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# Durability Related Functional Units for Life Cycle Assessment of High-Volume Fly Ash Concrete

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## ABSTRACT

High-volume fly ash (HVFA concrete) concrete has been developed in order to reduce the environmental impact of cement production. By replacing at least 50% of ordinary portland cement with pozzolanic fly ash, an important reduction in greenhouse gas emissions can be achieved. However, this proposition is only valid if HVFA concrete is as durable as portland cement concrete. In order to deal with differences in durability, the amount of concrete needed in a 1 m³ structure with a service life of 50 years is choosen as functional unit for life cycle assessment (LCA). This way, additional emissions caused by structure replacement over time are included. Accelerated test procedures for carbonation as well as freezing and thawing with deicing salts were used. Based on long-term damage prediction, a significant reduction in environmental impact is observed for concrete mixtures with 50% fly ash when subjected to carbonation. However, freezing/thawing resistance is poor.

## INTRODUCTION

Since the end of the twentieth century, more and more attention is being paid to the environmental impact of goods and services provided by all types of industries, including the concrete manufacturing business. In 2002, a research report indicated that 5% of the global anthropogenic $CO_2$ emissions were at the expense of the cement industry [Humphreys and Mahasenan 2002]. In Belgium 57% of these emissions are the result of decarbonation during clinker production in cement kilns [Febelcem 2006]. The amount of this process bound $CO_2$ can be reduced only if the demand for ordinary portland cement (OPC) diminishes in the future. Imposing a restraint on the use of concrete however, is not realistic. Therefore, the concrete manufacturing business is being encouraged to replace more OPC with by-products from other industries, such as blast-furnace slag, silica fume and fly ash. Because of the large application of concrete worldwide, even small reductions in cement use can make a major difference [Flower and Sanjayan 2007]. In 1986, Malhorta made an important contribution with the development of high-volume fly ash concrete in which at least 50% of the binder material consists of fly ash with a low calcium content [Malhorta and Mehta 2005].

However, little research has been conducted on environmental impact quantification of concrete with high cement replacment levels. In order to evaluate the product correctly a life cycle assessment (LCA) approach is recommended. This methodology normally requiresa full description of the product from the cradle to the grave, i.e. from extraction of raw materials to waste treatment. However, a cradle-to-gate approach is sometimes used within companies [Desmyter and Martin 2001], since the emphasis in that case is on optimizing the production process and not the product life cycle and the end-of-life scenarios. For this research, a variant of the latter approach has been used. Traditionally, a LCA consists of four major steps: definition of goal and scope, inventory analysis, impact analysis and interpretation [ISO 14040 2006]. The first one is a very important one since it includes the definition of the functional unit. This reference unit needs to be chosen in such a way that objective comparison between concrete mixtures with a different durability is possible. In this article, the amount of concrete needed in a 1 m³ structure with a service life of 50 years, is adopted for this unit. This kind of approach is considered to be more correct than simply comparing 1 m³ OPC concrete with 1 m³ HVFA concrete, since the latter does not include additional concrete manufacturing due to structure replacement over time. Values for the functional unit can be obtained from durability tests which are representative for specific realistic environments. This research comprises the evaluation of several concrete mixtures with a varying fly ash content (0, 35, 50, 55 and 67%) when exposed to carbonation as well as to freezing and thawing with deicing salts. In case of an acceptable durability the environmental impact of the mixtures was calculated with the LCA software SimaPro 7.1.

## CONCRETE MIXTURES

In total, seven concrete mixtures were tested for durability: one reference, one mixture with 35% (F35), three mixtures with 50% (F50-1, F50-2, F50-3), one mixture with 55% (F55) and one mixture with 67% (F67) of the cement CEM I 52.5 N replaced by fly ash. All mixture proportions together with admixture dosage, initial air content, slump and compressive strength class are given in Table 1. F0, F35, F50-1 and F67 were manufactured first. Based on the experimental results, the mix design for HVFA concrete was adjusted to improve its durability performance. With this purpose, F50-2, F50-3 and F55 were made.

