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An environmental evaluation of geopolymer based concrete production: reviewing current research trends

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ABSTRACT

In this study we carry out a detailed environmental evaluation of geopolymer concrete production using the Life Cycle Assessment methodology. The literature shows that the production of most standard types of geopolymer concrete has a slightly lower impact on global warming than standard Ordinary Portland Cement (OPC) concrete. Whilst our results confirm this they also show that the production of geopolymer concrete has a higher environmental impact regarding other impact categories than global warming. This is due to the heavy effects of the production of the sodium silicate solution. Geopolymer concrete made from fly ashes or granulated blast furnace slags based require less of the sodium silicate solution in order to be activated. They therefore have a lower environmental impact than geopolymer concrete made from pure metakaolin. However, when the production of fly ashes and granulated blast furnace slags is taken into account during the life cycle assessment (using either an economic or a mass allocation procedure), it appears that geopolymer concrete has a similar impact on global warming than standard concrete. This study highlights that future research and development in the field of geopolymer concrete technology should focus on two potential solutions. First of all the use of industrial waste that is not recyclable within other industries and secondly on the production of geopolymer concrete using a mix of blast furnace slag and activated clays. Furthermore geopolymer concrete production would gain from using waste material with a suitable Si/Al molar ratio in order to minimise the amount of sodium silicate solution used. Finally, by taking into account mix-design technology, which has already been developed for OPC concrete, the amount of binder required to produce a geopolymer concrete could be reduced.

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1. Introduction

Concrete is the most commonly used construction material. Its use by communities across the globe is second only to water. Customarily, concrete is produced by using Ordinary Portland Cement (OPC) as the binder; a highly energy intensive product which releases carbon dioxide (CO_2). Due to a world-wide increase in the demand for OPC (Capros et al., 2001), cement production could represent nearly 10% of total anthropogenic CO_2 emissions in the close future. Numerous studies have dealt with mitigation perspectives in the cement industry (von Bahr et al., 2003; Huntzinger and Eatmon, 2009; Liu et al., 1995; Szabó et al., 2006; Taylor et al., 2006; Worrell et al., 2000). A recent study showed that it is possible to reduce by half the 1990 CO_2 emission level in developed countries by improving current cement technology (Habert et al., 2010). However without a technological turn around, the goals recommended by the Intergovernmental Panel Group for Climate Change (IPCC) that is to say a reduction by a factor 4 of CO_2 emissions will not be reached (Habert et al., 2010). New low-CO_2 binders are therefore needed to meet the demand for concrete and still reach the CO_2 reduction goals. Among these new binders it is commonly accepted that sulfo-aluminate clinkers and geopolymers are highly potential solutions.

However, although geopolymers are presented by many authors as a solution for "green" concrete, few studies have quantified the environmental impact of geopolymers (Davidovits, 1999; Duxson et al., 2007) and to our knowledge only one environmental evaluation has been based on the Life Cycle Assessment (LCA) method (Weil et al., 2009). It evaluated two geopolymer mix-designs and focused on a few environmental impact categories (global warming, energy and resource depletion).

The objective of the present study is to perform a detailed environmental impact assessment of standard geopolymer

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concrete production and compare it with the production of OPC based concrete. This study uses geopolymer concrete mix-designs found in the literature. A distinction is made between three types of geopolymer concrete made from different materials: fly ash, blast furnace slag and metakaolin. This distinction allows us to identify the most promising environmental mix-design trend.

2. Materials and methods

The LCA method is divided into 3 main stages (ISO, 2006). First the *functional unit*, and the *system boundaries* have to be defined. Secondly, the inventory phase covers the identification and the quantification of energy and material consumption, as well as waste production and emissions. Finally, once the Life Cycle Inventory has been put together, different impact indicators are used to define the *environmental impact* of geopolymer concrete within each category. These three stages are detailed in the below section.

2.1. Functional unit and system boundaries

The studied system is reduced to the production of the concrete constituents. Therefore, the analysis does not include every stage of the product's life cycle (*cradle to grave*) but ends at an intermediate stage (*cradle to gate*) as shown in Fig. 1. This can be done when one analyses a production, such as concrete, which has multiple specific applications in civil engineering (beams, pillars, pavements, houses, bridges, etc.) and therefore disallows a unique life cycle to be defined. This type of partial analysis is useful for the further construction of complete life cycles for specific concrete end-products on a larger scale. Furthermore, it can be assumed that, once concrete is cast in the structure, the impacts during the rest of the life cycle (maintenance and demolition) will be similar for a geopolymer concrete or an OPC concrete. Actually both concrete can be considered as inert material for their disposal. Therefore as long as the compared materials display similar functional properties in terms of behaviour in the fresh state, durability and mechanical strength the assumption of a reduction of the study to a cradle to gate evaluation is valid.

As both geopolymers and OPC based concretes are mineral suspensions, the organic polymer technology can be applied to both materials to adjust their fresh properties as long as the polymers chosen are able to resist to the alkaline solution. Furthermore, these chemical admixtures have negligible environmental impacts (Flower and Sanjayan, 2007; Habert and Roussel, 2008) compared to the other components of the concrete. That is why fresh concrete properties have not been considered in the definition of the functional unit. The durability aspects can have large consequences on the results. Actually an alternative product that last only half as long as the reference will need to be used twice more which seriously modifies the environmental evaluation. The concrete durability has been investigated since decades and can be now finely modelled (Baroghel-Bouny et al., 2007; Baroghel-Bouny et al., 2009). Investigations on geopolymer concrete durability are less abundant but the first results concerning reinforcement corrosion, which is the main problem for the durability of concrete structures, seem to show similar comportments than OPC based concrete (Bastidas et al., 2008; Miranda et al., 2005). Geopolymer concrete structures can resist even better than ordinary concrete structures against fire or acid attacks (Bakharev, 2005; Cheng and Chiu, 2003; Fernandez-Jimenez et al., 2007). Therefore, the durability has not been considered in the functional unit and we choose here to reduce the *functional unit* to 1 cubic metre of concrete with a given compressive strength in the hardened state. Furthermore, as standard concretes are currently made with an average substitution of 30% of OPC by mineral additions such as Fly Ash (Habert and Roussel, 2009), in this paper, geopolymer based concrete from literature are compared with cement based concrete with the same mechanical strength and with a binder made with either only OPC or 30% clinker substitution.

