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Review Article

Toxicity and environmental and economic performance of fly ash and recycled concrete aggregates use in concrete: A review

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Abstract

This paper presents an overview of previous studies on the environmental impact (EI) and toxicity of producing recycled concrete aggregates (RCA), fly ash (FA), cement, superplasticizer, and water as raw materials, and also on the effect of replacing cement and natural aggregates (NA) with FA and RCA, respectively, on the mentioned aspects. EI and toxicity were analysed simultaneously because considering concrete with alternative materials as sustainable depends on whether their risk assessment is high. Therefore, this study mainly focuses on the cradle-to-gate EI of one cubic meter of concrete, namely abiotic depletion potential (ADP), global warming potential (GWP), ozone depletion potential (ODP), photochemical ozone creation (POCP), acidification potential (AP), eutrophication potential (EP), non-renewable energy (PE-NRe) and renewable energy (PE-Re). In terms of toxicity, leachability (chemical and ecotoxicological characterization) was considered. The results also include the economic performance of these materials, and show that the incorporation of FA in concrete significantly decreases the EI and cost of concrete. Thus, the simultaneous incorporation of FA and RCA decrease the EI, cost, use of

landfill space and natural resources extraction. Nonetheless, the leaching metals of FA decrease when they are incorporated in concrete. Relative to FA, the incorporation of RCA does not significantly affect the EI and cost of concrete, but it significantly reduces the use of landfill space and the need of virgin materials.

Keywords: Materials science, Environmental science, Industry, Economics, Safety engineering

1. Introduction

The Life Cycle Assessment (LCA) methodology was introduced in 1991 by SETAC (Society for Environmental Toxicology and Chemistry). According to Pinheiro [1], LCA intends to: (i) evaluate the environmental impacts (EI) of a product, process or activity by identifying and quantifying their environmental emissions and consumption of energy and materials; (ii) identify and evaluate the opportunities of making environmental improvements.

As shown in Table 1, the LCA boundaries of a construction material can be specified from “cradle-to-gate”, “cradle-to-grave” or “cradle-to-cradle”. According to the Environmental Protection Agency (EPA), the life cycle of a product comprises four main steps: (i) obtaining the raw materials - including the consumption of resources, materials and energy in the extraction, production and transport activities; (ii) production - including the raw material’s transformation, product fabrication and its conditioning and transport to final destiny; (iii) use, reuse and maintenance - where the activities and consumptions resulting from the use and maintenance of the product are quantified; (iv) recycling and waste treatment - where the impact of the activities associated with the disposal of the product, as well as the impact of the resulting waste, are evaluated.

Table 1. Detailed life cycle stages of building materials classification based on European Standards [2].

LCA boundaries			Life cycle stages/LCA information modules	Life cycle stage designation and description
Cradle to cradle	Cradle to grave	Cradle to gate	Product stage (A1–A3)	A1 Raw material extraction and processing, processing of secondary material input
				A2 Transport to the manufacturer
				A3 Manufacturing

(continued on next page)

Table 1. (Continued)

LCA boundaries			Life cycle stages/LCA information modules	Life cycle stage designation and description	
Cradle to cradle	Cradle to grave	Gate to grave	Construction process stage (A4–A5)	A4	Transport to the building site
				A5	Installation into the building
			Use stage - information modules related to the building fabric (B1–B5)	B1	Use or application of the installed product
				B2	Maintenance
				B3	Repair
				B4	Replacement
				B5	Refurbishment
			Use stage - information modules related to the operation of the building (B6–B7)	B6	Operational energy use
				B7	Operational water use
			End-of-life stage (C1–C4)	C1	De-construction, demolition
C2	Transport to waste processing				
C3	Waste processing for reuse, recovery and/or recycling (3R)				
C4	Disposal				
Cradle to cradle		Benefits and loads beyond the system boundary (D)	D	Reuse, recovery and/or recycling (3R) potentials	

Generally, the LCA of materials is carried out based on standards ISO 14040-14044 [3]. Frequently, researchers only complete the LCA from “cradle-to-gate”, and more rarely consider the “Use” and “End of life” stages. LCA studies usually rely on commercial software tools suitable for any product, process or activity (e.g. GaBi, SimaPro or openLCA software), where various EI assessment methods can be used to determine EI indicators according to EN 15804 [4]. Each method has a limited range of impact categories and CML “from the Centre of Environmental Science - Leiden University” [5] is the one prescribed in EN 15804 [4] for Environmental Product Declaration (EPD) development (Table 2). The majority of the studies and consider most of the following eight environmental categories defined in Table 2 (ADP, AP, EP, GWP, ODP and POCP, PE-Re and PE-NRe). The CML baseline method is normally used to quantify the impacts for the six first categories, and Cumulative Energy Demand method for the last two.

Table 2. Studied EI indicators and assessment methods [6].

Impact indicator	Unit	Standard	Method
		EN 15804 [4]	CML [7]
Abiotic depletion (ADP)	kg Sb eq	X	X
Acidification (AP)	kg SO ₂ eq	X	X
Eutrophication potential (EP)	kg PO ₄ ⁻³ eq	X	X
Global warming (GWP)	kg CO ₂ eq	X	X
Ozone layer depletion (ODP)	kg CFC ⁻¹¹ eq	X	X
Photochemical ozone creation potential (POCP)	kg C ₂ H ₄ eq	X	X
Non-renewable primary energy resources (PE-NRe)	MJ	X	X
Renewable primary energy resources (PE-Re)	MJ	X	X

Generally, LCA assessment enables evaluating the EI of materials or a service during their entire life cycle, from cradle-to-grave (extraction of raw materials → production and use stages → disposal in nature). There are several methods to assess EI within LCA, and evaluating the impacts of a given product can be made using different categories of EI. The most recent environmental impact assessment methods for LCA include ecotoxicology in the EI categories.

Ecotoxicology is one of the branches of toxicology that focuses on the toxic effects caused by natural or artificial substances present in the macro environment (water, soil and air) in living organisms. Therefore, to increase construction sustainability, this EI category (ecotoxicology) may have a strong contribution because it evaluates the potential environmental risk associated with the products to be used in the construction sector. For that purpose, it is required to evaluate their ecotoxicity using leaching tests, chemical analyses, and (eco) toxicity tests.

To assess the toxicity risks of construction materials, a common way is the determination of the leachability of their potentially harmful constituents. Aqueous eluates are produced and characterized by chemical analysis and (eco) toxicity testing. For this purpose, leaching batch tests [8] and measurement of heavy metals, sulphate, NO_x, SO_x, phenol index (carboxyl, halogen, hydroxyl, methoxyl or sulfonic acid), TOC (total organic carbon), pH and conductivity should be carried out. In addition and for chemical analysis (e.g. of cement and FA), researchers usually obtain the concentration ratio of the following elements: Al (Aluminium), As (Arsenic), B (Boron), Ba (Barium), Be (Beryllium), Ca (Calcium), Cd (Cadmium), Co (Cobalt), Cr (Chromium), Cu (Copper), Fe (iron), Ge (Germanium), Hg (Mercury), Mn (Manganese), Mo (Molybdenum), Ni (Nickel), Pb (Lead), Rb (Rubidium), Sb (Antimony),

Se (Selenium), Si (Silicon), Sn (Tin), Sr (Strontium), Th (Thorium), U (Uranium), V (Vanadium) and Zn (Zinc), and the major elements Al_2O_3 (aluminium oxide), CaO (calcium oxide), Fe_2O_3 (ferric oxide), K_2O (Potassium oxide), MgO (Magnesium oxide), Mn_2O_3 (Manganese III oxide), Na_2O (Sodium peroxide), P_2O_5 (Phosphorus pentoxide), SiO_2 (Silicon dioxide, or “silica, quartz”), SO_3 and TiO_2 (Titanium dioxide “rutile”), and Loss on ignition (LoI). Some of the mentioned elements are heavy metals, which are potentially toxic to the biological system, including Cd, Pb, As, Hg, Zn [9], Cr [10], Co, Ni, Cu, Sb and Zn [11].

It has been reported in many studies that EI of concrete can be decreased by using supplementary cementitious materials (SCM), e.g. fly ash (FA), and/or recycled aggregates (RA), e.g. recycled concrete aggregates (RCA). It is concluded that most of the studies focused on the technical performance [12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22] and there are few studies related with the LCA of concrete made with the mentioned SCM and/or RA [23, 24, 25, 26, 27, 28]. However, it is not correct to consider non-conventional concrete as a sustainable solution without their risk assessment [29]. In fact, the replacement of these non-conventional materials with traditional components is scarcely studied, in environmental and toxicity terms, and literature review studies regarding concrete with incorporation of both RCA and FA are absent.

2. Discussion of the literature

2.1. Toxicity of raw materials

The basic raw materials necessary to produce concrete are natural aggregates (NA), water and cement, but other materials (e.g., FA and RCA) can be incorporated for strength, durability and/or sustainability reasons. In addition, for chemical analysis (e.g. of cement and FA), researchers generally obtain the contents of the following elements: Al, As, B, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, Ge, Hg, Mn, Mo, Ni, Pb, Sb, Se, Si, Sn, Sr, Th, U, V and Zn, and of the major elements Al_2O_3 , CaO , Fe_2O_3 , K_2O , MgO , Mn_2O_3 , Na_2O , P_2O_5 , SiO_2 , SO_3 and TiO_2 , and LoI.

Hillier *et al.* [30] conducted a leaching procedure with acetic acid to characterise the toxicity of cement samples from 97 cement plants in North America. The results showed As, Be, Cd, Cr, Hg, Ni, Pb, Sb, Se and Th leached in detectable concentrations.

Barbudo *et al.* [31] studied the leaching potential of NA and RA from construction and demolition waste (CDW). The results showed that none of these aggregates release detectable quantities of heavy metals. However, high concentrations of SO_3 compounds, which can cause pollution of superficial and/or ground water, were found in mixed RA containing either ceramic particles or gypsum.

