and 217.4 (M6), with the lowest values obtained for concretes with a w/b ratio of 0.40–0.50 and binders with more CaO content. The highest values were held by concretes with $w/b = 0.60$ and active SCM with low CaO content. The relatively high $Ai_{carb,7}$ of M2 and M3 concretes is attributed to their high k_c (Table 1), whereas for M6 and M9, it was attributed to their porosity. Comparable concretes (M4-M5 and M7-M8) with a w/b ratio of 0.40 or 0.50 had markedly lower $Ai_{carb,7}$, explained by the increased porosity that facilitated CO_2 ingress and led to high k_c.

At 28 days, Ai_{carb} decreased, particularly for M2, M3, M6, and M9, which can be attributed to a reduction in pore size and connectivity resulting from the pozzolanic reaction. Although this reaction reduces the availability of $Ca(OH)_2$ for the formation of $CaCO_3$ and subsequent segmentation of the pore structure, it lowers the k_c.

Fig. 7 presents the support framework for sustainability assessment for the service limited by carbonation scenario for concretes cured for 7 (Fig. 7a) and 28 days (Fig. 7b). Concretes with w/b ratio ≤ 0.50 and with SCM that do not consume $Ca(OH)_2$ or with moderate CaO content (GGBS) were found to be the most sustainable in this scenario.

Particularly, M7 and M8 were among the most sustainable concretes in this scenario, even though NP reduced the $Ca(OH)_2$ availability by the pozzolanic reaction. However, since the NP content was lower than 15%, and natural pozzolans are typically SCM of moderate reactivity, the $Ca(OH)_2$ that does not react with NP contributes to reducing the k_c and, consequently, Ai_{carb}.

Discussion

When evaluating the sustainability of binders and concretes from different perspectives, some differences appear in the impact assessment.

Firstly, both the ECO_2eq of binders and concretes are mainly influenced by the amount of clinker per ton of binder or per m^3 of concrete. It is well known that ECO_2eq decreases when SCM replaces OPC. The reduction depends on the replacement level and the type of SCM used. The SCM included in this study have different preparation or production processes. Although the calcination temperatures of LKC, RIC, and OIC, especially for RIC and OIC, they are still lower than the temperature for clinker production, and the clays do not release CO_2 by decarbonization. These factors significantly decrease the ECO_2eq with respect to Portland clinker.

At this level of analysis, increasing the replacement level of OPC by SCM is the most sustainable approach. Nevertheless, it usually leads to a lower compressive strength at 28 days due to the dilution effect and the pozzolanic reaction that occurs at long term. It results in increased energy intensity (ei_{cs}) and carbon intensity (ci_{cs}) (Fig. 5). When the reduction in compressive strength is significant, blended concretes with lower ECO_2eq/m^3 than the corresponding OPC concrete exhibit higher ei_{cs} and ci_{cs} than that without SCM.

This calls for a debate on two aspects regarding the compressive strength of concretes with and without SCM. The first point has to do with the conformity age of concrete. Binders with active SCM develop good compressive strength beyond 28 days, achieving similar or higher compressive strength than the corresponding OPC concrete at 90 days. The Concrete Center published a guidance for specifying sustainable concretes in 2020 [52]. In terms of compressive strength, this guidance recommends not over-specifying strength and considering extending the conformity age from 28 to 56 days whenever possible. It involves a collaborative design project between the concrete designers and the

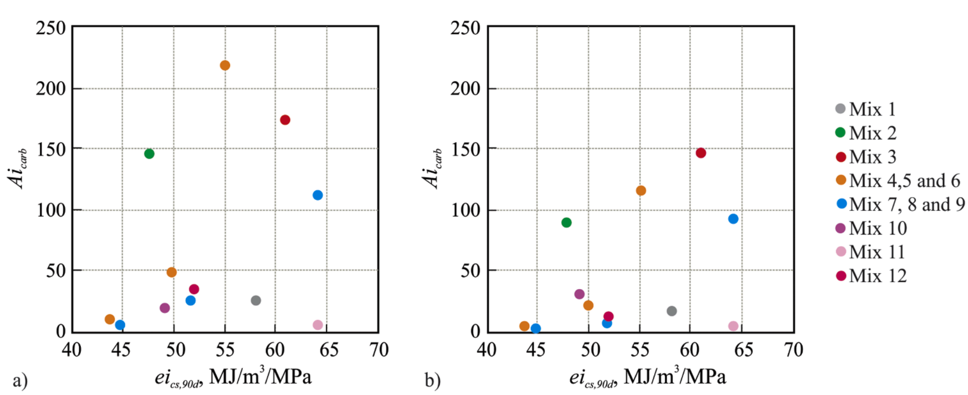

Fig. 7. Decision framework based on the *A-index* and energy intensity ($ei_{cs,90}$) for carbonation of concretes cured for a) 7 days and b) 28 days.

contractors, which would allow raising the replacement level of OPC by SCM without compromising their performance.

On the other hand, the American Concrete Institute (ACI) included sustainability principles in its code to design reinforced concrete structures [8]. This code recommends using higher-strength concrete, which usually has higher ECO_2eq/m^3 due to the greater total binder content (TBC) [5,6,52,53]. Nevertheless, the reduction in the volume of the structural elements obtained by increasing the strength offsets the increase in ECO_2eq/m^3 of concrete [5,6].