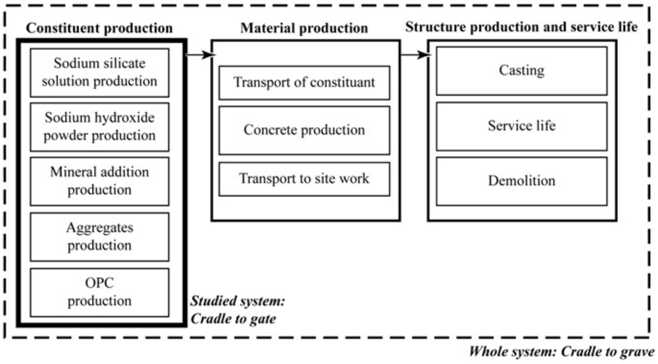

Fig. 1. Schematic description of concrete life cycle, which is divided in three main steps: the production of the concrete constituents, the production of concrete itself and the use of concrete within a structure. The boundaries of the system studied here are highlighted. It is restricted to the production of the constituents used in concrete.

2.2. Environmental and technical data collection

Geopolymer concrete mix-designs come from literature (Buchwald et al., 2007; Collins and Sanjayan, 1999; Dombrowski et al., 2007; Hardjito and Rangan, 2005; Kong and Sanjayan, 2010; Latella et al., 2008; Lee and van Deventer, 2002; Meliani, 2010; Olivia et al., 2008; Pacheco-Torgal et al., 2005; Rangan et al., 2005; Rovnaník, 2010; Sathonsaowaphak et al., 2009; Sumajouw et al., 2007; Weil et al., 2009; Yang et al., 2008) and are presented in Table 1. The mix-design for one cubic metre was either reported directly from literature or calculated from the above studies. The alkali-solutions are made with sodium silicate solution and sodium hydroxide. The aluminosilicate is either Fly Ash (FA), Blast Furnace Slags (GBFS) or metakaolin (MK).

To make the comparison meaningful, it is necessary to assess concrete with equivalent mechanical strengths. The compressive strength of the OPC concrete can be adjusted by varying the water to cement ratio of the paste (De Larrard, 1999). Indeed, the compressive strength of OPC concrete can be related to the cement content through the Féret equation:

$$f_c \approx K.Rc_{28} \cdot \left(\frac{V_{cement}}{V_{paste}}\right)^2 \tag{1}$$

Where f_c is the compressive strength, K a parameter that characterises the aggregate quality, Rc_{28} the specific mechanical strength of cement, V_{cement} the volume of cement and V_{paste} the volume of the paste that includes air, water and cement. Furthermore, the aggregate to paste volume ratio of the geopolymer concrete is used for the OPC concrete to which it will be compared. For concrete mix-design Rc_{28} have been set to 52.5 MPa and K have been adjusted with OPC concretes coming from the same studies as those used for geopolymer (Collins and Sanjayan, 1999; Olivia et al., 2008). For mortars, which are mix-design made with no coarse aggregates, equation (1) has been used and the K factor has been adjusted by considering that when characteristic strength of cement (Rc28) is 52.5 MPa it means that the 28 day compressive strength of a mortar made with 1350 g of sand, 450 g of cement and 225 g of water is 52.5 MPa (CEN, 2006). Finally, for mix-designs

Table 1
Mix-design for geopolymer based concrete. Data are from: Rangan et al., 2005; Olivia et al., 2008; Latella et al., 2008; Sumajouw et al., 2007; Lee and van Deventer, 2002; Pacheco-Torgal et al., 2005; Yang et al., 2008; Kong and Sanjayan, 2010; Meliani, 2010; Sathonsaowaphak et al., 2009; Collins and Sanjayan, 1999; Rovnaník (2010); Weil et al., 2009; Buchwald et al., 2007; Dombrowski et al., 2007; Hardjito and Rangan, 2005. Equivalent cement content is calculated from equation (1) (see text for details).

Ref	Gravel	Sand	Filler	Mineral addition	NaOH powder	Na Silicate solution	Water	Admixture	Compressive strength	Cement equivalent
Fly ash based geopolymers										
			Kaolin	FA						
1	1294	554		408	11	103		6	35	348
2	1201	647		408	17	103	26	6	35	347
2	1201	647		408	17	103	21	6	41	378
2	1201	647		408	17	103	17	6	68	483
2	1201	647		408	17	103	36	6	25	296
2	1170	630		444	18	111	26	6	48	435
2	1248	672		356	15	89	26	6	25	266
2	1292	554		408	17	103	26	6	36	354
16	1294	554		476	31	48			17	242
16	1294	554		477	13	120			57	444
16	1294	554		478	48	192			48	407
16	1294	554		479	19	264			68	485
16	1294	554		408	21	103	17		42	381
16	1294	554		408	21	103	17	4	41	376
16	1294	554		408	21	103	17	8	41	376
16	1294	554		408	21	103	17	16	36	353
16	1201	647		408	17	103	18	6	43	385
16	1201	647		408	15	103	14	6	38	362
16	1201	647		408	11	103		8	63	467
16	1294	554		408	17	103	11	8	59	452
16	1201	647		408	11	103		6	44	390
16	1201	647		408	13	103	8	6	55	436
16	1201	647		408	15	103	14	6	53	428
16	1201	647		408	17	103	21	6	51	420
16	1201	647		408	18	103	27	6	45	394
16	1201	647		408	17	103	21	6	47	403
4	1202	647		404	17	102	17	6	60	455
5			50	450	55	110			52	481
5			50	450	63	110			35	429
6	1756			476	13	120			60	515
7		1386		357		105	231		8	170
7		1374		331		127	229		8	173
7		1363		306		148	227		9	187
8		1312		735	70	175			72	561
8	1190	793		444	42	106			72	403
8	1209	806		451	43	107			62	353
10		1505		547	29	141			50	633
10		1505		547	36	170			46	608
10		1505		547	41	196			46	608
10		1505		547	49	233			42	581
10		1505		547	73	93			18	380
10		1505		547	61	131			32	507
10		1505		547	51	163			42	581
10		1505		547	41	196			46	608
10		1505		547	29	233			39	559
15				77	6		17		20	55
15			6	71	6		17		40	70
15			15	62	6		17		35	66
GBFS based geopolymers										
				GBFS						
7		1498		462		38	250		22	218
7		1480		432		62	247		44	322
7		1462		401		86	244		51	361
7		1445		373		109	241		52	380
7		1428		344		132	238		51	388
11	1090	801		347		78	174		50	390
11	1091	802	35	313		79	174		60	426
			FA							
11	1096	805	35	314		79	175		55	403
			Slag							
11	1077	791	34	308		78	172		48	396
			FA					Waste		
13	1878		57	230	48	33	99	83	39	351
			MK							
14			31	31	5		32		31	67
14			17	51	4		29		29	61
14			0	73	3		24		35	60

(*continued on next page*)

Table 1 (*continued*).