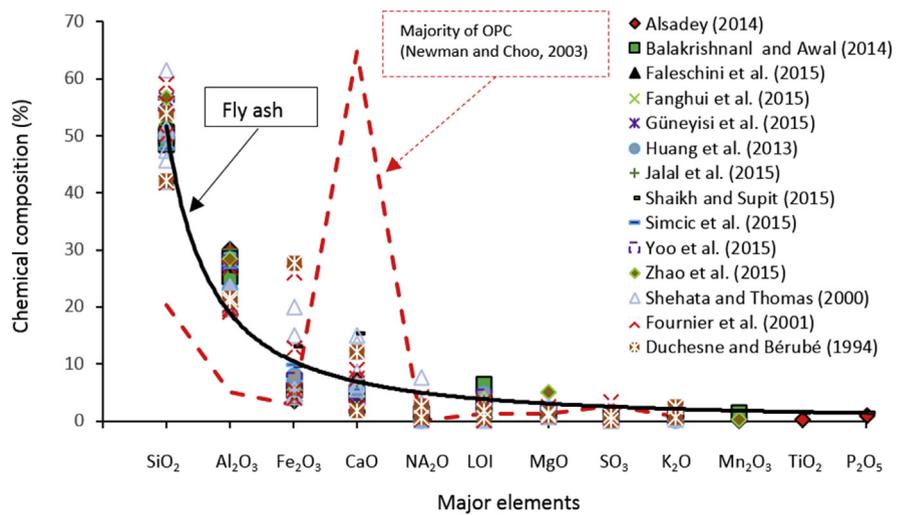


Fig. 1. Major element concentrations and LoI values of OPC and FA (type F).

A study [32] on the leaching characteristic of unbound RCA showed that leached heavy metals did not exceed the Norwegian drinking water criteria. The only influence on the soil was the existence of a greater quantity of calcium, which resulted in an insignificant increase in the soil’s pH.

Anderson [33] reported that the concentration of hazardous substances in superplasticizer (SP) is usually very low. The European Federation of Concrete Admixture associations [34] has made a LCA regarding the impact of concrete admixtures on the environment, which showed that, for 1 kg of SP, the hazardous waste, non-hazardous waste and radioactive waste disposed at the product stage (raw material supply, transport and manufacturing “A1–A3”) is only 0.00517, 25.6 and 0.9 grams, respectively.

Fig. 1 shows the average of the main elements of OPC and FA calculated from the results of several studies [35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49], and the results show that the major elements of FA (type F) are Al₂O₃, CaO, Fe₂O₃, K₂O, MgO, Mn₂O₃, Na₂O, P₂O₅, SiO₂, SO₃, TiO₂, and LoI, which consists of contaminant unburnt fuel. It is mainly SiO₂, but can also contain significant quantities of Al₂O₃ (for these studies, the average values of each major element and their standard deviation are shown in Table 3). The amount of CaO is limited but highly variable depending on the type of FA.

Table 3. Average major element concentrations and LoI values (%) of OPC and FA (type F).

Elements (%)	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	Na ₂ O	LOI	MgO	SO ₃	K ₂ O	Mn ₂ O ₃	TiO ₂	P ₂ O ₅	
Fly ash	Average	50.23	25.24	10.86	4.07	0.70	2.87	1.98	0.66	1.45	0.55	0.87	1.20
	STDEV	5.13	3.89	8.37	1.95	0.52	1.29	1.43	0.30	0.78	0.66	0.81	0.28
OPC	Average	20.40	5.10	2.90	64.80	0.11	1.30	1.30	2.70	0.77	–	–	–

Moreno et al. [50] studied most of FA (type F) produced in the European Union and indicated the following trace elements: Sr, Ba, V, Zn, B, Ni, Cr, Cu, Pb, RB, As, Co, Th, Ge, Be, Se, U, Mo, Sb, Sn, Cd and Hg (Fig. 2). In addition, the range of the trace elements concentrations obtained by Moreno *et al.* [50] is similar with that determined for most FA produced in the United Kingdom [51] (for this study, the average values of each trace element and their standard deviation are shown in Table 4).

A study on the potential metal leaching and toxicity of FA, when used as binder in soil stabilization, showed significant differences in leaching characteristics with respect to heavy metals. In this study, FA with high pH showed considerable leaching of heavy metals (Palumbo *et al.*, 2005). Similar results were observed in other studies [52, 53], in which the authors also concluded that, because of the relatively high concentration of heavy metals and corresponding potential environmental risk, FA must be regarded as hazardous materials. In fact, a study [53] showed that leaves and roots accumulate significant amounts of heavy metals, which may contaminate any food grown in that area.

The process of FA leaching includes physical and chemical transport/leaching. The metals deposition rate depends on the characteristics of the solid (nature inorganic oxide coating, particle size, zero point charge of the solid, temperature and organic carbon content), as well as on the properties of the liquid, including the pH and total dissolved metal concentrations. In natural environment, the heavy metals in solid material namely FA, can be classified in: (i) water-soluble; (ii) acid-soluble; (iii) oxide; (iv) difficultly reducible; and (v) residual [55]. This approach was employed to study the behaviour of FA leaching and mobility in environmental conditions (Table 5). Similar conclusions can be found in Sočo and Kalembkiewicz [56] and Landsberger *et al.* [57].

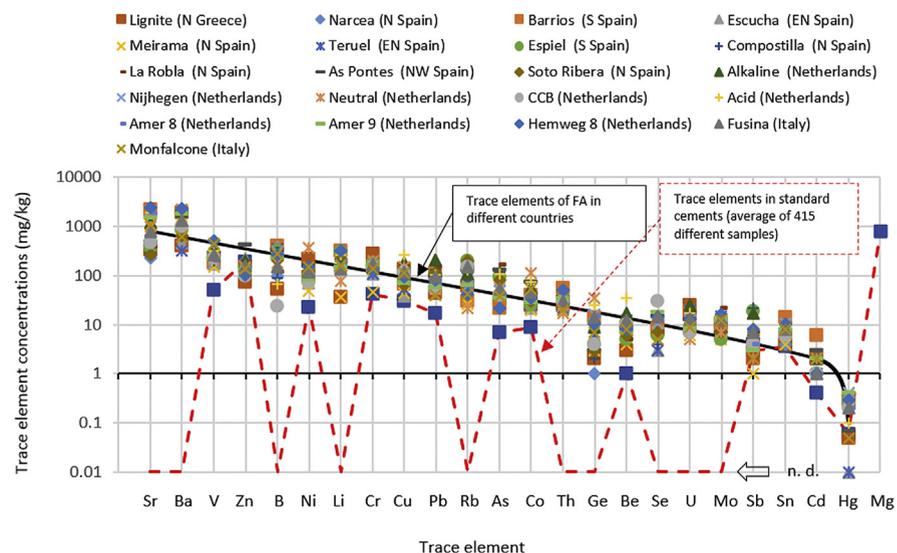


Fig. 2. Trace element concentrations (mg/kg) of FA (type F) produced in European Union (adapted from Moreno et al., [50]; VDZ, [54]), North (N) South (S) West (W) and East (E), and not determined (n. d.).

Table 4. Average trace element concentrations (mg/kg) OPC and of FA (type F) produced in European Union.

Elements (mg/kg)	Sr	Ba	V	Zn	B	Ni	Li	Cr	Cu	Pb	Rb	As	Co	Th	Ge	Be	Se	U	Mo	Sb	Sn	Cd	Hg	Mg	
FA	Average	3913	5363	255	161	216	123	187	153	101	88	105	69	41	31	9	9	10	13	11	7	8	2	0	n. d.
	STDEV	726	685	90	70	115	71	82	44	50	40	61	39	22	10	9	6	7	6	4	6	3	1	0.1	n. d.
OPC	Average	n. d.	n. d.	50	192	n. d.	23	n. d.	41	31	17	n. d.	7	9	n. d.	n. d.	1	n. d.	n. d.	n. d.	3	4	0.4	0.06	759

Table 5. Elemental speciation (%) in FA obtained by sequential extraction [55].

Fractions (%)	Si	Al	Ca	Fe	As	Ba	Cr	Mn	V	Zn
Water soluble	0.02	0.19	26.96	0.01	0.85	4	0.12	1.07	0	1.19
Acids soluble	0.08	0.62	16.81	0.64	12	6.47	2.34	0.86	3.43	1.65
Oxides	0.15	1.77	22.67	0.97	9.3	3.66	1.53	1	6.49	3.35
Difficultly reducible	0.18	2.65	12.46	1.09	6.67	6.94	1.32	0.84	3.14	7.31
Residual	99.56	94.77	21.1	97.28	71.18	78.93	94.69	96.22	91.9	80.23

FA is considered a serious issue for land disposal due to its environmental importance [58, 59] that may cause ground water contamination near the ash disposal area [29, 60, 61]. However, the final impact of each trace element depends on various factors, including the leaching time and FA source [62, 63]. Acidity has also a serious influence, since higher acidity causes greater rate and quantity of leaching elements. While high alkalinity seems to be characteristic of most ashes, leachates can vary from alkaline (pH = 12.4) to acidic (pH = 4.2) [64]. The amount of major elements also affects the leaching rate. The reaction between CaO and H₂O results in Ca(OH)₂, giving FA a pH in the range of 10–12.

Kadir *et al.* [65] reported that the concentration of heavy metal elements, namely Ni, Cr, Cu and FeO₃, in FA and bottom ash (BA) is higher than that of ordinary Portland cement (OPC). The results indicated that Ni, Cr, Cu and FeO₃ in FA are 560%, 460%, 420%, 390 and 40%, and in BA are 460%, 330%, 50% and 70% higher than that of OPC, respectively (Fig. 3). The lower hazardous heavy metal's

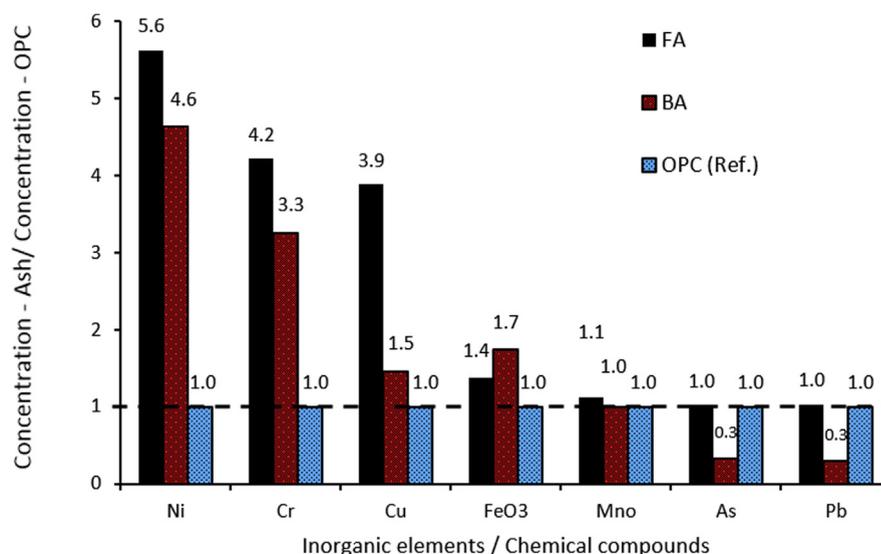


Fig. 3. Chemical composition of OPC, BA and FA produced in Malaysia [65]. The concentration of Ni, Cr, Cu, FeO₃, MnO, As and Pb in OPC sample was 19, 54, 26, 30200, 800, 37 and 60 mg/L, respectively.

concentration in BA than in FA is because BA mainly includes inert and non-combustible fractions of solid waste, which has lower concentrations of heavy metals. Frequently, the higher reactive elements are burned, resulting in ashes that are collected as FA, while inert and non-combustible materials settle in the furnace bottom and are defined as BA. Therefore, FA has a higher risk of hazardous heavy metal's leaching.