**Table 1. Mixture Proportions and Properties**

| | **F0** | **F35** | **F50-1** | **F50-2** | **F50-3** | **F55** | **F67** |
|---|---|---|---|---|---|---|---|
| **Sand 0/4 (kg/m³)** | 686 | 668 | 660 | 771 | 652 | 768 | 652 |
| **Aggregate 2/8 (kg/m³)** | 451 | 437 | 432 | 414 | 470 | 412 | 427 |
| **Aggregate 8/16 (kg/m³)** | 694 | 678 | 668 | 602 | 613 | 600 | 660 |
| **CEM I 52.5 N (kg/m³)** | 400 | 260 | 200 | 200 | 212.5 | 180 | 132 |
| **Fly ash (kg/m³)** | 0 | 140 | 200 | 200 | 212.5 | 220 | 268 |
| **Water (kg/m³)** | 160 | 160 | 160 | 150 | 160 | 150 | 160 |
| **W/CM (-)** | 0.400 | 0.400 | 0.400 | 0.375 | 0.375 | 0.375 | 0.400 |
| **AEA (ml/kg CM)** | 0.7 | 0.9 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 |
| **SP (ml/kg CM)** | 2.1 | 1.2 | 1.2 | 5.2 | 5.9 | 5.2 | 0.9 |
| **Air content (%)** | 5.6 | 4.1 | 5.5 | 3.4 | 3.2 | 3.7 | 4.1 |
| **Slump** | S4 | S4 | S3 | S1 | S3 | S1 | S3 |
| **Strength class** | C45/55 | C30/37 | C25/30 | C30/37 | C40/50 | C35/45 | C16/20 |

The mixtures F50-1, F50-2 and F50-3 mutually differ in cementitious material (CM) content and water-to-cementitious material ratio (W/CM). The mixtures were air entrained using the MICRO-AIR 103 con. 4% admixture (AEA). Because the concrete compositions are characterised by a low water content, GLENIUM 51 con. 35%, a polycarboxylate type superplasticizer (SP), was also added.

## CONCRETE ENVIRONMENTS

Depending on a structure's environment, concrete is being exposed to different deterioration processes. For this reason the concept of exposure classes has been proposed within the standards NBN EN 206-1 [2001] and NBN B15-001 [2004]. With a view to concrete composition design, the required minimum cement content ($C_{min}$), water-to-cement ratio (W/C), strength class and air content for each exposure class have also been defined in NBN B15-001 [2004]. The concrete mixtures investigated in this paper were subjected to durability tests which are representative for a XC1 and a XF4 environment. The first one includes permanently dry atmospheres as can be found inside buildings with a low air humidity. The second one represents an environment with a high water saturation and exposure to deicing salts, such as road and bridge decks. The prescribed concrete types for these environments are given in Table 2. The table shows that mixture F0 meets the criteria of both exposure classes and can thus be seen as an appropriate reference for comparison.

### Table 2. Concrete Types for XC1 and XF4 Environments According to NBN B15-001 (2004)

| **Exposure class** | **Concrete type** | **$C_{min}$ ($kg/m^3$)** | **W/C (-)** | **Strength class (-)** | **Air content (%) 11.2 mm $\leq$ $D_{max} \leq$ 16 mm*** |
|---|---|---|---|---|---|
| XC1 | T(0.65) | 260 | 0.65 | C16/20 | - |
| XF4 | T(0.45) | 340 | 0.45 | C35/45 | 5 |
| | T(0.50)A | 320 | 0.50 | C25/30 | 5 |

* the required air content depends on the maximum grain size $D_{max}$ of the aggregate

On the other hand, the system of concrete types is not applicable for HVFA mixtures since the maximum fly ash content in NBN B15-001 [2004] is strongly limited by the k value concept. As a consequence HVFA concrete is only accepted as a worthy alternative to OPC concrete if proof exists of an equivalent concrete performance in comparison with its proper reference.