Ref	Gravel	Sand	Filler	Mineral addition	NaOH powder	Na Silicate solution	Water	Admixture	Compressive strength	Cement equivalent
MK based geopolymers										
			Filler	MK						
3				34		63	3		70	92
9		1250	0	365	66	356	74		44	477
9		1250	18	347	63	339	71		44	481
9		1250	55	310	56	303	63		39	454
9		1250	92	274	49	267	56		35	427
9		1250	0	365	66	356	74		46	491
9		1250	19	347	63	339	71		45	487
9		1250	57	310	56	303	63		45	486
9		1250	95	274	49	267	56		35	425
9		1250	0	365	66	356	74		44	478
9		1250	19	347	63	339	71		42	469
9		1250	58	310	56	303	63		34	424
9		1250	96	274	49	267	56		27	378
12		1350		450		372	100		62	489
			GBFS							
14			0	55	7		38		5	39
14			31	31	5		32		31	67
14			51	17	4		29		29	61

made exclusively with paste, the equivalent paste made with OPC has been calculated with equation (2) (De Larrard, 1999).

$$f_{c\ \mathrm{paste}} \approx 11.4 \cdot Rc_{28} \cdot \left(\frac{V_{\mathrm{cement}}}{V_{\mathrm{paste}}}\right)^{2.85} \tag{2}$$

The environmental data for 1 kg of various concrete components used for OPC and geopolymer concrete products such as sand, gravel, soda powder, mineral additions are presented in Table 2. The life cycle inventory of cement production has been built by Chen (2009) and presented in Chen et al. (2010a). Data for aggregates production have been built by Chen (2009) with primary data from Martaud (2008) and the Ecoinvent database (Kellenberger and Althaus, 2003). Data for sodium powder manufacture have been calculated with the original system boundary of Althaus et al. (2007) and data for sodium silicate solution come from Fawer et al. (1999). Finally, for metakaolin there is a lack of controlled data. The data used by Duxson et al. (2007) come from industrial sources (Engelhard, 2009) and only gives CO_2 emissions without any comment on the viability of the measure. In this study, it has then been chosen to model the environmental impact of metakaolin by using an industrial report on the implementation cost of a metakaolin plant (NLK, 2002) that evaluates the energy demand for the production. To this energy consumption, the inventory of clay mining from ecoinvent has been added. The gas used for heating clay has been considered to come from biogas as it is done in a metakaolin plant in France (AGS, 2009). The results presented in Table 2 are in agreement with the industrial data from Engelhard (2009) for the global warming potential but cover all the other impact categories.

2.3. *Allocation procedure*

Industrial by-products, such as blast furnace slags (GBFS) and coal combustion fly ashes (FA) have lower environmental impact than cement if they are considered as waste from other industries (Gartner, 2004) or if their impacts are reduced to the energy and consumption required for their treatment (Kawai et al., 2005; Flower and Sanjayan, 2007). However, a recent European Union directive (EU, 2008) note that: *"a substance or object, resulting from a production process, the primary aim of which is not the production of that item, may be regarded as not being waste […] but as being a by-product only if the following conditions are met: a) Further use of the substance or object is certain; b) the substance or object can be used directly without any further processing other than normal industrial practise; c) the substance or object is produced as an integral part of a production process; and d) further use is lawful, i.e. the substance or object fulfils all relevant product, environmental and health protection requirements for the specific use and will not lead to overall adverse environmental or human health impacts."* This directive corresponds exactly to the context of use of supplementary cementitious materials such as GBFS and FA.

Actually, their further use is certain. It fulfils condition (a) of the European directive as in some part of Europe such as France, the production GBFS is fully used by cement industry and the use of FA in cement industry is equal to 130% of its yearly production as there exist FA stock.

GBFS are made from the extraction of iron from iron ore in blast furnace, whereas it is not possible to produce iron without producing GBFS. FA is made of the unburnt particulates (mainly siliceous components) that are released in exhaust gas when coal is burnt in coal power plants. For sanitary reasons, these gases have to be cleaned from ashes which are removed and concentrates to form FA. Thus both materials are produced as an integral part of a production process and then fulfil condition (b).

GBFS are vitrified with water and grinded. Fly ashes are only dried.Consequently, they can be used directly without any further processing other than normal industrial practise thus fulfilling condition (c).

Finally, the cement industry uses only GBFS and FA which comply with the existing standards regarding their suitability in terms of mechanical performance, risk for concrete durability and risk for the environment such as NF EN 450-1 standard for FA (CEN, 2007) and EN-197-1 for GBFS (CEN, 2001). Thus, condition (d) is fulfilled for the materials used in nowdays cement technology. Other GBFS and FA are still considered as waste.