Numerous toxic elements show high enrichment in the fine particles of coal FA [66]. In fact, the concentration of volatile elements, such as, Cd, Pb and Zn, increases with the decrease of FA particle size from coarse to fine [67, 68]. Moreover, the particles of FA have a large surface area in comparison to mass [69]. The smaller particles have higher surface areas and contain significant surface concentrations of potentially toxic trace elements [70]. According to Roy *et al.* [71], the leachability of elements (P, Fe, Al, B, K, and Ca) decreases for longer ages. The authors also sorted the relative concentrations of leached elements in 3 pH levels: (i) Alkaline: Se > B > Cr > Ni > Cu > Ba > As > Zn > Al; (ii) Neutral: B > Cd > As > Se > Zn > Ni > Mn > Cu > Ba; and (iii) Acidic: B > Zn > Ca, F > Na > Mg, Co > Ni, Sr > Be > Cu, Pb, Al > Si, Fe, K. Based on a study by Theis and Gardner [72], the aqueous solubility of FA ranges is about 0.5–3% of total original mass. In spite of the insignificant total amount of leachate, the content needs to be precisely investigated and compared to the corresponding regulations.

2.2. Toxicity of concrete with traditional and non-traditional raw materials

Generally, long-term leaching from well-cured concrete produced with OPC and NA does not release detectable concentrations of toxic metals. However, it was also found that poorly cured mixes may release detectable concentrations of V [30].

Siong and Cheong [73] studied the leaching potential of BA and concrete by using the Toxicity Characteristic Leaching Procedure (TCLP) in accordance with the Environmental Protection Agency (EPA) Regulatory Method 1311. Results showed that FA exhibits a high leaching potential and is unable to meet the strict drinking water requirements. However, heavy metal's concentrations leached from concrete containing FA are significantly inferior and very close to the EPA drinking water standard (except for Cr and Zn, which were slightly over the limits) (Fig. 4). This indicates that the solidification process prompted by the hydration of cement is effective in keeping heavy metals existing in FA, thus reducing the leaching of harmful elements, which was also found in another study [74].

Regennitter [75] reported that FA leaching does not cause public health risks, and FA from different concrete applications does not add potential Hg leaching to concrete.

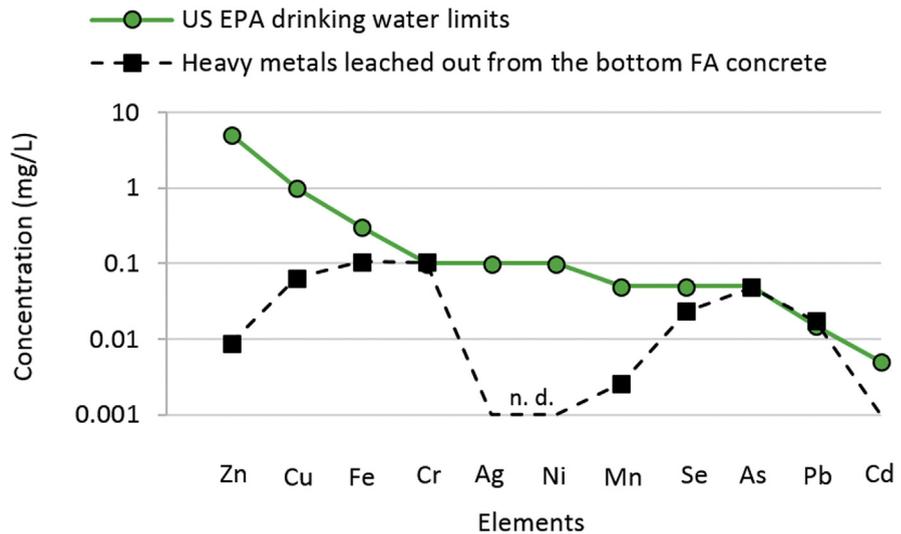


Fig. 4. Concentration of heavy metals leached out from bottom FA concrete vs US EPA drinking limits [73].

The leaching potential of RCA and NA is not considered a concern [32], but mixed RA containing either gypsum or ceramic particles may include high concentrations of SO_3 compounds [31].

Anderson [33] carried out a toxicological study on hazardous substances leached from concrete. The results showed that it may release detectable quantities of harmful elements, when used as a construction material, and the problem greatly increases when this material is crushed and reused in road banks.

Kadir *et al.* [65] made leachability tests in self-compacting concrete (SCC) with FA and BA using toxicity characteristic leaching procedure. 10%, 20% and 30% of cement was replaced with FA, and a similar incorporation ratio was repeated for BA and for the blend of both ashes “FA + BA”. After compressive strength tests, concrete samples were crushed and sieved to be less than 9.5 mm in size and in order to be used for toxicity characteristic leaching procedure. As shown in Table 6, the level of heavy metal’s leachate from control concrete and FA concrete was lower than the standard limit, except for As, whose level was above the limit of 5 mg/L allowed by US-EPA. However, this does not necessarily represent the true nature of the leachates featured by crushed SCC. The low pH of the solution (2.88) used as an extraction fluid may cause this metal to significantly leach out. Similarly to FA concrete and control concrete, BA concrete, as well as FA and BA concrete, showed similar behaviours.

It is therefore important to collect and systematize environmental and toxicity data on traditional/non-traditional components of CBM to minimize the risks of product developers and users.

Table 6. Result for heavy metals in SCC specimens using TCLP (mg/L) [65].

Heavy metal	Concentration limits ^a	Control	FA (%)			BA (%)			FA and BA (%)		
			10	20	30	10	20	30	10	20	30
As	5	26.52	23.89	25.19	25	27.82	26.94	24.8	23.51	24.49	27.47
Cr	5	0.033	—	—	—	—	—	—	—	—	—
Pb	5	0.447	0.578	0.646	0.695	0.783	0.847	0.909	0.94	1.028	1.024
Zn	500	0.173	0.173	0.202	0.222	0.145	0.144	0.16	0.172	0.177	0.181
Cu	100	0.026	0.029	0.026	0.031	0.03	0.03	0.031	0.033	0.034	0.035
Ni	1.34	0.135	0.187	0.211	0.221	0.26	0.132	0.064	0.287	0.061	—
Fe	—	0.095	0.074	0.083	0.083	0.053	0.034	0.017	0.136	0.037	—
Mn	—	0.033	0.027	0.105	1.235	0.007	—	—	—	—	—

^aUnited States Environmental Protection Agency [76].

A study of Kurda [77] complemented a toxicity analysis made by study of Rodrigues *et al.* [29], Table 7 summarizes the classification obtained for leachate samples of the raw materials and concrete mixes. Generally, the chemical analysis shows that the lowest potential hazard level was registered for the eluate samples of RCA and conventional concrete (M1), followed by sample M2 (high volume of FA), M3 (high volume of FA and 100% of RCA) and, finally, sample FA has the highest potential hazard level. In terms of ecotoxicological characterization, there is no evidence to classify the leachate samples of RCA, FA and M1 as eco-toxic, since the results

Table 7. Classification of raw and construction materials [77, 80].

Materials	Chemical characterization		Ecotoxicological characterization		
	EC Decision 2003/33/EC Directive No. 1999/31/CE		CEMWE (ADEME, 1998)		TCS (Persoone <i>et al.</i> , 2003)
	Classification	Parameter	Classification	Parameter	
NA	Inert	—	No evidence of ecotoxicity	n.a.	n.a.
Cement	—	—	No evidence of ecotoxicity	n.a.	n.a.
RCA	Non-dangerous	TDS	No evidence of ecotoxicity	n.a.	Class III
FA	Dangerous	Se	No evidence of ecotoxicity	n.a.	Class III
M1 (RCA 0% and FA 0%)	Non-dangerous	TDS	No evidence of ecotoxicity	n.a.	Class III
M2 (RCA 0% and FA 60%)	Non-dangerous	TDS, Cr, Mo	Ecotoxic	<i>DM microcrustaceans</i>	Class IV
M3 (RCA 100% and FA 60%)	Non-dangerous	TDS, Cr, Mo	Ecotoxic	<i>DM microcrustaceans</i>	Class IV

n.a. - not applicable; Class III - Acute Toxicity; Class IV - High Acute Toxicity.

of the ecotoxicological and chemical characterizations comply with the threshold established in the French proposal CEMWE [78]. However, the results obtained for samples M2 and M3 allow classifying these materials as ecotoxic, since the minimum value defined in the French proposal CEMWE for “daphnia magna” microcrustaceans is exceeded [78]. However, according to TCS [79], RCA, FA and conventional concrete (M1) were classified with acute toxicity, and FA concrete with or without RCA were considered with high acute toxicity. Furthermore, a study of Rodrigues [29] reported that, based on the previous studies, there is no danger or evidence of ecotoxicity in NA and cement leachate samples in terms of chemical and ecotoxicological characterizations, respectively.

Based on data of studies [80, 81, 82], it is suggested that the alkaline pH of the FA and FA concrete with and without RCA granulated concrete may be relevant in this respect and may contribute to possible environmental risks. Such risks can be particularly relevant if eluates or leachates formed from FA or concrete are produced in landfills and/or during building service (e.g. due to rain) and can reach freshwater ecosystems leading to water alkalinisation.