Furthermore, the results indicate that prolonging the curing time of concretes with blended cements leads to an improvement in sustainability by reducing the coefficients of chloride migration and carbonation. Notably, this does not necessarily entail using wet curing for a longer period, which would increase on-site water consumption and the environmental impact of concrete. Various techniques are currently available to extend the curing time and guarantee good mechanical properties and durability performance of the coating concrete. Nonetheless, further research is required to determine the optimal on-site curing time/technique to achieve adequate durability comparable to the laboratory-measured properties of long curing times.

In addition, it was observed that concretes with higher compressive strength showed lower k_c and D_{nssm} values (Table 1), as both durability and strength parameters are closely related to pore size [35,40,41,54]. Thus, increasing the concrete compressive strength can lead to the production of more sustainable concretes, which requires an increase in the TBC or extension of curing (or the conformity age) when active SCM are used.

The study also showed that concretes with GGBS and LKC exhibited the best performance in terms of ECO_2eq and energy intensity and sustainability for the scenario of service limited by durability from 28 days (Figs. 5, 6, and 7). This can be attributed to their reactivity starting earlier than 28 days, thereby reducing pore size and connectivity, increasing compressive strength, and improving resistance to CO_2 and chloride ions ingress. In contrast, concretes with calcined illitic clays and NP show poor performance, attributable to their slower pozzolanic reaction.

However, it is crucial to note that promoting the use of binders with GGBS and LKC and avoiding using those with calcined illitic clays (RIC or OIC) or NP may be a wrong conclusion. GGBS and LKC have limitations regarding their use. For instance, GGBS has limited availability due to the steel production process and the growth of steel production from scrap [55]. Additionally, LKC has severe workability problems due to its high-water consumption [56], which may limit its use for general-use concrete. Furthermore, the impact of transportation cannot be ignored, as LKC's transport can increase from 1.01 to 9.62% ECO_2eq/m^3, depending on the transport distance and the replacement level.

For example, replacing 25% of OPC with LKC decreases ECO_2eq by 12% compared to M1, but transporting it from the Patagonia to the central part of the country would increase ECO_2eq by 6.94%. If it were transported to the northern part of the country, the increase in ECO_2eq associated with transportation would be 9.62%, practically offsetting the benefit of using SCM. GGBS would reduce ECO_2eq by 12–19% compared to OPC concrete, but transporting it from the east to the west part of the country would increase the ECO_2eq up to 5.49%, depending on the level of OPC replacement by GGBS. However, the primary limitation of using GGBS is related to its availability. Indeed, its share in the Portland cement market in the Metropolitan Region of Buenos Aires is less than 21%, considering both compound Portland cement (CEM V/A) and blast-furnace cement (CEM III/A) [44].

Based on the nature and distribution of the locally abundant materials, it can be concluded that sustainable concretes with good compressive strength and durability performance can be obtained using local materials. In regions where materials are scarce or have slow/no reactivity, the sustainability of concrete can still be improved by reducing the *w/b* ratio (e.g., M4 and M7) or extending the curing time (e.g., M3). These findings highlight the potential for using local materials in concrete production and the importance of optimizing the mix design for maximum sustainability.

Concluding remarks

An analysis of concrete sustainability from various perspectives reveals the following insights:

- The analysis of compositional changes of concrete or cement per unit of volume or weight falls short when analyzing the sustainability of these materials. It is necessary to go further and integrate performance parameters (strength and/or durability) if the aim is to contribute to reducing the effects of climate change.
- As is well known, replacing ordinary Portland cement (OPC) by supplementary cementitious materials (SCM) reduces the embodied energy and ECO_2eq per ton of cement and m^3 of concrete.
- For relatively short transport distances (<500 km) and levels of cement replacement by SCM up to 25%, the influence of transport on ECO_2eq/m^3 emissions is almost negligible ($\leq$1.12%). However, for long transport distances (>2500 km), the ECO_2eq/m^3 related to SCM transport reaches 9.62%, which could meet the reduction of ECO_2eq/m^3 due to the use of SCM and obtain a net ECO_2eq/m^3 decrease that is virtually zero.
- Sustainability assessment based on performance parameters suggests the most sustainable concretes are those with the highest compressive strength and the best durability performance.
- The significance of using locally available materials should be emphasized, even in the absence of active SCMs, as sustainable concretes can be produced using 20% of limestone filler and low levels of calcined clay by reducing the water/binder ratio.

This study confirms the importance of including performance parameters in the analysis of the life cycle of concrete, such as compressive strength and durability. Nevertheless, the research should be extended by considering any other parameters proposed where performance attributes are involved. In addition, certain issues, such as on-site curing and its influence on durability, must be studied further, especially for concretes requiring extended curing times, as well as different environmental conditions (i.e., different chloride and CO_2 concentrations and moistness). In addition, it is necessary to adopt frameworks for analyzing the sustainability of concrete to achieve or approach as closely as possible zero net emission concrete, providing housing and infrastructure for the population and contributing to slowing down climate change.

Declaration of Competing Interest

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

Data availability

Data will be made available on request.

Acknowledgments

The authors acknowledge the support received by CONICET and the Universidad Nacional del Centro de la Provincia de Buenos Aires. They also are grateful to Cementos Avellaneda SA and Loma Negra CIASA for the materials supplied.

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