These mineral additions must then be considered, in a European perspective, as by-products and not waste anymore and thus be affected by an allocation coefficient. Indeed, in LCA when a production system produces several products, material and energy flows and the associated environmental burdens must be partitioned between them (including the by-products) in order to accurately reflect their individual contribution to the environmental impacts. A recent study has evaluated the influence of different allocation procedures on the environmental impact of GBFS and FA when they are used as a replacement of clinker in blended cement (Chen et al., 2010b). As no specific method seems to be fully adequate (Ekvall and Finnveden, 2001), and as the ISO standard for LCA (ISO, 2006) states that when several alternative allocation procedures seem applicable, a sensitivity analysis should be conducted to illustrate the influence of the procedure on the results, it has been chosen in this study to test three allocation procedures.

i) In the first one, FA and GBFS are respectively considered as waste from coal power and iron industries. Their environmental burdens are therefore limited to the specific treatments needed for their use in concrete (grinding, drying and stock). As already stated, this is not appropriate in France and more generally in Western Europe. This method is however used in most recent studies dealing with environmental evaluation of FA and GBFS used in concrete (Gartner, 2004; Kawai et al., 2005; Flower and Sanjayan, 2007).
ii) The second allocation procedure is based on the relative mass ratio between the products and the co-products. Although SETAC strongly recommends to rely for the allocation procedure primarily on physicochemical considerations (Lundie et al., 2007), this procedure is not always usable as co-products have often similar impacts as the main product.
iii) The third allocation procedure is based on the economical values of products and by-products. This procedure is the one that is often preferred in allocations studies (Schuurmans et al., 2005) as it reflects the reality of the industrial process where the main products (iron and electricity) are the ones that form the main purpose of the industrial processes compared to the by-products (GBFS and FA respectively). With this allocation procedure, the main part of the environmental impact is affected to the main products and a small part on the by-products (Chen et al., 2010b).

To build the inventory, a distinction is made between the production of both products and by-products (Iron industry, coal power plants), and the specific treatments used for the by-products for their introduction in concrete. Input and output data are from Althaus (2003) for iron production and Dunlap (2003) for GBFS treatment, Doka and Hischier (2005) and Dones et al. (2007) for coal power plants process and Surschiste (2009) for FA treatment. For GBFS, the prices of iron were obtained from Dahlström and Ekins (2006) and Metal Bulletin (2010) and assumed to be equal to 450 €/t of crude iron. The prices for GBFS were fixed at 45 € (Ecocem, 2010; Vinci, pers. com). For FA, relative prices of electricity and fly ashes were obtained from EDF (2009) and Vinci (pers.com.) and fixed at 0.12 €/kWh and 25 €/t for electricity and FA respectively. More detailed explanations are provided by Chen et al. (2010b). The results of the various allocation procedures for 1 kg of FA and BFGS production are presented in Table 2.

2.4. Environmental impact calculation

In the present paper, environmental impacts are evaluated according to the baseline method of CML01 (Guinée et al., 2002) that evaluates 10 environmental impacts (abiotic depletion, global warming, ozone layer depletion, fresh and marine water ecotoxicity, terrestrial ecotoxicity, human toxicity, eutrophication, acidification and photochemical oxidation).

No classification aiming to assign inventory results to different impact categories has yet been done. This can introduce potential double counting and magnify the impacts of a particular burden (Reap et al., 2008). However, these classifications need a spatial differentiation (Finnveden and Nilsson, 2005), which is difficult in all inclusive studies. A site-generic impact modelling where all sources are considered to contribute to the same generic receiving environment has then been chosen (Guinée et al., 2002). The environmental impacts for the different materials and for the three allocation methods for FA and GBFS are presented in Table 2.

3. Results

In this section, the environmental impacts of geopolymer concrete types are first presented by considering GBFS and FA as waste. The effects of the allocation procedure on the environmental benefit of geopolymer concrete are studied afterwards.

3.1. Environmental analysis of fly ash based geopolymer concrete compared to OPC concrete

The mix-design given in Table 3 represents a mean value extracted from 49 mix-designs of Fly Ash based geopolymer concrete (Dombrowski et al., 2007; Hardjito and Rangan, 2005; Kong and Sanjayan, 2010; Lee and van Deventer, 2002; Olivia et al., 2008; Pacheco-Torgal et al., 2005; Rangan et al., 2005; Sathonsaowaphak et al., 2009; Sumajouw et al., 2007; Yang et al., 2008). A concrete made with OPC and displaying an equivalent mechanical strength shall contain 354 kg m^{-3} of cement. The environmental impact of these mix-designs is presented in Table 4. It is obvious that the use of a sodium silicate solution is responsible for the major part of the environmental impact in the case of geopolymer concrete (Table 4). When this type of geopolymer concrete is compared with hydraulic cement based standard concrete, it is obvious that this new type of binder allows for a strong reduction of the global warming potential. From 306 kg of equivalent CO_2 per m^3 for OPC based concrete, the geopolymer concrete releases only 169 kg of equivalent CO_2 per m^3, which represents a saving of 45%. However, it is interesting to note, that this value is not so different from CO_2 emission reduction reached with an improvement in cement technology efficiency, where a 50% of reduction can be achieved by using existing technologies (Gäbel and Tillman, 2005; Habert et al., 2010). Therefore, even if this new technology provides an important reduction in CO_2 emission, it is not significantly different from solutions where no radical technical changes are needed and where only technological improvements and clinker substitution are promoted. These eco-efficiency solutions, rather than revolutionary options, are traditionally preferred in construction industry where there exists an understandable conservative approach with regard to new products. Therefore, Fly Ash based geopolymer concrete as manufactured today does not represent the breakthrough technology, which could allow the concrete industry to reduce CO_2 emissions by a factor 4.

Concerning the others environmental impact categories, Fig. 2 shows that geopolymer concrete systematically shows higher impacts than OPC concrete, due to the use of sodium silicate solution. The use of sodium silicate solution in concrete to substitute OPC shall then induce a pollution transfer from global warming considerations towards all other environmental impacts.

3.2. Environmental profile of different geopolymer concrete types made with fly ash, slag or metakaolin

The environmental impact of 49, 13 and 17 geopolymer concrete made respectively with FA, GBFS and MK have been studied (Buchwald et al., 2007; Collins and Sanjayan, 1999; Dombrowski et al., 2007; Hardjito and Rangan, 2005; Kong and Sanjayan, 2010; Latella et al., 2008; Lee and van Deventer, 2002; Meliani, 2010; Olivia et al., 2008; Pacheco-Torgal et al., 2005; Rangan et al., 2005; Rovnaník, 2010; Sathonsaowaphak et al., 2009; Sumajouw et al., 2007; Weil et al., 2009; Yang et al., 2008). The

Table 2
Environmental impact for 1 kg of various concrete components. Calculations are made with CML01.