2.3. LCA of raw materials

Water is one of the major components required for the production of concrete and its components. Therefore, it is important to understand the EI of the production of potable water. For that purpose, most of researchers consider a “cradle-to-gate” approach that ends in the final consumer (Fig. 5). Morales -Pinzón *et al.* [83] and Braga [84] estimated these EI by using the Ecoinvent v3.0 database included in SimaPro software. This process includes water treatment and transportation to the final consumer. Cabejšková [85] performed a study by using the Gabi 4 software and CML 2002 characterisation method from two water treatment plants in the Czech Republic. The results show that the total EI of Želivka was four times higher than that of Hrdějovice. This can be explained by the fact that the water treatment plants of Želivka were built 40 years before those of Hrdějovice (Table 8). Broadly speaking, the most important impact categories in the last study are found to be Global Warming Potential, Acidification Potential and Abiotic Depletion of fossil resources, due to energy consumption at water treatment plants.

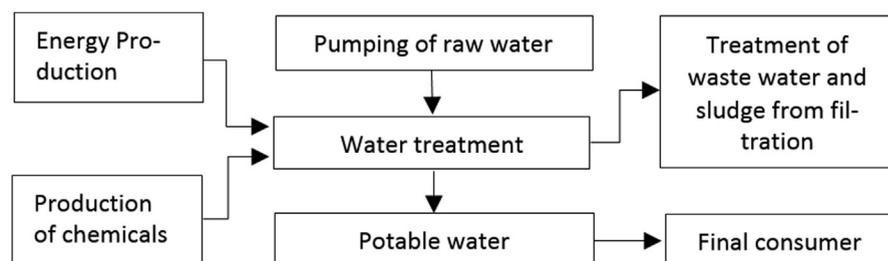


Fig. 5. System boundaries of the production of potable water [85].

Table 8. EI resulting from the life cycle of production of 1 kg of tap water.

Impact category	Unit	Cabejšková [85]		Braga [84]	Morales-Pinzón <i>et al.</i> [83]	
		Czech Republic		Portugal	Colombia	
		Želivka	Hrdějovice	–	Pereira	Bogota
ADP	kg Sb eq	3.07E-11	1.18E-12	1.57E-11	8.05E-05	7.27E-05
GWP	kg CO ₂ eq	1.88E-04	7.82E-05	1.33E-04	1.27E-02	1.18E-02
ODP	kg CFC-11 eq	2.93E-11	1.18E-11	5.93E-12	1.29E-08	1.28E-08
AP	kg SO ₂ eq	7.12E-07	1.47E-07	3.87E-08	8.71E-05	8.30E-05
EP	kg PO ₄ -3 eq	5.83E-08	7.02E-09	9.70E-07	–	–
POCP	kg C ₂ H ₄ eq	5.59E-08	6.72E-09	4.99E-08	–	–
PE-Re	MJ	–	–	1.80E-05	–	–
PE-NRe	MJ	–	–	1.80E+01	–	–
Abiotic Depletion (fossil)	MJ	1.91E-03	4.71E-04	–	–	–
Freshwater Aquatic Ecotoxicity Potential	kg DCB eq	3.66E-07	6.55E-08	–	–	–
Human Toxicity Potential	kg DCB eq	3.38E-06	3.89E-07	–	8.83E-03	–

With a general content between 0.8% and 4.0% by weight of cement, SP increases the workability and compaction of concrete due an increase of dispersion of the cement particles. Moreover, SP's use can decrease water content significantly in a range between 15% and 40% [86]. However, a reduction in the w/b does not seem to be an interesting solution to decrease the EI of concrete [84].

EFCA [87] made an Eco-profile for all main groups of SP (Sulphonated naphthalene formaldehyde, Sulphonated melamine formaldehyde, Vinyl copolymers and Poly carboxylic ethers). The production of 1 kg of SP was studied according to ISO 14040 series on LCA (including “A1: Production of preliminary products”, “A2: Transport to the plant” and “A3: Production including provision of energy, production of packaging as well as auxiliaries and consumables and waste treatment” sub-stages, in a “cradle-to-gate” approach), without including transportation to the concrete plant production (Table 9).

Serres *et al.* [6] calculated similar data in an Eco-profile for 1 kg of SP according to the NFP01-010 standard (data collected by SYNAD). Braga [84] modelled the production of SP using EFCA (2006) data and considering that the company producing SP is 19.55 km away from the concrete plant. More recently, EFCA [34] presented a new LCA for the production of 1 kg of SP with EI significantly changed from the previous ones (Table 10). Moreover, Sjunnesson [88] showed that SP makes a contribution of 6.0 % of POCP, 2.1 % of AP, 0.4 % of GWP and 0.7 % of EP, in ordinary concrete.

Table 9. Eco-profile for the production of 1 kg of SP [87].

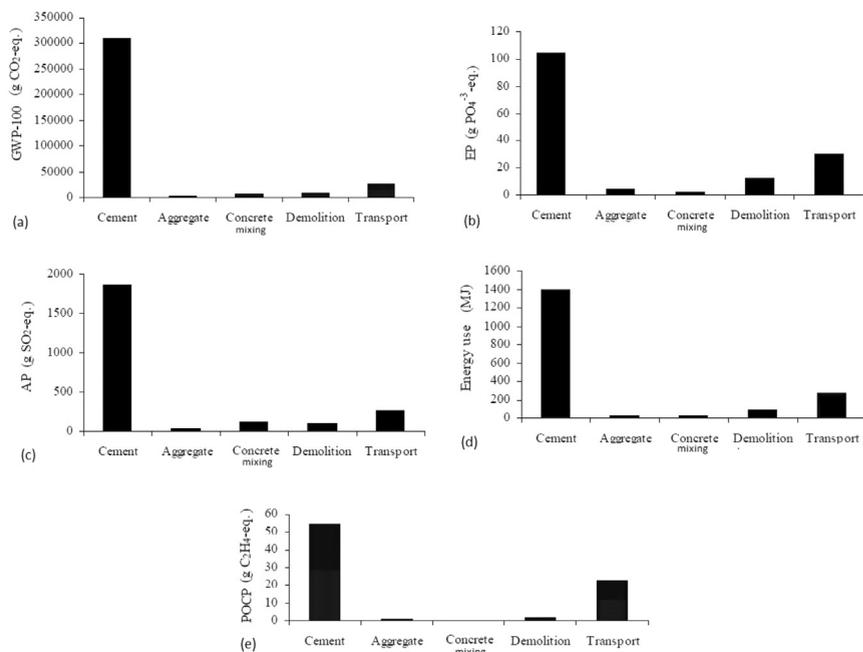
Input/output	Unit	Value	Unit	Value
<i>Raw materials - input</i>				
Coal, brown	g	82	Crude oil	kg
Coal, hard	g	51	Natural gas	m ³
<i>Emissions to air</i>				
CO ₂	kg	0.72	Acetic acid	mg
CO	g	0.55	Ammonia	g
SO _x	g	3.6	As	μg
No _x	g	1.8	Chromium VI (Cr)	μg
N ₂ O	mg	67	heavy metals	mg
Methane	g	1.2	Hg	μg
Butane	mg	11	Ni	mg
Pentane	MG	14	V	mg
Methanol	MG	60	Dioxins	ng
Ethane	mg	8.9	CFC-10	μg
Benzene	mg	7.4	CFC-114	μg
Non-methane VOC	g	0.29	Halon-1211	μg
PAH	ug	39	Halon-1301	μg
<i>Emissions to water</i>				
Chemical oxygen demand	g	2.6	Oils, unspecified	g
PAH's	ug	67	Ni	MG
Barite	mg	51		
<i>Emissions to soil</i>				
Chromium VI (Cr)	mg	0.22	Oils, unspecified	g
<i>Solid waste</i>				
Non-hazardous waste	g	21	Hazardous waste	g
<i>Hazardous waste</i>				
<i>Total Energy</i>				
Total energy	MJ	18.3		

Table 10. EI for the production of 1 kg of SP.

References	Baseline CML method						Cumulative Energy Demand	
	ADP	GWP	ODP	POCP	AP	EP	PE-NRe	PE-Re
	kg Sb eq	kg CO ₂ eq	kg CFC-11 eq	kg C ₂ H ₄ eq	kg SO ₂ eq	kg PO ₄ ⁻³ eq	MJ	MJ
Braga (2015)	3.88E-11	0.771	8.78E-08	5.68E-05	4.26E-03	1.05E-03	18	1.80E-05
EFCA (2015)	1.10E-06	1.88	2.30E-10	3.12E-04	2.92E-03	1.03E-03	31.4	1.51

OPC “CEM I” is one of the common types used worldwide and is a major contributor to the high EI values of concrete production (Fig. 6), as can be confirmed in other studies [88, 89].

This can be explained by the need of blending together raw materials (clay and limestone), which are then fed into a rotating kiln with a temperature about 1450 °C [90], to produce cement. Apart from high energy consumption due to the heating process, the emissions to air due to this chemical process ($\text{CaCO}_3 + \text{heat} \rightarrow \text{CaO} + \text{CO}_2$) are also relevant, despite the former being the most harmful stage in cement production (Fig. 7). The European Cement Research Academy [91] has developed an EPD for OPC (CEM I) produced in Europe, while Blengini (2006) calculated the EI of the production in Portugal of different types of OPC. The results show that, by

**Fig. 6.** Impact categories, namely (a) GWP, (b) EP, (c) AP, (d) energy use and (e) POCP per functional unit for the production of 1 m³ of concrete in Serbia [24].

increasing the strength of cement from 42.5 to 52.5, EI (ADP, GWP, AP, EP, POCP, PE-NRe and PE-Re) increase 4%, 3%, 15%, 9%, 2%, 11%, 18% and 5 %, respectively. Moreover, Braga [84] modelled the production of different types and grades of cements using data from Blengini (2006) and added the transportation stage (it was considered that the company producing the cement is 100 km away from the concrete plant), finding that ADP, GWP, AP, EP, POCP and PE-Re only increased slightly while PE-NRe increased 6.5% (Table 11). In addition, most of the impact category values shown in Table 11 are similar except for the results of Teixeira *et al.* [92]. This can be explained by the fact that the authors only estimated the EI based on the environment report of the company, while the results of other studies were obtained based on the detailed investigation on LCA. Also, some different values can be seen in the results of ECRA [91]. This was due to a significant methodological difference that occurred in ECRA recently. In addition, Babor *et al.* [93] concluded that cement is a major contributor to CO₂ in the atmosphere and the most energy-intensive material in the construction sector. Moreover, Peshkova *et al.* [94] focused on the Russian cement industry development based on the experience of developed countries and showed that a key factor that may ensure the sustainable development of the construction industry is a cross-sectoral cooperation, which will allow organizing a low-waste production cycle with a minimum costs of raw materials.