## EXPERIMENTAL DETAILS

After concrete manufacturing, all cast specimens were kept in a climate chamber with a temperature of 20 °C and a relative humidity of 95%. Demoulding took place the next day whereupon the specimens were stored in the climate chamber again until the appropriate testing age was reached. Carbonation experiments were conducted at age of 28 and 91 days. Salts scaling resistance was only evaluated after 28 days of curing.

## Carbonation

An accelerated carbonation test on three (n = 3) cubes with a 100 mm side has been adopted for all mixtures. After 21 and 84 days, an impermeable coating was put on top of 5 of the 6 cube surfaces in order to establish an unidirectional flow throughout the specimens during the experiment. Then, after 28 and 91 days, three cubes of each composition were stored in a carbonation chamber with a 10% $CO_2$ concentration by volume and a relative humidity of 60% at 20°C. At different time intervals (after 1, 3, 6, 10 and 14 weeks of exposure) a 10 mm thick slice was taken from all cubes perpendicular to the exposed surface, whereupon the carbonation front was visualized with a phenolphtalein solution. The color of this indicator turns from purple to colorless when the pH drops below the point of transition situated between pH 8.2 and 10. Since reinforced concrete requires a pH between 9 and 13 in order to maintain the protective passivation layer upon the steel reinforcements, the use of phenolphtalein gives a more or less adequate carbonation measurement.

Plotting the carbonation depth x (mm) in function of the square root of time t (weeks) results in a pronounced linear correlation. The equation slope counts as a measure for the accelerated carbonation velocity and is defined as the carbonation coefficient $A_{acc}$. This square-root-t-law is generally used to compare carbonation resistance of concrete [Khunthongkeaw et al. 2006].

However, long-term carbonation prediction of steel reinforced concrete can not be solely based on this value since the normal carbon dioxide concentration in air is much lower – about 0.03% by volume under normal circumstances [Sisomphon & Franke, 2007] – than during the experiment. A similar value has been assumed in Audenaert [2006] and Meyer, et al. [1967]. Furthermore, a value of 0.3% by volume has been adopted for an urban environment. In Sisomphon and Franke [2007] the depth of carbonation under real environmental conditions is approximated from the results of the penetration in accelerated tests using Fick’s law of diffusion. In the end, the carbonation coefficients are expressed in terms of carbon dioxide concentrations alone. Given this relation, the carbonation coefficient $A_{env}$ in a normal concrete environment, is calculated from the following equation (1):

$$\frac{A_{acc}}{A_{env}} = \frac{\sqrt{c_{1,acc}}}{\sqrt{c_{1,env}}} \quad (1)$$

with $c_{1,acc}$ the experimental $CO_2$ concentration (10% by volume) and $c_{1,env}$ the concentrations in a normal (rural) and urban environment, which respectively amount to 0.03 and 0.3% by volume. Then, the carbonation depth after 50 years $x_{50\ years}$ (mm) is obtained from the square-root-t-law mentioned above with $A_{acc}$ replaced by $A_{env}$:

$$x_{50\,\text{years}} = A_{env}\sqrt{50\ \text{years}} \quad (2)$$

When this depth exceeds the concrete cover over the steel reinforcements and carbonation initiated corrosion may be feared, the structure will need replacement within the 50 years life span. If so, additional concrete manufacturing will have to be included within the LCA.