CML 2001		Fly Ash			Blast Furnace Slag Granulated			Silica Fume		
		No allocation	Mass allocation	Economic allocation	No allocation	Mass allocation	Economic allocation	No allocation	Mass allocation	Economic allocation
Abiotic depletion	kg Sb eq.	2.02×10^{-4}	1.95×10^{-2}	1.79×10^{-3}	2.88×10^{-4}	1.21×10^{-2}	1.71×10^{-3}	1.99×10^{-6}	3.43×10^{-2}	1.0×10^{-2}
Global warming potential	kg CO_2 eq.	5.26×10^{-3}	2.51	2.10×10^{-1}	1.69×10^{-2}	1.25	1.67×10^{-1}	3.13×10^{-4}	4.12	1.20
Ozone layer depletion	kg CFC-11 eq.	3.35×10^{-9}	2.43×10^{-8}	5.07×10^{-9}	4.11×10^{-9}	2.45×10^{-8}	6.57×10^{-9}	1.21×10^{-11}	2.59×10^{-7}	7.56×10^{-8}
Human toxicity	kg 1,4-DB eq.	1.58×10^{-3}	5.01×10^{-1}	4.25×10^{-2}	8.24×10^{-3}	3.99×10^{-1}	5.56×10^{-2}	1.39×10^{-1}	1.74	5.08×10^{-1}
Fresh water aquatic ecotox.	kg 1,4-DB eq.	1.76×10^{-4}	3.17×10^{-2}	2.76×10^{-3}	1.92×10^{-3}	2.01×10^{-1}	2.60×10^{-2}	1.84×10^{-4}	5.69×10^{-2}	1.66×10^{-2}
Marine aquatic ecotoxicity	kg 1,4-DB eq.	1.93	2.97×10^{-3}	2.45×10^{-2}	10	5.20×10^{-2}	7.18×10^{-1}	2.50×10^{-1}	1.80×10^{-4}	5.25×10^{-3}
Terrestrial ecotoxicity	kg 1,4-DB eq.	1.68×10^{-5}	4.48×10^{-4}	3.83×10^{-4}	1.42×10^{-4}	3.35×10^{-3}	5.31×10^{-4}	4.59×10^{-6}	1.38×10^{-2}	4.05×10^{-3}
Photochemical oxidation	kg C_2H_4 eq.	1.93×10^{-6}	6.62×10^{-4}	5.60×10^{-5}	1.59×10^{-5}	8.39×10^{-4}	1.16×10^{-4}	7.08×10^{-8}	2.26×10^{-3}	6.59×10^{-4}
Acidification	kgSO_2 eq.	3.32×10^{-5}	1.92×10^{-2}	1.60×10^{-3}	3.46×10^{-4}	4.85×10^{-3}	8.91×10^{-4}	1.90×10^{-6}	2.42×10^{-2}	7.07×10^{-3}
Eutrophication	kg PO_4^{3-} eq.	4.94×10^{-6}	1.06×10^{-3}	9.12×10^{-5}	1.05×10^{-5}	6.77×10^{-4}	9.11×10^{-5}	1.29×10^{-7}	1.87×10^{-3}	5.48×10^{-4}

environmental impact of these materials are calculated and compared with a 100% OPC concrete displaying an equivalent mechanical strength and also with a concrete made with 30% of substitution of the cement with the same mineral addition (FA, GBFS and MK). For each type of geopolymer concrete, a mean impact and a standard deviation are evaluated. The impact of standard OPC concrete is considered as a reference and set at a 100% value and relative values are calculated for each mix-design. Results are presented in Fig. 3. These results show that FA and GBFS based geopolymer concretes have a lower global warming impact than MK based geopolymer concretes. This result can be understood by the fact that to reach sufficient mechanical strength, the Si/Al molar ratio has to be around 2 (Rowles and O'Connor, 2003) whereas this ratio is close to 1 in MK. This leads to the addition of an important quantity of sodium silicate in the solution. On the contrary, Si/Al molar ratios of FA or GBFS are higher, which allows for a reduction in sodium silicate use, when these geopolymer concretes are compared with concretes in which 30% of the cement is substituted by FA, GBFS or MK.

Table 3
Mix-design of a standard FA based geopolymer concrete. These are mean values from the works of Lee and van Deventer (2002); Hardjito and Rangan (2005); Pacheco-Torgal et al. (2005); Rangan et al. (2005); Dombrowski et al. (2007); Sumajouw et al. (2007); Olivia et al. (2008); Yang et al. (2008); Sathonsaowaphak et al. (2009); Kong and Sanjayan (2010).

Gravel	Sand	FA	NaOH powder	Na Silicate solution	Water	Admixture	Compressive strength	Cement equivalent
1292	554	408	17	103	26	6,1	36	354

Moreover BFGS based geopolymer, is the only geopolymer type that has a lower impact than pure OPC concrete for more than the global warming potential (acidification and ozone layer depletion). It is also the only geopolymer type that still has a significant lower global warming potential impact than blended cement. However, it is known that the use of GBFS is not limited to 30% substitution as it has been done for blended cement in Fig. 3 and can be increased up to 90% and some type of cement are already commercialised and standardised with this amount of substitution (CEM III, CEN, 2001). In these conditions, it can be questioned whether if it is a true long term perspective to promote GBFS geopolymers since GBFS is already used as a hydraulic binder and is not available in large quantities in european regions. This aspect will be discussed at the end of the present paper.

3.3. *Influence of the allocation procedure on the impact of different types of geopolymer concrete*

Normalisation procedure which is an optional step in LCA (ISO, 2006), can be used to highlight the most important impact categories. In that case, results can be presented by dividing the impact of concrete production by the impact from all activities of a European citizen during one year. When the activities of Western Europe in 1995 are used (Huijbregts et al., 2003), the four impact categories

Table 4
Environmental impact for a standard FA based geopolymer concrete. Details of the different component are shown. It is compared with a 100% OPC concrete displaying the same mechanical strength.