Table 12 shows the impact assessment results for the production of 1 kg of different types of aggregates from cradle to gate in different countries. The results of the studies were different mainly due to the transportation scenario.

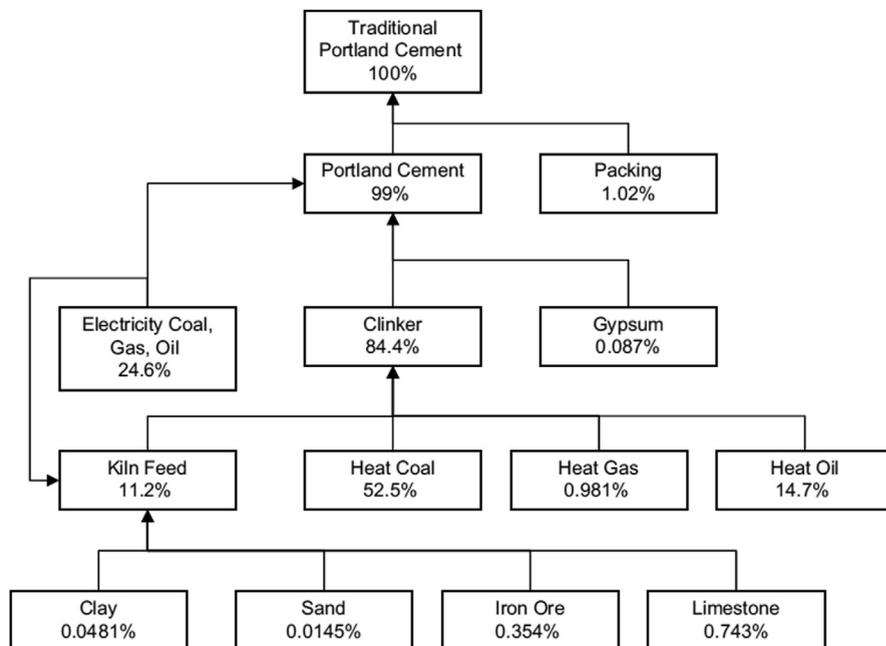


Fig. 7. EI for each step of Portland cement manufacturing process [95].

Table 11. EI for the production of 1 tonne cement.

Sources	Country	Material	Baseline CML method						Cumulative Energy D.	
			ADP	GWP	ODP	POCP	AP	EP	PE-NRe	PE-Re
			kg Sb eq	kg CO ₂ eq	kg CFC-11 eq	kg C ₂ H ₄ eq	kg SO ₂ eq	kg PO ₄ ⁻³ eq	MJ	MJ
ECRA [91]	EU	CEM I	0.001	898	1.21E-07	0.142	1.48	0.211	222	3700
Blengini [96]	Portugal	CEMI 42.5	3.83	926	9.47E-05	0.0748	2.54	0.35	203	5641
		CEMI 52.5	3.99	951	1.09E-04	0.0831	2.76	0.36	240	5907
Braga [84] based on Blengini [96], with transport to concrete plant	Portugal	CEMI 32.5	3.36	804	8.49E-05	0.0665	2.25	0.31	222	4970
		CEMI 42.5	3.83	927	9.47E-05	0.0752	2.55	0.35	218	5640
		CEMI 52.5	3.99	952	1.09E-04	0.0834	2.76	0.36	255	5910
Teixeira <i>et al.</i> [92]	Portugal	CEM I	–	1790	2.58E-05	0.214	9.24	2.09	–	–
De Schepper <i>et al.</i> [89]	Netherlands	CEM I	1.6	830	2.40E-05	0.045	1.2	0.275	–	6870
Marinković <i>et al.</i> , [23]	Serbia	CEM I	–	887	–	0.156	5.3	0.3	–	1255
Chen <i>et al.</i> [97]	France	CEM I	1.59	844	2.28E-05	0.0426	1.15	0.173	–	6420

Table 12. Impact assessment results for the production of 1 kg of different types of aggregates.

Source	Type	Country	ADP	GWP	ODP	POCP	AP	EP	PE-NRe	PE-Re
			kg Sb eq	kg CO ₂ eq	kg CFC ⁻¹¹ eq	kg C ₂ H ₄ eq	kg SO ₂ eq	kg PO ₄ ⁻³ eq	MJ	MJ
Natural aggregate										
Braga [84]	River sand	Portugal	3.37E-10	9.87E-03	1.71E-11	2.80E-06	4.58E-05	1.08E-05	1.35E-01	1.56E-04
	Crushed sand		1.24E-09	2.79E-02	2.26E-10	9.06E-06	1.59E-04	3.54E-05	3.92E-01	4.52E-04
	Granitic coarse aggregate		1.09E-09	2.44E-02	2.43E-10	7.83E-06	1.44E-04	3.18E-05	3.44E-01	3.81E-04
	Limestone coarse aggregate		1.39E-09	3.14E-02	2.09E-10	1.03E-05	1.75E-04	3.90E-05	4.41E-01	5.23E-04
Tošić <i>et al.</i> , [27]	River aggregate	Serbia		1.43E-03		2.78E-07	1.64E-05	2.02E-06	1.48E-05	
	Crushed stone aggregate			2.12E-03		4.15E-07	2.42E-05	3.01E-06	2.19E-05	
Korre and Durucan [98]	Crushed rock aggregates	UK		9.30E-04	1.06E-10	4.58E-07	5.85E-06		4.35E-07	
	Crushed rock aggregates			3.29E-03	4.50E-10	1.20E-06	1.89E-05		1.07E-06	
	Land won gravel aggregate			2.16E-03	3.19E-10	7.35E-01	1.20E-05		6.87E-07	
	Land won sand aggregate ^d			1.85E-03	2.14E-10	9.85E-07	1.03E-05		5.90E-07	
	Marine gravel aggregates			3.79E-02	8.50E-06	5.40E-05	6.77E-04		1.04E-04	
	Marine sand aggregates			3.80E-02	1.78E-10	5.40E-05	6.77E-04		1.04E-04	
Marinković <i>et al.</i> [99]	Natural aggregate	Serbia		1.56E+00		3.09E-04	1.79E-02	2.22E-03		1.62E-02
Sjunnesson [88]	Crushed stone aggregate	Sweden		1.60E-03		1.70E-06	7.80E-07		3.00E-02	2.00E-02
	River aggregate			7.00E-04		3.80E-10	5.00E-05		1.24E-03	2.40E-03
Average			1.01E-09	1.16E-01	8.50E-07	4.90E-02	1.33E-03	1.96E-04	1.68E-01	5.73E-03
Standard deviation			4.68E-10	2.98E-01	4.25E-06	4.25E-06	1.18E-03	4.29E-05	1.93E-01	8.28E-03

(continued on next page)

Table 12. (Continued)

Source	Type	Country	ADP	GWP	ODP	POCP	AP	EP	PE-NRe	PE-Re
			kg Sb eq	kg CO ₂ eq	kg CFC ⁻¹¹ eq	kg C ₂ H ₄ eq	kg SO ₂ eq	kg PO ₄ ⁻³ eq	MJ	MJ
Recycled aggregate										
Braga [84]	Coarse RCA	Portugal	2.12E-10	7.44E-03	1.60E-10	2.14E-06	4.05E-05	9.28E-06	1.08E-01	9.61E-05
Tošić <i>et al.</i> , [27]	32.5% coarse and 35% fine river aggregate, and 32.5% coarse RCA	Serbia		2.28E-03		7.03E-07	2.49E-05	3.01E-06	2.59E-05	
	65% Coarse RCA and 35% fine river aggregate			3.38E-03		1.18E-06	3.61E-05	4.34E-06	3.95E-05	
Korre and Durucan [98]	RCA	UK		2.42E-03	2.83E-10	8.00E-07	1.21E-05	7.06E-07		
Marinković <i>et al.</i> [23]	RCA	Serbia		1.64E+00		3.21E-04	1.88E-02	2.33E-03		1.70E-02
Average			2.12E-10	3.42E-01	2.22E-10	3.20E-04	1.36E-03	4.33E-06	3.60E-02	9.14E-03
Standard deviation				7.56E-01	8.70E-11	7.12E-04	2.98E-03	3.62E-06	6.23E-02	1.28E-02

From 2012 to 2014, an increase of 5.2% (up to 40.20 billion tonnes) per year was expected in the international market of aggregates, and this increment can reach 60.3 billion tonnes by 2022 [100]. To reduce the EI of aggregates, one option is to use RA from CDW. According to estimations, RA still represent only 3% of the total aggregates consumption [101]. CDW corresponds to 1/3 of wastes generated in Europe, but significant differences can be seen in the percentage of recycling between countries [102]. In terms of the total CO₂ emissions, aggregates have small contribution of concrete production (~15%), which essentially result from their processing/extraction [103]. However, since aggregates take about 70% of the total concrete volume, the incorporation of RA may reduce the EI of concrete [102].

The economic and environmental advantages of using RA are highly dependent on transportation distances [84]. Both RA cost and ecologic footprint can significantly increase depending on the demolition site locations, thus decreasing the interest to consumers. However, it is possible to use mobile recycling plants that practically eliminate the need of transport operations, depending on the target application and availability of raw materials [104].