### Freezing and thawing with deicing salts

This experiment was performed for six (n = 6) cylindrical concrete specimens per testing age and per composition. After 21 and 84 days three cores with a diameter of 100 mm were drilled out of three cubes with a 150 mm side. Then, two 50 mm high cylinders containing a moulded surface of the original cube were sawn from each core and glued into a piece of insulated PVC tube with epoxy. At the age of 28 and 91 days, a 5 mm thick 3% NaCl solution was put on top of the test surfaces whereupon the six samples were subjected to 28 freezing and thawing cycles in a freezing chamber. The applied temperature regime varied from 20 to -18°C over 24 hours in correspondance with NBN EN 1339 [2003]. Every 7 days, scaled off material was collected, dried and weighed in order to determine the mass loss per unit area (Δm) as a function of time. According to the European standard the overall mass loss after 28 days should be less than 1 kg/m². If not, the structure will need replacement within its life span. A concrete life span prediction should be based upon the average number of severe freezing days a year. However, such an estimation for HVFA concrete is only useful if the salt scaling resistance does not differ too much from the reference concrete. If not, the environmental performance is obviously poor and attention should be paid first to optimalisation of the mixture design.

## LCA METHODOLOGY AND RESULTS

### Definition of goal and scope

This analysis was conducted in order to quantify the environmental benefits of an OPC replacement with fly ash. Special attention was paid to its influence on the reduction of greenhouse gas emissions, since this is the reason why HVFA concrete was developed in the first place. The obtained results are representative for concrete with steel reinforcements. The use of HVFA concrete in unreinforced structures has already been proven successful. Malhorta and Mehta [2005] give the example of the BAPS Temple and Cultural Complex in Chicago with 250 unreinforced, cast-in-place concrete caissons in HVFA concrete. The main goal of this study however, is an environmental evaluation of the material in more demanding steel reinforced applications.

As only the total amount of concrete needed for the 1 m³ structure with a 50 years life span was taken into account, the scope is characterised by a pseudo cradle-to-gate approach. Environmental impacts attributed to implementation of the material on site, end-of-life scenarios and structure replacement over time were not included. Emissions originating from the production of all concrete constituents were incorporated in the analysis, except for fly ash. This by-product from coal-fired electrical power plants was not included, since its environmental impact is at the expense of the electricity companies. Only transport from the power plant to the concrete manufacturer was taken into account. The functional unit was chosen as stated in the introduction section and requires experimental results from durability testing.

### Carbonation

Carbonation coefficients representative for both a rural (0.03% $CO_2$) and urban (0.3% $CO_2$) environment after 28 and 91 days of curing are presented in Table 3.

**Table 3. Carbonation Coefficients A (mm/√years) for Rural and Urban Environments after 28 and 91 Days of Curing (n = 3)**

| A (mm/√years) | F0 | F35 | F50-1 | F50-2 | F50-3 | F55 | F67 |
|---|---|---|---|---|---|---|---|
| **28 days** | | | | | | | |
| rural | 0.00 | 0.69 | 2.34 | 1.79 | 1.55 | 2.86 | 5.26 |
| stdv | 0.00 | 0.02 | 0.28 | 0.02 | 0.10 | 0.06 | 0.19 |
| urban | 0.01 | 2.17 | 7.41 | 5.67 | 4.90 | 9.05 | 16.64 |
| stdv | 0.00 | 0.05 | 0.89 | 0.06 | 0.32 | 0.20 | 0.60 |
| **91 days** | | | | | | | |
| rural | 0.01 | 0.22 | 1.60 | 1.48 | 0.92 | 2.06 | 4.72 |
| stdv | 0.00 | 0.06 | 0.07 | 0.15 | 0.16 | 0.16 | 0.19 |
| urban | 0.02 | 0.69 | 5.07 | 4.69 | 2.92 | 6.52 | 14.94 |
| stdv | 0.00 | 0.20 | 0.22 | 0.49 | 0.50 | 0.51 | 0.60 |

Standard deviations (stdv) on the individual values are also given. The results clearly show that carbonation rates increase with increasing fly ash content. Comparison between carbonation coefficients obtained at the age of 28 and 91 days indicates that for fly ash concrete an important reduction in carbonation rate is realised with a longer curing period. Of all concrete compositions with a 50% fly ash content, mixture F50-3 with the higher CM content (425 kg/m³) and the lower W/CM ratio (0.375) shows the best carbonation resistance. The carbonation depth after 50 years was calculated using square-root-t-law (2) and the coefficients mentioned in Table 3. If the $CO_2$ ingress exceeds the assumed 35 mm concrete cover over the steel reinforcements, additional concrete manufacturing is required to replace the structure over time. The total amount of each concrete mixture (m³) needed for a service life of 50 years is shown in Figure 1.