	Abiotic Depletion kg Sb eq	Global warming potential kg CO_2 eq	Ozone layer depletion kg CFC-11 eq	Human toxicity kg 1,4-DB eq	Fresh water ecotoxicity kg 1,4-DB eq	Marine ecotoxicity kg 1,4-DB eq	Terrestrial ecotoxicity kg 1,4-DB eq	Photochemical oxidation kgC_2H_4	Acidification kg SO_2 eq	Eutrophication kg PO_4^{2-} eq
Sand and gravel	4.72×10^{-2}	6.87	6.73×10^{-7}	4.64	1.05	2.79×10^{-3}	4.25×10^{-2}	1.60×10^{-3}	3.85×10^{-2}	6.97×10^{-3}
Fa	8.25×10^{-2}	2.14	1.37×10^{-6}	6.44×10^{-1}	7.18×10^{-2}	7.86×10^{-2}	6.84×10^{-3}	7.88×10^{-4}	1.35×10^{-2}	2.01×10^{-3}
NaOH powder	2.72×10^{-2}	3.71×10^{-1}	2.28×10^{-6}	15.84	3.98	7.87×10^{-3}	7.72×10^{-1}	7.67×10^{-3}	1.78×10^{-1}	1.34×10^{-2}
Na Silicate	7.44×10^{-1}	117.8	9.08×10^{-6}	82.75	21.84	3.42×10^{-4}	9.23×10^{-1}	2.51×10^{-2}	5.37×10^{-1}	5.10×10^{-2}
Water	4.50×10^{-9}	3.99×10^{-3}	3.51×10^{-10}	2.55×10^{-3}	1.28×10^{-3}	4.75	4.46×10^{-5}	2.57×10^{-6}	3.79×10^{-5}	2.61×10^{-6}
Admixtrure	5.22×10^{-2}	4.56	5.17×10^{-7}	1.58	6.83×10^{-2}	2.53×10^{-2}	3.07×10^{-2}	1.40×10^{-3}	5.23×10^{-2}	6.28×10^{-3}
OPC	5.64×10^{-1}	299.1	8.07×10^{-6}	14.26	1.46	6.89×10^{-3}	4.16×10^{-1}	1.51×10^{-2}	4.09×10^{-1}	6.13×10^{-2}
Geopolymer concrete	1.19	168.5	1.39×10^{-5}	105.4	27.01	4.59×10^{-4}	1.77	3.65×10^{-2}	0.82	7.96×10^{-2}
OPC concrete	0.61	305.9	8.74×10^{-6}	18.90	2.52	9.68×10^{-3}	0.45	1.67×10^{-2}	0.45	6.83×10^{-2}

Soda, powder	Sodium silicate solution (37%)	CEMI	Limestone filler	Metakaolin	Gravel	Sand	Water	Admixture
1.64×10^{-2}	7.22×10^{-3}	1.59×10^{-3}	2.02×10^{-4}	1.68×10^{-4}	2.95×10^{-5}	1.64×10^{-5}	1.93×10^{-6}	8.56×10^{-3}
2.24	1.14	8.44×10^{-1}	3.51×10^{-2}	9.24×10^{-2}	4.29×10^{-3}	2.40×10^{-3}	1.55×10^{-4}	7.49×10^{-1}
1.38×10^{-7}	8.82×10^{-8}	2.28×10^{-8}	3.04×10^{-9}	1.52×10^{-9}	4.08×10^{-10}	2.63×10^{-10}	1.36×10^{-11}	8.48×10^{-8}
9.57×10^{-1}	8.03×10^{-1}	4.02×10^{-2}	1.77×10^{-2}	2.36×10^{-2}	2.90×10^{-3}	1.61×10^{-3}	9.87×10^{-5}	2.59×10^{-1}
2.40×10^{-1}	2.12×10^{-1}	4.14×10^{-3}	4.54×10^{-3}	3.28×10^{-3}	6.83×10^{-4}	3.15×10^{-4}	4.95×10^{-5}	1.12×10^{-2}
4.75×10^{-2}	3.32×10^{-2}	1.94×10^{-1}	1.05×10^{-1}	4.59	1.85	7.20×10^{-1}	1.84×10^{-1}	4.15×10^{-1}
4.66×10^{-2}	8.96×10^{-3}	1.17×10^{-3}	2.04×10^{-4}	3.23×10^{-4}	2.85×10^{-5}	1.02×10^{-3}	1.73×10^{-6}	5.04×10^{-3}
4.63×10^{-4}	2.43×10^{-4}	4.26×10^{-5}	6.47×10^{-6}	1.09×10^{-5}	1.01×10^{-6}	5.26×10^{-7}	9.98×10^{-8}	2.29×10^{-4}
1.07×10^{-2}	5.22×10^{-3}	1.15×10^{-3}	1.61×10^{-4}	3.24×10^{-4}	2.34×10^{-5}	1.49×10^{-3}	1.47×10^{-6}	8.58×10^{-3}
8.10×10^{-4}	4.95×10^{-4}	1.73×10^{-4}	2.87×10^{-5}	4.89×10^{-5}	4.15×10^{-6}	2.90×10^{-6}	1.01×10^{-7}	1.03×10^{-3}

that are relevant for concrete are: abiotic depletion, global warming potential, marine ecotoxicity and acidification (Chen et al., 2010a). To clarify our point and facilitate the reading we decided to select these four impact categories in the next section.

In Fig. 4, the impacts of geopolymer and blended cement concrete types are presented relatively to a pure OPC concrete for the 3 allocation procedures. The results show that for 3 out of 4 environmental impact categories (abiotic depletion, marine ecotoxicity and acidification), whatever the allocation procedure, the environmental impact of the production of the geopolymer concrete is higher than for the production of a blended cement based concrete with the same compressive strength. For the global warming potential, FA based geopolymers have lower environmental impacts only if FA is considered as a waste (no allocation), which will probably soon not be the case anymore in Europe (EU, 2008). GBFS based geopolymers are the only concrete that still have a lower global warming impact than usual concretes even when an economic allocation is assumed. However, they have a much higher impact if a mass allocation procedure is assumed.

Therefore, geopolymer concrete seem much more sensitive to allocation procedure than blended cement based concrete. This can be explained by the fact that the main component of the geopolymer paste is the mineral addition (FA or GBFS) whereas for blended cement, the main component is still clinker (70%) and the mineral addition only represents 30% of the cement. MK based concrete are not affected by this allocation because MK is not a by-product but is an industrial product itself with no associated by-products. However, as shown previously MK based geopolymer have higher impacts than standard concrete.