Estanqueiro *et al.* [102] carried out a calculation of the EI of coarse NA (Scenario i) and coarse RCA in the manufacture of concrete using, for the latter, a recycling fixed (Scenario ii) and mobile plant (Scenario iii) (Fig. 8). SimaPro software was used to model LCA of coarse NA and RCA and site-specific data were supplied from Portuguese companies. It was found that RA (if all fine RA are sent to landfill) is not more convenient than the use of NA (Scenario i) in terms of a single score,

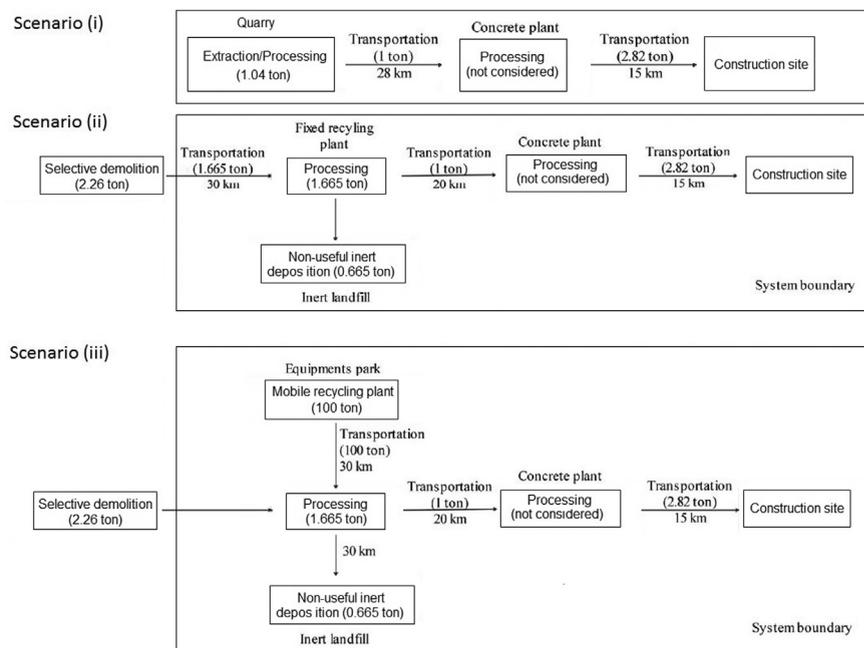


Fig. 8. Life cycle of the scenarios studied in Portugal [102].

even if a mobile recycling plant is used (scenario 3). S_{ii} and S_{iii} display a maximum benefit over S_i in land use category. These authors also reported, however, that coarse RA can show a better environmental performance than natural ones if fine RA are also used in concrete production instead of being sent to a landfill (Fig. 9). Once again, these results are mainly dependent on transportation distances. In addition, Estanqueiro *et al.* [102] showed that the incorporation of RA in concrete is more beneficial than the incorporation of NA only in terms of land use and respiratory inorganics impact categories, resulting essentially from the exploitation of quarry.

Globally, the lion's share of coal (86%) is consumed in thermal generation, largely by pulverized coal combustion [105]. Worldwide production of coal was 3,830 Mt in 2015 [106]. Asia pacific (namely China) is the largest coal producing region, followed distantly by North America, Europe and Eurasia, Africa, South and Central America, and the Middle East. The majority of the coal is consumed in the country of origin, with about 16% of hard coal production traded on the international coal market [107] to countries that are not self-sufficient in terms of coal production but consume it for power generation (e.g. Portugal, Serbia, Switzerland, etc.). CO_2 represent the greatest quantity (98–99 %) of the air emissions of this process, and their majority (96%) is released from the combusted coal of the power plant [108].

Fig. 10 shows a typical electrical power plant where the major input material is coal. It burns inside the furnace by injection with air and fuel to generate heat and increase the temperature of the pipe located in the furnace. Thus, the water inside the pipe is transformed into high pressure steam and moves ahead to the turbine, where it is converted into mechanical energy, and transmitted to the generator in order to produce electricity. The secondary input materials are NH_3 (Ammonia) and CaO “lime”, to remove NO_x and SO_2 , respectively. Concerning output materials, Heidrich *et al.* [107] reported 85% FA, <15% BA, and a small percentage of Cenospheres (hollow ash particles), flue gas De-sulfurization (FGD) and boiler slag. In Europe (European Union 15), the European Coal Combustion Products Association [109] reported use rates around 43% and 46%

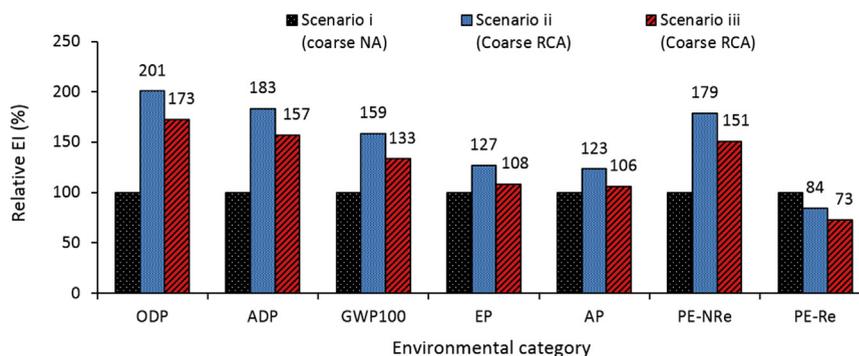


Fig. 9. Relative EI of the scenario using CML Baseline 2000 [102].

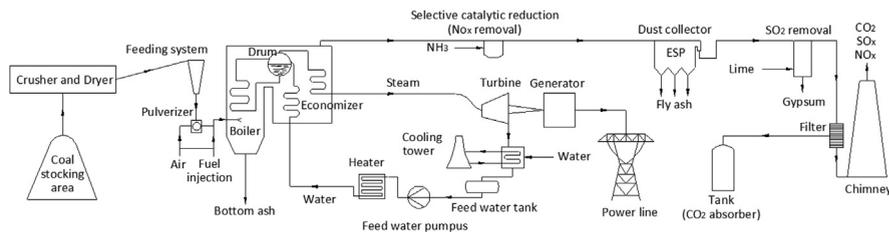


Fig. 10. A typical coal power plant.

for FA and BA, respectively, in the year 2010 (excluding restoration and reclamation), 100% for boiler slag and 78% for FGD gypsum [110]. Worldwide use of ashes was reported to be 53% of total production in 2011, with extreme values of 96.4% and 10.6% [105]. In addition, Jovanovic [111] showed that generating 1 kWh of electricity consumes 1.290 kg of coal and produces 0.194 kg and 0.013 of FA and BA, respectively. However, this value is higher than the results of Chen *et al.* [97]. This is due to the efficiency of the coal power plant.

Chen *et al.* [97] studied the EI of FA production using the LCA methodology with three allocation procedures. The first was “no allocations”, which is the current practice, with FA considered as waste and only including impacts from secondary processes. The second procedure was “Mass allocation”, where impacts of primary process were allocated between the main product and by-product according to the ratio of their masses. The third procedure was “Economic allocation”, where impacts of primary process were allocated between the main product and by-product according to the ratio of their revenues. For that purpose, they obtained data from Sokka *et al.* [112] and Dones *et al.* [113] for coal power plants (primary process) and Surschiste [114] for FA treatment (secondary process) and ATILH [115] for French CEM I production (Table 13).

Fig. 11 presents the results of Chen *et al.* [97] in percentages relatively to CEM I, with a logarithmic scale, using the values shown in Table 14. These results show that for “no allocation procedure”, where FA is considered as waste, its EI can be negligible compared to CEM I (all environmental indicators are less than 25% of the impacts of OPC). When “allocation by mass” procedure is considered, all EI are much higher than those of OPC. When an “economic allocation” is followed, EI of three indicator categories are higher (AP, POCP and ADP), and only one is lower (GWP) than that of OPC.

Teixeira *et al.* [92] obtained the EI of FA produced from a major Portuguese coal power plant (located in the centre of the country) using SimaPro 7.3.3 and an economic allocation procedure. The results show that the EI of coal FA are lower than those of cement for the majority of the impact categories. The Danish Technological Institute [116] reported, in an EPD in accordance with EN ISO 14025 [117] and EN 15804 [4], that the EI of FA are not significant (Table 14).

Table 13. Input and outputs for production and treatment of 1 kg FA [97].

		1 kWh electricity Coal-fired power plants	1 kg FA Drying + stock			1 kWh electricity Coal-fired power plants	1 kg FA Drying + stock
Input							
Raw materials	Water (m ³)	0.035					
Energy	Hard coal (kg)	0.432		Gaz (MJ)		0.29	
	Electricity (kWh)		6.82E-03	Fuel (m ³)	0.000156		1.03E-06
Transport	Train (t km)	4.27366E-06		Truck (t km)			0.003
Installation	Coal thermal plant	1.33E-11					
Output							
Waste	Mud “Boues”(kg)		8.48E-05	Ashes (kg)	25.8		
Air emission	Heat (MJ)	5.62		Manganese	1.01E-07		
	Ashes (kg)	3.38E-04	3.23E-05	Methane	1.04E-05		
	Chloride hydrogen	1.26E-04		NOx (kg)	1.96E-03	1.75E-05	
	SOx (kg)	4.54E-03	9.13E-08	CO (kg)	8.16E-05	9.05E-06	
	Lead	2.32E-07		Dioxins (kg)	7.14E-14		
	Nickel	1.07E-07		CO ₂ (kg)	0.95		
Products	Electricity (kWh)	1		FA (kg)	0.052		1
	Bottom ash (kg)	0.014					

2.4. LCA of concrete with traditional and non-traditional raw materials

There are still few LCA studies on LCA of RAC. Hendriks and Janssen [118] compared the eco-costs/value ratio (EVR) of conventional concrete with that of RAC. The EVR model is a business-oriented definition that connects the thinking of the modern manager to the need for a more sustainable society. A low EVR means that a product is suitable in a future sustainable society and a high EVR means that the economic value/cost ratio will worsen over time. The results showed that the use of RA instead of NA is very beneficial.

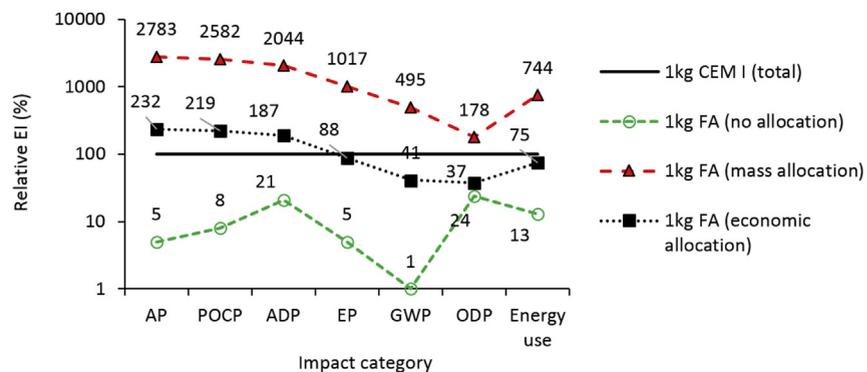


Fig. 11. Converting the EI (for the different CML indicators) of the 1 kg of cement CEM I to 1 kg eq. FA [97].

Table 14. IE for producing 1 kg of FA in different countries.