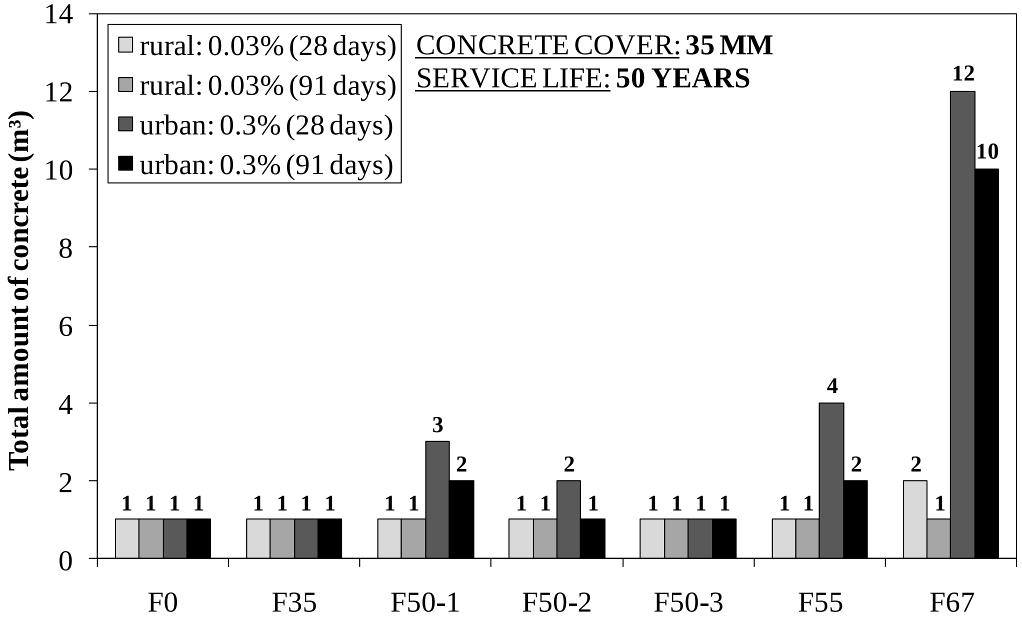

**Fig. 1. Total Amount (m³) of Each Concrete Mixture Needed for a Service Life of 50 Years after 28 and 91 Days of Curing**

A reduction in environmental impact due to the use of fly ash is obvious when the needed amount of concrete equals that of the reference mixture F0. Results again show that cement replacement levels of 50% are beneficial in both rural and urban environments, although a higher CM content (425 kg/m³) and a lower W/CM ratio (0.375) are required. A longer curing period results in an equal amount of concrete for mixture F50-2 with a CM content of 400 kg/m³ in an urban environment. The same is true for mixture F67 when used in a rural environment. In case of mixtures F50-1, F55 and F67 a longer curing period also results in a lower amount of concrete. However in urban environments, this amount is still higher than the amount needed for reference concrete, which implicates that the beneficial effect of cement replacement is canceled out.

## Freezing and thawing resistance with deicing salts

Regarding freezing/thawing resistance, experimental results are normally given in terms of total mass loss per unit area Δm (kg/m²). This amount is the sum of the scaled off material collected after 7, 14, 21 and 28 freezing and thawing cycles. In Figure 2 these values (including stdv) are presented for all seven mixtures after 28 days of curing. Values obtained for the mixtures with a CM content of 400 kg/m³ and a W/CM ratio of 0.400 (F0, F35, F50-1 and F67) clearly show that Δm significantly increases with increasing fly ash content. In comparison with F50-1, mixture F50-2 with a lower W/CM ratio is characterised by a lower salt scaling resistance. By reducing the water content of a 55% fly ash mixture (F55), an even higher Δm is measured than for the mixture with 67% fly ash (F67). With respect to mixture F50-3 a reduction in W/CM ratio (0.375) together with an increased CM content (425 kg/m³ instead of 400 kg/m³) gives rise to more salt scaling in comparison with F50-1. Although both parameters are responsible for a significant increase in compressive strength, they are far from improving the freezing/thawing resistance of the concrete. After 28 cycles Δm is much higher than the maximum permitted value of 1 kg/m².