4. Discussion and perspectives

This study shows that within current mix-design trends, geopolymer concrete made from FA and GBFS results in lower CO_2 emissions than OPC concrete. However this reduction is not sufficient enough to achieve the factor 4 objectives. Within the literature there are a few cases of geopolymer concrete made from GBFS which reach these objectives. However they only do so by not taking into account the impact allocation of the by-products used, in this case GBFS. This study also highlights that the environmental impact of geopolymer concrete stems from the use of the sodium silicate solution. If, in order to reduce global warming, standard OPC concrete was replaced by geopolymer concrete, the sodium silicate solution it contains would in fact lead to a pollution transfer within all of the other environmental impact categories.

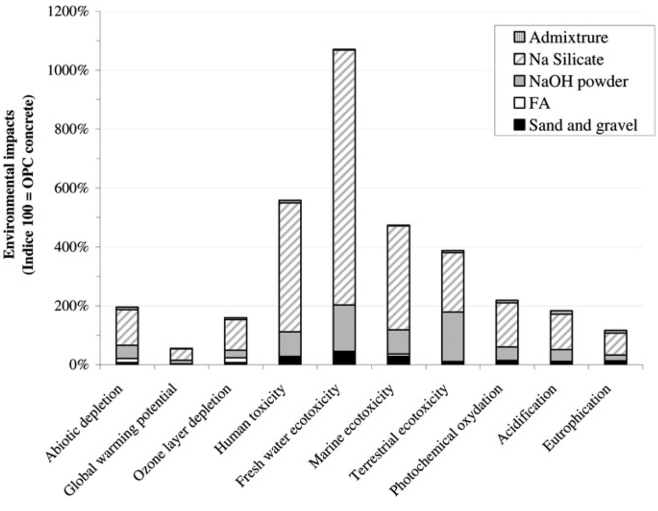

Fig. 2. Eco-profile of Fly Ash based geopolymer concrete compared to OPC based concrete. The pure OPC concrete binder is made exclusively with CEM I whereas current concrete binder is prepared with 70% CEM I and 30% fly ash.

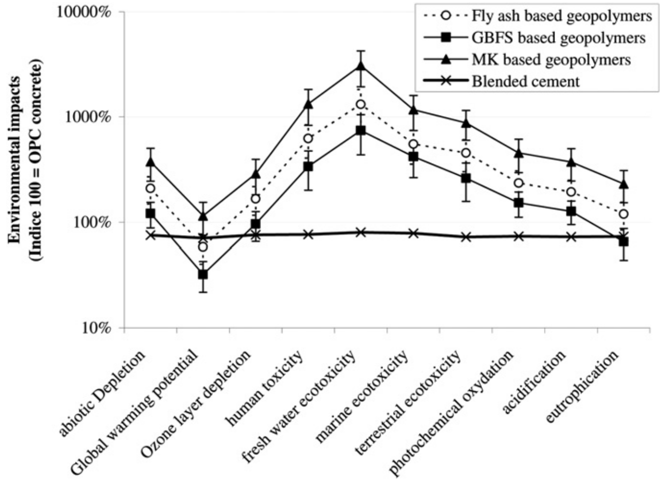

Fig. 3. Eco-profile of different geopolymer concrete types compared to OPC based concretes. The pure OPC concrete binder is made exclusively with CEM I whereas current concrete binder is on average prepared with 70% CEM I and 30% mineral addition.

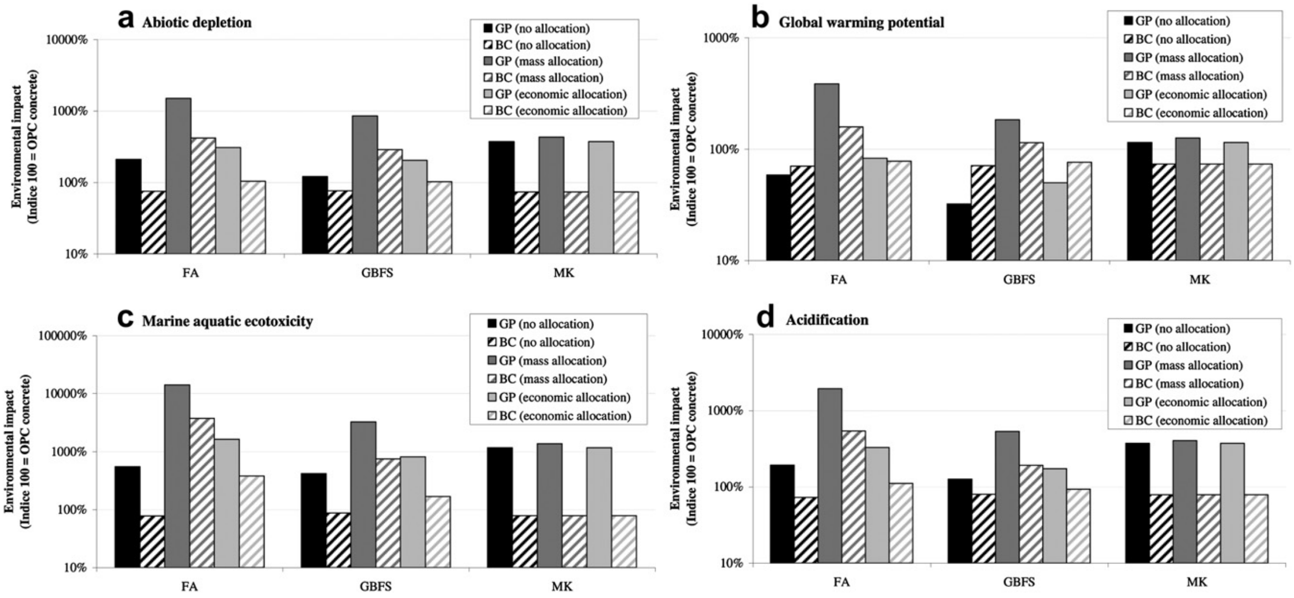

Fig. 4. Comparison of the impact of alternative concretes: Geopolymer (GP) and blended cement (BC) based concrete types, for the different allocation procedure (no allocation, economic and mass allocation). The four environmental impacts are: a) abiotic depletion, b) Gobal warming potential, c) Marine ecotoxicity, d) acidification. Fly Ash (FA), blast furnace slags (GBFS) and metakaolin (MK) are the studied mineral additions.