Impact category	Country	Allocation	ADP	GWP	ODP	POCP	AP	EP	Energy use
Unit			kg Sb eq	kg CO ₂ eq	kg CFC ⁻¹¹ eq	kg C ₂ H ₄ eq	kg SO ₂ eq	kg PO ₄ ⁻³ eq	MJ
Chen <i>et al.</i> [97]	France	No	3.37E-04	8.77E-03	5.58E-09	3.22E-06	5.53E-05	8.23E-06	0.833
		Economic	2.98E-03	3.50E-01	8.45E-09	9.34E-05	2.67E-03	1.52E-04	4.84
		Mass	3.25E-02	4.18	4.06E-08	1.10E-03	3.20E-02	1.76E-03	49.70
Teixeira <i>et al.</i> [92]	Portugal	Economic		1.01E-02	7.16E-11	9.52E-07	2.07E-05	2.56E-07	0.199
DTI [116]	Danish	Economic	3.29E-10	3.92E-03	9.88E-13	5.49E-07	7.26E-06	1.05E-06	0.058

Braunschweig *et al.* [119] performed a LCA in order to study and identify ecological optimization potentials of aggregate production and developed scenarios for ecologically optimal production of aggregates and concrete for construction projects. The reuse of RCA in the production of concrete proved to have higher environmental benefits than their disposal. However, the feasibility of producing RCA concrete mainly relies on the wastes' transport distances.

The LCA results of de Schepper *et al.* [89] suggest that, when compared to conventional concrete, the use of RA can reduce the global warming potential of this material.

Sjunnesson [88] concluded that the GWP of concrete is highly affected by raw material production and, along with other indicators (EP, AP and POCP), depends on transportation operations. He also showed that the environmental load is linearly related to transport distances. By decreasing the transport distances by 40%, the transport operation's EI decreased by 63%, which became approximately equivalent to the EI of the raw material production. Estanqueiro *et al.* [102] performed a LCA on RA used in the manufacture of ready-mixed concrete, and also the calculation and comparison of the corresponding EI. However, the results also showed that the assessment was very sensitive to the transportation distances.

Evangelista and de Brito [120] obtained the EI of fine NA and RCA concrete at the production and construction stages, use and end of life, in a "cradle-to-grave" LCA (Fig. 12) by using EcoConcrete software. Since this software is limited in some specific stages, they used an interactive Excel-spreadsheet to obtain reliable results. They found that the EI (ADP, GWP, ODP, AP, EP and POCP) decrease 6–8% and 19–23%, when 30% and 100% of fine NA are replaced with fine RCA, respectively.

Braunschweig *et al.* [119] showed that the EI of high-quality RCA concrete with 25% of RCA and of NA concrete are similar if the cement content of the former is just slightly higher. They also showed the contribution of NA and RA production for GWP, AP, respiratory effects, energy use, gravel use and land use is below 10% in concrete. Weil *et al.* [121] compared NA concrete to RCA concrete with 35% and 50% of RCA and different cement contents. The conclusions were similar to those of

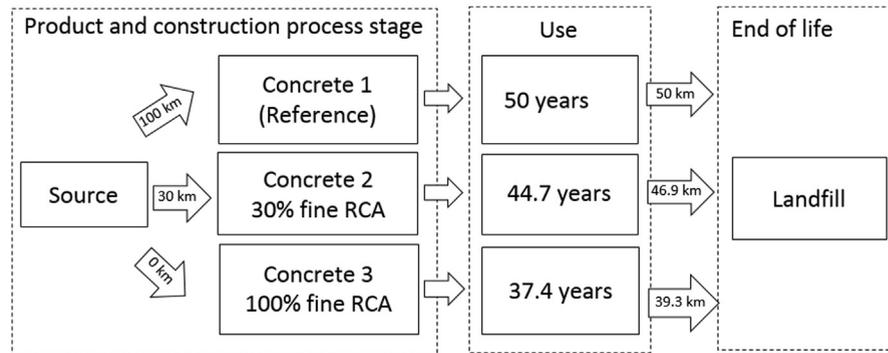


Fig. 12. Life cycle of NA and FRA concrete [120].

Braunschweig *et al.* [119]. According to Knoeri *et al.* [122], impacts from NA and RA concrete mixes can be similar when the additional cement content of RA concrete is below 10%. Furthermore, these authors accounted for the advantages from recovered steel scrap and eliminated impacts of waste disposal into the LCA to make the analysis more beneficial for RCA concrete.

Marinković *et al.* [23] compared the EI caused by the production of two types of ready-mixed concrete (one using NA, and the other RCA) with two transport scenarios. The distance between concrete plant and NA and cement production sites was considered to be 100 km or 150 km. The distance between concrete plant and RCA production sites was considered to be 15 km in the first scenario and 100 km in the second one. It was found that, for the Serbian context, the EI of NA and RCA concrete are highly dependent on travel distances and transport type of aggregates between the recycling plants and construction sites. Moreover, Marinković *et al.* [23] reported similar EI for NAC, and RCA concrete with 100% of coarse RA and 3% additional cement, when transport distances of RA are smaller than those of NA.

Taranu *et al.* [94] compared and evaluated the EI of the four common concrete classes used in Romania, and determined the best one by using LCA methodology, and the GaBi ts software.

Tošić *et al.* [27] complemented the study of Marinković *et al.* [23] by applying a normative multi-criteria optimization method (VIKOR method) developed at the University of Belgrade, Faculty of Civil Engineering [123] in order to find out the optimal solution. They concluded the incorporation of 50% of coarse NA by coarse RCA is optimum in terms of economic and from an EI point of view (Table 15). However, other studies [23, 27] showed that EI slightly increases as the incorporation of RCA grows (Table 15), despite RCA having clear advantages in terms of mineral resources, environmental load, depletion and waste production [27].

Braga [84] analysed the LCA of coarse NA and RA concrete in terms of their environmental and economic impacts. It was established that the use of cement type II is

more convenient than that of cement type I, due to the reduction in cost of 12% and 7% for cement 32.5 and 42.5, respectively. Contrary to expectations, higher concrete strengths do not necessarily lead to a higher EI. As for selecting aggregates, it is advised to use limestone coarse NA and rolled fine sand. Using limestone instead of granite aggregates results in 50% cost reduction while using coarse RCA causes 80% savings. Mixes with maximum EI do not contain SP in their composition, but is also advised to use SP to decrease cement content, which is the main responsible for the EI. However, the increase of SP rises the cost of concrete. In addition, the w/b reduction does not show a significant improvement in EI. The benefits of coarse RCA is higher when it is used for lower strength concrete. It was also concluded that the use of coarse RCA results in a reduction of costs and EI. Some of the mentioned conclusions are briefly presented in Table 16.

Prusinski *et al.* [124] studied the EI of concrete containing blast furnace slag as partial replacement for OPC. The results showed that energy consumption, and CO₂ and other emissions, are significantly reduced when blast furnace slag is used.

Table 15. EI of the production of 1 m³ of concrete in Serbia.

Impact category	Sources	Process	Energy use	GWP	EP	AP	POCP
			(MJ)	kg CO ₂ eq	kg PO ₄ ⁻³ eq	kg SO ₂ eq	kg C ₂ H ₄ eq
100% fine and coarse river aggregate	Tošić et al. [27]	Concrete	20	5,718	1.7	110	0.1
		Transport	160	11,995	14.3	123	8.6
		Total	1,617	333,844	124.5	2,139	64.2
34% fine and 66% coarse crushed stone aggregate		Concrete	20	5,718	1.7	110	0.1
		Transport	195	14,495	16.2	141	7.8
		Total	1,783	364,121	136.9	2330	68.4
32.5% coarse and 35% fine river aggregate, and 32.5% coarse RCA		Concrete	20	5,718	1.7	110	0.1
		Transport	166	12,434	14.5	125	8.1
		Total	1,642	335,660	126.2	2,155	64.4
65% coarse RCA and 35% fine river sand		Concrete	20	5,718	1.7	110	0.1
		Transport	183	13,645	15.2	133	7.4
		Total	1,723	348,281	132.2	2,238	66.2
NA concrete (scenario (i): transport distances of the NA was 100 km)	Marinković et al. [23]	Concrete	20	5,718	1.7	110	0.1
		Transport	267	19,958	22.8	198	15.6
		Total	1,570	307,612	121.6	2,009	65.2
RA concrete (scenario (i): transport distances of the RCA was 15 km)		Concrete	20	5,718	1.7	110	0.1
		Transport	249	18,542	20.2	179	15.0
		Total	1,613	319,626	123.7	2,071	67.0
RA concrete (scenario (ii): transport distances of the RCA was 100 km)		Concrete	20	5,718	1.7	110	0.1
		Transport	558	41,612	44.1	395	37.1
		Total	1,923	342,696	147.6	2,287	89.1

The results of Kurda [77] show that the cost of the concrete mixes without VAT decreased slightly with the incorporation of RCA. Contrary to the incorporation of RCA, the use of a small percentage of SP increased the cost significantly. However, the results show that, by incorporating 60% of FA in NA concrete or incorporating 30% of FA in RCA concrete, the high cost of concrete with SP can be offset. In addition, by incorporating RCA in FA concrete, even when SP is used, the cost is lower than that of the reference concrete. In addition, transportation also affects the cost of concrete. For example, Golgota et al. [125] obtained a sustainable and low-cost concrete produced by local materials and their impact in everyday life in Albania.

In another study, O'Brien *et al.* [126] performed a LCA on embodied greenhouse gas emissions of concrete as a function of FA content and its transportation distance. The results showed that there is a reduction in greenhouse gas emissions if FA replaces cement and comes from less than 100 km from the concrete plant.

The CO₂ emissions can be decreased from 18% to 57% by increasing the FA content from 24% to 70% [127], and 54% by replacing 50% of OPC with FA [128]. Moreover, Malin [129] reported that for producing concrete with compressive strength of 20 MPa at 28 days, CO₂ emission can be decreased by 28% by replacing 35% of OPC with FA. Flower and Sanjayan [103] showed that the use of FA may reduce the CO₂ emissions by 13–15%, compared with conventional concrete. Turk *et al.* [130] concluded that the use of FA may reduce the EI in about 75%.

An EPD report [131] showed that with replacement rates of 20%, 30% and 40% of OPC with FA, the EI (namely, GWP, ODP, ADO, EP, POCP, NR and RE) of concrete mixes decrease between 9–14%, 13–22% and 18–30%, respectively, regardless of their target strength. On the other hand, the EI of the same incorporation levels of FA increase 6–13%, 21–38%, 46–125%, 54–133% and 77–156%

Table 16. Influence of each raw material in the economic and environmental performance of concrete [84].