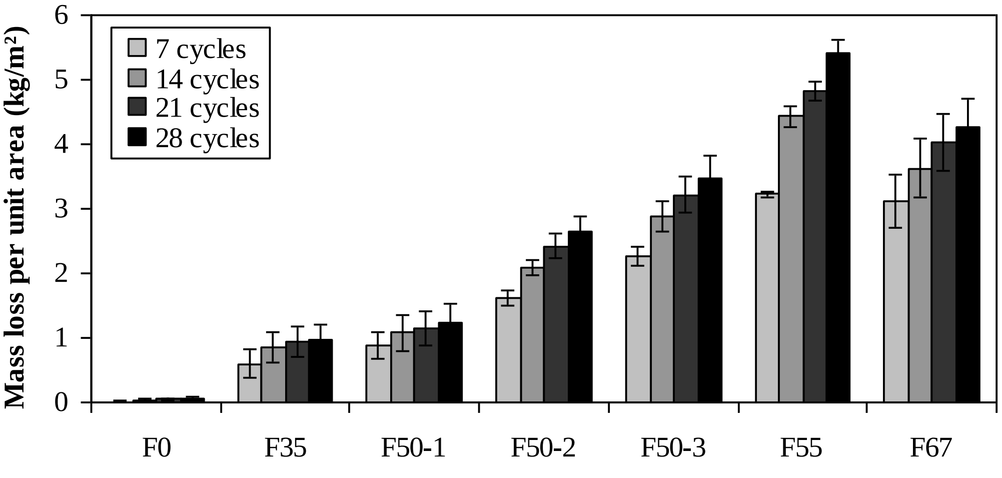

**Fig. 2. Cumulated Mass Loss per Unit Area (kg/m²) of All Concrete Mixtures after 7, 14, 21 and 28 Freezing and Thawing Cycles (n = 6)**

However, according to Table 1 the initial air content of F50-2, F50-3 and F55 is lower than the values obtained for the other fly ash containing compositions. Therefore, a second batch of these three mixtures (F50-2’, F50-3’ and F55’) was made. In order to increase the initial air content, a higher AEA dosage was applied. Adding more SP resulted in a more fluid consistency. The used amounts of each admixture together with the corresponding air content and slump are presented in Table 4. The freezing and thawing resistance (inclusive stdv) after 28 days of curing is shown within the same table. As can be seen, adjustments in admixture dosage did not result in a Δm below 1 kg/m².

**Table 4. AEA and SP Dosage, Initial Air Content, Slump and Δm after 28 Freezing and Thawing Cycles of F50-2’, F50-3’ and F55’**

| | **F50-2’** | **F50-3’** | **F55’** |
|---|---|---|---|
| **AEA (ml/kg CM)** | 2.0 | 2.0 | 2.0 |
| **SP (ml/kg CM)** | 6.2 | 8.5 | 6.8 |
| **air content (%)** | 3.3 | 3.9 | 3.1 |
| **slump** | S3 | S3 | S5 |
| **Δm (kg/m²)** | 3.72 | 3.61 | 5.02 |
| **stdv (kg/m²)** | 0.49 | 0.50 | 0.86 |