The best way for the concrete industry to reach its current CO_2 objectives, would be to produce geopolymer concrete from raw material recognised as industrial waste and therefore not considered to have an allocation impact. Furthermore the industry should choose a waste material with a suitable Si/Al molar ratio in order to reduce the use of sodium silicate solution when producing geopolymer concrete.

In fact, geopolymer technology allows us to use waste that is unsuitable in other industries and that can therefore be considered as veritable waste instead of a by-product from an LCA point of view. For example while magnesium iron slags (Zosin et al., 1998), ferronickel slags (Komnitsas et al., 2007) or tungsten mine waste mud (Pacheco-Torgal et al., 2007) are of little or no benefit in blended cement technology, they can be used successfully as geopolymeric binders. Slag based geopolymer concrete only requires small amount of sodium silicate and therefore has the lowest environmental impact. Furthermore, when using the waste materials listed above, it avoids their disposal and the associated environmental impacts such as toxic leakages of mine waste.

Concerning metakaolin based geopolymer concrete it has been shown that due to the low Si/Al ratio in MK a high amount of sodium silicate is required and induces a high environmental impact. One research perspective involves using other thermally activated clays with a higher Si/Al ratio than MK (Buchwald et al., 2009; Mackenzie, 2009) or combining MK with slag which acts as geopolymer precursor (Davidovits, 2009). The solution, proposed by Davidovits, has the advantage of using less slag than pure GBFS geopolymer concrete. This is beneficial from an environmental point of view if one considers GBFS as a by-product which more so is not easily available in Western Europe. This alternative also requires less sodium silicate compared with pure MK based geopolymer, this is illustrated in Fig. 5. The figure plots the relative impact of the different GBFS based geopolymer concrete tested in this study compared with blended cement concrete (with 30% mineral addition). In doing so for three different allocation procedures it compares the relative environmental advantage or disadvantage of using geopolymer technology instead of the current hydraulic model. This relative environmental advantage is shown as a percentage whilst the numbers allocated to the various geopolymer mix-designs reproduce the order in which they are presented in Table 1. It is interesting to note two trends: geopolymer concretes such as GP1 GP2 or GP13 and a second trend exemplified by GP11 and GP12. The environmental impact of the geopolymer concrete in the first case increases when instead of using “no allocation” a “mass allocation” procedure is used. The second trend shows concretes with a lower environmental impact than blended cement based concrete once a mass allocation procedure is applied. The concretes included in this second trend are made with MK and slags. For example GP11 which has the lowest environmental impact as soon as an economic or mass allocation procedure is used, is made up of 50% MK and 50% GBFS (Table 1). As shown above using MK and GBFS reduce the environmental impact because it has a lower content of GBFS than a pure GBFS geopolymer (GP1) and furthermore a much lower content in sodium silicate solution than a pure MK based geopolymer.

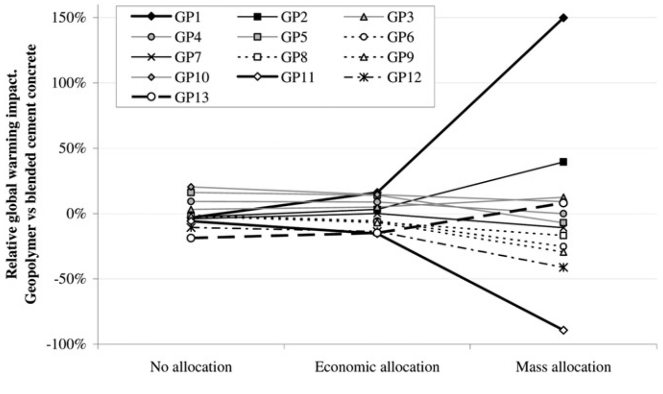

Fig. 5. Influence of the allocation procedure on the relative global warming impact of the different GBFS geopolymer concretes tested in the study compared with a blended cement concrete (with 30% of GBFS).

Another research perspective would be to use particle technology more fruitfully. As Provis et al. (2010) recently noted, most of what we know about the mix-design of OPC based concrete from particle technology could also be used to improve geopolymer concrete. For example, improving the granular distribution within the geopolymer material would increase the granular packing, the material would therefore require less active binder. These binders could also be partially replaced with more environmentally friendly filler particles as it has been shown that secondary phases do not affect the geopolymerisation reaction (Zibouche et al., 2009). Finally sodium silicate solution can also be replaced with sodic slags. This has already been developed in the Geocistem (1997) as has the use of specific sodic waste (Laldji and Tagnit-Hamou, 2007).

5. Conclusion

This study used the Life Cycle Assessment methodology to carry out a detailed environmental evaluation of the production of geopolymer concrete. Our results show that the production of most standard types of geopolymer concrete has a slightly lower impact on global warming than standard Ordinary Portland Cement (OPC) concrete. However they also reveal that the production of geopolymer concrete has a higher environmental impact regarding other impact categories than global warming. This is due to the heavy effects of the production of the sodium silicate solution. Geopolymer concrete made from fly ashes or granulated blast furnace slags based require less of the sodium silicate solution in order to be activated. They therefore have a lower environmental impact than geopolymer concrete made from pure metakaolin. However, when the production of fly ashes and granulated blast furnace slags is taken into account during the life cycle assessment (using either an economic or a mass allocation procedure), it appears that geopolymer concrete has a similar impact on global warming than standard concrete. This study highlights that future research and development in the field of geopolymer concrete technology should focus on two potential solutions. First of all the use of industrial waste that is not recyclable within other industries and secondly on the production of geopolymer concrete using a mix of blast furnace slag and activated clays. Furthermore geopolymer concrete production would gain from using waste material with a suitable Si/Al molar ratio in order to minimise the amount of sodium silicate solution used. Finally, by taking into account mix-design technology, which has already been developed for OPC concrete, the amount of binder required to produce a geopolymer concrete could be reduced. It is only by adopting these directions that geopolymer concrete could allow us to achieve the current objectives for a long term reduction of CO_2 emissions.

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