Raw materials	€	EI	€ and EI
CEM I	–	–	–
CEM II	+	+	+
River aggregate	+	+	+
Crushed aggregate	–	–	–
Granitic coarse aggregate	–	+	–
Limestone coarse aggregate	+	–	+
Coarse RCA	++	++	++
SP	–	+	+

Note: + represents a reduction of impact; ++ represents a significant decrease of impact; – represents an increase of impact.

when increasing the target strength 0–17.24 MPa, 17.25–20.68 MPa, 20.69–27.58 MPa, 27.59–34.47 MPa, 34.48–41.37 MPa and 41.38–55.16 MPa, respectively (Table 17). This behaviour was attributed to the fact that increasing target strength requires higher cement content, which is a major contributor to the high EI values of concrete production [88, 89].

Marinković *et al.* [25] performed a LCA of concrete mixes made with 0%, 19%, and 38% of FA and with coarse NA fully replaced by coarse RCA. Since the EI of FA depends on the allocation procedure, the authors obtained the EI of concrete mixes considering three allocation approaches. In the ‘no allocation’ case, all impacts of RCA concrete with FA are lower than those of RCA concrete with no FA, but the results are different for the remaining approaches. This is due to the large quantities of airborne pollutants emitted from coal power plants in the process of electricity production, where a small allocation coefficient can significantly influence FA impact indicators [97]. Thus, the ‘mass allocation’ approach of RCA concrete with FA, due to the relatively great mass of FA generated during electricity production, presents significantly higher impacts than RCA concrete with no FA, except for ODP. In the ‘economic allocation’ case, results are more positive for RCA concrete with

Table 17. Cradle to gate LCA of the production of 1 m³ of FA concrete with different strength classes at 28 days [131].

Indicator/LCI Metric unit (equivalent)	FA	GWP	ODP	ADP	EP	POCP	NRE	RE
	%	kg CO ₂	kg CFC-11	kg SO ₂	kg N	kg O ₃	MJ	MJ
0–17.24 (MPa)	0	302.3	5.3E-06	1.36	0.15	17.66	2,384	17
	20	261.2	4.6E-06	1.2	0.14	16.14	2,103	15
	30	239	4.2E-06	1.11	0.13	15.33	1,951	14
	40	215.6	3.8E-06	1.02	0.12	14.47	1,792	13
17.25–20.68 (MPa)	0	337.1	5.8E-06	1.5	0.16	19.02	2,624	19
	20	290.5	5.0E-06	1.31	0.15	17.31	2,305	17
	30	265.3	4.6E-06	1.21	0.14	16.38	2,132	15
	40	238.9	4.2E-06	1.11	0.13	15.4	1,951	14
20.69–27.58 (MPa)	0	416.1	7.1E-06	1.81	0.19	22.12	3,167	22
	20	357.1	6.17E-06	1.58	0.17	19.95	2,763	20
	30	325.2	5.6E-06	1.45	0.16	18.77	2,544	18
	40	291.6	5.04E-06	1.32	0.15	17.53	2,315	17
27.59–34.47 (MPa)	0	509.1	8.6E-06	2.92	0.28	33.78	3,809	27
	20	435.6	7.4E-06	2.63	0.25	31.07	3,305	23
	30	395.9	6.74E-06	2.47	0.23	29.61	3,033	21
	40	353.9	6.0E-06	2.3	0.21	28.06	2,746	19
34.48–41.37 (MPa)	0	536.1	9.0E-06	3.03	0.29	34.92	3,999	28
	20	458.5	7.7E-06	2.72	0.26	32.06	3,467	24
	30	416.5	7.0E-06	2.55	0.24	30.51	3,180	22
	40	372.1	6.3E-06	2.38	0.22	28.87	2,876	20
41.38–55.16 (MPa)	0	624.1	1.0E-05	3.38	0.32	38.37	4,605	32
	20	532.6	8.93E-06	3.02	0.28	34.99	3,978	27
	30	483.2	8.1E-06	2.82	0.26	33.17	3,640	25

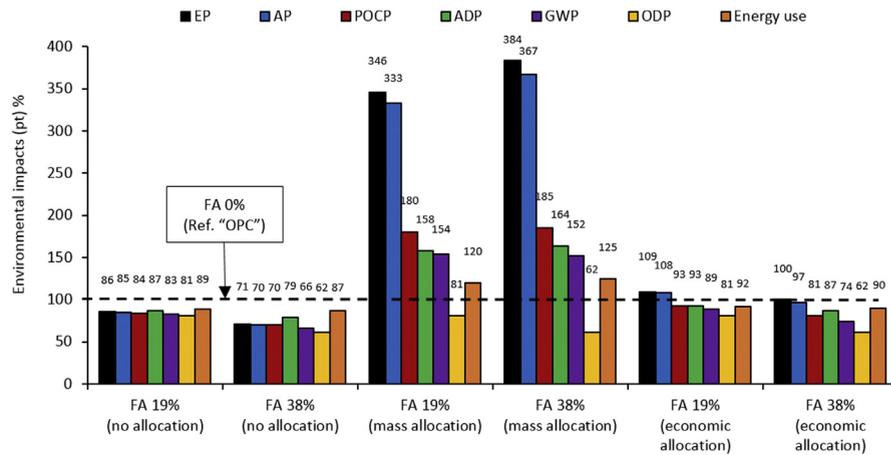


Fig. 13. Impact category indicators of coarse RA concrete incorporating various ratios of FA in ‘no allocation’, ‘mass allocation’ and ‘economic allocation’ cases [25].

FA mainly due to the very low price of FA in Serbia. In this case, all impact indicators of RCA concrete with 38% of FA (except eutrophication where results are approximately the same) are lower than those of RCA concrete with no FA (Fig. 13).

Tait and Cheung [132] summarized the results of EPD 2008 for NA concrete (cement type CEM I) and compared it to FA concrete (cement type CEMII/B-V, 35% FA), obtaining results similar to Marinković *et al.* [25] (FA 38%, no allocation). However, a small difference can be noted between the studies, which can be explained by the fact that Tait and Cheung [132] used only NA (Fig. 14).

Al-Ayish [133] showed that SCM have a dual influence on the GWP of reinforced concrete structures. First, it decreases the greenhouse gases through cement clinker replacement, and secondly it significantly increases chloride ion penetration resistance. However, this is not considered in the regulations, which makes it difficult to predict in LCA at early design stages.

Latawiec *et al.* [134] obtained the mechanical strength and durability performance of FA concrete mixes, as well as their LCA. The study shows how to estimate the

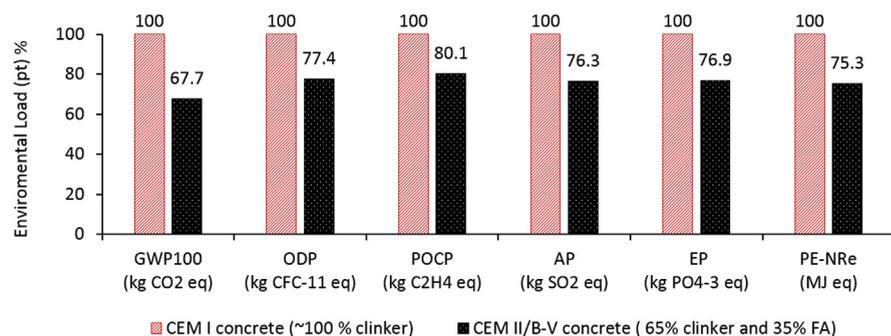


Fig. 14. EPD 2008 impact assessment: characterization [132].

combined effect of the modification in terms of both some of the technical features and CO₂ emissions of concrete.

3. Conclusions

The scope of this literature review included an examination of the effects of fly ash (FA) and recycled concrete aggregates (RCA), and of some traditional raw materials, on the environmental, economic and toxicological performance of concrete. The main findings were:

- Ordinary Portland cement (OPC) is one of the materials most used globally and it can be considered as a major contributor to the high environmental impacts (EI) values of concrete production;
- The superplasticizer (SP) content does not significantly affect the EI of concrete. But, since the cement content (major contributor to the EI of concrete) can be decreased by using SP, this action is highly advisable in terms of EI. SP's use can decrease the water content significantly between 15% and 40%, which is positive in terms of EI. However, due to the low EI of water, a reduction in the w/b does not seem to be an difference-making solution to decrease EI of concrete;
- Aggregates give a small contribution to the total CO₂ emissions of concrete production, which essentially result from their processing/extraction. However, since aggregates take up about 70% of the total concrete volume, the incorporation of RA may reduce the EI of concrete. Furthermore, the incorporation of FA in concrete mixes is more efficient than that of RA to reduce the CO₂ emissions;
- The reuse of RCA in the production of concrete proved to have higher environmental benefits than their disposal. However, the environmental feasibility of producing RCA concrete mainly relies on the wastes' transportation distances;
- The economic advantages of using RA are highly dependent on transportation distances;
- The use of SP significantly increases the concrete cost. However, the incorporation of FA in concrete significantly decreases the cost of concrete. Thus, the simultaneous incorporation of FA and RCA decrease the cost of concrete, and the higher cost of concrete with SP can be offset.
- Neither natural aggregates (NA) nor RA from construction and demolition waste (CDW) release detectable quantities of heavy metals, except RA containing either ceramic particles or gypsum;
- The concentration of hazardous substances in superplasticizer (SP) is usually very low;
- It has been argued that FA concrete showed considerable leaching of heavy metals and must be regarded as hazardous materials while other studies show

that heavy metal's concentrations in leachates from concrete containing FA are significantly lower and very close to the EPA drinking water standard limit;

- Long-term leaching from well-cured concrete produced with OPC and neither RCA nor NA release detectable concentrations of toxic metals;
- The leaching metals of FA decrease when they are incorporated in concrete. This reduction of the heavy metals' leaching between the FA powder and FA concrete may be related to the cement's ability to solubilize/stabilize the concentration of heavy metals due to chemical retention processes that allow the incorporation of the elements in the cement matrix, and physical retention by encapsulation. However, this study still suggests avoiding the use of FA concrete for drinking water tank and architectural concrete applications.

Declarations

Author contribution statement

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The authors declare no conflict of interest.

Additional information

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