Despite the doubled AEA dosage, the initial air content of F50-2’ and F55’ is even lower than before (Table 1). Only mixture F50-3’ seems to contain a little more air than the original batch. Normally, the still unsufficient air content should not be attributed to the higher SP dosage, since polycarboxylate-type superplasticizers are known for their air-entraining effect [Hanehara and Yamada 2008]. Yet, in real plants excessive air is often not entrained in such a way, so the use of an AEA is still necessary. Also according to Hanehara & Yamada [2008], the selection of this AEA is very important because some polycarboxylate-type superplasticizers drastically reduce the freezing/thawing resistance. Whether the SP really causes the lower AEA induced air content and therefore the poor salt scaling resistance, is still under investigation. Further research is necessary before this viewpoint can be subscribed. Because of the poor freezing/thawing performance of the fly ash containing mixtures, it is obvious that the environmental advantage is non-existing in a XF4 environment and further calculations are redundant. In contrast with this conclusion, carbonation resistance of some HVFA concrete mixtures was acceptable. In this case it is useful to go through the whole LCA procedure.

### Inventory analysis

Data regarding all concrete constituents, except for the AEA [European Federation of Concrete Admixture Associations 2005] and the SP [European Federation of Concrete Admixture Associations 2006], were taken from the Ecoinvent 2.0 database [Frischknecht & Jungbluth 2007]. An overview of the data assigned to material input (kg), transport (km) and processing (kWh) is given in Table 5. Regarding the Ecoinvent data the proper short description as mentioned in the database is shown.

## Table 5. Overview of Material Input, Transport and Processing

| Material input | Material data | Amount (kg) |
|---|---|---|
| Aggregates | Gravel, round, at mine/CH S | Table 1 |
| Sand | Sand, at mine/CH S | Table 1 |
| Cement | Portland cement, strength class Z 52.5, at plant/CH S | Table 1 |
| Fly ash | - | Table 1 |
| Water | Tap water, user/CH S | Table 1 |
| AEA | European Federation of Concrete Admixture Associations (2005) | Table 1 |
| SP | European Federation of Concrete Admixture Associations (2006) | Table 1 |
| **Transport** | **Transport data** | **Distance (km)** |
| Aggregates | Transport, barge/RER S | 192.7 |
| | Transport, van <3.5t/RER S | 2.3 |
| Sand | Transport, barge/ RER S | 192.7 |
| | Transport, van <3.5t/RER S | 2.3 |
| Cement | Transport, van <3.5t/RER S | 113.0 |
| Fly ash | Transport, van <3.5t/RER S | 38.2 |
| Water | - | - |
| AEA | Transport, van <3.5t/RER S | 118.0 |
| SP | Transport, van <3.5t/RER S | 118.0 |
| **Processing** | **Processing data** | **Energy (kWh)** |
| Mixing | Electricity, low voltage, production BE, at grid/BE S | 3.83 |

With respect to material input, the weights (kg) from Table 1 need to be multiplied with the total required amount of concrete (m³) presented in Figure 1 in order to calculate a durability related environmental impact. No data regarding emissions, use of resources and energy were assigned to fly ash, since they are at the expense of the electricity companies.

### Impact analysis and interpretation

The actual reduction of greenhouse gas emissions attributed to cement replacement level in concrete was quantified using IPCC 2007 GWP 100a, an impact method developed by the Intergovernmental Panel on Climate Change with a timeframe of 100 years. Within this method, all greenhouse gas emissions are converted into kilograms $CO_2$ equivalent (kg $CO_{2eq}$). The total amount is a measure for the product’s impact on global warming and climate change. The output of this calculation is presented in Figure 3.

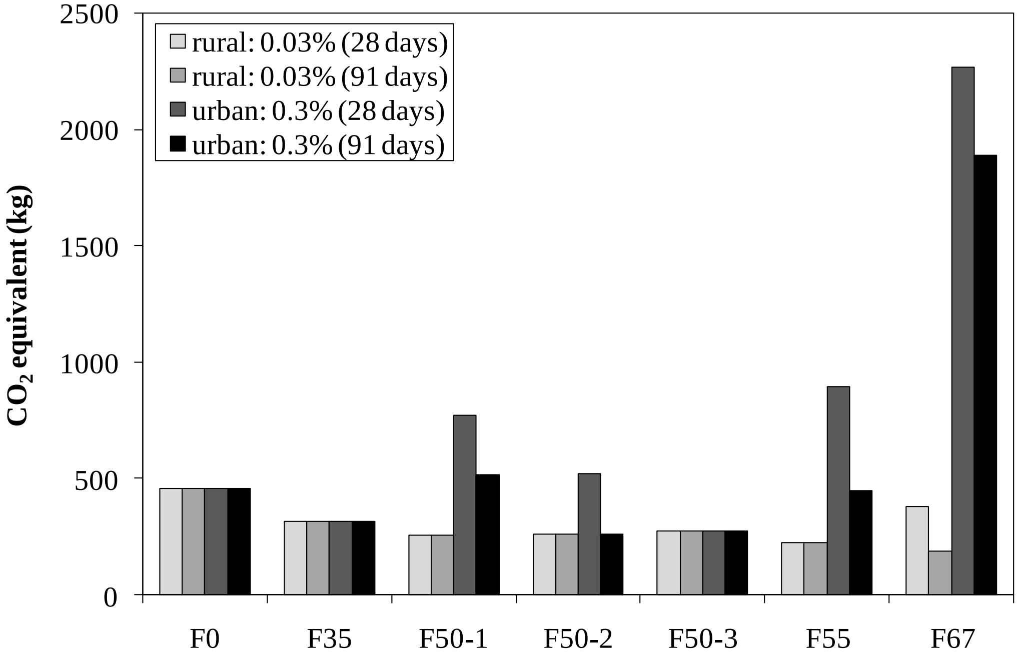

**Fig. 3. Environmental Impact of All Concrete Mixtures According to the IPCC 2007 100a Impact Method**

Only if the amount of concrete required for a 50 years life span (Figure 1) equaled the amount of reference concrete, a significant reduction in environmental impact (F35: 30.6%, F50-1: 43.8%, F50-2: 43.2%, F50-3: 40%, F55: 51% and F67: 58.6%) is detected. This reduction does not simply correspond with the fly ash content, since a fly ash concrete which is durable in an urban environment, requires a significant higher admixture dosage (F50-2, F50-3 and F55) and a higher CM content (F50-3) in case of a shorter curing age.

## CONCLUSIONS

This article shows that sufficient knowledge of HVFA concrete durability is required before the environmental impact can be calculated correctly. If not, the environmental benefit of replacing more cement with fly ash can be canceled out due to additional concrete manufacturing over time. Obviously, a structure made with a less durable HVFA concrete, will sooner need replacement than a superior OPC concrete structure. In order to deal with these differences, the total amount of concrete necessary for a 1 m³ structure with a 50 years service life is to be adopted as functional unit for LCA. Values for this unit can be obtained only from durability tests which are representative for the environment where the concrete is to be applied.

Carbonation resistance of HVFA steel reinforced concrete with a fly ash content of 50% proves to be acceptable in both a rural (0.03% $CO_2$) and urban (0.3% $CO_2$) XC1 environment, although a higher CM content (425 kg/m³) and a lower W/CM ratio (0.375) are required. With the impact on climate change expressed in terms of kilograms $CO_2$ equivalent, the amount for the HVFA concrete composition is about 40% less in comparison with the reference OPC concrete.

However, HVFA mixtures with a low water content which require a much higher SP dosage, have a poor salt scaling resistance in a XF4 environment. In comparison with the reference concrete Δm after 28 freezing/thawing cycles is at least 52 times higher, so the environmental benefit of using fly ash under these circumstances is clearly non-existing. Doubling the AEA dosage, does not result in a significant higher initial air content and lower Δm. Possibly, a higher amount of polycarboxylate-type SP in HVFA concrete partially reduces the air entraining effect of the AEA. However, further research on this matter is still imperative.

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